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Mechanistic evaluation of the reservoir engineering performance for the underground hydrogen storage in a deep North Sea aquifer

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HIGHLIGHTS

- Numerical compositional simulation utilising CMG-GEM of underground hydrogen storage in a North Sea aquifer.
- Hydrogen recovery efficiency improves as the storage cycles increase.
- Heterogeneity and hysteresis are the most impactful mechanisms on underground hydrogen storage.
- Hydrogen solubility and diffusion in water have a minimal impact on the storage process when studied separately.
- Solubility and diffusion effects become more pronounced when studied in combination with each other and hysteresis.

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ABSTRACT

Underground hydrogen storage (UHS) in aquifers, salt caverns and depleted hydrocarbon reservoirs allows for the storage of larger volumes of H₂ compared to surface storage in vessels. In this work, we investigate the impact of aquifer-related mechanisms and parameters on the performance of UHS in an associated North Sea aquifer using 3D numerical compositional simulations.

Simulation results revealed that the aquifer's permeability heterogeneity has a significant impact on the H₂ recovery efficiency where a more homogenous rock would lead to improved H₂ productivity. The inclusion of relative permeability hysteresis resulted in a drop in the H₂ injectivity and recovery due to H₂ discontinuity inside the aquifer which leads to residual H₂ during the withdrawal periods. In contrast, the effects of hydrogen solubility and hydrogen diffusion were negligible when studied each in isolation from other factors. Hence, it is essential to properly account for hysteresis and heterogeneity when evaluating UHS in aquifers.

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1. Background

Hydrogen energy is expected to play an essential role in the energy transition from fossil fuel to renewable forms of energy in the quest for reducing greenhouse gases and limiting climate change. Burning of hydrogen gas can provide energy without the production of carbon dioxide or greenhouse gases. The role of hydrogen in the energy transition can be seen as an energy store for the excess energy produced from other renewable energy sources. During low energy demand, the excess energy produced from wind and solar can be converted to hydrogen and stored for use during peak energy demand periods [1,2,3,4]. Hydrogen itself is a major driver for achieving the “Net-Zero” energy targets in addition to decarbonisation via the subsurface CO₂ storage in the depleted reservoirs combined with the enhanced oil and gas recovery schemes, CO₂ sequestration and storage in saline aquifers, and subsea, and arctic permafrost conditions in the forms of gas hydrates alone or combined with the methane-CO₂ replacement in the natural gas hydrate reservoirs [5,6,7,8,9].

Hydrogen can be produced through steam reforming of natural gas or water electrolysis using excess renewable energy [10,11,12]. The produced hydrogen gas can then be stored underground in suitable geological formations. These formations can be aquifers, salt caverns, or depleted hydrocarbon reservoirs. This process can be carried out through the injection of hydrogen gas utilising a drilled well into the specific formation [13,14,15]. Underground storage formations provide more suitable options for hydrogen storage compared to surface storage due to the considerably larger volumes of hydrogen that can be stored underground. The storage formation should be a porous and permeable reservoir rock where gas can be stored in the presence of a good sealing cap rock that would limit the escape of hydrogen upward to the surface [13,16,17].

Aquifers provide a suitable underground hydrogen storage option owing to their availability at suitable pressures housing large volumes of hydrogen gases. Several factors determine the successful operations for the aquifer storage of hydrogen that includes the injection and production rates, number of wells, and number and length of storage cycles [18,19,20]. On the other hand, the reservoir heterogeneity, relative permeability hysteresis hydrogen solubility in brine and hydrogen diffusion play an important role in defining the efficiency of underground hydrogen storage (UHS) in aquifers [18,21].

Hydrogen gas-water relative permeability hysteresis reflects the difference in the behaviour of hydrogen gas and water flow in porous media depending on whether imbibition or drainage is underway. This phenomenon is expected to lead to residual hydrogen gas being left behind due to during the withdrawal (production) phase after its injection. The change in flow behaviour causes some of the trailing hydrogen gas phase to get disconnected from the rest of the leading edge of the connected gas phase during withdrawal. Hydrogen gas solubility and diffusion may also lead to a potential loss of the amount of hydrogen gas stored inside the aquifer. CMG-GEM based numerical simulation study by Delshad et al. [18] revealed that relative permeability hysteresis improves hydrogen gas injectivity and reduced hydrogen gas recovery

from aquifers. Molecular diffusion and solubility showed a negligible effect on the results. Amid et al. [22] also reported that the hydrogen solubility in brine through geochemical and biochemical evaluations is minimal (<0.1%) during underground hydrogen storage. Moreover, sulfate reduction poses a greater challenge to underground hydrogen storage compared to methanation. However, hydrogen loss due to dissolution was limited to less than 0.1%. Feldmann et al. [23] used numerical modelling through DuMu^x to investigate hydrodynamics and gas mixing during UHS in an aquifer. Gravity override and viscous fingering phenomena were found to hinder the UHS process. The mixing of hydrogen with other gases is influenced by mobility ratios, densities, molecular diffusion and mechanical dispersion. Mechanical dispersion amplifies the gas mixing process and is more pronounced than molecular diffusion. In another work, Heinemann et al. [19] investigated the role of cushion gas in anticline saline aquifers using the commercial compositional simulator CMG-GEM at varying three geological parameters (reservoir depth, trap shape, reservoir permeability). They concluded that cushion gas does not expand the storage capacity, however, it can be used to exploit the existing capacity efficiently. The cushion gas required is dependent on the desired amount of working gas, hence, studies should be conducted early in projects for determining cushion gas requirements to avoid any unnecessary investment losses. Moreover, their simulation results showed that hydrogen storage in geological structures at greater depths with higher reservoir permeabilities requires lower amounts of cushion gas. They also concluded that tighter anticlines make the injection process more difficult without affecting the production and more open anticlines result in increasing water production. Saeed et al. [24] utilised the compositional simulation using CMG-GEM to understand the effects of different cushion gases on the performance of underground hydrogen storage in a North Sea aquifer. The hydrogen recovery efficiency was found to be related to the cushion gas density where CH₄ resulted in the highest recovery followed by N₂ and CO₂. Hydrogen gas recovery of up to 80% was observed when using CH₄ as a cushion gas within a studied period of 10 years. Moreover, the use of a cushion gas during underground hydrogen storage was preferred to storing hydrogen gas without utilising a cushion gas.

The effects of interactions between hydrogen, rock, brine, and cushion gases on the underground hydrogen storage process have been investigated in several studies. Aslannezhad et al. (2023) [25] highlighted the importance of understanding the wettability of geological formations for safe and efficient hydrogen storage. They emphasized the need for a comprehensive review on hydrogen wettability, considering parameters such as salinity, temperature, pressure, surface roughness, formation type, and the influence of organic material. Meanwhile, Ali et al. [26] examined the effects of organic acid concentrations and types on hydrogen wettability using mica surfaces. They found that increasing organic acid concentration led to higher hydrophobicity, potentially impacting hydrogen containment security. Hosseini et al. [27] analysed the hydrogen wettability of shales and evaporites under different conditions, including organic acid concentrations. They discovered that pressure, organic acid concentration, and total organic content influenced wettability, sealing efficiency,

and storage capacity. Yakeen et al. [28] focused on clay-gas interfacial tension and highlighted the scarcity of data on clay-hydrogen, clay-N₂, and clay-CO₂ systems. Their calculations revealed that hydrogen exhibited higher interfacial tension with clay minerals compared to N₂ and CO₂, suggesting the suitability of nitrogen and carbon dioxide as cushion gases. Alanazi et al. [29] investigated hydrogen storage efficiency and cushion gas selectivity in organic-rich carbonate-rich Jordanian source rock samples. They found that CO₂ demonstrated preferential behaviour as a cushion gas, indicating the importance of considering the presence of organic residuals in hydrogen storage. Saeed et al. (2023) [30] employed geochemical modelling to assess the impact of geochemical reactions on underground hydrogen storage performance. They validated the model against experimental data and observed minimal hydrogen loss over 30 years, while CO₂ losses were more significant. They highlighted the need for remedial CO₂ injections to maintain reservoir pressure and discussed the effects of temperature, pressure, and CO₂ on rock properties and gas storage capacity. Yekta et al. [31] have emphasized the potential for geochemical interactions involving stored hydrogen gas, cushion gas, reservoir fluids, and reservoir minerals during UHS. These interactions can lead to hydrogen conversion into gases like methane and hydride sulphide, resulting in potential losses in stored hydrogen. Additionally, interactions with other gases in the reservoir can reduce the purity of the hydrogen gas [32,33]. Understanding the impact of these geochemical processes on hydrogen storage is crucial for successful UHS implementation.

In this work, we focus on investigating the effects of aquifer-related factors and mechanisms on the performance of underground hydrogen storage in an associated North Sea aquifer. The investigated parameters in this study are aquifer heterogeneity, relative permeability hysteresis, hydrogen

solubility, and hydrogen diffusion. Further details on the methodology utilised to carry out this study are outlined next.

2. Methodology

In this study, we employed numerical simulation to model the process of storing hydrogen in a deep North Sea aquifer using the multicomponent compositional simulator CMG-GEM™ [34] and investigate the impact of aquifer rock parameters and fluid-fluid mechanisms on the overall performance of underground hydrogen storage in the aquifer.

2.1. Aquifer model description

The model used in this study (Fig. 1) is of an aquifer in the North Sea that is associated with a depleted oil reservoir and is located to the South of the reservoir and is at a depth of 10,000 feet. The depressurization of the oil field and its associated aquifer occurred due to the field's oil production, therefore, the pressure support resulting from the aquifer was gradually lost since 1997 when oil production increased significantly to 200,000 STB of oil. In this study, underground hydrogen storage in three sandstone formations in the aquifer will be evaluated. The depleted oil reservoir currently has a present pressure of about 2030 psi and a temperature of 110 °C. These same conditions are assumed for the aquifer. In this study, underground hydrogen storage in three sandstone formations in the aquifer will be evaluated. The original salinity of the aquifer water is 24,000 parts per million (ppm), which is quite low. A summary of aquifer properties is detailed in Table 1. The fluid compositional model consists of water and hydrogen gas and used the Peng-Robinson Equation of State to calculate the fluid properties. Henry's Law was utilised to include the

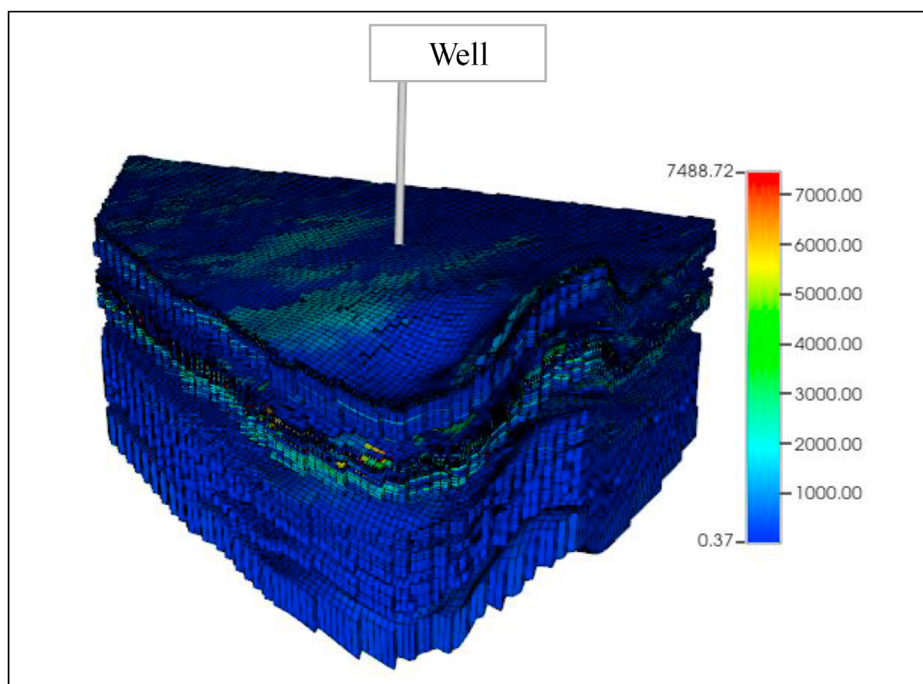


Fig. 1 – A 3D compositional aquifer model showing distribution of permeability and position of operating well.

Table 1 – Summary of aquifer properties.

Property	Value
Depth (ft)	10,000
Temperature (°C)	110
Initial Pressure (psi)	6000
Current Pressure (psi)	2000
Salinity (ppm)	24,000
Water initially in place (Billion ft ³)	2.1
Average porosity (%)	19
Aquifer gross thickness (depth 10,000 ft to 10,500 ft)	500

effects of hydrogen gas solubility in the water, assuming that the hydrogen gas and the aqueous phase are in thermodynamic equilibrium. The hydrogen diffusion coefficient in water was assumed to be 8.5×10^{-5} cm²/s. The reservoir's dependence of rock compressibility on pressure and rock compressibility are 6.12×10^{-12} psi⁻² and 3.5×10^{-6} psi⁻¹, respectively. The H₂-water relative permeability curves (see Fig. 2) were adapted from the experimental investigations of Yekta et al. [35]; accommodating the hysteresis effect. Based on relative permeability curves the irreducible water saturation during drainage is 0.15 and the trapped gas saturation

between the drainage and imbibition processes is 0.18. In this work, the effects of geochemical reactions were not included in the simulations.

2.2. Case studies

Six cases were run to investigate the effects of aquifer's permeability heterogeneity, H₂-water relative permeability and capillary pressure hysteresis, hydrogen solubility and diffusion on the injection and production of hydrogen gas during UHS in the aquifer. In the base case (Case Aq - 1), a single well is used for both hydrogen injection and production during seven cycles. The first cycle consists of injecting hydrogen gas at a rate of 35 MMscf/d for seven months and producing hydrogen gas at a rate of 20 MMscf/d for seven months. The cycles from the second to the seventh are then identical to each other and they involve hydrogen gas injection at 35 MMscf/d for 5 months and producing it back for 7 months at 20 MMscf/d. The seventh cycle is then followed by 3 years extended period of production to exhaust the aquifer of its pressure. Both the hydrogen gas injection and production are carried out through the top perforations of each sandstone formation. The injection is limited by the maximum injection

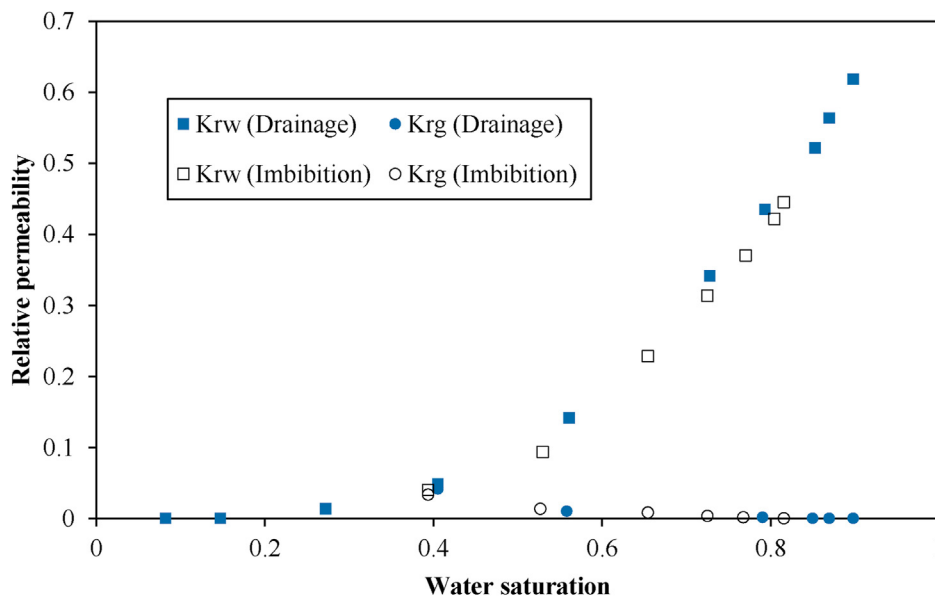


Fig. 2 – (a) H₂-water relative permeability (b) capillary pressure curves for drainage and imbibition deployed in the model [35].

Table 2 – Summary of simulated modelling cases and respective studied effects.

Case	Parameters/mechanisms investigated
Case Aq - 1	Base Case: The original heterogeneous model without solubility or hysteresis or diffusion effects.
Case Aq - 2	Homogeneous model (permeability modified from Case Aq-1)
Case Aq - 3	Relative permeability hysteresis
Case Aq - 4	Solubility of H ₂ in water
Case Aq - 5	Diffusion of H ₂ in water
Case Aq - 6	Hysteresis, solubility, and diffusion combined effect
Case Aq - 7	Effect of number of storage cycles (14 cycles case) in the combined hysteresis, solubility, and diffusion effect

rate and a maximum bottomhole pressure of 4500 psi to avoid reaching the formation's fracture pressure of 5100 psi. Similarly, the production was restricted by the maximum production rate and a minimum bottomhole pressure of 1900 psi to avoid problems with lifting aquifer fluids to the surface.

The effects of aquifer heterogeneity, hysteresis, hydrogen gas solubility and diffusivity on the performance of the underground hydrogen storage in aquifers were studied both in

isolation of each other and combined. Case Aq – 1, is the base case, an original heterogeneous aquifer, vertical to horizontal permeability ratio is approximately 40% and does not take into account the effects of hysteresis, solubility and diffusion. Case Aq – 2 represents a 500 mD homogeneous aquifer model while keeping all other properties unchanged. This case is then compared to the base case (Case Aq - 1) to evaluate the effect of aquifer heterogeneity on the UHS performance. Relative

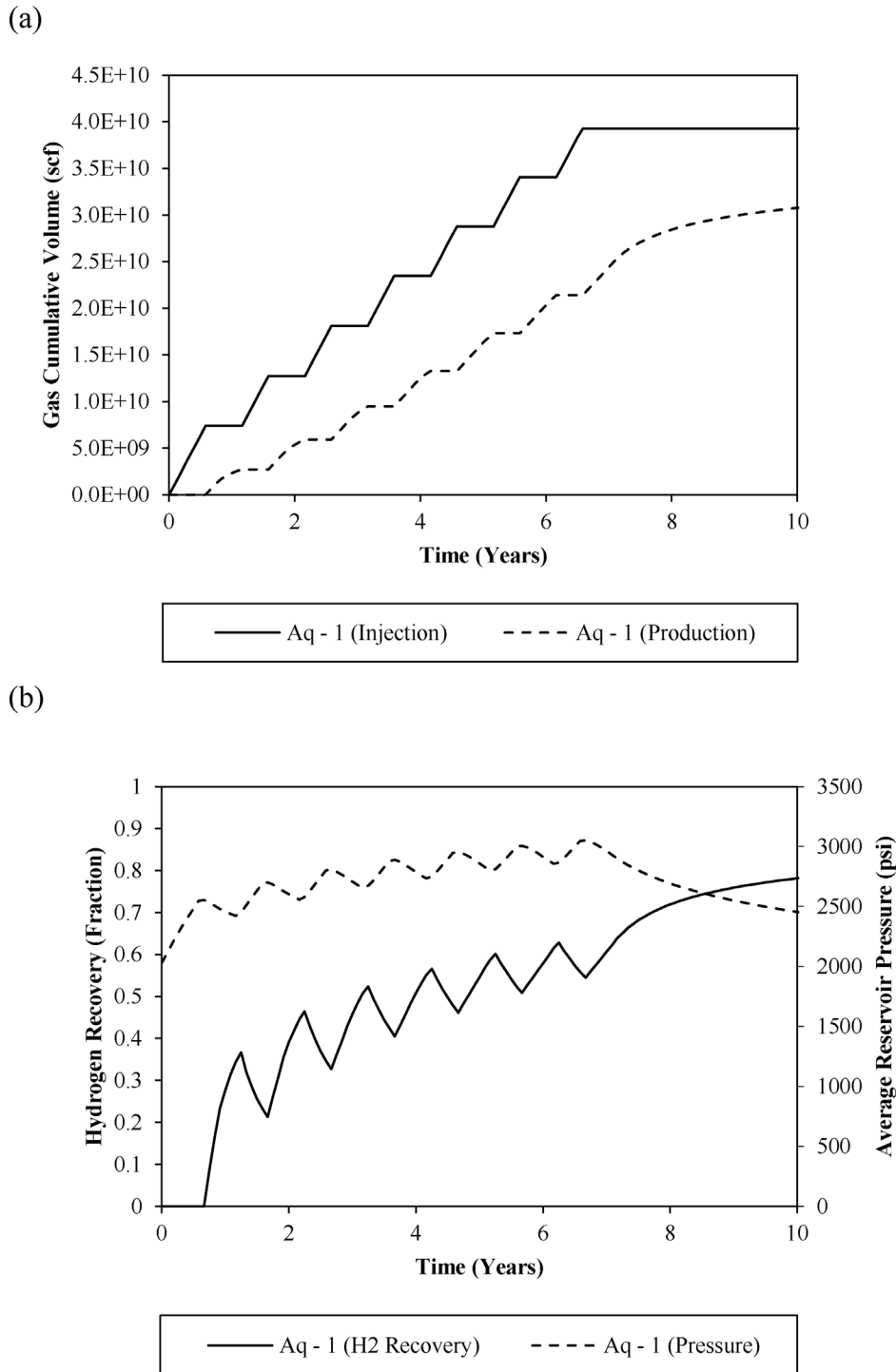


Fig. 3 – (a) Cumulative H₂ injected and produced, and (b) H₂ recovery and average aquifer pressure predicted for base case of UHS in aquifer.

permeability and capillary pressure hysteresis were invoked using both the imbibition and drainage curves as shown in the hydrogen gas injection and recovery, Case Aq – 3 was run while considering Fig. 2. The isolated effects of hydrogen gas solubility and diffusivity on the underground hydrogen storage process were evaluated in Cases Aq – 4 and Aq – 5, respectively. Another scenario, Case Aq – 6, was run to investigate the collective effects of hysteresis, solubility, and diffusivity on underground hydrogen storage in the studied aquifer. Finally, Case Aq – 7 was run using a similar configuration to Case Aq – 6 with an increased number of cycles, 14 cycles in Case Aq – 7 and 7 cycles in Case Aq – 6. Details of the simulation cases indicating the effects evaluated in this study are given in Table 2.

3. Results and discussion

3.1. Base case

The base case simulation (Case Aq-1) in this study involves seven cycles of injecting hydrogen at a rate of 35 MMscf/d and producing it at a rate of 20 MMscf/d from a single well. Over a period of ten years, 39.5 Bscf of hydrogen gas was injected during the injection cycle, followed by 30.7 Bscf of hydrogen recovery with a 78% recovery factor while

retaining 22% of hydrogen (see Fig. 3). The hydrogen recovery does, however, continue to improve with the increasing number of storage cycles from 36.6% in the first cycle to 62.8% at the end of 6th cycle and then 78.1% in the final cycle., suggesting that the hydrogen recovery would be higher if the storage cycles were increased (presented later in Case Aq – 7). The injected hydrogen gas occupies a larger space in the aquifer and spreads outward away from the injection well as the cycles progress. This can be observed from the 2D map presented in Fig. 4 where the hydrogen gas saturation is shown at the end of the production stages of the 1st, 4th, and 7th cycles and after the extended production period. The average aquifer pressure (see Fig. 3b) is initially at 2030 psi and it increases with the initial injection of the hydrogen in the first cycle to 2534 psi followed by 82 psi pressure drop to 2452 psi due to the hydrogen production in the first cycle. The pressure inside the aquifer continues to increase and drop during injection and production stages, respectively, with an overall rising trend as a result of the residual hydrogen gas volume accumulating as the cycles continue. Eventually, the built-up pressure inside the aquifer reached a maximum of 3038 psi by the end of the seventh cycle's injection stage. This pressure is then utilised to produce hydrogen gas for a continuous three years depleting the aquifer's pressure down to 2500 psi by the end of the extended production period.

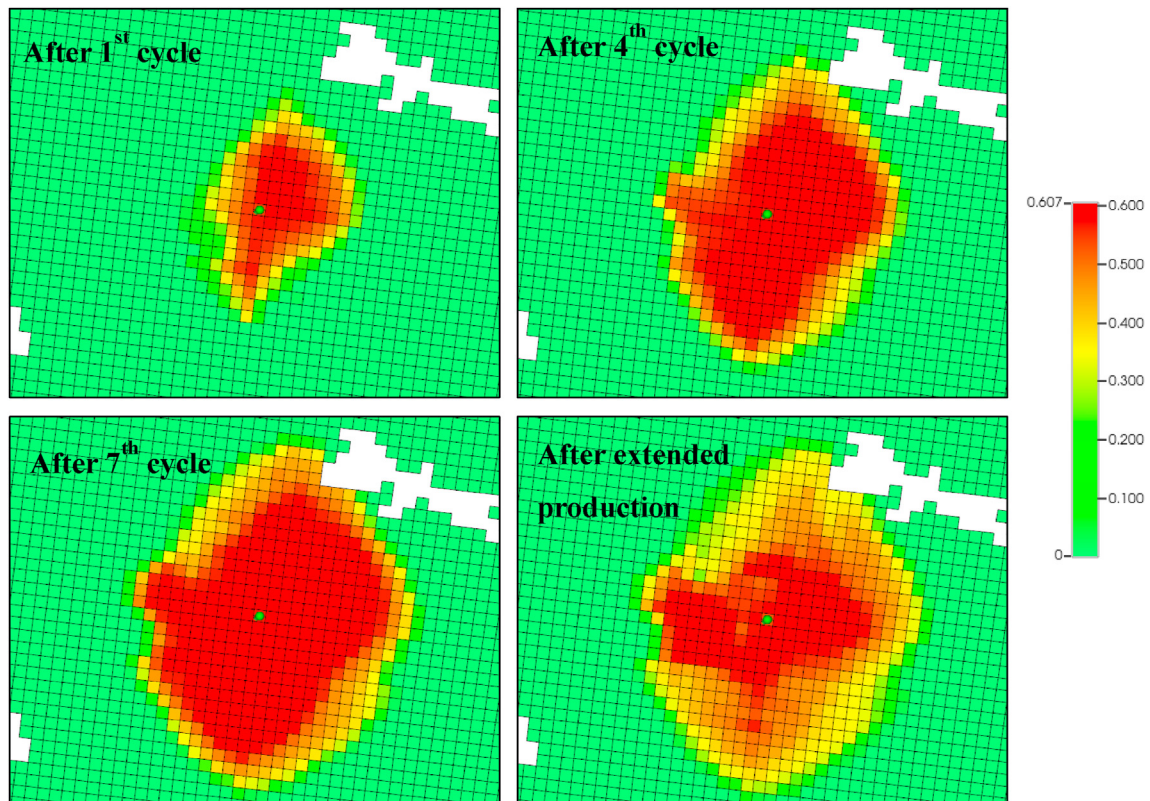


Fig. 4 – Hydrogen gas saturation distribution in the bottom formation at the end of the production stage of the 1st, 4th, and 7th cycles and after the extended production period. The green dot at the centre indicates the injection and production (withdrawal) cycles respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

3.2. Reservoir heterogeneity effect

The effect of aquifer's heterogeneity on the performance of UHS in aquifers is evaluated by comparing the original heterogeneous reservoir model (base case Case Aq – 1) with Case Aq – 2 wherein the aquifer's vertical and horizontal permeability was set to a fixed value of 500 mD. In both cases, the maximum injection and production rates remained similar (35 and 20 MMscf/d, respectively). Injected rates shown in Fig. 5a decline earlier (than the complete cycle duration) in cycles 5, 6 and 7 than the previous cycles, whereas the production rates start to decline earlier right from the first cycle

which then delays in the consecutive cycles reaching the full cycle length by cycle 7. This is attributed to the 500 mD uniform permeability in Case Aq – 2.

As shown in Fig. 5a, the total volume of hydrogen injected in the homogenous case (Case Aq – 2), was 39.53 Bscf which was slightly higher than that of the heterogenous case (Case Aq – 1), 39.5 Bscf. The amount of recoverable hydrogen is also higher in the homogenous Case Aq – 2, 33.53 Bscf, compared to the heterogenous Case Aq – 1, 30.38 Bscf. Moreover, average aquifer pressure remained above the initial aquifer pressure of 2000 psi and below 3200 psi throughout the seven storage cycles as depicted in Fig. 5c in both cases.

Permeability in heterogeneity affects the fluid flow through the storage formation, thus impacting the hydrogen injection (injectivity), its movement through the pore flow paths (migration) and the subsequent withdrawal during the production. The higher cumulative H₂ injection and production volumes can be attributed to the better injectivity, and productivity experienced in the homogenous Case Aq – 2, owing to the uniform vertical and horizontal permeability of 500 mD. This has resulted in the bottomhole pressure (BHP) rising or dropping more gradually compared to the heterogenous Case Aq – 1 (see Fig. 5) and consequently avoids violating the injection and production BHP constraints. This allows for higher volumes of hydrogen gas injection and consequent production. As a result, it can also be observed from Fig. 5c that the hydrogen recovered as a percentage of total hydrogen injection in the homogenous case (85.34%) is higher than in the heterogenous case (78.3%). This is a result Therefore, the UHS in a homogeneous aquifer is more favoured in terms of storage capacity and stored hydrogen recoverability to heterogeneous aquifers with an incremental hydrogen recovery of up to 7% expected in the homogenous aquifer. It is also observed from Fig. 5c that the average aquifer pressure in Case Aq – 2 trends lower compared to the average pressure in Case Aq – 1 due to the lower residual gas volume in Case Aq – 2 (6 Bscf) in comparison to Case Aq – 1 (9.12 Bscf).

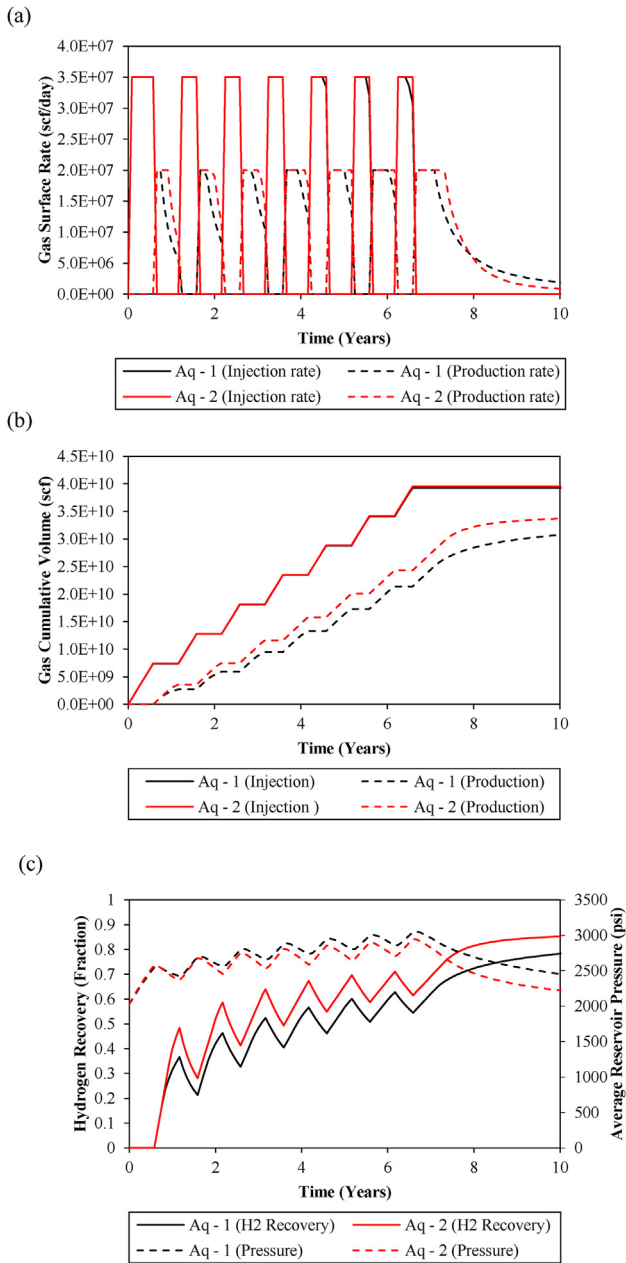


Fig. 5 – Effect of aquifer heterogeneity on UHS in aquifer on (a) H₂ injection and production rates, (b) cumulative H₂ injected and produced, and (c) H₂ recovery and average aquifer pressure.

3.3. Effect of the hydrogen-water relative permeability hysteresis

Relative permeability hysteresis affects the fluid flow in porous media during both hydrogen injection and withdrawal periods. This would consequently affect the overall injectivity and productivity of hydrogen in the aquifer. In this study, we examine the effect of relative permeability on the UHS process using Case Aq-3 and then compare it with the no-hysteresis first scenario (Case Aq – 1). Fig. 6a shows the effect of relative permeability hysteresis on the hydrogen gas production rates. The production rate in the earlier cycles is not affected by the hysteresis effect. However, at later stages and especially during the extended period of production after the seventh cycle, the production rate declines as a result of the inclusion of the hysteresis effect. A comparison of the cumulative volumes in Fig. 6b indicates that the relative permeability hysteresis declined the cumulative hydrogen injection and production volumes in Case-3 by 0.5 Bscf (39.5 Bscf to 39 Bscf) and 1.24 Bscf (30.7 Bscf to 29.46 Bscf) respectively against the no-hysteresis case (Case Aq-1). It is apparent

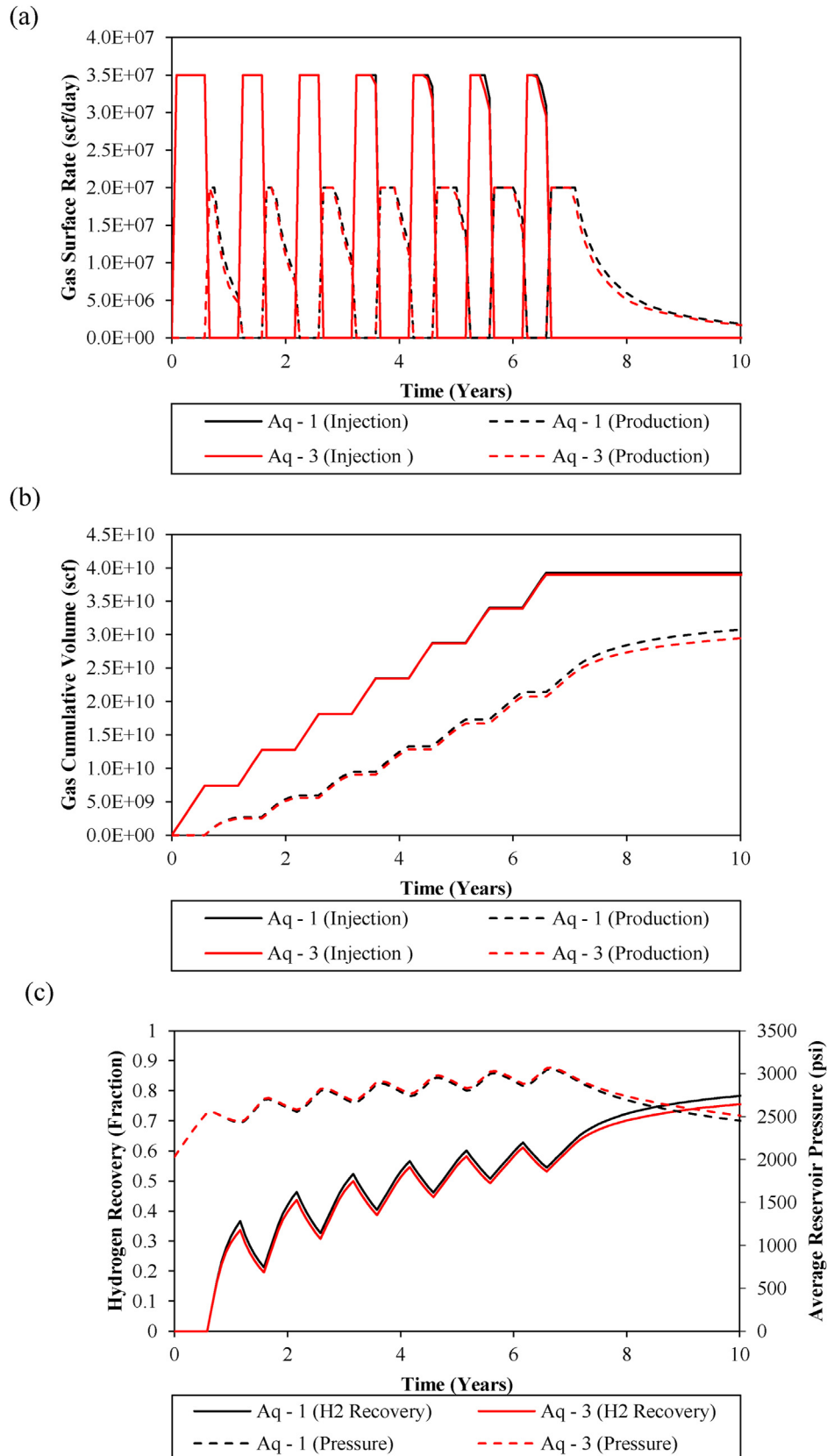


Fig. 6 – Effect of relative permeability hysteresis on UHS in aquifer on (a) H₂ injection and production rates, (b) cumulative H₂ injected and produced, and (c) H₂ recovery and average aquifer pressure.

from this comparison that the inclusion of relative permeability hysteresis has a significant impact on the underground hydrogen storage process. This is also reflected in the reduced hydrogen recovery efficiency from 78.72% to 75.91% (see Fig. 6c) This decline is attributed to the hydrogen gas entrapment that occurs due to the discontinuity of hydrogen gas during the withdrawal period. The disconnected hydrogen gas is then separated from the mobile gas at the front of the migrating plume and is left immobile inside the aquifer. This consequently reduces the amount of recoverable hydrogen gas from the storage aquifer formation. The respective pressure profile continually keeps rising in each successive cycle, with minimal but noticeable higher values than the no hysteresis case. Hysteresis trapping makes more and more hydrogen molecules left behind in the pores leading to higher pressure compared to the no-hysteresis case.

This can further be observed by examining Fig. 7a and b where the gas saturation distribution inside the aquifer at the end of the studied period for Cases Aq – 1 and Aq – 3 are shown. The inclusion of relative permeability hysteresis resulted in higher trapped volumes of residual hydrogen gas as shown in Fig. 7b. A comparison (see Fig. 7c and d) of the change of hydrogen gas saturation during the storage cycles in two localised (shown in Fig. 7a and b) reveals that the hydrogen saturation continued to rise in each successive cycle

in those cells since the advent of the gas front, whereas the extended production period clearly shown the higher gas saturation due to the continuous hydrogen production. These results highlight the importance of considering relative permeability hysteresis when simulating and designing an underground hydrogen storage process in aquifers.

3.4. H_2 solubility effect

The hydrogen gas is expected to dissolve in aquifer water depending on the pressure and temperature conditions and salinity of the formation water. Hence, this affects the inventory of hydrogen stored inside the aquifer. This was studied by comparing the results of Case Aq – 1 where hydrogen solubility in water was ignored to the results of Case Aq – 4 where solubility was modelled. Fig. 8a exhibits the injection and production rates for the two cases where the maximum injection and production rates were maintained similarly which were 35 and 20 MMscf/d, respectively. The cumulative produced and injected volumes of hydrogen gas for Cases Aq – 1 and Aq – 4 are depicted in Fig. 8b. Furthermore, the hydrogen recovery efficiency and average aquifer pressure results are given in Fig. 8c. By examining the comparative modelling results, we can observe that both cases gave almost identical results which highlight the

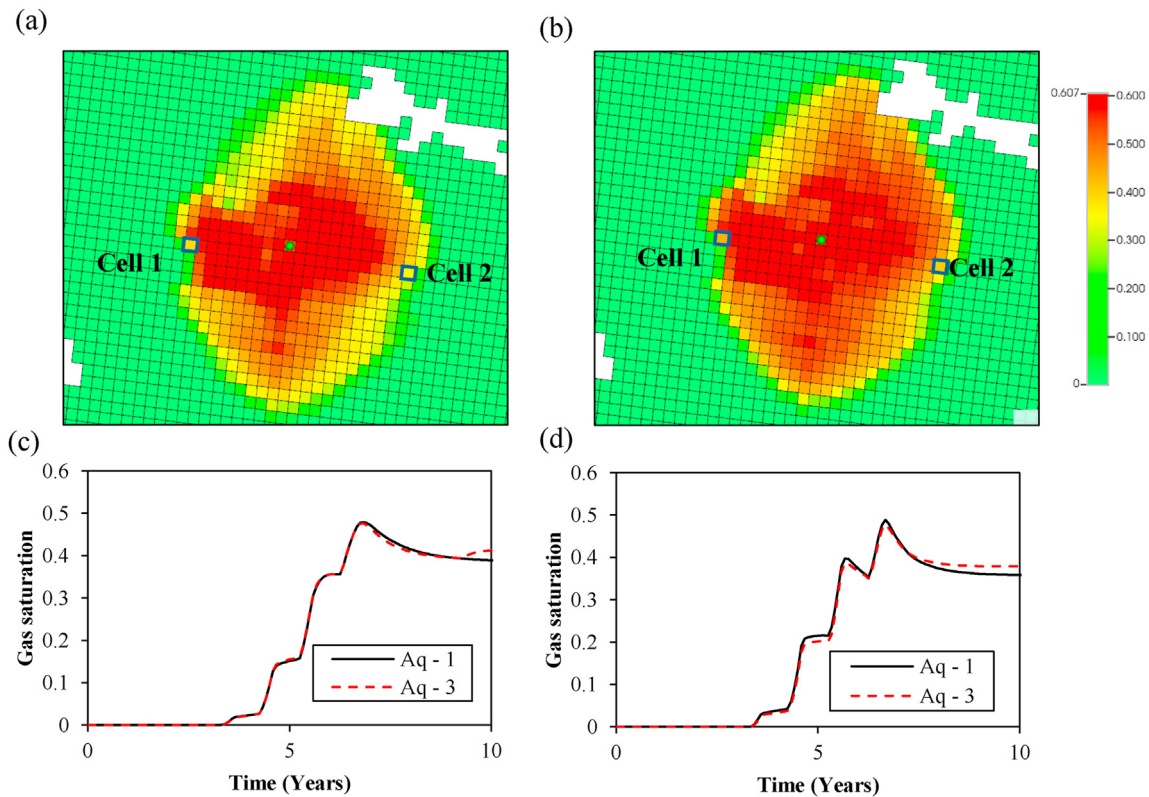


Fig. 7 – The hydrogen gas saturation at the end of the extended production period in (a) the base case (Case Aq – 1) and (b) the relative permeability hysteresis case (Case Aq – 3). Hydrogen gas saturation change as a function of time for Cases Aq – 1 and Aq – 3 captured in (c) Cell 1 and (d) Cell 2. The green dot in (a) and (b) at the centre indicates the injection and production (withdrawal) cycles respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

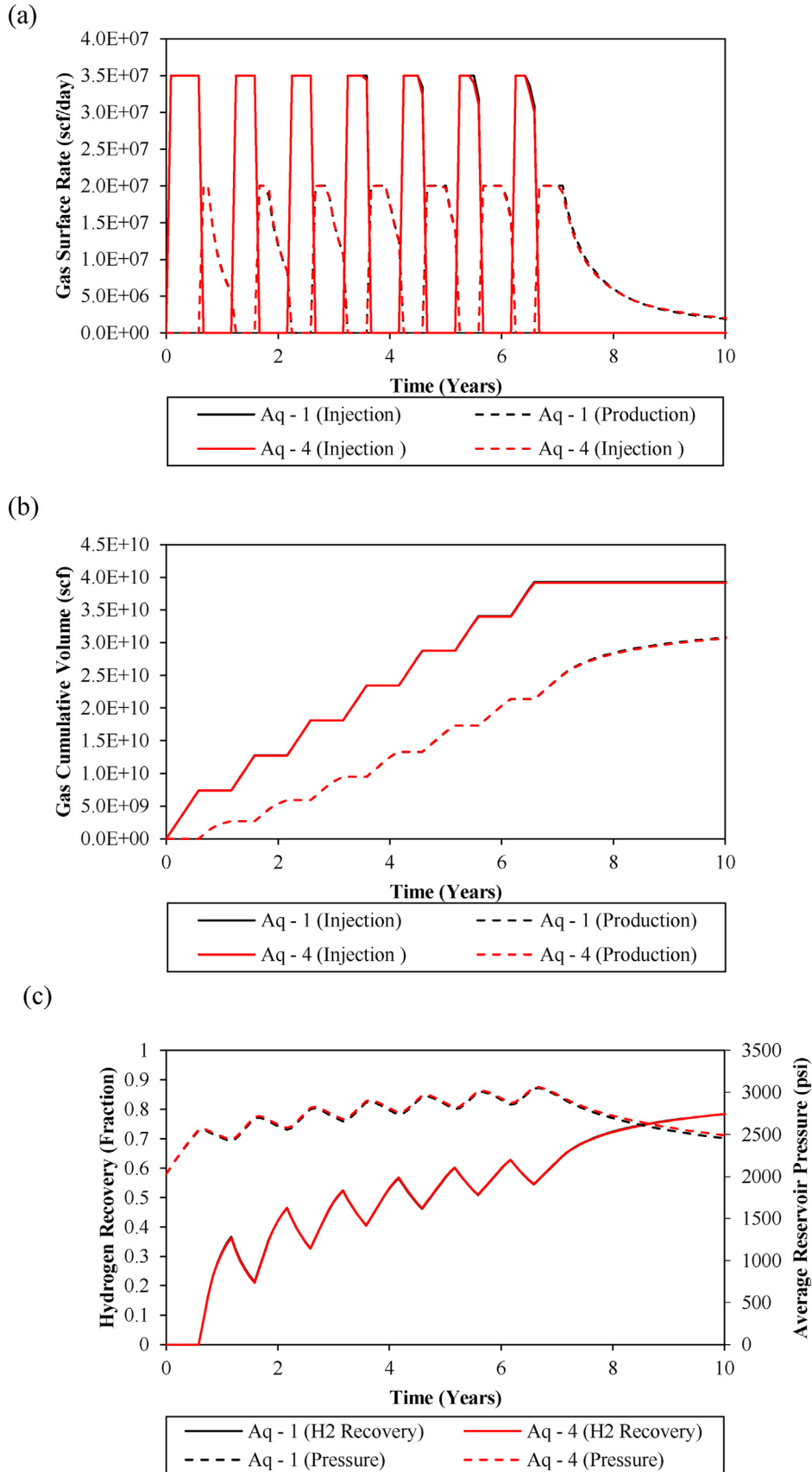


Fig. 8 – Effect of hydrogen solubility on UHS in aquifer on (a) H₂ injection and production rates, (b) cumulative H₂ injected and produced, and (c) H₂ recovery and average aquifer pressure.

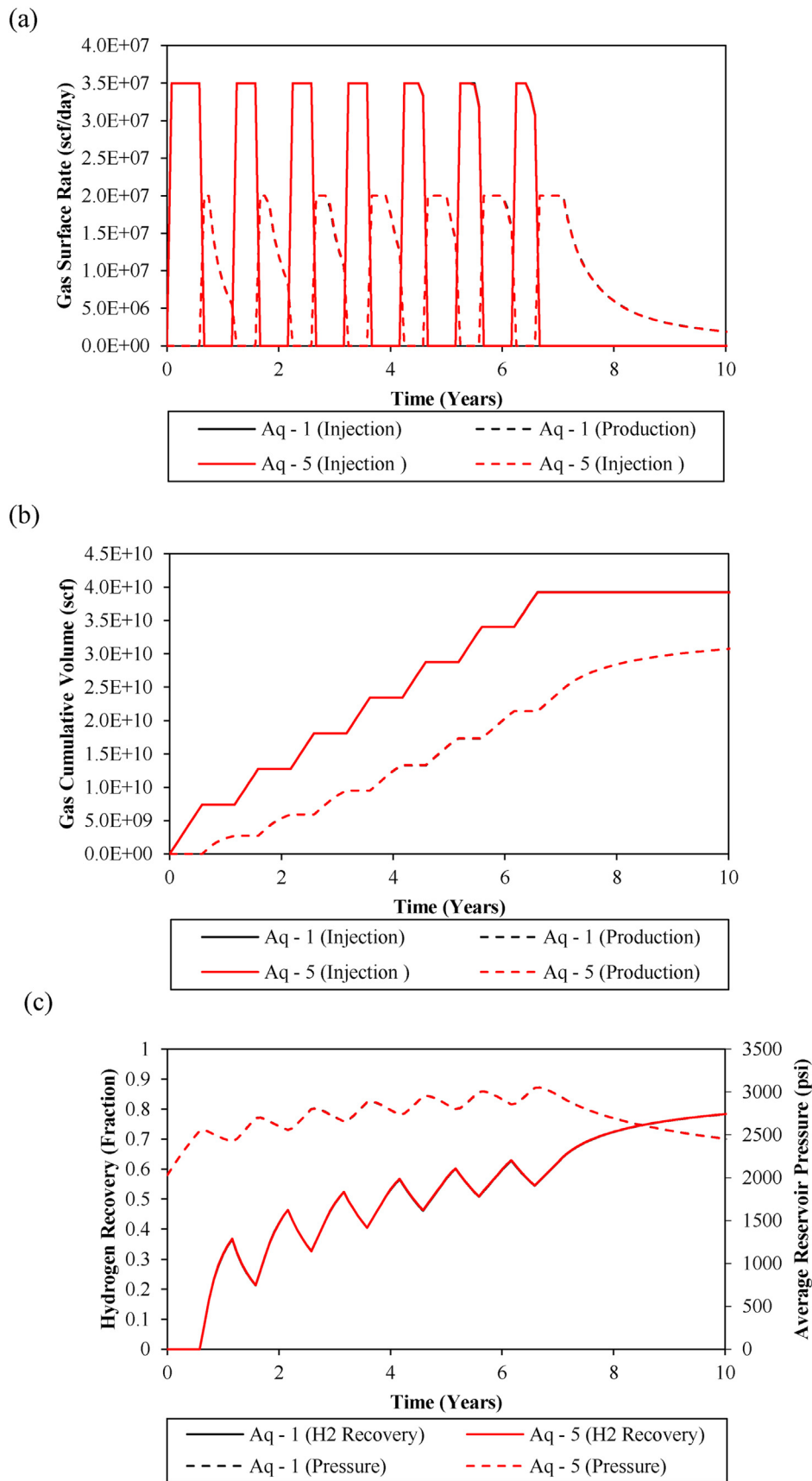


Fig. 9 – Effect of hydrogen diffusion on UHS in aquifer on (a) H₂ injection and production rates, (b) cumulative H₂ injected and produced, and (c) H₂ recovery and average aquifer pressure.

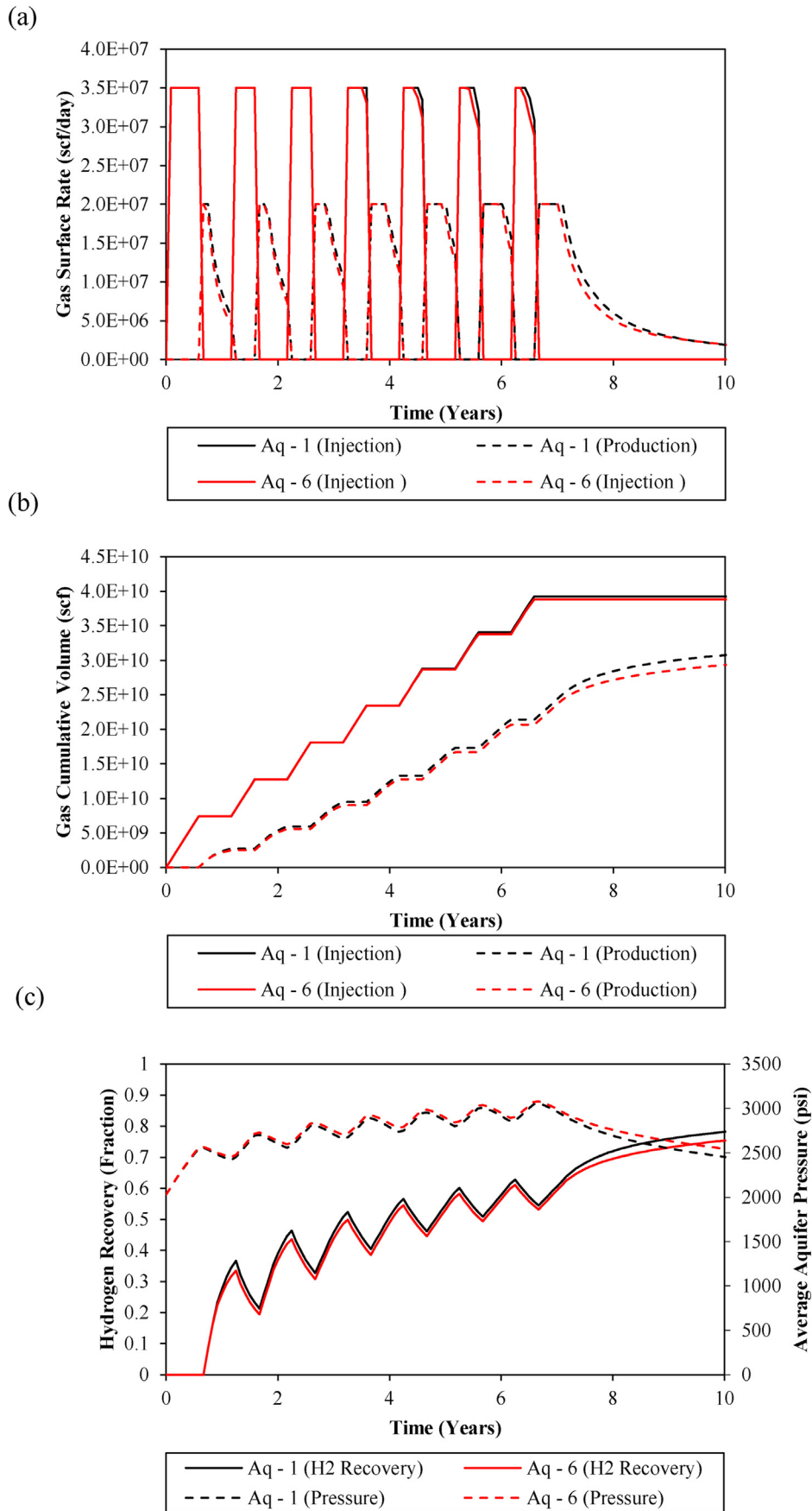


Fig. 10 – Combined effect of relative permeability hysteresis, hydrogen solubility and diffusion on UHS in aquifer on (a) H_2 injection and production rates, (b) cumulative H_2 injected and produced, and (c) H_2 recovery and average aquifer pressure.

negligible effect of hydrogen gas solubility in aquifer water. This observation is in line with previous studies where they also concluded that hydrogen gas solubility had a minimal impact on the UHS performance [22,18].

3.5. H₂ diffusion effect

Hydrogen diffusion in water is another factor that is expected to affect the performance of UHS in aquifers. It is predicted that it will increase the amount of hydrogen gas trapped in aquifer water. To understand the effect of diffusion on the underground hydrogen storage process, we compare the results of the base case, Case Aq – 1, with the results of Case Aq – 5, which considers the effect of diffusion. The H₂ injection and production rates and cycles for the two cases are shown in Fig. 9a. A comparison between the cumulative hydrogen gas volumes produced and injected (see Fig. 9b), shows that diffusion has a minimal effect on the overall performance of the underground hydrogen storage. This is also reflected in the overall hydrogen recovery efficiency and average aquifer pressure as shown in Fig. 9c where the differences are negligible. These results align with the findings from Ref. [22] where they found that losses of hydrogen due to dissolution and diffusion amount to less than 0.1% of the hydrogen gas inventory.

3.6. Hysteresis, solubility, and diffusion combined effect

To understand the overall impact of hysteresis, solubility, and diffusion on the UHS in the aquifer, a scenario (Case Aq – 6) was run while considering these mechanisms together. The results from Aq – 6 were compared with the results of base case Aq – 1. A comparison of the cumulative results over the 7 cycles shown in Fig. 10a, reveals that the overall injected hydrogen gas volume decreased from 39.5 Bscf to 38.85 Bscf as

a result of the combined effects of hysteresis, solubility and diffusion. It is also observed that the cumulative hydrogen gas produced decreased from 30.8 to 29.34 Bscf. This drop in the produced hydrogen gas resulted in a decrease in the hydrogen recovery efficiency from 78.7% to 75.8%. A comparison of the average aquifer pressures in Case Aq – 1 and Aq – 6 as exhibited in Fig. 10c shows that the pressure in the two cases remains similar throughout the first seven cycles with the pressure in Case Aq – 6 slightly higher than Case Aq – 1. This is due to the higher residual hydrogen gas volume inside the reservoir as a result of the combined effects of hysteresis, solubility, and diffusion. During the extended production period, the difference in the pressure in the two cases increases to 100 psi whereby at the end of the production period the average pressure in Case Aq – 6 is 2554 psi and in Case Aq – 1 2454 psi. This increase in the pressure difference between the two cases is the result of the higher residual gas saturation due to the hysteresis effect that becomes more pronounced during the extended production period as observed previously in Fig. 6.

To evaluate the contribution of each mechanism on the overall UHS performance, we have a look at Fig. 11 which displays the isolated impact of each mechanism on the cumulative hydrogen gas injected and produced. The most impactful aquifer rock parameter or mechanism is the reservoir heterogeneity, such that an aquifer with a homogenous permeability result in a 0.5% increase in cumulative volume injected compared to the heterogenous case (Case Aq – 1). It also leads to a 9.26% higher volume of hydrogen gas production over the same period of time. This signifies the importance of selecting a storage site with lower permeability heterogeneity to be able to enhance the hydrogen gas storage capacity and recovery efficiency. The inclusion of relative permeability and capillary pressure hysteresis effects in Case Aq – 3, resulted in a 0.76% reduction in the total hydrogen gas

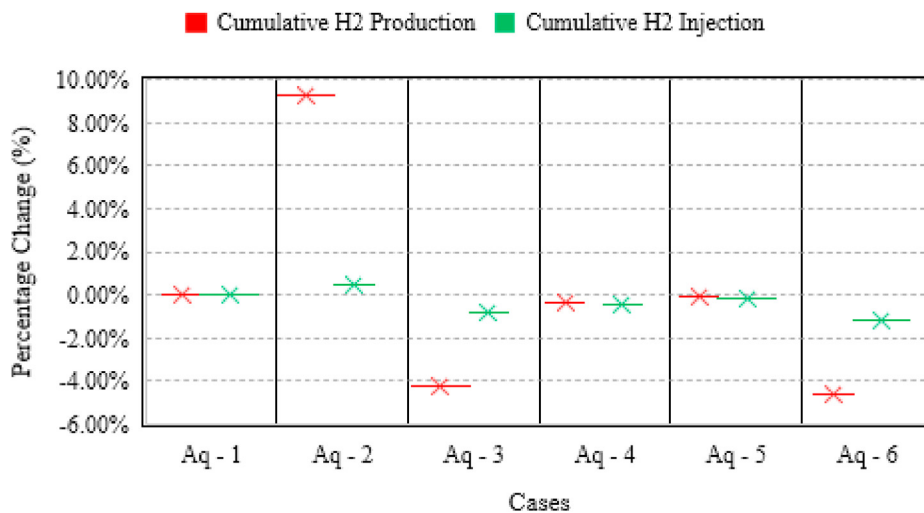


Fig. 11 – Effect of aquifer homogeneity (Aq – 2), relative permeability hysteresis (Aq – 3), hydrogen solubility (Aq – 4), hydrogen diffusion (Aq – 5) and combined effect of hysteresis, solubility and diffusion (Aq – 6) on cumulative hydrogen production and injection.

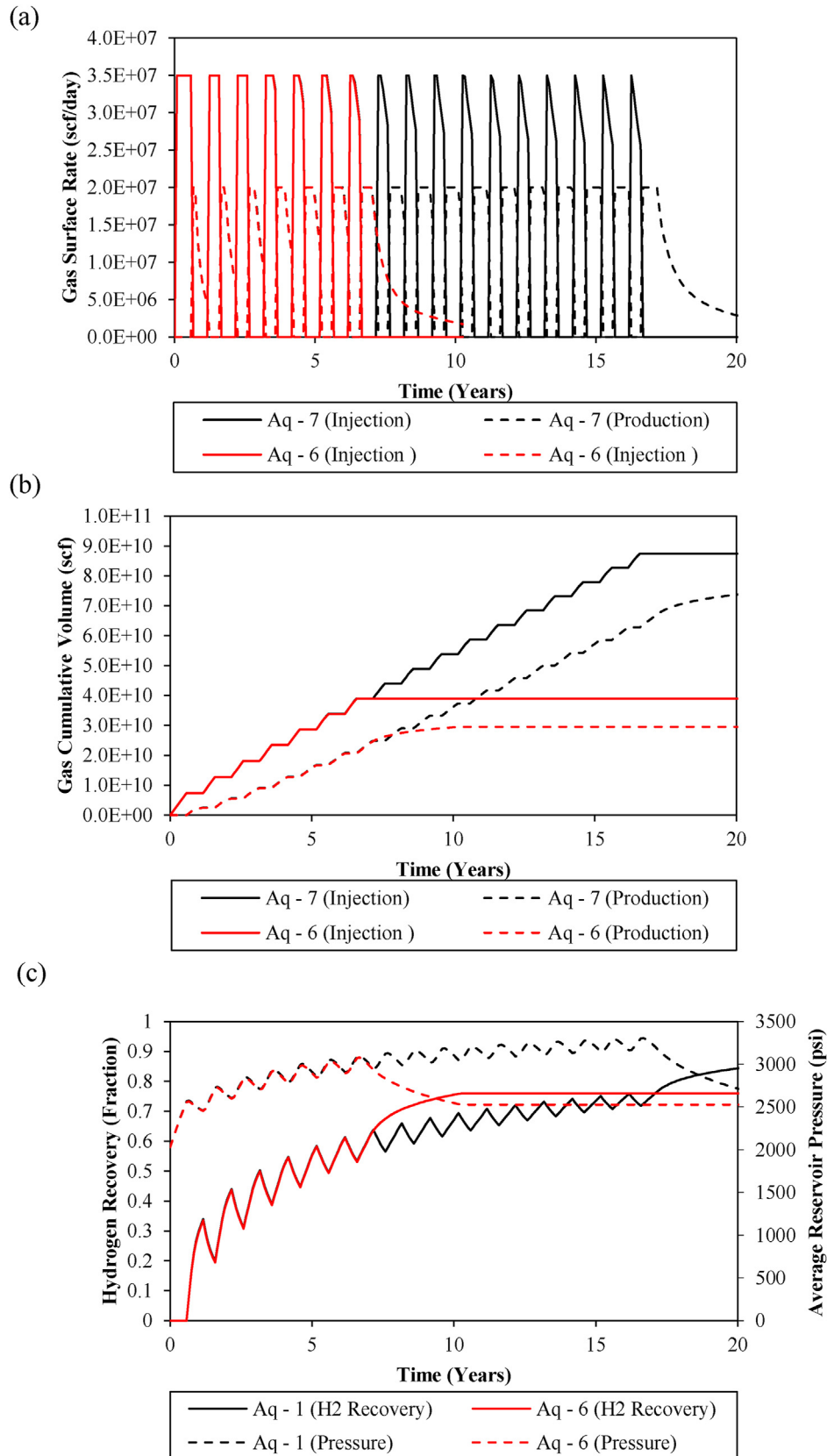


Fig. 12 – Comparison between Case Aq – 6 where seven cycles were used and Case Aq – 7 where 14 cycles were simulated in terms of (a) H₂ injection and production rates, (b) cumulative H₂ injected and produced, and (c) H₂ recovery and average aquifer pressure.

volume injection and a 4.26% decrease in the hydrogen gas volume produced compared to the base case. Therefore, it is crucial to include the hysteresis effect in any modelling study during UHS process design to avoid overestimating the aquifer's H_2 storage capacity and recovery efficiency. Both hydrogen solubility (Case Aq - 4) and diffusion (Case Aq - 5) amount to very negligible per cent changes in the overall injected and produced volumes compared to the base case. However, the effects of solubility and diffusion became more apparent when combined together with the hysteresis effect in Case Aq - 6. Where the combined effect of the three mechanisms showed a 1.15% decrease in the total hydrogen gas injection and a 4.58% drop in the cumulative hydrogen gas produced compared to the base case. And if we compare this impact with the isolated impact of hysteresis studied in Case Aq - 3, we can conclude that solubility and diffusion do have an impact on the overall injected and produced volumes of hydrogen gas. Hence, studying these effects in isolation from each other may lead to the slightly misleading conclusion of the minimal impact of each mechanism, especially, solubility and diffusion.

3.7. Increasing the number of storage cycles

As observed from the previous results, the hydrogen recovery efficiency increases with the number of storage cycles. Therefore, in this section, we investigate the effect of doubling the number of storage cycles on the overall performance of underground hydrogen storage in the studied aquifer. Case Aq - 7 was run with the same configuration as Case Aq - 6 except that the number of storage cycles (before the extended 3 years period) was 14 cycles compared to 7 cycles in Aq - 6. Hydrogen gas maximum injection and production rates are shown in Fig. 12a.

A comparison between the cumulative injection and production volumes of the hydrogen gas in the two cases is shown in Fig. 12b where the cumulative volumes injected and produced in Case Aq - 7 were 87.4 and 72.4 Bscf, respectively. The injected and produced hydrogen gas volumes in Case Aq - 6 were 38.9 and 29.5 Bscf. In this section, the important comparison is the comparison of the hydrogen recovery efficiency in the two cases Aq - 6 and Aq - 7 as depicted in Fig. 12c. Results show that the hydrogen recovery efficiency of the underground hydrogen storage improved in Case Aq - 7 where the hydrogen recovery at the end of the studied period was 84% which is higher than the 75.9% hydrogen recovery obtained in Case Aq - 6. This observation highlights the importance of timescale on the technical and economic feasibility of underground hydrogen storage projects. Moreover, this calls for predicting the maximum hydrogen gas recovery expected in an underground hydrogen storage project to properly account for the economical aspect of the project. This maximum hydrogen recovery efficiency or Ultimate Hydrogen Recovery (UHR) can be defined by extending the simulation process and storage cycles until no further increase in recovery efficiency is observed.

4. Conclusion

In this study, we evaluated the impact of aquifer rock parameters and mechanisms on the performance of underground hydrogen storage in aquifers. Numerical simulations of underground hydrogen storage were conducted to evaluate the impact of these factors. The following reservoir parameters and mechanisms were considered: aquifer rock's heterogeneity, relative permeability hysteresis, hydrogen gas solubility in brine, and hydrogen gas diffusion. Results obtained from the numerical simulation revealed the following:

- The hydrogen gas recovery efficiency improves with the increasing storage cycles; hence the overall efficiency of the process is expected to improve with time.
- Aquifer permeability heterogeneity has a significant impact on the total volume of hydrogen produced where the cumulative volume of hydrogen produced in the homogenous case, was 33.53 Bscf which was higher than that of the heterogenous case, 30.38 Bscf. This resulted in an incremental hydrogen recovery of 7% in the homogenous case compared to the heterogeneous case. This was attributed to the better productivity expected in the homogenous aquifer model.
- The inclusion of the effects of relative permeability hysteresis led to the lower total injected and produced volumes of hydrogen gas compared to the base case where the hysteresis effect was ignored. A loss of 0.5 Bscf (39.5 Bscf to 39 Bscf) in the injected hydrogen volume and 1.24 Bscf (30.7 Bscf to 29.46 Bscf) in the recoverable hydrogen volume is observed due to relative permeability hysteresis effect. This is a result of the hydrogen gas entrapment that occurs due to the discontinuity of hydrogen gas during the withdrawal period. The disconnected hydrogen gas is then separated from the mobile gas and is left immobile inside the aquifer.
- When each parameters evaluated in isolation from the others, hydrogen gas solubility and diffusion resulted in minimal effects on the overall performance of underground hydrogen storage in aquifers. However, when combined and with the hysteresis effect, the impact of solubility and diffusion is more pronounced. Therefore, it is important to study these mechanisms collectively rather than in isolation for any underground hydrogen storage evaluation.

CRediT authorship contribution statement

Prashant Jadhawar: Writing - original draft, Visualization, Investigation, Methodology, Software, Data curation, Formal Analysis, Funding Acquisition, Writing - review & editing. Motaz Saeed: Writing - original draft, Visualization, Investigation, Methodology, Software, Data curation, Writing - review & editing, Formal Analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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REFERENCES

- [1] Abe JO, Popoola API, Ajenifuja E, Popoola OM. Hydrogen energy, economy and storage: review and recommendation. *Int J Hydrogen Energy* 2019;44(29):15072–86. <https://doi.org/10.1016/j.ijhydene.2019.04.068>.
- [2] Carden PO, Paterson L. Physical, chemical and energy aspects of underground hydrogen storage. *Int J Hydrogen Energy* 1979;4(6):559–69. [https://doi.org/10.1016/0360-3199\(79\)90083-1](https://doi.org/10.1016/0360-3199(79)90083-1).
- [3] Gallo AB, Simões-Moreira JR, Costa HKM, Santos MM, Moutinho dos Santos E. Energy storage in the energy transition context: a technology review. *Renew Sustain Energy Rev* 2016;65:800–22. <https://doi.org/10.1016/j.rser.2016.07.028>.
- [4] McCay MH, Shafiee S. Hydrogen: an energy carrier. *Future Energy: Improved, Sustainable and Clean Options for Our Planet* 2020:475–93. <https://doi.org/10.1016/B978-0-08-102886-5.00022-0>.
- [5] Abhishek R, Dąbrowski K, Klimkowski Ł, Somehsaraei HN, Assadi M, Jadhawar P. Opportunities and challenges in aquifer underground natural gas storage with a CO₂ cushion. *Challenges and Recent Advances in Sustainable Oil and Gas Recovery and Transportation* 2023:347–69. <https://doi.org/10.1016/B978-0-323-99304-3.00005-4>.
- [6] Green DW, Willhite GP. *Enhanced oil recovery (vol. 6)*. Henry L. Doherty memorial fund of AIME, society of petroleum engineers 1998.
- [7] Jadhawar P, Mohammadi AH, Yang J, Tohidi B. Subsurface carbon dioxide storage through clathrate hydrate formation. *Advances in the Geological Storage of Carbon Dioxide* 2006:111–26. https://doi.org/10.1007/1-4020-4471-2_11.
- [8] Jadhawar P, Yang J, Chapoy A, Tohidi B. Subsurface carbon dioxide sequestration and storage in methane hydrate reservoirs combined with clean methane energy recovery. *Energy Fuel* 2021;35(2):1567–79. https://doi.org/10.1021/ACS.ENERGYFUELS.0C02839/ASSET/IMAGES/LARGE/EF0C02839_0011.JPEG.
- [9] Oldenburg C, Stevens S, Benson S. Carbon sequestration in natural gas reservoirs: enhanced gas recovery and natural gas storage. *Greenhouse Gas Control Technologies - 6th International Conference* 2003:691–6. <https://doi.org/10.1016/B978-008044276-1/50110-0>.
- [10] Das D, Veziroğlu TN. Hydrogen production by biological processes: a survey of literature. *Int J Hydrogen Energy* 2001;26(1):13–28. [https://doi.org/10.1016/S0360-3199\(00\)00058-6](https://doi.org/10.1016/S0360-3199(00)00058-6).
- [11] Davison J, Arienti S, Cotone P, Mancuso L. Co-production of hydrogen and electricity with CO₂ capture. *Int J Greenh Gas Control* 2010;4(2):125–30. <https://doi.org/10.1016/j.jggc.2009.10.007>.
- [12] Holladay JD, Hu J, King DL, Wang Y. An overview of hydrogen production technologies. *Catal Today* 2009;139(4):244–60. <https://doi.org/10.1016/j.cattod.2008.08.039>.
- [13] Flesch S, Pudlo D, Albrecht D, Jacob A, Enzmann F. Hydrogen underground storage—petrographic and petrophysical variations in reservoir sandstones from laboratory experiments under simulated reservoir conditions. *Int J Hydrogen Energy* 2018;43(45):20822–35. <https://doi.org/10.1016/j.ijhydene.2018.09.112>.
- [14] Tarkowski R. Underground hydrogen storage: characteristics and prospects. *Renew Sustain Energy Rev* 2019;105:86–94. <https://doi.org/10.1016/j.rser.2019.01.051>.
- [15] Zivar D, Kumar S, Foroozesh J. Underground hydrogen storage: a comprehensive review. *Int J Hydrogen Energy* 2021;46(45):23436–62. <https://doi.org/10.1016/j.ijhydene.2020.08.138>.
- [16] Pan B, Yin X, Ju Y, Iglauer S. Underground hydrogen storage: influencing parameters and future outlook. *Adv Colloid Interface Sci* 2021;294. <https://doi.org/10.1016/j.cis.2021.102473>.
- [17] Raza A, Arif M, Glatz G, Mahmoud M, Al Kobaisi M, Alafnan S, Iglauer S. A holistic overview of underground hydrogen storage: influencing factors, current understanding, and outlook. *Fuel* 2022;330. <https://doi.org/10.1016/j.fuel.2022.125636>.
- [18] Delshad M, Umurzakov Y, Sepehrnoori K, Eichhubl P, Batista Fernandes BR. Hydrogen storage assessment in depleted oil reservoir and saline aquifer. *Energies* 2022;15(21):8132. <https://doi.org/10.3390/EN15218132>.
- [19] Heinemann N, Scafidi J, Pickup G, Thaysen EM, Hassanpouryouzband A, Wilkinson M, Satterley AK, Booth MG, Edlmann K, Haszeldine RS. Hydrogen storage in saline aquifers: the role of cushion gas for injection and production. *Int J Hydrogen Energy* 2021;46(79):39284–96. <https://doi.org/10.1016/j.ijhydene.2021.09.174>.
- [20] Jafari Raad SM, Leonenko Y, Hassanzadeh H. Hydrogen storage in saline aquifers: opportunities and challenges. *Renew Sustain Energy Rev* 2022;168. <https://doi.org/10.1016/j.rser.2022.112846>.
- [21] Pan B, Liu K, Ren B, Zhang M, Ju Y, Gu J, Zhang X, Clarkson CR, Edlmann K, Zhu W, Iglauer S. Impacts of relative permeability hysteresis, wettability, and injection/withdrawal schemes on underground hydrogen storage in saline aquifers. *Fuel* 2023;333. <https://doi.org/10.1016/j.fuel.2022.126516>.
- [22] Amid A, Mignard D, Wilkinson M. Seasonal storage of hydrogen in a depleted natural gas reservoir. *Int J Hydrogen Energy* 2016;41(12):5549–58. <https://doi.org/10.1016/j.ijhydene.2016.02.036>.
- [23] Feldmann F, Hagemann B, Ganzer L, Panfilov M. Numerical simulation of hydrodynamic and gas mixing processes in underground hydrogen storages. *Environ Earth Sci* 2016;75(16). <https://doi.org/10.1007/S12665-016-5948-Z>.
- [24] Saeed M, Sayani JKS, Jadhawar P. Evaluating the performance of various cushion gas types for underground hydrogen storage in an aquifer. *1st International Conference on Green Hydrogen for Global Decarbonisation 2023*. March 18.
- [25] Aslannezhad M, Ali M, Kalantariasl A, Sayyafzadeh M, You Z, Iglauer S, Keshavarz A. A review of hydrogen/rock/brine interaction: implications for Hydrogen Geo-storage. *Prog Energy Combust Sci* 2023;95:101066. <https://doi.org/10.1016/j.pecs.2022.101066>.

- [26] Ali M, Yekeen N, Pal N, Keshavarz A, Iglauer S, Hoteit H. Influence of organic molecules on wetting characteristics of mica/H₂/brine systems: implications for hydrogen structural trapping capacities. *J Colloid Interface Sci* 2022;608:1739–49. <https://doi.org/10.1016/J.JCIS.2021.10.080>.
- [27] Hosseini M, Ali M, Fahimpour J, Keshavarz A, Iglauer S. Assessment of rock-hydrogen and rock-water interfacial tension in shale, evaporite and basaltic rocks. *J Nat Gas Sci Eng* 2022;106. <https://doi.org/10.1016/j.jngse.2022.104743>.
- [28] Yekeen N, Al-Yaseri A, Negash BM, Ali M, Giwelli A, Esteban L, Sarout J. Clay-hydrogen and clay-cushion gas interfacial tensions: implications for hydrogen storage. *Int J Hydrogen Energy* 2022;47(44):19155–67. <https://doi.org/10.1016/J.IJHYDENE.2022.04.103>.
- [29] Alanazi A, Rasool Abid H, Usman M, Ali M, Keshavarz A, Vahrenkamp V, Iglauer S, Hoteit H. Hydrogen, carbon dioxide, and methane adsorption potential on Jordanian organic-rich source rocks: implications for underground H₂ storage and retrieval. *Fuel* 2023;346:128362. <https://doi.org/10.1016/J.FUEL.2023.128362>.
- [30] Saeed M, Jadhawar P, Bagala S. Geochemical effects on storage gases and reservoir rock during underground hydrogen storage: a depleted North Sea oil reservoir case study. *2023 Hydro* 2023;4:323–37. <https://doi.org/10.3390/HYDROGEN4020023>. 4(2), 323–337.
- [31] Yekta AE, Pichavant M, Audigane P. Evaluation of geochemical reactivity of hydrogen in sandstone: application to geological storage. *Appl Geochem* 2018;95:182–94. <https://doi.org/10.1016/J.APGEOCHEM.2018.05.021>.
- [32] Bo Z, Zeng L, Chen Y, Xie Q. Geochemical reactions-induced hydrogen loss during underground hydrogen storage in sandstone reservoirs. *Int J Hydrogen Energy* 2021;46(38):19998–20009. <https://doi.org/10.1016/J.IJHYDENE.2021.03.116>.
- [33] Hassannayebi N, Azizmohammadi S, De Lucia M, Ott H. Underground hydrogen storage: application of geochemical modelling in a case study in the Molasse Basin, Upper Austria. *Environ Earth Sci* 2019;78(5):1–14. <https://doi.org/10.1007/S12665-019-8184-5/FIGURES/11>.
- [34] **Computer Modelling Group. CMG user manual. Computer Modelling Group; 2022.**
- [35] Yekta AE, Manceau JC, Gaboreau S, Pichavant M, Audigane P. Determination of hydrogen–water relative permeability and capillary pressure in sandstone: application to underground hydrogen injection in sedimentary formations. *Transport Porous Media* 2018;122(2):333–56. <https://doi.org/10.1007/S11242-018-1004-7>.