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Citation for published version:
https://doi.org/10.1016/j.fuel.2022.126516

Digital Object Identifier (DOI):
10.1016/j.fuel.2022.126516

Link:
Link to publication record in Edinburgh Research Explorer

Document Version:
Peer reviewed version

Published In:
Fuel

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Impacts of relative permeability hysteresis, wettability, and injection/withdrawal schemes on underground hydrogen storage in saline aquifers

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Abstract

Underground hydrogen storage (UHS) is a key strategy in the implementation of a large-scale hydrogen (H₂) economy and promotion of renewable energy development/utilization. For UHS in water-wet saline aquifers, H₂ displaces *in-situ* brine during injection; during well shut-in and H₂ withdrawal, brine imbibes back into the flow paths where it displaces some H₂. These processes are influenced by H₂-brine transport physics, H₂-brine-rock interactions and injection/withdrawal schemes, which, in turn, determine H₂ storage capacities and injection/withdrawal efficiency. However, these effects are poorly understood. Therefore, this work focuses on the impact of relative permeability hysteresis (RPH), wettability, and H₂ withdrawal rate on UHS performance in a saline aquifer. Furthermore, differences between UHS and CO₂ geo-storage (CGS) are examined.

The primary findings include: 1) RPH results in a smaller H₂ withdrawal factor ($H_{2-WF}$), but a larger H₂ withdrawal purity ($H_{2-WP}$); 2) $H_{2-WF}$ increases with rock hydrophobicity, while $H_{2-WP}$ is mostly insensitive to rock wettability; 3) under similar storage conditions, $H_{2-WF}$ and $H_{2-WP}$ are approximately 10% less than $CO_{2-WF}$ and $CO_{2-WP}$.

These insights demonstrate the significance of RPH and rock wettability on UHS performance and provides guidance on H₂ injection/withdrawal scheme optimization. This study aids in the implementation of an industry-scale hydrogen economy.
Keywords: Underground hydrogen storage; Saline aquifers; Relative permeability hysteresis; Wettability; Injection/withdrawal scheme.

1. Introduction

Commercial development of renewable and sustainable energy resources are required to accelerate energy transition, mitigate global warming, and accomplish carbon neutrality [1–3]. However, these energy resources (e.g., wind, solar, and tide) are time-, season-, weather- and/or region- dependent, which limits their stability, reliability and large-scale economic implementation [4–6]. To overcome these drawbacks, underground hydrogen storage (UHS) is considered as a promising solution [7–11]. When energy supply is larger than energy demand, excess renewable and sustainable energy can be converted to hydrogen (H₂) through water electrolysis as a green energy carrier, and the H₂ can be injected into the subsurface for storage; when energy demand is high, H₂ can be withdrawn again from the subsurface for usage [12–17]. Potential subsurface storage sites include deep coal seams [18,19], depleted hydrocarbon reservoirs [20,21], aquifers [22,23] and salt caverns [15,24]. Salt caverns are suitable for frequent cyclic H₂ injection and withdrawal, but salt cavern storage capacities are usually small (around 50 × 10⁴ Sm³ [15,24,25]). Depleted hydrocarbon reservoirs are often well characterized with the necessary geological information, and substantial surface/subsurface infrastructure are in place [20,21]. However, microbial activity may be high, which causes both serious H₂ loss and H₂ purity reduction (e.g. via the reactions C₂H₆ + H₂ → 2 CH₄ or H₂ + S → H₂S...
In contrast, saline aquifers have the largest storage capacity and relatively weak microbial reactions, and thus attract significant attention [7,27]. Heinemann et al. [7] discussed the scientific challenges to enabling large-scale hydrogen storage in saline reservoirs while Pan et al. [27] reviewed all available experimental data related to UHS in saline aquifers.

Numerical simulation is a cost-effective and rapid method for large-scale UHS evaluation, which should be conducted prior to field-scale pilot tests. Pfeiffer et al. [28,29] used numerical simulations to predict UHS performance in the Rhaetian deposit, Germany, and the results demonstrated that up to 7700 × 10^4 Sm^3 (equal to 245 MW electricity) could be stored. Feldmann et al. [30] simulated 5 years of continuous H2 injection and subsequent 5 years of seasonal cyclic H2 injection/withdrawal into a depleted gas reservoir; the authors found that the H2 withdrawal purity (H2−WP) and withdrawal factor (H2−WF) reached 82% - 85% and 39%, respectively. Sainz-Garcia et al. [22] conducted a three-dimensional multiphase numerical simulation for three annual H2 injection/withdrawal cycles in Castilla-Leon, Spain. The stored H2 (67400 × 10^4 Sm^3) was able to supply 15% of the electric consumption for a population of 175,000 over 3 months. Additionally, Lubon and Tarkowski [23] utilized numerical simulations to predict seasonal H2 cyclic injection/withdrawal scenarios in a deep aquifer at Suliszewo, Poland - it was discovered that water coning was the main obstacle for UHS performance. Recently, Heinemann et al. [11] investigated the role of cushion gas for H2 injection and withdrawal in saline aquifers, demonstrating that the produced
H$_2$ was equal to 1.625 TWH from a three dimensional anticline reservoir model with
the ratio of cushion gas to working gas at 1.27 [11].

In the above-mentioned UHS simulations, either CO$_2$-brine relative permeability
curves were used as input parameters [11,28,29], or H$_2$-brine relative permeability
hysteresis (RPH) was not considered [22,23,30] (though CO$_2$ RPH strongly influences
capillary CO$_2$ trapping in saline reservoirs in CGS schemes [31]). To mitigate this
uncertainty, and to address the fact that H$_2$ is a very different molecule than CH$_4$ or CO$_2$
[27,32], UHS reservoir simulations with real H$_2$ input parameters and H$_2$ transport
physics are required.

Therefore herein, the impact of RPH, rock-H$_2$ wettability, and
injection/withdrawal schemes on UHS performance in saline aquifers is systematically
evaluated. Further, the acquired UHS results are compared with CGS data. This work
will provide important information supporting large-scale UHS implementation and the
decarbonization of energy supply chains.

2. Methodology

2.1 Geological model

The commercial reservoir simulator IMEX from Computer Modelling Group
(CMG) was used to simulate UHS in a synthetic PUNQ-S3 geological model. This
simulation is based on the classic black-oil model following the mass conservation
principles [33]. PUNQ-S3 is a three-dimensional, geometrically complicated and
heterogeneous geological model (a central dome + 5 layers of sand/shale) [34]. This
model was previously used for oil production forecasting [34] and CO₂ geo-storage simulation [31]. The average aquifer thickness is 15 m, and the entire domain is discretized into $19 \times 28 \times 5$ grid blocks (1761 of them active). Each cell has a length of 180 m in the horizontal direction. The average horizontal permeability and porosity are 100 mD and 0.2, respectively, with their spatial distributions shown in Figure 1.

Figure 1. (a) Horizontal permeability and (b) porosity distributions in the PUNQ-S3 geological model (Modified after [31]). For simplicity, only one well was used for gas injection and withdrawal.

A single well was drilled at the structurally highest location for gas injection and withdrawal. Similar to [31], the pore volume around the geological boundaries is set ~1000 times larger than the area of interest so that in-situ brine could be displaced during the gas injection; the displaced brine is imbibed back again during well shut-in (in case of water-wet rock) and simultaneously pumped out of the subsurface with gas during gas withdrawal.

2.2 Input parameters

Information about H₂ density, H₂ viscosity and H₂ expansion factor at UHS conditions are tabulated in Table 1. Currently, only one H₂-brine relative permeability
curve was measured for the process of \( \text{H}_2 \) injection into a brine saturated water-wet sandstone [35] (Figure 2). Using pore network modelling, \( \text{H}_2 \)-brine relative permeability curves (for drainage and imbibition) were also predicted for additional rock wettabilities [i.e., brine contact angles (\( \theta \)) of 51° and 83°] [36] (Figure 2). Information about \( \text{CO}_2 \) properties and relative permeability curves (which were collected from previous literature [31]) are not shown here for simplicity.

Table 1. \( \text{H}_2 \) properties at 40 °C and various pressures (compiled from [7,27]).

<table>
<thead>
<tr>
<th>Pressure [MPa]</th>
<th>Density [kg/m(^3)]</th>
<th>Expansion factor [-]</th>
<th>Viscosity [mPa(\cdot)s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>0.089</td>
<td>1</td>
<td>0.0092</td>
</tr>
<tr>
<td>10</td>
<td>8</td>
<td>89.9</td>
<td>0.0094</td>
</tr>
<tr>
<td>20</td>
<td>14</td>
<td>157.3</td>
<td>0.0096</td>
</tr>
<tr>
<td>30</td>
<td>20</td>
<td>224.7</td>
<td>0.0098</td>
</tr>
<tr>
<td>40</td>
<td>24</td>
<td>269.7</td>
<td>0.01</td>
</tr>
<tr>
<td>50</td>
<td>29</td>
<td>325.8</td>
<td>0.0104</td>
</tr>
</tbody>
</table>
Figure 2. H$_2$-brine relative permeability curves (modified after [35,36]; curve fitting was conducted to smooth the raw experimental and simulation data based on the least squares method). Experimental data is from [30]; simulated curves using pore network modeling are from [31].

2.3 Simulation scenarios

Four separate scenarios were simulated to explore the impact of RPH, rock wettability, and injection/withdrawal scheme on UHS performance, Table 2. In addition, two scenarios were simulated for CGS to provide a comparison.

Table 2. Scenarios simulated in this work (NA means that CO$_2$ wettability was unknown in the simulations).

<table>
<thead>
<tr>
<th>Gas</th>
<th>Case</th>
<th>Relative permeability hysteresis</th>
<th>Wettability</th>
<th>Injection/withdrawal scheme</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 (base case)</td>
<td>No</td>
<td>35º</td>
<td></td>
</tr>
<tr>
<td>H$_2$</td>
<td>2</td>
<td>Yes</td>
<td>51º</td>
<td>Same as case 1</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>Yes</td>
<td>83º</td>
<td>Same as above</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>4</td>
<td>Yes</td>
<td>51º</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5 (base case)</td>
<td>No</td>
<td>NA</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>Yes</td>
<td>NA</td>
<td>Same as case 5</td>
</tr>
</tbody>
</table>
3 Results and discussion

3.1 Impact of relative permeability hysteresis and wettability

For UHS, $H_2$ injection into a water-wet aquifer is dominated by the forced drainage (of the resident formation water), while $H_2$ withdrawal is dominated by the spontaneous and forced imbibition [37,38]. Therefore, it is necessary to assess the impact of RPH and rock wettability on UHS performance.

Figure 3. Effect of relative permeability hysteresis and rock wettability on the (a) actual $H_2$ withdrawal rate during the 1st withdrawal cycle, (b) $H_2$ withdrawal factor (the ratio of the accumulated $H_2$ withdrawal volume during a specific $H_2$ withdrawal cycle to the total $H_2$ in-place volume prior to this withdrawal cycle) and (c) $H_2$ withdrawal purity (the ratio of $H_2$ withdrawal mass to water production mass) at the prescribed withdrawal rate of $100 \times 10^4$ Sm$^3$/day).
RPH and a strongly water-wet state cause the actual H₂ withdrawal rate ($H_{2WR}$) to deviate from the prescribed $H_{2WR}$ value and to reach zero earlier, Figure 3(a).

During the 1st H₂ withdrawal cycle, the actual $H_{2WR}$ started to deviate from the prescribed 100 × 10⁴ Sm³/day on the 54th, 36th and 45th day, while it reached zero on the 81st, 49th and 62nd day, for the case of no RPH, case with RPH at $\theta = 51^\circ$, and the case with RPH at $\theta = 83^\circ$, respectively. Further, by the end of each withdrawal cycle, the H₂ withdrawal factor ($H_{2WF}$, the ratio of the accumulated H₂ withdrawal volume during a specific H₂ withdrawal cycle to the total H₂ in-place volume prior to this withdrawal cycle) follows the order: case with no RPH > case with RPH at $\theta = 83^\circ$ > case with RPH at $\theta = 51^\circ$. $H_{2WF}$ was 38%, 33% and 30%, respectively for the above-mentioned three scenarios at the end of the 2nd withdrawal cycle, Figure 3(b).

Moreover, at the beginning of each H₂ withdrawal cycle, RPH causes a larger H₂ withdrawal purity ($H_{2WP}$, the ratio of H₂ withdrawal mass to water production mass) than without RPH (e.g., 86% - 88% versus 55% - 65%), though the wettability impact is insignificant, Figure 3(c). In addition, with the H₂ withdrawal cycle increase, $H_{2WF}$ decreased at the end of each cycle of withdrawal, while $H_{2WP}$ increased at the beginning of each cycle of withdrawal, Figure 3(b) and (c), consistent with the previous literature study [28].

Note that RPH and rock wettability influence pore-scale gas-brine two phase flow characteristics and therefore determine reservoir-scale gas injection/withdrawal efficiency [38–43]. In the absence of RPH, the injected gas exists as a continuous gas plume, and capillary trapping is relatively weak [31]. If RPH is present, the trailing
edges of the gas plumes tend to convert into discontinuous phases, and capillary trapping are relatively strong (which is favorable for CGS because of reduced leakage risk) – however, it is unfavorable for UHS because of the more difficult gas re-mobilization [27]. Therefore, $H_{2-WF}$ was higher in the absence of RPH. Further, a more water-wet state leads to more snap off events [44,45], and therefore a more serious H$_2$ loss and a smaller $H_{2-WF}$. Moreover, during well shut-in, discontinuous H$_2$ bubbles can exert strong resistance for the spontaneous imbibition of in-situ formation brine [46,47] – therefore, if RPH is present, the initial H$_2$ concentration is higher around the wellbore region (which again results in a larger $H_{2-WP}$ at the beginning of each H$_2$ withdrawal cycle). The observed $H_{2-WF}$ and $H_{2-WP}$ response to the withdrawal cycle is because 1) at the end of each H$_2$ injection cycle, more H$_2$ will be in place than the earlier injection cycle; and 2) at the end of each H$_2$ withdrawal cycle, more H$_2$ will be lost to the subsurface than the earlier withdrawal cycle [28].

3.2 Impact of H$_2$ withdrawal rate

To operate a field-scale UHS project efficiently, the H$_2$ injection/withdrawal scheme [7] should be optimized, especially $H_{2-WR}$. Therefore, the impact of $H_{2-WR}$ on UHS performance was investigated in this section.
Figure 4. Effect of the prescribed $H_2$ withdrawal rate on (a) actual $H_2$ withdrawal rate during the 1st withdrawal cycle, (b) $H_2$ withdrawal factor and (c) $H_2$ withdrawal purity for the case of relative permeability hysteresis and brine contact angle of 51º.

Clearly, a larger prescribed $H_{2-WR}$ causes the actual $H_{2-WR}$ to deviate from the pre-set value and reach zero earlier in the simulated cases, Figure 4(a) and (b). For example, during the 1st $H_2$ withdrawal cycle (for the prescribed $H_{2-WR} = 100 \times 10^4$ Sm$^3$/day and $H_{2-WR} = 200 \times 10^4$ Sm$^3$/day), the actual $H_{2-WR}$ started to deviate from the prescribed value on the 38th and 18th day, respectively, while it reached zero on the 50th and 36th day, respectively. This is due to the faster pressure depletion caused by the larger $H_{2-WR}$ [48]. Therefore, it is suggested that sufficient $H_2$ is stored and sufficiently high reservoir pressure is maintained for continuous $H_2$ withdrawal at an expected withdrawal rate. Furthermore, a larger $H_{2-WR}$ caused a larger $H_{2-WF}$ and a more serious water production problem. For example, for the prescribed $H_{2-WR} = 100 \times 10^4$ Sm$^3$/day and $H_{2-WR} = 200 \times 10^4$ Sm$^3$/day, by the end of the 5th $H_2$
withdrawal cycle, \( H_{2-WF} \) was 25% and 29% respectively, and cumulative water production reached \( 7.5 \times 10^4 \) Sm³ and \( 13.1 \times 10^4 \) Sm³, respectively.

In principle, larger \( H_2 \) injection rates result in higher viscous forces, which can override capillary forces (analogue to \( \text{CO}_2 \) flooding [49]), and suppress lateral \( H_2 \) migration beneath the caprock, resulting in a larger \( H_{2-WF} \) [31]. However, to avoid water production problems, an optimized \( H_2 \) withdrawal rate is required; to determine this optimum \( H_2 \) withdrawal scheme, it is suggested that a balance between initial gas in place, transient \( H_2 \) demand, and gas purification/separation ability should all be considered [23].

### 3.3 Impact of gas type

During the past decades, CGS has been investigated comprehensively (e.g., [31,40,42,43,50–54]). In contrast, UHS is a relatively new technology which is still in its infancy [27,55–57]. Whether previous learnings from CGS can be directly used in UHS is still uncertain. Therefore, in this section, UHS and CGS are compared (under the prescribed gas withdrawal rate of \( 100 \times 10^4 \) Sm³/day and RPH conditions, Table 3 and Figure 5).

**Table 3.** Comparisons between underground hydrogen storage (UHS) and \( \text{CO}_2 \) geo-storage (CGS) during the first cycle of withdrawal, under the prescribed gas withdrawal rate of \( 100 \times 10^4 \) Sm³/day and relative permeability hysteresis conditions.
<table>
<thead>
<tr>
<th>Gas</th>
<th>Plume areal coverage</th>
<th>Deviation from the prescribed withdrawal rate</th>
<th>Withdrawal factor by the end</th>
<th>Withdrawal purity at the beginning</th>
</tr>
</thead>
<tbody>
<tr>
<td>UHS</td>
<td>Large: 35.22 km²; 30.5%</td>
<td>Early: 38 days</td>
<td>Low: 31%</td>
<td>Low: 84%</td>
</tr>
<tr>
<td></td>
<td>Small: 14.58 km²; 12.6%</td>
<td>Late: 50 days</td>
<td>High: 44%</td>
<td>High: 96%</td>
</tr>
</tbody>
</table>

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(a) After the 1st H₂ injection cycle  (b) After the 1st CO₂ injection cycle  

(c) After the 1st H₂ well shut-in  (d) After the 1st CO₂ well shut-in  

(e) After the 1st H₂ withdrawal cycle  (f) After the 1st CO₂ withdrawal cycle  

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Figure 5. (a-f) Gas saturation distribution during the 1st H₂ storage cycle; (g) the actual gas withdrawal rate during the 1st withdrawal cycle; (h) gas withdrawal factor during the continuous 5 withdrawal cycles and (i) gas withdrawal purity during the continuous 5 withdrawal cycles under the prescribed withdrawal rate of 100 × 10⁴ Sm³/day and relative permeability hysteresis conditions.

Clearly, UHS and CGS exhibit significant differences in gas saturation distribution, actual gas withdrawal rate, gas withdrawal factor and gas purity, Table 3 and Figure 5. After the initial gas injection for 9 months, the H₂ plume was ~ 2 - 5 times larger than CO₂, Figure 5(a) and (b) - this difference is caused by different gas viscosity and diffusivity [27]; after a well shut-in for 3 months, H₂ migrated significantly upward and accumulated beneath the caprock, while CO₂ only migrated slightly upward, Figure 5(c) and (d) - this was mainly caused by the difference in gas-brine density [27,58].

Furthermore, as shown in Table 3, Figure 5(h) and (i), in the same timeframe, \( H₂₋WF \) and \( H₂₋WP \) were smaller than \( CO₂₋WF \) and \( CO₂₋WP \).

Note that sandstone rocks are more water-wet in a H₂ environment than in a CO₂ environment [55,57,59,60], therefore gas bubble snap-off is more favored for H₂ than
for CO$_2$, which has led lower $H_{2-WF}$ than $CO_{2-WF}$ [61,62]). Meanwhile, especially during the gas injection stage, viscous fingering was predicted to be more pronounced for H$_2$ than for CO$_2$ [27], and that H$_2$ moves farther away from the wellbore region than CO$_2$ [57].

4 Conclusions and Recommendations

Underground hydrogen storage (UHS) is a promising technology which could aid the development of a large-scale hydrogen economy [12–17]. For UHS in saline aquifers, H$_2$-multi-cycle injection/withdrawal schemes are influenced by the energy supply and demand [23,63]. H$_2$-brine two phase flow physics, and H$_2$-brine-rock interactions determine UHS performance [7,27]. Therefore, in this work, the impact of relative permeability hysteresis, rock wettability, and injection/withdrawal schemes are systematically studied, and the results for UHS are then compared with those for CO$_2$ geo-storage (CGS). The following conclusions are reached:

1) H$_2$-brine relative permeability hysteresis results in a lower H$_2$ withdrawal factor, but a higher purity of withdrawn gas.

2) More water-wet rocks have lower H$_2$ withdrawal efficiencies.

3) Larger H$_2$ withdrawal rates increase H$_2$ withdrawal efficiency, but also increase water production.

4) UHS and CGS demonstrate significant differences and direct correlations should be avoided.
This study provides important information to aid in the implementation of a large-scale hydrogen economy, and therefore also supports the decarbonization of energy supply chains. For future work, it is suggested to further analyze the pore-scale H$_2$-brine two phase flow physics, and to establish a better understanding of meso-scale parameters (such as the H$_2$-brine relative permeabilities for cyclic drainage and imbibition processes and how they vary with wettability). Such improved input data leads directly to improved prediction of UHS performance [64,65].

Acknowledgements

Bin Pan thanks the initiative funding from the University of Science and Technology Beijing. Weiyao Zhu thanks the funding support from the National Natural Science Foundation of China (No. 51974013 and No. 11372033) and Open Research Foundation (NEPU-EOR-2019-003). Yang Ju acknowledges the funding support from the National Natural Science Foundation of China (No. 52121003). Christopher R. Clarkson thanks the sponsors of the Tight Oil Consortium (TOC) and Ovintiv and Shell for support of his Chair position in Unconventional Gas and Light Oil Research in the Department of Geoscience at the University of Calgary. Stefan Iglauer would like to thank the Australian Research Council for financial support (under grant DP220102907).

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