Feasibility Analysis of Hydrogen Storage in Depleted Natural Reservoirs Through a Multi-Phase Reservoir Simulator

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Summary

Unstable supply of renewable energy arises with the inevitable seasonal dependency, which contradicts with periodic energy demand. As hydrogen shows high energy density and mobility, yet low solubility and residual saturation, underground hydrogen storage (UHS) becomes a promising solution of scalable energy storage to rebalance demand and supply. Depleted gas reservoirs (DGR) are one of the most appropriate options for UHS because of the integrity of their caprock and storage system. In this study, we developed a numerical model based on TOUGH+RGB simulator (code) to simulate the flow and thermal transport during UHS in reservoirs such as DGR. Given the different transport and thermodynamic properties of hydrogen, different Equation-of-State (EOS) for modeling the phase behavior of hydrogen-included mixtures are calibrated with literature (lab) data, and further are coupled with the simulator. This benefits our numerical experiments to examine various cushion gas pre-injection strategies for pressure maintenance, boundary conditions, and potential hydrogen leakage into caprock. Hence, we can comprehensively assess the seasonal gas recovery factor of hydrogen stored in DGR. The calculated density of hydrogen-methane mixture based on GERG-2008 EOS and Soave-Redlich-Kwong (SRK) EOS is in perfect agreement with experimental data, while that from Peng-Robinson EOS is not quite consistent. Due to the accuracy and
efficiency, SRK EOS is employed in our simulator. Hydrogen injection-idle-withdrawal operation is simulated in a synthetic heterogeneous anticline DGR. Due to gravity segregation, we observe that hydrogen displaces pre-existing methane and resides at the top of the storage zone. When the caprock permeability ranges from $10^{-5}$ to $10^{-3}$ mD, only 0.05% of the injected hydrogen at maximum leaks into the caprock. Besides, an open boundary condition connecting with the storage zone helps the pressure maintenance in the storage and lowers the leakage, since with a close boundary condition the leakage rises to 0.35%. Further, about 1% of injected hydrogen is dissolved into the aqueous phase. Those results demonstrate that UHS in DGR has become a feasible choice. Nevertheless, only about 75% amount of hydrogen can be withdrawn if the bottom-hole pressure of producing well is 2MPa below the reservoir pressure. Therefore, cushion gas is necessary for the UHS project to increase hydrogen recovery. This work provides an in-depth investigation of various physics important to UHS, including EOS, hydrogen transport, capillary pressure, mixing, and dissolution. We quantitatively evaluated the hydrogen loss problem, including leakage to caprock, dissolved in water, and mixing with other gas molecules, which is the first-of-its-kind analysis in literature to the authors’ best knowledge. The modeling study is useful for the feasibility analysis of hydrogen storage in the depleted gas reservoir.

**Introduction**

The impact of the environmental consequences due to fossil fuel utilization has made the energy consumption issue a hot topic. Increased attention is paid to avoiding some issue caused by fossil fuels with an emphasis on carbon dioxide emission. Especially after the United Nations framework convention on climate change (UNFCCC)’s 2015 Paris agreement. The agreement aims limited global warming to well below 2, preferably to 1.5 degree Celsius, compared to preindustrial levels by reducing carbon dioxide emissions (Rogelj et al. 2016). In this context, the energy produced by renewable sources instead of fossil fuel-based energy sources attracts much attention globally to meet the Paris agreement. Renewable energy helps slow climate change because it produces only limited levels of greenhouse gases emission (Energy 2020). In practice, the share of renewable energy sources in the energy mix of Germany and Denmark is already significant (Energy 2014).

However, compared to conventional energy sources that are capable to produce electricity constantly, power generation of renewable energy, including but not limited to wind and solar, is subject to natural fluctuations governed by the weather conditions, resulting in renewable energy often excesses or deficits to meet the demand (Heinemann et al. 2021). In the context of the energy transition, intermediate energy storage is required to deal with the temporary mismatches between energy supply and demand. One promising answer is to use excess renewable energy to produce hydrogen gas, which can be subsequently stored and re-electrified if required (Mouli-Castillo et al. 2021, Sgobbi et al. 2016). The concept of energy storage in form of hydrogen was proposed in around the middle of 1970(Yartys and Lototsky 2005). Today, hydrogen is an attractive energy storage option for decarbonization and for making energetic systems more flexible with the development of hydrogen production technology (Luboń and Tarkowski 2020). More importantly, the properties of hydrogen also make it an ideal candidate as a carrier for energy storage. Hydrogen has a high specific energy capacity of 120 MJ/kg and has a clean combustion product (Muhammed et al. 2022). As an energy carrier, hydrogen has a low density of 0.089 kg/m$^3$ at standard conditions. To effectively balance the mismatch between market demand and supply, large-scale volumes of hydrogen, much beyond the scope of surface-based storage facilities, are needed to store energy on the scale of gigawatt hour to terawatt hour (Hashemi et al. 2021).

Therefore, underground space is suitable for such large scale of energy storage in the form of hydrogen since it supplies ample volume and high pressure. In addition, gas immobilization mechanisms such as structural, residual/capillary, mineral, and dissolution trapping make hydrogen stored for a mid to long time scale (Ali et al. 2021, Kalam et al. 2021). Although
each kind of underground space has its specific properties and geological characteristics, all of them should guarantee that the injected hydrogen can be recovered in the greatest amount possible, with minimal loss by leakage (Sørensen 2007). Therefore, potential sites of underground hydrogen storage (UHS) include saline aquifers, depleted hydrocarbon deposits, caverns, and coal seams (more recent) (Iglauer et al. 2021). Among them, the depleted gas reservoirs represent the best choice for large-scale UHS because of its well-known geological structure, proper compactness and integrity of the cap-rock, and pre-existence of surface facilities and infrastructures. Therefore, most UHS operations worldwide were situated in depleted hydrocarbon reservoirs (476), then in aquifers (82), and salt caverns (76) (Fig. 1) (Tarkowski 2019).

![Figure 1](image)

Fig. 1—Share of worldwide UGS by storage type in 2010 (Tarkowski 2019)

The interest in UHS is constantly increasing both in industry and academia. Several authors deal with various aspects of the topic, including technical aspects, evaluation of suitable sites, and economic evaluation (Tarkowski 2019). Numerical modeling and simulation are essential research approaches because laboratory experiments can only be used on a small scale and in limited situations, and field tests are extremely expensive. So far, mathematical models of UHS can account for thermal, multiphase, and multicomponent flow, and the actual field conditions, so they are capable to answer most of the questions in UHS, such as the gas distribution evolution when hydrogen is injected into an aquifer and the factor that affects the efficiency of the injection and withdrawal cycles (Wallace et al. 2021, Zivar et al. 2021, Muhammed et al. 2022). Several professional reservoir simulation software, CMG, ECLIPSE, COMSOL, DUMUX, OpenGeoSys, and TOUGH+, have been used to investigate the performance of UHS. Wallace et al (2021) wrote a comprehensive review of the details of software for UHS, along with the advantages and constraints of each software. However, only limited authors built their mathematical model to investigate the UHS. Cai et al (2022) developed a simulation for modeling grid-scale hydrogen and gas mixture storage in a cavern, deep saline aquifers, and depleted gas reservoirs. Regarding the UHS simulation, interested readers could find more details from Nasiru Salahu Muhammed et al (2022) and Davood Zivar et al (2021). They summarize recent modeling studies of UHS concerning the controlling parameter, storage medium, objectives, and major findings of each model. It should be noticed that most of the software is developed for other applications, which limited the validation of the UHS simulation (Cai et al. 2022). Besides, one common assumption made through models is that of an impermeable caprock to reduce complexities in the simulation, which results in a missing investigation on hydrogen leakage during the storage process. Due to the density, viscosity, and extremely low molecular size of the hydrogen, the leakage problem is noteworthy to consider in UHS modeling (Carden and Paterson 1979).

In practice, a whole UHS operations period includes the pre-injection of cushion gas followed by the hydrogen injection. The cushion gas serves as a buffer for pressure maintenance, namely, it undergoes alternate compression and expansion during the hydrogen injection and withdrawal process to keep the required pressure and the deliverability rate (Lord et al. 2014). Several kinds of gas, including nitrogen, carbon dioxide, methane, and even hydrogen itself, have been proposed as the potential cushion gas (Feldmann et al. 2016, Pfeiffer and Bauer 2015, Oldenburg 2003) because of their unique properties and benefit for UHS. For example, nitrogen has a lower investment cost, and it also facilitates the displacement of water to
achieve optimum storage space. In comparison to the aquifer UHS option, the depleted gas reservoir also has the advantage of the pre-existence of the remaining gas to prevent a massive amount of cushion gas needed for UHS operation. Only 50% to 60% of the entire reservoir capacity for cushion gas is required in the depleted gas reservoir as compared to 80% of the capacity in aquifers (Lord et al. 2014, Wallace et al. 2021). However, the mixing of hydrogen with cushion gas or native gas is one of the critical issues causing hydrogen loss. Besides, the production of cushion gas together with hydrogen can also increase the cost of UHS because it needs extra effort, equipment, processes, and energy to remove these from producing gas.

Our study aims to evaluate the feasibility of UHS via Tough+RealGasBrine (T+RGB) developed by Moridis et al (2014). T+RGB can describe the non-isothermal two (for pure water) or three (for brine with solid phase) flow of an aqueous phase and a real gas mixture in a gas-bearing medium, with a particular focus on ultra-tight reservoirs. Up to 12 individual real gases can be simulated, including hydrogen. Therefore, it meets simulation requirement for UHS by accounting for coupled flow and thermal effects, real hydrogen behavior, Darcy and non-Darcy flow (for the tight system such as caprock), hydrogen solubility into the water, and so on. The choice of a depleted gas reservoirs was based on its aforementioned advantages for UHS. Two aspects with limited information in previous studies will be discussed. First, how much is the hydrogen loss as a percentage of the total hydrogen injection amount? Three main parts of hydrogen loss are considered in our studies, namely, leakage into caprock, dissolution in the water, and staying in the reservoir after the withdrawal process. Second, evaluate the effect of two different injected cushion gas (nitrogen/carbon dioxide) on the hydrogen recovery and hydrogen losses in detail. The results allow us to figure out the possibilities of the UHS and optimize its operation.

**Methodology**

**Method.** Before the simulation, we compared the hydrogen gas mixture density calculated by three different equation of state (EOS) models, including the Peng-Robinson equation (Peng and Robinson 1976), Soave-Redlich-Kwong equation (Soave 1972), and GERG-2008 model (Kunz and Wagner 2012), with experimental data to determine the most accurate model. According to the comparison, the EOS model that balances accuracy and ease of computation will be applied in the simulation of our work. Then, the three basic UHS scenarios with different cap-rock properties and no-cushion gas injection were simulated first. After, two other cases involving different cushion gases were investigated.

Depending on the energy demand of the market and the fluctuation of renewable energy, the UHS project generally follows a cyclic operation with an alternate period of injection, idle, and withdrawal. In our study, hydrogen injection was performed for 150 days (about 5 months), and idle for 60 days (about 2 months) followed by 150 days (about 5 months) of hydrogen withdrawal. The injection rate increases or decreases linearly during the initial or end stage of the injection period, to prevent a steep pressure change in the reservoir. The hydrogen injection flow rate was set to 0.053 mscf/d (1.4 × 10^{-3} kg/s) according to the size of our simulation domain (25% of total pore volume). The producing method is the constant bottom-hole pressure method. For the simulation cases involving the cushion gas, an initial 180 days (about 6 months) of cushion gas injection followed by 180 days of the idle period was performed before the hydrogen injection. The cushion gas injection mass rate was set to 864 kg/d. Table.1 reports the cushion gas (if involved) and hydrogen injection/withdrawal schedule during the whole cycle. Both gas injection and withdrawal are assumed to be performed in the same well located at the summit of the slope reservoir.

In our work, the average pressure, temperature, and saturation of the storage zone and caprock, the amount of each gas part in different phases, and the production rate of each component were recorded during the simulation and further used to evaluate the feasibility of the UHS project.
Table 1. The schedule of cushion gas/hydrogen injection and withdrawal

<table>
<thead>
<tr>
<th>Process</th>
<th>Period</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Injection cushion gas</td>
<td>0-180 days</td>
<td>Constant rate 864 kg/day</td>
</tr>
<tr>
<td>Idle</td>
<td>180-360 days</td>
<td></td>
</tr>
<tr>
<td>Injection hydrogen</td>
<td>360-390 days</td>
<td>Rate increases from 0 to 121.62 kg/day</td>
</tr>
<tr>
<td>Injection hydrogen</td>
<td>390-480 days</td>
<td>Constant rate 121.62 kg/day</td>
</tr>
<tr>
<td>Injection hydrogen</td>
<td>480-510 days</td>
<td>Rate decreases from 121.62 kg/day to 0</td>
</tr>
<tr>
<td>Idle</td>
<td>510-570 days</td>
<td></td>
</tr>
<tr>
<td>Withdrawal</td>
<td>570-720 days</td>
<td>Constant bottomhole pressure 9.0d6 pa</td>
</tr>
</tbody>
</table>

**Constructed reservoir model.** A conceived depleted gas reservoir with a heterogeneous permeability distribution was considered as the simulation domain. The native gas is methane only. The constructed simulation domain covers an area of 500 m × 450 m, and the unstructured grid was generated based on the division of cells following the Voronoi method, giving a total of 7277 grid blocks. The domain was divided into four parts, namely, overburden or caprock (cyan zone), storage zone (blue zone), under burden (yellow zone), and a vertical well on the right side of the storage zone, as shown in Fig. 2. The thickness of the storage zone is about 100 m, and the left side of the storage zone was assumed to connect an infinite aquifer. The permeability distribution of the storage zone was randomly generated following Gaussian random permeability field and reconstructed by Karhunen-Loeve (KL) expansion (Zhang and Lu 2004), while the permeability of under burden is uniform for all cases. To investigate the effect of the caprock on hydrogen leakage, three different overburden permeability (corresponding entry capillary pressure) were used in our work. In the modeling of gas phase and aqueous phase flow, widespread characteristics were used: a modified version of stone’s model (Stone 1970) regarding relative permeability, and the Van Genuchten function (Van Genuchten 1980) regarding capillary pressure. Before UHS implementation, the initialization of pressure, temperature, and gas saturation distribution was performed based on the properties listed in the Table. 2. The other major properties used in our study can also be found in the Table. 2.

![Fig. 2—The grid block distribution of the simulation domain.](image-url)
Table 2. Properties of constructed reservoir domain in all cases

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure at z = 300 m</td>
<td>10^7 Pa</td>
</tr>
<tr>
<td>Temperature at z = 300 m</td>
<td>50.0°C</td>
</tr>
<tr>
<td>WGC location</td>
<td>Z = 300 m</td>
</tr>
<tr>
<td>Geothermal gradient</td>
<td>30.0°C/km</td>
</tr>
<tr>
<td>Methane mole fraction</td>
<td>100%</td>
</tr>
<tr>
<td>Injected gas temperature</td>
<td>50.0°C</td>
</tr>
<tr>
<td>Injected gas pressure</td>
<td>10^7 Pa</td>
</tr>
<tr>
<td>Bottomhole pressure</td>
<td>9.0 × 10^6 Pa</td>
</tr>
<tr>
<td>Porosity of storage zone</td>
<td>0.25</td>
</tr>
<tr>
<td>Porosity of overburden/under burden</td>
<td>0.05</td>
</tr>
<tr>
<td>Permeability of underburden</td>
<td>10^{-3} mD</td>
</tr>
<tr>
<td>Permeability of overburden</td>
<td>10^{-3}/10^{-4}/10^{-5} mD</td>
</tr>
<tr>
<td>Entry capillary pressure</td>
<td>2.0 × 10^6/6.325 × 10^5/2.0 × 10^6 Pa</td>
</tr>
</tbody>
</table>

**Numerical experiments and discussion**

Accurate predictions of the properties of gas mixtures containing hydrogen are important for the UHS simulation results. Here, the comparison between different gas EOS models is presented first in this section. Then, the effects of caprock permeability and cushion gas are discussed in detail based on the simulation results of different scenarios.

**Comparison of the gas mixture density calculated by three EOS models.** We compared three EOS models, namely the PR model, SRK model, and GERG-2008 model, applicable to hydrogen gas mixtures. Among them, PR and SRK models are traditional cubic EOSs. They are widely used in the petroleum field because of their simplicity and reliability (Abdollahi-Demneh et al. 2010); the GERG-2008 model is a non-cubic EOS that was proposed as a high-accuracy reference model. The density of gas mixture containing hydrogen, including hydrogen + methane, hydrogen + nitrogen, and hydrogen + carbon dioxide, was calculated by T+RGB (PR and SRK models are included) and an open-source code of GERG-2008 model provided by the NIST (National Institute of Standards and Technology) (https://github.com/usnistgov/aga8). Besides, we used published experimental data of the aforementioned binary gas system from the literature (Hernandez-Gomez et al. 2017, Hernandez-Gomez et al. 2018, Sanchez-Vicente et al. 2013) to statistically analyze and assess the reliability and accuracy of three EOS models.

The comparison results of four representative cases with different gas mole fractions, including 50% methane+50% hydrogen, 95% methane+5% hydrogen, 50% nitrogen+50% hydrogen, and 90% carbon dioxide+10% hydrogen, are shown in Fig. 3. Obviously, the results show that the GERG-2008 model and experimental data are in perfect agreement for all hydrogen mixtures. Then, the SRK model gives better accuracy in comparison with the PR model; especially for the case of 50% methane+50% hydrogen and 50% nitrogen+50% hydrogen, in which cases the densities calculated by the GERG-2008 model and the SRK model are almost the same as the experimental data. Besides, for the gas mixture of carbon dioxide + hydrogen, we also find there are deviations between the thermodynamic model and experimental data when pressure is extremely high. However, this deviation is not a problem because the pressure range involved in our study is from 9 MPa to 12 MPa.

In terms of simplicity of the EOS models, the accuracy of the GERG-2008 model is accompanied by increased mathematical complexity, resulting in increased computational time. We did a simple numerical test: for a simulation with 1000 grid blocks and the real simulation time is 60 days, it takes twice as long to complete using GERG-2008 model compared to that by using traditional cubic EOS model; the elapsed time ratio will be higher with the increase of reservoir grid number. This is because the calculation of the molar density of the GERG-2008 model requires an iterative calculation.
step, which consumes a lot of computational time (Baladão and Fernandes 2018, Kunz and Wagner 2012). Therefore, the SRK model is recommended for the application connected with hydrogen. In the following section, the SRK model is used to describe the properties of gas mixtures in all simulations.

![Graphs showing density of gas mixtures](image)

**Fig. 3**—Thermodynamic modeling and experimental results of the density of (hydrogen + methane, hydrogen + nitrogen, hydrogen + carbon dioxide) mixture at a range of temperature and pressure

**Effect of caprock permeability on hydrogen leakage** Hydrogen leakage into the caprock is a principal issue for UHS operation, but there is limited literature discussing it. A comparison between the three base models without cushion gas injected can demonstrate the effect of caprock properties on hydrogen leakage. In our simulation, three cases with different caprock permeability (as listed in table 2) were simulated. At the same time, the capillary pressure entry was scaled according to the permeability by the Leverett j-function:

\[ P_e = \frac{\sqrt{\frac{k_{ref}/\phi_{ref}}}{k/\phi}} P_{ref} \]  

(1)  

Where \( P_{ref} = 2.0 \times 10^6 \text{ Pa} \), \( k_{ref} = 10^{-20} \text{ m}^2 \), \( \phi_{ref} = 0.05 \). The corresponding capillary entry pressure for different permeability is also listed in Table 2.

Take the case of caprock permeability of \( 10^{-3} \text{ mD} \) as an example. **Fig. 4** shows the profile of the gas saturation and methane mole fraction, and hydrogen mole fraction in the gas phase on day 10, day 150, day 210, and day 360, which
correspond to the end of injection, idle, and withdrawal periods, respectively. The gas saturation distribution on day 10 is almost the same as the initial gas saturation distribution; the gas-water contact is at z=300 m. With hydrogen injection, the gas gradually spreads downwards to the deep storage zone along the edge of the caprock. From the methane mole fraction distribution shown in Fig. 4 row(b), we can observe the injected hydrogen displace the pre-existing methane. The black dotted circle indicates the gas leakage in the caprock. After hydrogen injection, hydrogen mainly accumulates in the upper part of the storage zone because of the gravity segregation driven by the density difference between the hydrogen and methane, as shown in Fig. 4 row(c). Then, since the gas phase is much lighter than the aqueous phase, the gas phase front moves back a little because of the buoyancy during the idle period. After day 210, the hydrogen begins to withdraw at a constant bottom-hole pressure. The gas phase front shrinks further. Since the hydrogen accumulates near the well after the injection and idle process, most of the injected hydrogen can be recovered, which is demonstrated by the result that the methane mole fraction in the gas phase is larger than 90% almost everywhere while the hydrogen mole fraction in the gas phase is small, as shown in Fig. 4 row(b) and row(c). However, the profile of day 360 indicates that some hydrogen remains in the storage zone and a portion of methane is extracted along with the hydrogen. The quantitative results will be discussed below.

Fig. 4—Gas saturation (a), methane mole fraction (b), and hydrogen mole fraction (c) distribution at day 10, day 150, day 210, and day 360 in each row.

Fig. 5 shows variations of average pressure, temperature, and gas saturation of storage zone with time under different scenarios. For all scenarios, the pressure first builds up because of hydrogen injection. With increasing pressure, the average pressure increase rate gradually slows down with hydrogen injection. This is because the pressure at the left boundary of the
storage zone keeps the same as the initial value, so the pre-existing water flows out of the left boundary as the storage zone pressure rises. Therefore, after about 70 days, there is a balance between the hydrogen injection and water flowing out, resulting in the average pressure being nearly constant. After, the average pressure begins to decrease when the hydrogen injection rate drops during the last 30 days of the injection period. During the hydrogen injection, the average temperature increases first and then decreases because our injection temperature is 50°C. Obviously, the average gas saturation increases all time because we injected gaseous hydrogen into the storage zone.

After entering the idle period, the average pressure continues to decrease because of the effect of the boundary condition, caprock, and under burden. At the same time, the average gas saturation also keeps increasing although there is no injected gas. This is because the gas mixture is less dense for lower pressure conditions, which indicates the gas volume increases with the pressure decreasing. Finally, the average pressure, temperature, and gas saturation suddenly decrease from the beginning of the withdrawal period. Subsequently, the average pressure and temperature increase again because the deeper water with high pressure and temperature flows back toward the production well.

Another noteworthy result in Fig. 5 is there is almost no difference between the three cases with different caprock permeability. That means the system has only a very small amount of gas leakage, even though 10^{-3} mD is already a very high permeability for the caprock (Armitage et al. 2011). More details about the leakage amount will be shown later.

![Fig. 5—Average pressure (a), temperature (b), and gas saturation (c) of the storage zone profile for the UHS without cushion gas.](image)

Obviously, larger caprock permeability results in more water and gas invasion into the caprock rather than moving into the deep storage zone, which leads to higher average pressure and gas saturation of caprock, as shown in Fig. 6. Besides, the caprock with high permeability has a low capillary entry pressure. Therefore, the gas is more likely to invade the caprock with high permeability. Fig. 6 (b) shows the caprock average gas saturation begins to increase from zero on day 10, day 25, and day 33 under three different cases. Due to the high irreducible gas saturation of the caprock, the average gas saturation of the caprock does not decrease during the withdrawal period; on the contrary, it keeps increasing because of gas expansion caused by pressure decreasing.
The variation of hydrogen amount in the storage zone and caprock is shown in Fig. 7. According to the injection schedule, the total injected amount of hydrogen is about 14594.4 kg. From Fig. 7, most of the injected hydrogen accumulates in the storage zone in gaseous form, while only a limited amount of hydrogen dissolves in water, up to about 190 kg (Fig. 7(a)). These dissolved gas accounts for only about 1.3% of the total injected amount. The low solubility of hydrogen promotes less hydrogen loss due to dissolution (Amid et al. 2016). The hydrogen amount dissolved in water increases a little bit during the idle period due to the increase in gas-water contact area caused by the fact that the space occupied by the gas phase expands during this period. After, the hydrogen either in the gas phase or aqueous phase is recovered. However, not all injected hydrogen can be extracted. On day 360, there is still about 4000 kg (27% of the total injected hydrogen) of hydrogen remaining in the storage zone. The amount of hydrogen remaining in water is too little to be worth considering.

The hydrogen amount in caprock represents the hydrogen leakage amount. Obviously, the hydrogen leakage amount increases with the permeability-increasing, as shown in Fig. 7(c)&(d). However, even though the permeability of caprock is $10^{-3}$ mD (a very large permeability for caprocks), the hydrogen leakage amount is only about 8 kg (4 kg in the gaseous phase and 4 kg dissolved in water). Therefore, there is almost no difference between the hydrogen amount in the storage zone in the three cases. Fig. 8 shows the ratios of hydrogen leakage to the total injected amount, which more intuitively shows that the hydrogen leakage amount is insignificant. For the caprock with a permeability of $10^{-3}$ mD, only about 0.05% of the injected hydrogen leaks into the caprock. The ratio would predictably become lower in the case of lower permeability.
According to the results above, we plot Fig. 9 to show where the injected hydrogen goes throughout the whole process for the case with a caprock permeability of $10^{-3} \text{ mD}$. The hydrogen in the reservoir is divided into four parts, namely existing in the gaseous phase in the storage zone, dissolution in water, leakage into caprock, and producing hydrogen. Fig. 9 demonstrates that the amount of hydrogen leakage and dissolution is insignificant, below 1% of the total injected amount all time. This proves that a depleted gas reservoir is indeed a suitable site for UHS because of its integrity. The capillary entry pressure plus the very small permeability of the caprock reduces the ability of the buoyant gas to percolate thus high storage capacity is achieved. However, as the case is shown in Fig. 9, only about 74% of the total injected hydrogen is recovered.
after the withdrawal period. Therefore, strategies need to be implemented to enhance hydrogen recovery, such as cushion gas. The next subsection will illustrate the effect of cushion gas on the UHS operation.

**Fig. 9—Variation of hydrogen distribution proportion**

**Effect of cushion gas on hydrogen storage.** The mechanics of primary recovery relies on the expansion of the fluids in the reservoir. Since the gas has higher expansion than water, to obtain a high primary recovery, as much gas as possible should be kept in the reservoir. This is why the injection of cushion gas will improve the economy of the UHS project. Therefore, the effect of cushion gas, including nitrogen and carbon dioxide, was investigated.

**Fig. 10** shows the comparison of the gas saturation profile when the cushion gas injection is complete. Carbon dioxide has a much higher density than nitrogen under the same thermo-physical conditions. Therefore, for the same injection mass of different cushion gas, the nitrogen occupies much more space. Compared to the initial gas saturation distribution, the gas phase extends a lot of space in the case of nitrogen, while there is only a small change in gas phase distribution in the case of carbon dioxide. Besides, the high solubility of carbon dioxide decreases the available gaseous carbon dioxide, which will be discussed in detail later.

Moreover, such significant different physical properties, such as density and mobility, also result in a distinct difference in each component's mole fraction distributions in the gas phase. **Fig. 11** shows the profile of cushion gas mole fraction in the gas phase on day 180 (the end of cushion injection), day 360 (the beginning of hydrogen injection), day 520 (the end of hydrogen injection), and day 720 (the end of withdrawal period). Both nitrogen and carbon dioxide have a larger density than
pre-existing methane. After injecting cushion gas, they all expand to the storage zone along the gas-water contact face or the lower edge of the storage zone. However, carbon dioxide is much denser than methane, so the gravity separation effect is more obvious, as shown in row (b) of Fig. 11. Besides, since two cases have the injected mass rate, and nitrogen has a smaller molar mass, the maximum mole fraction of nitrogen in the gas phase is approximately 95%, while the maximum mole fraction of carbon dioxide is only about 80%. Subsequently, the cushion gas is displaced to the deep storage zone by the injected hydrogen. All the cushion gas in the upper part of the storage zone near the injection well is replaced by hydrogen, except for the gas trapped by the caprock, as shown on day 520. Finally, the cushion gas returns to the vicinity of the well after the withdrawal period.

Fig. 11—Cushion gas profile at day 180, day 360, day 520, and day 720; row(a): nitrogen; row(b): carbon dioxide.

Fig. 12 shows the profile of methane mole fraction in the gas phase on day 180, day 360, day 520, and day 720. As the original gas in the storage zone, methane is always the main component at the front of the gas-water interface. It is displaced by the injected cushion gas; nitrogen displaces methane like a piston because of their close density, while carbon dioxide pushes methane up because it is much denser, as shown at day 360. After, the hydrogen further displaces the methane to the deep storage zone in the case of nitrogen. However, in the case of carbon dioxide, because the methane accumulates in the upper of the storage zone after the cushion gas injection, the disturbance caused by the hydrogen injection mix carbon dioxide and methane very well. Therefore, both methane and carbon dioxide mole fractions are very close and not extremely high after the hydrogen injection, as shown in row(b) of Fig. 11 and Fig. 12. Similarly, the methane ultimately flows back to the production well.
Fig. 12—Methane profile at day 180, day 360, day 520, and day 720; row (a): nitrogen; row (b): carbon dioxide.

Besides, nitrogen could increase the storage zone pressure to higher levels than the one with carbon dioxide because of its higher gas compressibility factor under the same thermodynamic conditions (Fig. 13 (a)). Also, the average gas saturation is higher when injecting nitrogen (Fig. 13(b)). Further, the average pressure gradually drops because of the effect of the infinite aquifer, as discussed before. At the beginning of the hydrogen injection, the average pressure of the storage zone with cushion gas of carbon dioxide is almost the same as the initial value. Then, the circumstances of the change in average pressure and gas saturation after the hydrogen injection are the same as before. During the hydrogen storage process after day 360, there is no significant difference between the average pressure in the three cases, while the average gas saturation is much higher in the case with nitrogen as cushion gas, compared to the rest of the two cases.

Fig.13—Average pressure (a), and average gas saturation (b) of the storage zone changes with time for the UHS with cushion gas.

The philosophy behind the necessity of cushion gas is that it can improve the hydrogen recovery factor, as we mentioned before. To quantify the improvement of cushion gas on the hydrogen factor, Fig. 14 shows the changes in the amount of the three gas components in the storage zone. According to Fig. 14 (a) and (d), on day 720, the remaining amount of hydrogen in the gaseous phase in the storage zone decreased from about 4000 kg to 2725 kg in the case of carbon dioxide, and 1276 kg in the case of nitrogen. This result definitely demonstrates that although the injected cushion gas has a limited effect on the
pressure buildup, the high compressibility of gas shows higher efficiency during the constant bottom-hole pressure production, compared to the original reservoir fluid, namely water with low compressibility. We also noticed that the hydrogen amount dissolved in water also decreases because of the existence of cushion gas, but the difference can be ignored.

The displacement of nitrogen enhances the methane-water contact area (Fig. 12) so that there is more methane dissolved in water, as indicated in Fig. 14(b). At the same time, the methane is displaced by injected nitrogen and eventually accumulates in the deep storage zone (Fig. 12). Therefore, the methane is hard to withdraw. According to Fig. 14(e), there is still about 8.8d4 kg of methane in the storage zone after the withdrawal period in the case of nitrogen, while there is only about 8.0d4 kg of methane in the cases of no cushion and carbon dioxide. The remaining methane is not only beneficial for the next UHS cycle but also means that the hydrogen purity of the produced gas is better.

The large solubility of carbon dioxide causes a significant amount of injected carbon dioxide to be dissolved in water, as shown in Fig. 14(c). Focusing on the cushion gas-injected scenarios, the total amount of injected cushion gas is 155.5 tons in our simulation; about 18% of injected carbon dioxide is dissolved in water, while only about 0.6% of nitrogen is dissolved in water at the end of the second idle period. This is another advantage of nitrogen as the cushion gas; the expansion of gaseous nitrogen can displace hydrogen during the withdrawal period. During the hydrogen injection period, the increased pressure and contact area led to more carbon dioxide dissolution. Finally, either nitrogen or carbon dioxide, part of it can accompany the hydrogen that is being produced, thereby affecting the purity of the produced hydrogen.

Regarding the effect of cushion gas on the hydrogen leakage problem, Fig. 15 shows that the ratio of hydrogen leakage amount to total injection amount drops from 0.05% in the base case (no-cushion gas injection) to 0.033% in the case of nitrogen as cushion gas. This influence exists but is almost negligible. Except for the integrity of the caprock, the pre-existing gas is also a reason for such a low leakage ratio in depleted gas reservoirs. After the cushion gas or hydrogen injection, the increased pressure in the storage zone drives the gas under the caprock to leak. Therefore, pre-existing gas, namely methane or cushion gas, leaks out firstly and occupies the pore volume of the caprock near the interface. At the same time, the capillary pressure within the caprock increases with the leakage of pre-existing gas, which prevents the subsequent leakage of hydrogen.

![Fig. 14—The variation of hydrogen, methane, and cushion gas amount in the storage zone.](image-url)
Although injected cushion gas can improve the hydrogen recovery factor, it also inevitably increases the impurity in the produced gas, especially for constant bottomhole pressure production. Fig. 16 presents the share (mass fraction) of three gas components, hydrogen, methane, and cushion gas, out of total gas production during the withdrawal period. In all scenarios, the gas produced in the early stage of production is mainly hydrogen because the hydrogen accumulates in the vicinity of the production well. As withdrawal proceeds, more and more associated gases were produced along with hydrogen. The hydrogen purity in the base case without cushion gas is the highest because there are only two gas components in produced gas; half of the final produced gas is hydrogen. For the case of cushion gas as nitrogen, about 20% of the produced gas is hydrogen and 75% of the produced gas is nitrogen. It is worth mentioning that the share of methane is very low, which means the produced gas can be regarded as two components (except for water vapor). However, the produced gas in the case of cushion gas as carbon dioxide is a different scenario. As mentioned earlier, the methane and carbon dioxide mix very well during the cushion gas and hydrogen injection period (Fig. 11 and Fig. 12). Therefore, both the mass fraction of carbon dioxide and methane are very high, about 30% and 45% respectively. This highlights the need for implementing several purification steps for the hydrogen retrieval process, which increases the costs.

The above results show that injection of cushion gas prior to the hydrogen injection can improve the UHS performance in terms of the hydrogen recovery factor. Compared to carbon dioxide, in addition that nitrogen is cheap and easily available
gas, nitrogen shows a better performance as the cushion gas. However, the utilization of carbon dioxide, which is a kind of greenhouse gas, as a cushion gas has some environmental benefits. The good news is that in all scenarios, either with cushion gas or without cushion gas, the hydrogen loss during the UHS operation is ignorable. Besides, we also notice that the effect of boundary (infinite aquifer) is also an important factor that we should be paid attention to.

Conclusion

In our study, the potential of implementing underground hydrogen storage in a depleted gas reservoir was numerical evaluated. The noteworthy hydrogen loss issues and the effect of cushion gas injection were investigated in details. Based on the results, the following conclusions were drawn:

1. In the calculation of thermophysical properties involving hydrogen, the SRK model is a cubic EOS model that balances accuracy and computational efficiency. However, we also find that the classical PR model can cause a large error in the calculation of gas mixture density involving hydrogen, especially when the hydrogen mole fraction is high.

2. The depleted gas reservoir is indeed a suitable site for underground hydrogen storage because of its integrity. Besides, most injected hydrogen is stored in the storage zone in gaseous form due to its extreme low solubility in water. In our simulation cases, the hydrogen loss (either dissolved in water or leak into caprock) is below 2% of total injected amount during the whole cycle.

3. Injection of a cushion gas prior to the hydrogen injection can sweep the original water away from the injection well. The presence of more gas phase makes the reservoir fluids more expandable during the withdrawal period. Therefore, can significantly improve the UHS performance in terms of the hydrogen recovery factor.

4. Compared to carbon dioxide, nitrogen exhibited a better performance as the cushion gas not only in terms of hydrogen recovery factor, but also in terms of purity of the produced gas.

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