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Review article

Hydrogen storage in depleted gas reservoirs: A comprehensive review

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A R T I C L E I N F O

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A B S T R A C T

Hydrogen future depends on large-scale storage, which can be provided by geological formations (such as caverns, aquifers, and depleted oil and gas reservoirs) to handle demand and supply changes, a typical hysteresis of most renewable energy sources. Amongst them, depleted natural gas reservoirs are the most cost-effective and secure solutions due to their wide geographic distribution, proven surface facilities, and less ambiguous site evaluation. They also require less cushion gas as the native residual gases serve as a buffer for pressure maintenance during storage. However, there is a lack of thorough understanding of this technology.

This work aims to provide a comprehensive insight and technical outlook into hydrogen storage in depleted gas reservoirs. It briefly discusses the operating and potential facilities, case studies, and the thermophysical and petrophysical properties of storage and withdrawal capacity, gas immobilization, and efficient gas containment. Furthermore, a comparative approach to hydrogen, methane, and carbon dioxide with respect to well integrity during gas storage has been highlighted. A summary of the key findings, challenges, and prospects has also been reported.

Based on the review, hydrodynamics, geochemical, and microbial factors are the subsurface's principal promoters of hydrogen losses. The injection strategy, reservoir features, quality, and operational parameters significantly impact gas storage in depleted reservoirs. Future works (experimental and simulation) were recommended to focus on the hydrodynamics and geomechanics aspects related to migration, mixing, and dispersion for improved recovery. Overall, this review provides a streamlined insight into hydrogen storage in depleted gas reservoirs.

1. Introduction

Global warming and climate change have had tremendous negative impacts on our planet. The situation is getting worse due to the increasing greenhouse gas (GHG) emissions from the burning of fossil-based fuels in different industrial activities from transportation to building and power sectors [1,2]. While the COVID 19 pandemic is considered a global challenge, it significantly reduced carbon dioxide (CO2) emissions as the projected 36.8 billion tonnes of CO_2 emission in 2020 was significantly reduced by 8 % (i.e. 2.94 billion tonnes) [3]. This vast reduction (considered the steepest fall in CO_2 emission since the tail end of World War II) [3] is believed to result from the slowdown in global industrial activities during the pandemic. Nevertheless, as the international community moves toward a balance, the global energy outlook on anthropogenic CO_2 emissions is again projected to exceed 40 billion metric tons by 2030 [4].

Many countries around the globe have established long-term substantial emission reduction objectives to achieve a net-zero CO_2 emission by 2050 [5]. This is evident as the 2015 Paris agreement aimed at reducing the global CO_2 emissions well below pre-industrialized levels, to hit 1.5 °C, was reemphasized in the recent COP26 Climate Change Conference in Glasgow [6]. Also, the joint report from the National Aeronautics and Space Administration (NASA) and the National Oceanic and Atmospheric Administration (NOAA) highlights that in 2020, the earth's average temperature will hit 1.02 °C, which holds a direct risk to the planet's inhabiting species [7], as such, the emission of CO_2

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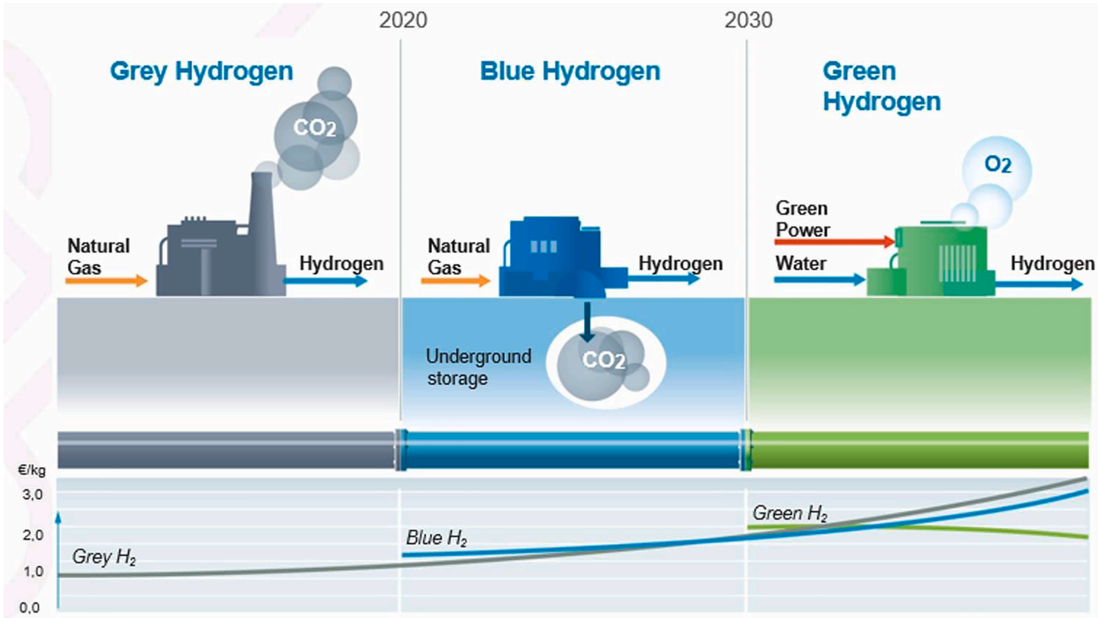


Fig. 1. The basic types of H_2 [17].

dramatically increased from 280 ppm to 410 ppm between 1750 and 2020 timescale of two centuries, respectively [4]. Therefore, it is highly significant to explore other approaches to reduce CO_2 emissions, such as CO_2 sequestration, carbon-free solar panels, wind power, geothermal energy, and hydrogen technology, to effectively mitigate the CO2 emission rate and its direct effect on the planet.

Hydrogen (H_2) is gaining popularity as a low-carbon energy carrier for decarbonizing transportation, heat and power, and fuel-intensive industries (chemical and steel) [8–10]. The United Nations Industrial Development Organization describes it as “a true paradigm shift in more efficient energy storage, especially for renewable energy on an industrial scale”, and it must play a significant role as a fuel substitute in energy-intensive industries to limit global warming [11]. Currently, natural gas reforming (i.e. steam methane reforming; SMR) is the typical way of producing blue and grey H_2 (note that CO_2 is captured and sequestered in blue H_2), Fig. 1, with an energy efficiency between 65 and 85 % [12]. While other fossil-based technologies such as partial oxidation, autothermal reforming, and coal gasification [13–15] exist. However, these emit harmful greenhouse gases into the atmosphere. In SMR, 1 mol of CO_2 is produced for every 4 mol of H_2, while for others (partial oxidation), the ratio is 1 mol of CO_2 per 3 mol of H_2 [16]. Thus, SMR seems to be more efficient among these technologies. H_2 can also be produced by water electrolysis (with 55–75 % energy efficiency based on electrolyzer capacity [12]), and this method is particularly suitable for renewable energy.

The production estimates for blue H_2 currently cost between $2.40/kg, whereas grey H_2 is $1.2/kg [18]. This indicates that blue H_2, even with capture, is close to that of grey without capture, implying that it should be the choice for sustainability. Moreover, blue H_2 cost is anticipated to decline further (Fig. 1). As for green H_2, an estimated cost

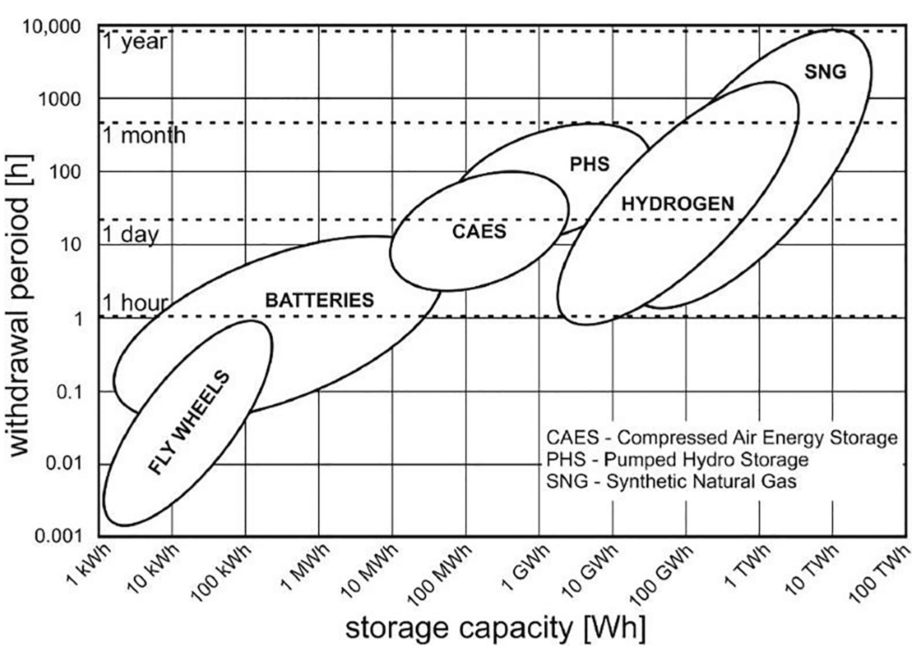


Fig. 2. Energy storage systems, their capacities, and withdrawals period [21].

of \$4.85/kg at an electricity costing rate of \$53/ MWh is reported [18]. With the recent cost reductions for renewable energy sources of electricity, it is projected that the green H_2 will catch up with the rest (\$2.50/kg) by 2030 [19]. However, this cost estimate is dependent on key parameters, such as renewable electricity price and the utilization factor (operating hours) [19]. Notwithstanding, steam reforming of natural gas remains the standard means of H_2 production, as fossil-based fuel's global energy supplies amount to 81.2 % compared to renewables (13.8 %) and nuclear energy (4.9 %) [13].

Besides the production challenges from fossil and renewable sources [13], H_2 storage for large and long-term effects are major bottleneck faced in recent times, and it remains untested mainly [8]. At typical conditions, H_2 has a high specific energy capacity of 120 MJ/kg and a low density of 0.089 kg/m^3 with clean combustion products [20]. As a result, large volumes are required (GWh to TWh) for community-level distribution and utilization, which is beyond the reach of the surface-based storage facilities, Fig. 2 [21]. However, this seems to be only possible if geological structures, such as salt caverns, aquifers, or depleted hydrocarbon reservoirs [22–24], which has the capacity for ample volumes for storing H_2 at high pressure (thus high energy densities) and more extended time. Fig. 3 is a schematic of the conventional geo-storage means. Once H_2 is injected, it is retained in the geological formation through trapping mechanisms like structural/stratigraphic, residual/capillary, solubility/dissolution, and mineral trapping [25,26].

Amongst geo storage options practised, the depleted hydrocarbon reserves represent the best choice for large-scale UHS because of their known geological structure, good compactness and source rock integrity, and pre-existence of surface and subsurface facilities [27]. Moreover, studies have proven them as the most cost-effective. It was found that salt caverns, depleted hydrocarbon reservoirs, and saline aquifers had a Levelized cost per kg of H_2 of 1.61, 1.23, and 2.77 USD, respectively [28]. However, converting a depleted hydrocarbon reservoir into a UHS site necessitates extensive research because the reservoir contains excess oil/gas that may facilitate the possibilities of specific chemical reactions, such as the injected H_2 turning into, for example, methane [29]. Again, the solubility/miscibility of the injected H_2 in/with the fluids (water, oil, gas) also significantly influences their loss, making research and simulation of these phenomena crucial. Thus, it becomes essential to understand the basics of depleted hydrocarbon (oil and gas) reservoirs employed for UHS operations.

As can be seen in Table 1, several reviews on UHS are available in the public domain ranging from geological standpoint [22–24,27,30–36], microbial activities [8,21,37–39], as well as parametric insights on hydrodynamics [8,22,24,36,40,41], geomechanics [8,30,42], geochemical interactions [8,34,38,39], and well integrity [39,43]. While these reviews (as summarized in Table 1) are considered timely, streamlined insights on the depleted gas reservoir are yet to be considered. We thus focus here on depleted gas reservoirs (because oil reservoirs have more complicated multiphase fluid flow interactions, which will result in pore space reduction and greater storage costs due to separation after withdrawal; reservoirs also do not have the added benefit of a cushion gas already in place [5] and data on H_2 storage in oil reservoirs are limited).

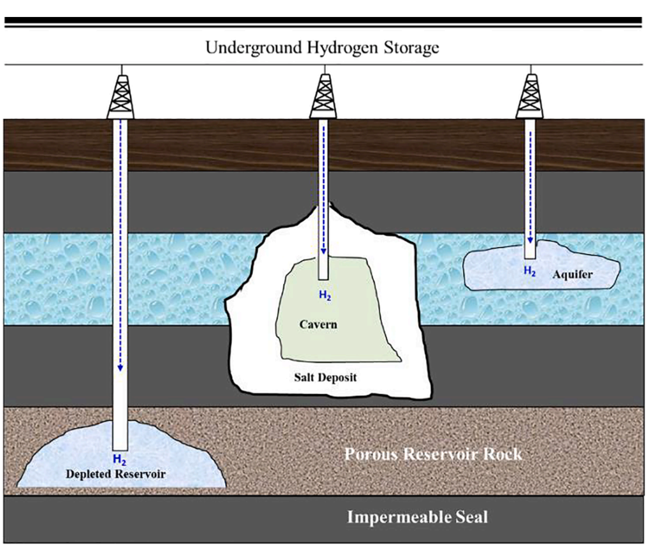


Fig. 3. Underground hydrogen storage schematics.

This study aims to provide comprehensive insight into H_2 storage in depleted gas reservoirs. This will be achieved by presenting a comparative characteristic of H_2 with CH_4 and CO_2 (as they are the conventional geo-storage gases) and their immobilization methods in the subsurface. Furthermore, an overview of the mechanism of H_2 storage in a gas reservoir, the current operating and potential sites, and some case studies will be discussed. In addition, H_2 storage with respect to hydrodynamics, geochemical and microbial factors will be reviewed as they affect the storage and withdrawal capacity and efficient gas containment. A systematic economic comparison between salt caverns, aquifers, and depleted reservoirs will also be analyzed before well integrity-related issues and knowledge gaps.

The following is a breakdown of how this review is structured. Following the introduction, **section 2** covers the physicochemical features of H_2 in comparison to methane and carbon dioxide, as well as gas trapping processes, while **section 3** covers a basic overview of depleted gas reservoirs, potential sites, and case studies. In **section 4**, data on experimental and modelling studies were critically analyzed before economic analysis (**section 5**). **Section 6** concluded with a comparison study (similarities and differences) of methane, carbon dioxide, and hydrogen in UGS, CCS, and UHS, respectively. In **sections 7 and 8**, the identified knowledge gaps and the review's learning outcomes were emphasized, respectively.

2. Background insights

This section briefly summarizes the physicochemical properties of H_2, and analog gases (CH_4 and CO_2) often used in geo-storage. The general retention mechanism (as presented in the introduction) for underground gas storage (caverns, aquifer, and depleted hydrocarbon) is also discussed.

2.1. Comparison of H_2 properties with CH_4 and CO_2

H_2 storage is similar to other gas storage systems, such as CH_4 and CO_2, but it confronts additional obstacles due to its characteristics and limited understanding. As the smallest and most abundant element, its energy content (120 MJ/kg) per unit mass is about 2.2 times that of CH_4 (55 MJ/kg) [44]. H_2 is about 8 and 22 times less dense than CH_4 and CO_2, respectively, implying that more storage space and pressure will be required to achieve the same mass amount of H_2 gas. Its low viscosity over CH_4 and CO_2 makes its retention a major challenge during geo-storage. Also, H_2 mobility and large diffusivity cause a lower residual amount of H_2 in the porous media during the withdrawal stage. Thus, a practical understanding of the best trapping mechanism for H_2 is essential to increase the containment of the stored gas. H_2 has low solubility in pure water at high pressure compared to CH_4, which is 15 times greater than CO_2 (though it varies with reservoir pressure). This implies that the loss of H_2 via dissolution is minor, whereas losses via diffusion and dispersion may be more due to its high diffusivity. Furthermore, hydrogens' high diffusivity and low molecular weight make it more subjective to leak through the overburdened layers [12,22,45] during storage.

The physical and chemical properties of H_2 concerning CH_4 and CO_2 influence the selection choice for cushion gas during the storage cycle. The cushion gas is needed for pressure maintenance during the injection

Table 1
Summary of review studies on underground hydrogen storage.

References	Topic	Focus
Reitenbach et al. [39]	Influence of added hydrogen on underground gas storage: a review of key issues	• Storage media integrity • Materials compatibility • Microbial Activity
Tarkowski [23]	Underground hydrogen storage: characteristics and prospects	• Geological • Technical • Environmental • Economic perspectives on UHS
Zivar et al. [22]	Underground hydrogen storage: a comprehensive review	• Thermophysical insights • Microbial and geochemical activities • Flow dynamics • Modelling and economic viability • Challenges and road map
Cyran [42]	The influence of impurities and fabrics on mechanical properties of rock salt for underground storage in salt caverns – a review	• Rock salt fabrics • Short-and long-term mechanical properties
Dopffel et al. [37]	Microbial side effects of underground hydrogen storage, knowledge gaps, risks, and opportunities for successful implementation	• Risk assessment • Monitoring and implications
AbuAisha and Billiotte [30]	A discussion on hydrogen migration in rock salt for tight underground storage with an insight into a laboratory setup	• Geochemical interaction • Modeling approach • Effect of temperature
Heinemann et al. [8]	Enabling large-scale hydrogen storage in porous media – the scientific challenges	• Hydrodynamics • Geochemical insights • Microbial aspects • Geomechanics insight • Storage integrity
Pan et al. [41]	Underground hydrogen storage: influencing parameters and outlook	• Solid properties • Fluid properties • Solid-fluid interactions
Thaysen et al. [38]	Estimating microbial growth and hydrogen consumption in hydrogen storage in porous media	• Homoacetogens • Methanogens • Sulfate reducers • Iron reducers • Controlling environmental factors (nutrient, temperature, salinity, brine complexity, pH, pressure, and inhibitors) affecting microbial growth
Wallace et al. [31]	Utility-scale subsurface hydrogen storage: UK perspectives and technology	• Challenges and barriers • Geological criteria and availability • Road map for UHS research
Aftab et al. [36]	Toward a fundamental understanding of geological hydrogen storage	• Abiotic and biotic reactions in salt caverns • Progress and influencing factors • Energy capacity relative comparison
Muhammed et al. [24]	A review on underground hydrogen storage: insight into geological sites, influencing factors, and outlook	• Global outlooks • Hydrodynamics/Geochemical • Microbial activities • Economics of UHS • Prospects and challenges
Sambo et al. [32]	A review on worldwide underground hydrogen storage operating and potential fields	• Global outlooks (operating and potential) on cavern, aquifer, and depleted reservoirs • Challenges and prospects
Ugarte and Salehi [43]	A review on well integrity issues for underground hydrogen storage	• Well integrity on gas storage • Well-integrity criteria, recommendations, and selection
Jafari Raad et al. [33]	Hydrogen storage in saline aquifers: opportunities and challenges	• Transport and storage mechanisms • Technical challenges • Economic perspective
Raj et al. [34]	A comprehensive review of the mechanisms and efficiency of underground hydrogen storage	• Global and geological insight • Technical outlooks • Monitoring mechanism • Ethical implication and challenges
Tarkowski and Uliasz-Misiak [35]	Towards underground hydrogen storage: a review of barriers	• Geological limitations • Technical limitations • Legal limitations
Molíková et al. [21]	Underground gas storage as a promising natural methane bioreactor and reservoir?	• Microbial versus thermogenic insights • Microbial processes and communities in UGS
Epelle et al. [27]	Perspectives and prospects of underground hydrogen storage and natural hydrogen	• Pros and cons between surface and subsurface technologies • Comparative insights on UGS, CCS, and UHS efficiency • Natural occurrence
Raza et al. [40]	A holistic overview of underground hydrogen storage: Influencing factors, current understanding, and outlook	• Influencing factors • Hydrodynamics • Geochemical • Economics of UHS • Prospects and challenges

and withdrawal stages. It has been reported that despite using Nitrogen (N_2) due to its low cost [46,47], the intensive mixing of H_2 and N_2 during the cyclic operation was a major challenge. Additionally, structural geometry plays a significant role in cushion gas selection due to the density and viscosity as fingering and gravity override can occur [48]. Moreover, the effect of its very low density and viscosity in the subsurface will result in more viscous fingering and poor conformance during injection, more rapid migration towards structurally high locations, and thicker columns of H_2 at a higher pressure than other gases trapped beneath the seals. Therefore, H_2 will be much more buoyant in the reservoir and have much higher mobility than CH_4 and CO_2.

In a comparative study on H_2 density and viscosity concerning CH_4 and CO_2, it was found that the densities and viscosities of H_2 and CH_4 are considerably smaller than CO_2; thus, H_2 and CH_4 have a broader potential storage sites coverage as they are not restricted by depth [45,49] but are determined based on the geological feature of the sites as it requires proper containment features to trap and seal the fluid. This implies that CH_4 storage sites can be used as a direct substitute for H_2 storage.

2.2. Trapping mechanisms

Gas storage in the depleted reservoir is an established concept. The first documented report dates back to 1915 in Welland County, Ontario, Canada, from a storage operation in an operating gas field [50]. Since then, scientists and engineers have leveraged analogies from storage operations with similar technical, geological, and hydraulic constraints, such as natural gas geo-storage and, to a lesser extent, CO_2 geo-storage, to better understand the behaviour of H_2 in the subsurface. However, several characteristics (physical and mainly chemical) that are peculiar to H_2 storage must be considered. As mentioned in the introduction section, there are four methods for retaining gas in geological formations: structural/stratigraphic, residual/capillary, solubility/dissolution, and mineral trapping.

In the contest of CO_2, Ali et al. noted that structural/stratigraphic trapping is more dominant in caprock and sedimentary formations. Whereas residual/capillary trapping is the most common for sedimentary formations. Both solubility/dissolution and mineral trapping dominate in basaltic and sedimentary formations (see refs. [4]). Albeit H_2 trapping via adsorption in coal seams has been documented [51–54], it is beyond the scope of this review. Thus, gas trapping mechanisms discussed in the next section are often reported for CO_2 geo-storage [55–57]. However, it can be extended to H_2 as they exhibit similar behaviour to a certain degree during gas subsurface immobilization (see Pan et al. [41]; see also Fig. 4).

2.2.1. Structural/stratigraphic trapping.

Structural or stratigraphic trapping is a time-dependent hydrological process in which supercritical (CO_2) or gaseous (CO_2 and H_2) gases are injected into and trapped in a geological formation beneath an impermeable caprock [2]. The presence of the impervious layer prevents the upward migration of buoyant gas (since it has a lower density than in-situ brine). This physical trapping process (thought to be the most prominent [59]) can hold the injected gas for extended periods and is influenced by the trap volume and caprock integrity [60,61]. Despite the trapping structures' well-defined features, there is still a risk of leakage since the stored gas under the sealing rock is highly mobile. During injection operation [55], H_2 gas, for instance, (which is highly mobile compared to CO_2), may increase the reservoir pressure resulting in the activation of new or existing faults and crack, which could lead to the escape of the mobile H_2 gas via slow diffusion. It's worth noting that a balance of viscous, capillary, and gravity forces controls the vertical and lateral distribution of injected gas via porous sedimentary storage rocks beneath the impermeable caprock [2].

The displacement characteristics of the two-phase flow (drainage and imbibition) are defined by the water receding (θ_r) and water advancing (θ_a) contact angles [62]. θ_r corresponds to H_2 injection (drainage – water displaced by H_2), and it's relevant for structural trapping whereas the θ_a implies H_2 withdrawal (imbibition – water displacing H_2) and its relevant for capillary trapping (see **section 2.2.2**).

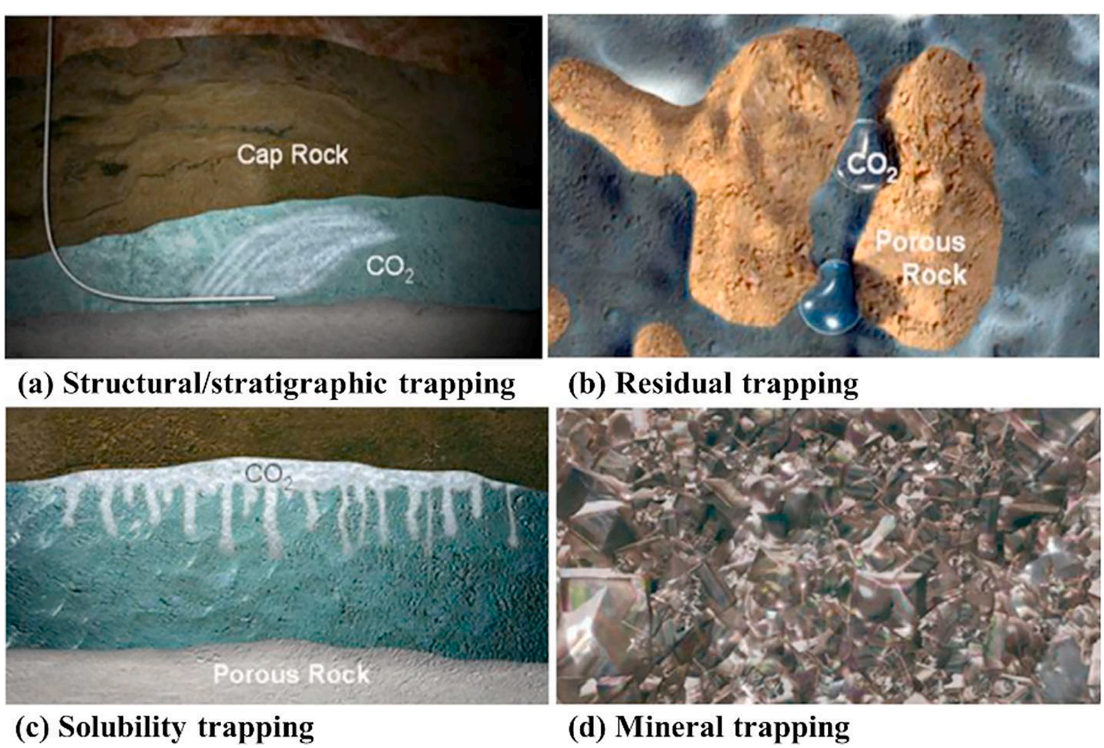


Fig. 4. Gas trapping mechanisms [58]. The figure represents CO_2 injection; however, a similar scenario can be extended to H_2 during injection and withdrawal.

In the injection phase, the gas is retained by the impermeable seal through a balance between the capillary and buoyancy forces. **Eq. (1)** is therefore known as the H_2 column height that can be permanently immobilized beneath the caprock based on the equilibrium balance between these forces [63,64].

$$h = \frac{2\gamma \mathrm{Cos}\theta}{h\rho g r} \tag{1}$$

herein, h is the permanently immobilized gas below the caprock, γ is the interfacial tension between the brine and the H_2 phase, θ is the gas/brine/rock receding contact angle of the system, Δρ is the density difference between the brine and gas phase, r is the effective pore radius corresponding to the narrowest pore throat along the entire flow path and ′g′ is the gravitational constant. Note that the interplay of these forces determines the exact fluid configuration on the rock surface, and it's characterized by Young's equation (i.e., the macroscopic force balance between the cohesive and adhesive forces, **Eq. (2)** [65].

$$\cos\theta = \frac{\gamma_{\mathrm{rb}} - \gamma_{\mathrm{rg}}}{\gamma_{\mathrm{bg}}} \tag{2}$$

here, 'θ' is the contact angle that governs the wettability, usually determined experimentally through the denser phase. γ_{rb}, γ_{rg} and γ_{bg} denote rock/brine, rock/gas, and brine/gas interfacial tensions, respectively.

2.2.2. Residual/capillary trapping

This trapping mechanism is based on the concept of wettability, where a rock exhibits a strong preference for a phase. For instance, when gases, such as CO_2 or H_2 are injected into the subsurface [66], which is often the non-wetting phase in many sedimentary rocks, first displaces the fluid as it moves through the porous rock, and then as the movement continues, the displaced fluid returns to imbibe the gas remaining within the pores at the trailing edge [67]**.** After such imbibition, the gas is left behind in the form of disconnected immobile bubbles and ganglia [68,69]. The rock wettability, particularly the θ_a affects the residual trapping and is a key factor in the identification of the H_2 interaction with reservoir brine and the storage rock. More precisely, it allows for understanding the distribution of H_2 through the porous rock. Besides the rock wettability, the capillary forces holding the buoyant H_2 gas in the capillary pores can be estimated using the Young-Laplace equation [62].

$$\mathrm{P_c} = \mathrm{P_{H_2}} - \mathrm{P_{brine}} = \frac{2\gamma\cos\theta}{\mathrm{r}} \tag{3}$$

here, $\mathrm{P_{H_2}}$ and $\mathrm{P_{brine}}$ denotes the nonwetting and wetting phase pressures, respectively. Note that **Eqs. (1) and (3)** assume an ideal cylindrical capillary tube and thus only provide a first approximation as the pore geometry plays a major role [63].

Unlike structural/stratigraphic trapping, this trapping mechanism relies entirely on hysteresis (a fundamental feature of multiphase flow in which repeated drainage and imbibition events are encountered in a porous media [70]). Relative permeability and capillary pressure as a function of saturation are the two constitutive relations used to represent the hysteresis behaviour of gas/brine systems as seen in the experimental investigation in a CO_2 brine system for relative permeability [71–73] and capillary pressure [70,74,75]. While the core flooding experiment of Yekta et al. [76] gave insight into the relative permeability and capillary pressure data relevant to UHS, the recent pore network modelling by Hashemi et al. [20] elaborated this work to present the hysteresis behaviour in an H_2 brine system for both relative permeability and capillary pressure. It is reported that this immobilization mechanism significantly helps determine the quantity of gas migration and distribution within the formation, which impacts the effectiveness of other trapping methods [2,77]. It is widely acknowledged as a key trapping mechanism for the security of stored gas. For instance, Hesse et al. [78] and Ide et al. [79] reported that this mechanism is the only option that can effectively achieve 100 % gas (CO_2) immobilization over time.

2.2.3. Solubility/dissolution trapping

This process occurs when injected gas comes in contact with the formation fluid (i.e., brine/gas) and dissolves afterward. As a result, the laden formation fluid (gas plus in-situ brine) becomes denser and sinks rather than rising [80–82]. While the laden formation fluid is less susceptible to leaks due to increased density, the effectiveness of this mechanism (i.e., the amount of gas that can be dissolved in the brine) depends on the physical properties, such as the reservoir pressure, temperature, and brine salinity [66,83]. Moreover, the possibilities of dissolution trapping of H_2 are low, especially when the storage scenario is short and non-permanent [66]. This phenomenon of trapping can take up to thousands of years under natural conditions [84].

Convection and dispersion generally govern the transport of dissolved gases. In gas mixing, the process is mainly influenced by dispersion, mobility ratio, and density difference [48,85]. Hydrodynamic dispersion involves both molecular diffusion and mechanical dispersion. In most cases, molecular diffusion (a slow process compared to advective/convective transport [85]) is induced by the concentration gradient independent of the fluid flow. Thus it is usually not considered in-field case simulations because the characteristic diffusion length is relatively smaller compared to the dimension of the gas plume [86]. On the other hand, mechanical dispersion is caused by variations in local velocity due to porous medium heterogeneity [87]. It also results from variations in the velocity, which can occur on various scales, from microscopic to reservoir scale [48,85]; as a result, both fluid velocity and reservoir rock properties (such as porosity and tortuosity) influence this process [80]**.**

2.2.4. Mineral trapping

Mineral trapping and dissolution trapping have certain similarities, as both entail fluid dissolving after gas injection. However, in this situation, the injected gas (e.g., CO_2) dissolves in the formation water to produce a weak carbonic acid (H_2CO_3), which causes rock minerals to dissolve/precipitate due to the pH drop, forming solid phases that effectively trap the injected gas [4,88,89]. These solid phases could be calcium carbonate or calcium sulfate, depending on the available anion [90]. This process takes thousands to billions of years (geological timescale), depending on the rock chemistry (reactivity) [59,84] and the formation of water [2]. Mineral trapping, however, is slower than CO_2 dissolution trapping because it involves the interaction of fluid and rock; additionally, it provides a greater long-term CO_2 sink and a stable CO_2 trapping mechanism in the form of carbonate minerals [2], although much faster reaction time has recently been observed in basalt (e.g. Matter et al. [89]).

Haven discussed the four standard gas trapping mechanisms. Recent experimental investigation often ascribes structural and residual trapping as the primary mechanism for H_2 retention in geo-storage [25,26].

It is vital to state herein that, in the subsurface, H_2 geochemical reactions are redox driven whereas CO_2 is acid-base driven [41]. These redox-driven H_2 reactions predominantly occur in the presence of iron-bearing minerals (e.g., hematite and goethite), iron-bearing clays and micas as well as in the presence of dissolves sulfur species or sulfur-bearing minerals [8,91]. H_2 also undergoes microbial-type reactions once the subsurface microorganisms are exposed to excess H_2. This reaction (microbial) type could be due to abiotic (non-living components such as water, rock minerals, and gases) and biotic (living components such as bacteria) factors which are majorly dependent on the environment (since the subsurface is characterized by combinations of factors such as elevated temperature, high pressure, high salt concentrations, reduced void space, and limited nutrient availability). The major difference between geochemical and microbial-type reactions is that microbial type leads to permanent loss of H_2 as the H_2 is technically converted to products like CH_4 or H_2S [8]. Detailed insights on the potential reactions of injected H_2 with pre-existing minerals, gases, ions, microorganisms, and other substances for safe (to reduce the risk of leakage) and successful operations (to avoid H_2 conversion or reduction in its purity) can be found elsewhere [38,92].

3. Depleted gas reservoirs

Hydrocarbon reservoirs (oil or gas) are geological traps that have undergone several diagenetic formation stages (source rock, migration, and time) to serve as a hydrocarbon storage medium. These traps are usually overlaid with an impermeable layer that is generally aquifer supported from the bottom or edges. A gas field is often transformed into a gas storage facility when it approaches the end of its productive life. Due to aquifer water displacement, a depleted (or about to be depleted) gas reservoir is generally characterized by low pressure and high-water saturation in the zone once occupied by the gas. As a result, gas saturation behind the water front varies from a minimum, which corresponds to residual gas saturation near the initial gas/water contact, to a maximum, which corresponds to gas saturation near the gas/water contact [93]. In other words, the depleted gas reservoir can be thought of as a part of an aquifer (geological traps) where only trace amounts of water exist within pores that are occupied mainly by trapped gas [31].

Depleted gas fields can be used to store gases because their imperviousness (resistance) over geological time has already been proven [94,95]. To date, these types of reservoirs are the most commonly employed for natural gas storage due to their well-identified geological features, the rigidity of the seal (trap), and the highly well-researched prior exploration, development, and production activities [96]. Fig. 5 is a conceptual depleted gas reservoir showing the key elements required for safety operations, namely (i) a porous and permeable formation for H_2 storage, (ii) an impermeable seal to inhibit vertical migration due to buoyancy, and (iii) an anticline trap to limit H_2 lateral movement for effective plume build-up. As seen, the underground gas storage inventory is made up of two types of gases: working gas and cushion gas [48,85]. The working gas (in this case, H_2) is typically injected and then withdrawn from storage during demand, whereas the cushion or base gas (e.g., CO_2, CH_4, N_2, and even H_2) is stored in the facility to maintain pressure throughout the process via compression and expansion during injection and withdrawal cycles [24]. Further, it prevents water inclusion for optimum storage space [97] as well as reduces the effect of contamination of the injected gas.

During operation, the injected H_2 displaces the in-situ fluids (brine and residual gas) in the pores after blending (mixing). It subsequently spreads underneath a low permeable seal capable of hosting the fluids [8]. The fluid is forced downward or sideways to generate storage space due to density changes between the injected H_2 and native fluid that occurs with increasing pressure. Because of the constant increase in pressure, an interface (liquid–gas) is formed during the injection process. The developed interface between the H_2 and the brine/gas in the pore spaces may disrupt the withdrawal process [22,47], especially for short-term storage. Since this phase will be miscible with the injected H_2, it is reasonable to expect that during the initial cycles of injection/withdrawal of H_2, the recovered gas will contain a proportion of the mixture, which will decrease with the number of storage cycles. However, the degree of mixing of native (e.g., CH_4, CO_2, N_2) and injected gas and the interaction between the gas and liquid phases is uncertain. Although previous experience with natural gas storage has shown piston-like behaviour and limited mixing between the injected and native gas [98], it is unknown whether this will be the case with H_2 when injected into the reservoir.

The cushion gas requirement for the depleted gas field is minimal (as compared to the aquifer) because the native gas in the reservoir allows for reduced cushion gas during injection [24]. Between 50 and 60 % cushion gas has been reported, compared to 80 % for aquifers [28,31]. Cushion gas, on the other hand, can be problematic if it compromises the purity of the injected H_2; as a result, caution must be used when selecting the best candidate gas for maintaining pressure during UHS projects.

Working gas and cushion gas can be of similar composition in H_2 storage operations as reported in the simulation works of Sainz-Garcia et al. [99] and Lubon and Tarkowski [100] while assessing the feasibility of UHS in San Pedro belt (Spain) and Suliszewo structure (Poland) respectively. Alternative non-H_2 gases such as CH_4, CO_2, and N_2 have also been used (more details in Table 4). Particularly, CO_2 is employed based on its physical properties (density and viscosity) at typical reservoir conditions as well as to reduce GHG emission whereas both CH_4 and N_2 are used based on costs [8].

3.1. Potential sites and UHS project outlooks

Many countries worldwide have been diligent in developing

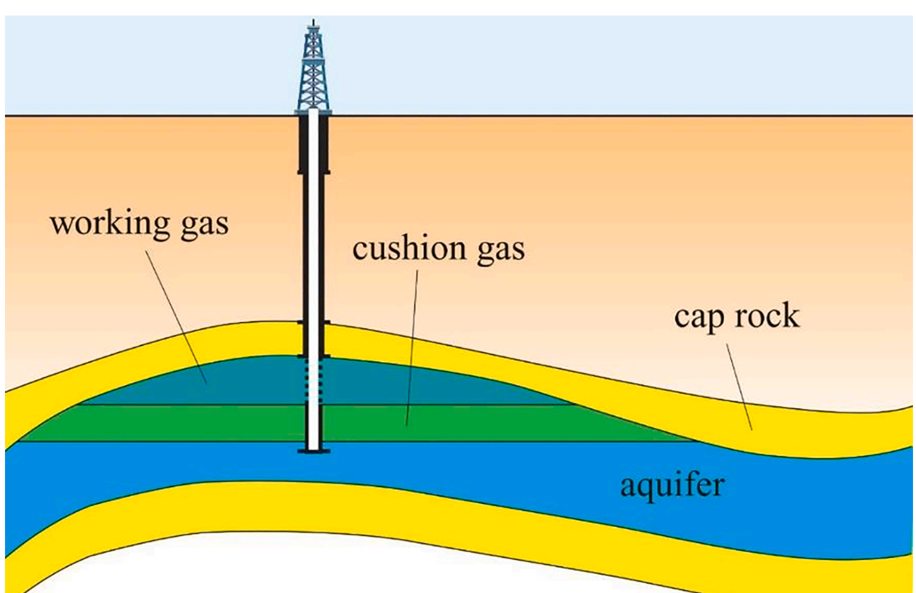


Fig. 5. Conceptual H_2 storage in the depleted gas reservoir.

practical UHS solutions, as seen by the rise in projects like Roads2HyCOM [101], HyStorPor [5,38], HyUnder [102], H_2STORE [103], ANGUS+ [104], SUN.STORAGE [105], Hychico [106], ADMIRE [20,107], just to mention a few.

The HyStorPor project [5], for example, focused on the geological storage of H_2 in geographically widespread porous rocks in the United Kingdom. Simon et al.'s paper [102] gave an outline of the European HyUnder project, which attempts to test the feasibility of large-scale UHS. The collaborative research projects: H2STORE [108] and ANGUS+ [109] looked into the feasibility of storing H_2 in porous geological formations [104]. The H2STORE and the European HyUnder project evaluated UHS as a potentially attractive solution from a technological, economic, and social angle. In Austria, the underground SUN STORAGE project investigated the effects of storing H_2 in a methane storage container [105]. The Hychico SA project in Patagonia, Argentina, aims to convert the power generated by the Diadema wind farm into H_2, which would then be injected into a depleted field that acts as a natural gas storage facility [106].

These projects demonstrate diverse research organizations' continued efforts to promote a carbon-free environment. Table 2 and Fig. 6 represent some reported operating and potential sites for H_2 storage in depleted oil and gas scenarios using different criteria. Though several of the pilot projects described (HyStorPor, SUN.STORAGE, and Hychico) are dedicated to porous medium with emphasis on depleted reservoirs, prior studies have highlighted some practical experience in caverns and aquifers. For example, a mixture of 95 % H_2 and 3–4 % CO_2 has been reported in a salt cavern in Teesside, UK. Similarly, the Gulf Coast of Texas in the United States reported 95 % H_2 and 3–4 % CO_2, as did the Kiel town gas project in Germany, which held roughly 60 % H_2 [32]. The geo-storage of H_2 in combination with other gases: CO_2, CH_4, CO, and N_2 in an aquifer has been recorded in the projects of Ketzin in Germany, Lobodice in the Czech Republic, and Beynes in France [22]. More details on worldwide operating and potential sites for UHS can be found elsewhere [32].

3.2. Case studies

To meet the seasonal demand for H_2, many countries have put forward long-term emission reduction goals by 2050. Despite the stated advances in achieving net-zero carbon emissions through greater energy efficiency, increased renewables and nuclear power, and a large move from coal to natural gas-fired power plants, the global prognosis on emissions remains uncertain [5]. One of this complexity is the use of heat in buildings, as related activities, such as the direct combustion of fuels in cookers and boilers, account for about 17.5 % of the world's emissions [5,117]. When it comes to using natural gas as a primary source of heat via direct combustion, the United Kingdom has the highest percentage (84 %), followed by the Netherlands (83 %), Italy (72 %), and Hungary 69 % [5]. This indicates that GHG emissions remain a challenge for most affluent countries worldwide, and urgent decarbonization measures are required to meet the 2050 vision and the CO_2 reduction goal.

The UK is known for its high seasonal gas demand, as documented by several authors [8,38]. More so, its offshore regions offer suitable UHS facilities [118]. The seasonal demand pattern for H_2 in the UK propelled the study by Mouli-Castillo et al. [5] to map out the geological H_2 storage capacity and regional heating demands. The authors selected four locations (Southern North Sea Basin, the Central Graben, the Viking Graben, and the East Irish Sea), each serving as a gas terminal. Followed by the H_2 storage capacity estimation, by replacing the gas field volumetric capacity occupied by natural gas (assuming methane properties) with H_2, and finally, the amount of H_2 storage required to balance the seasonal supply and demand was estimated. The study documented that, out of the general H_2 energy needed in the UK (77.9 TWh), the combined estimated gas storage capacity of the studied gas fields was 2661.9 TWh. This indicates that a single gas field alone is more than sufficient to store enough energy as H_2 to balance the fluctuating seasonal demand for UK domestic heating. Their study reveals that H_2 storage in depleted gas deposits will not affect other low-carbon subsurface storage applications as only a few fields are required out of the entire 41 selected offshore gas fields [5].

Previously, the Ontario province in Canada depended on coal-fired power plants. However, the continuous effort toward renewable energy space made a massive impact as it officially phased out all its coal-fired power plants in 2014 [116]. This decision was made easier because the wind and nuclear energy sectors generated around 6 % and 63 % of total energy in 2017, respectively, compared to 0.9 % and 53 % in 2008 [116]. Currently, the province's energy demand has declined by 11 % between 2008 and 2017, making it a key export hub with an estimated net energy of 12.3 TWh as of 2017 [116]. As a result, surplus energy must be managed using power-to-gas conversion, in which extra energy is used to make H_2 gas by electrolysis and then stored underground for seasonal demands. Though the number of UHS sites is currently restricted, researchers have proposed introducing a power-to-gas strategy [119,120] to promote a long-term H_2 economy in both domestic uses [121,122] and transportation industries [123]. As of the present literature, 35 depleted gas fields are found in the southwestern part of Ontario (major wind and supply plants region) with an estimated storage capacity of 6.9 billion m^3 [116].

As seen in the geological map (Fig. 7), over 340 oil and gas pools with an estimated production capacity of 37.6 billion (natural gas) and 90.4 million (oils) exist in the southern province of Ontario [116]. The principal oil and gas reservoirs grouped into five with their exact formation lithology and depth are presented in Table 3. It is believed that natural gas is stored currently in the pinnacle reef structures within the Silurian carbonates found in southwestern Ontario [124]. A preliminary assessment of CCS suitability has been established in these fields, implying that the depleted oil and gas reservoirs have the requisite effective trapping mechanisms; thus, H_2 storage in prospect should be ideal in these locations, provided the geological properties remain the same. Common challenges, such as those due to microbial archaea (methanogens, sulfate reducer, and those found during lixiviation of metallic ores) that cause H_2 loss via bacteria metabolism (H_2 oxidation), cannot be avoided [125], and as such, more investigation will be required.

Poland [115,126] is another coal-dependent country in terms of economics. However, as evidenced by several discoveries of H_2 potential sites (caverns, aquifer, depleted oil, and gas) as reviewed [97], increasing effort is being made to promote the transition to an H_2 economy [100,115,126,127]. Specifically, 19 locations from Upper Permian and Neogene (Miocene) formations and the Polish lowlands were suggested for caverns as UHS potential sites, whereas 19 prospective deep aquifer locations from Lower Cretaceous and Lower Jurassic formations previously used for CO_2 were proposed [97]. In the case of depleted oil and gas reservoirs, 39 locations from the Polish Lowlands, the Carpathians, and the Carpathian Foredeep, was suggested [97]. We also note that recent advances were made to utilize deep coal seams as H_2 storage reservoirs [51–54]. However, all these prospective sites are dependent on setting the requirement and storage conditions on a detailed geological analysis and reservoir engineering properties [128]. Several studies on the integrated UHS facilities (aquifer, caverns, and depleted oil and gas reservoirs) in Poland were analyzed to assess the strength and ranking framework [115]. According to the Analytical Hierarchical Process (AHP) and the authors' assumptions, it was found that only 47 of the recommended sites (77 locations) [97] met the UHS criterion, with 11 out of 19 for salt caverns, 14 out of 19 for aquifers, and 22 out of 39 for depleted oil and gas reservoirs. In particular, 18 UHS sites were found as a potential depleted gas fields for H_2 storage, Fig. 8 [115].

These case studies illustrate that research is growing to find geological formations to support a sustainable H_2 economy, which primarily refers to the use of H_2 as a zero-emission fuel for automobiles,

Table 2
Worldwide operating and potential H_2 storage sites based on depleted reservoir conditions.

Reservoir type	Country	Location(s)	Operating conditions				Others			Study outlook	Status	Composition (%)	Reference
			Pressure (psi)	**Temperature (°C)**	**Porosity (%)**	**Permeability (mD)**	**Capacity (MMm^3)**	**Depth (ft)**	**Salinity (ppm)**				
Gas	Austria	Molasse basin	1131	30 - 80	NR	NR	NR	3280	14 – 18,000	H_2 and methane storage assessment and sun conversion	Operating	H_2 = 10 - 20	[105]
	Germany	Gas field based on Röt formation, Buntsandstein and Zechstein	588	40	5 - 10	NR	NR	2188	NR	Potential risk study	Potential	NR	[110]
	Netherlands	Dutch geological structure	Properties based onBundsandstein stratigraphy				Properties based on Bundsandstein stratigraphy			Feasibility study	Potential	NR	[111]
	UK	Rough gas storage facility	725 - 1450	92	20	75	48	8999	NR	Seasonal storage via geochemical analysis	Potential	NR	[12]
	USA	California	NR				NR			Impact of H_2 mixing with natural gas	Potential	NR	[112]
Oil	China	Bohai Bay	3190	20	26	4.66	6240	6233	NR	Hybrid system for H_2 production and storage	Potential	NR	[113]
Oil and gas	Argentina	Diadema field	145	55	25	300 - 500	NR	2673	NR	H_2 underground storage and methanation	Not Reported	H_2 = 10	[22,114]
	Poland	Sudetes and Carpathians foredeep within Polish Lowland	Not reported, however, detailed characteristics and condition is presented by [97]				Not reported, however, detailed characteristics and condition is presented by [97]			Screening and ranking framework for H_2 storage based on geological setting	Potential	NR	[115]
	Scotland	D'Arcy-Cousland and Balgonie Anticlines of the carboniferous age within the Midland Valley	NR	NR	NR	60 - 80	NR	328 - 3280	NR	Introduced the concept of H_2 storage play	Potential	NR	[45]
	Norway	Norne field	1885 - 3916	NR	25 -30	20 - 2500	1500	8999	NR	Seasonal H_2 storage investigation	Potential	NR	[96]
	Canada	Ontario	NR	NR	NR	NR	NR	360 - 3937	NR	Preliminary assessment for H_2 storage	Potential	NR	[116]

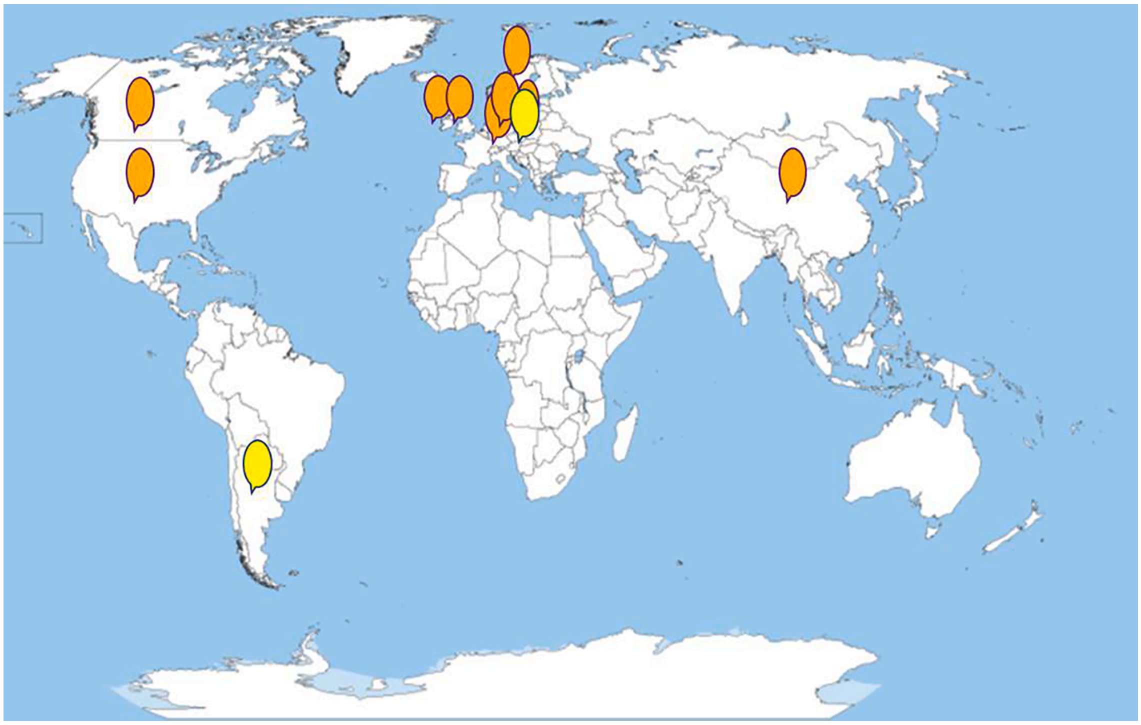


Fig. 6. Worldwide operating and potential depleted oil and gas reservoirs for H_2 storage. The yellow colour represents the operating sites, whereas the orange represents the potential sites for H_2 storage. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

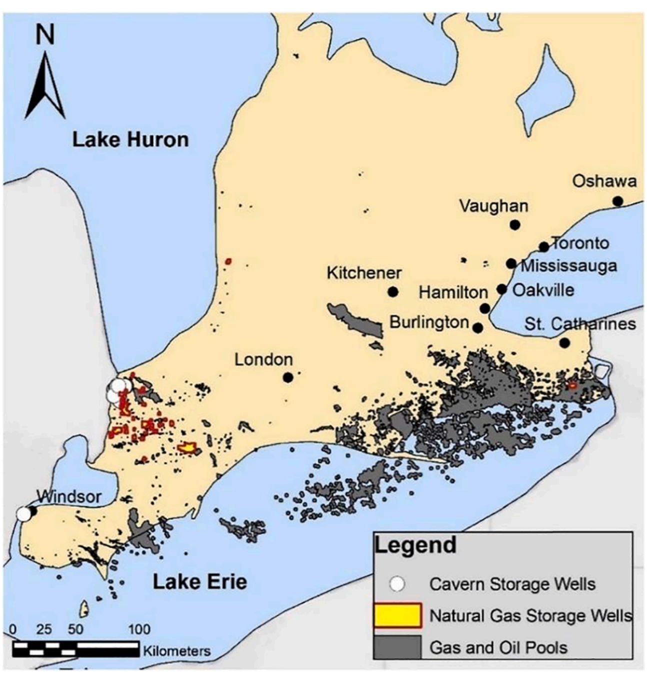


Fig. 7. Geological map for oil and gas pools for natural gas storage and potential UHS in the southwestern part of Ontario, Canada [116].

industries, and power plants in a safe, cost-effective, and long-term manner [14]. The H_2 pathway's most essential milestones must be focused mostly on increased awareness, research, and development programs in examining and investigating the prospects and problems. Furthermore, revisiting natural gas storage facilities will aid in improving the current understanding of UHS operation, as natural gas storage sites are frequently converted to UHS due to similar behaviour

Table 3
Geological division of the principal oil and gas reservoirs in Ontario. Modified from [116].

Geological time scales	Lithology	Depth (m)
Structural and stratigraphic traps in Cambrian	Sandstones and sandy dolomites	800 – 1200
Hydrothermal Ordovician reservoirs	Limestones	600 – 800
Stratigraphic traps in lower Silurian	Sandstones and carbonates	500 – 700
Reefs and structural traps in middle Silurian	Carbonates	350 – 450
Structural traps in fractured Devonian	Dolomitized carbonates and sandstones	110 – 140

and a pre-existing storage mechanism. Though the recent review by Tarkowski et al. [35] theorized that direct extrapolation of results from UGS to UHS should be avoided as CH_4 and H_2 show very different physical and chemical properties, we also hypothesized that while UHS and UGS both undergo a cyclic process (injection/withdrawal) in the subsurface as compared to CO_2 in CCS which is sequestered, a level of comparison between UGS and UHS is potentially possible. However, care must be exercised as the injected H_2 into the underground storage sites can interact with reservoir rocks, caprocks, and reservoir fluids [35,41].

In sum, essential factors and key parameters such as (i) the geological examination of the selected structure and its cap rock; (ii) an examination of its behaviour during the production phase, (i.e., drive mechanism), (iii) the use of mathematical models to dynamically simulate the reservoir behaviour during injection and withdrawal phases; (iv) using alternative dynamic pressure values at the wellhead to determine performance when the reservoir is filled to the initial pressure and beyond; and (v) determination of reservoir performance as a function of number and well type (vertical or horizontal), as well as the type of completion [93,129] from UGS can be used as a support system to effectively understand UHS.

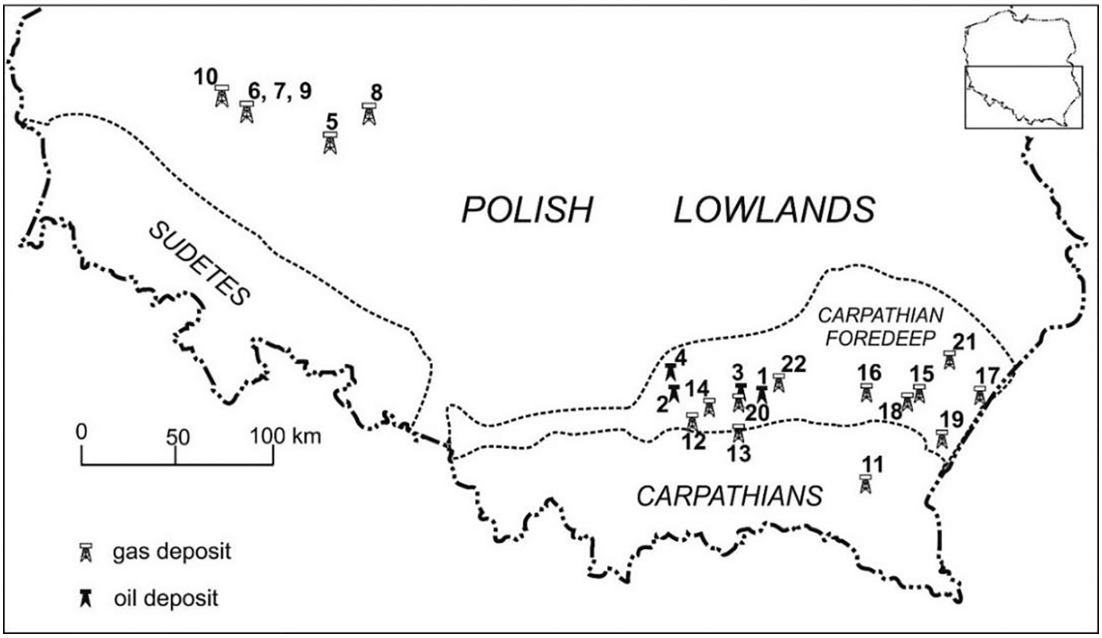


Fig. 8. UHS potential for depleted oil (1 – 4) and gas (5 – 22) deposits [115].

4. Experimental and modeling studies

Generally, black oil, crude oil, and gas are the most common petroleum fluid systems based on the phase diagram and prevailing reservoir conditions (physical properties, composition, gas-oil ratio, appearance, pressure, and temperature) [130]. Of these categories, researchers have recently focused on gas reservoirs which are divided into dry, wet, and condensate gas-bearing formations, as a preferable choice for underground gas storage, especially CO_2 due to the compressive nature [131–133]. This is evident as several studies have been conducted to assess the possibility of injecting CO_2 into dry gas [134–140] and condensate gas reservoirs [132,141–143] for CCS purposes. Findings from these studies reveal that the injection strategy, reservoir features, and operational parameters all play a role in CO_2 storage success. Similar observations have been reported for natural gas storage in depleted gas reservoirs [93,144–148], showing that deliverability, integrity, and reservoir quality significantly impact the underground gas storage efficiency.

In the case of H_2, where a new gas is injected into the subsurface, a similar analogy between natural gas and CO_2 can be extended to understand its behavior in a depleted reservoir scenario. Besides, there have been very few studies evaluating these reservoirs for storage. This could be owing to the limited understanding of important storage properties of these reservoirs, such as injectivity, storage capacity, trapping processes, and containment security. Furthermore, H_2 mixing with a cushion or native gas depends on the reservoir geometry, anisotropy, heterogeneity, injection rate, time, rock, fluid, and rock-fluid properties. Thus, in this review section, literature results on depleted gas reservoirs are discussed, leveraging the following mechanisms presented in Fig. 9. Although research data are limited, aquifer studies are presented for effecting comparison to illustrate the porous media's behaviour better.

4.1. Experimental works

Several reports have shown the importance of thorough geological analysis irrespective of the storage medium (caverns or porous media) due to the potential threats caused by microbes, rock, temperature, salinity, pressure, nutrient, pH, cementing materials, and lots more. In depleted gas reservoirs, some of these launched projects (see **section 3.1**), such as HyStorPor and HyUNDER have studied such scenarios. In the late'70s [149,150], partly theoretical consideration was given to this topic, even at that, it was based on single (mono) mineral phase reaction/interactions with H_2, and the experiment were carried out under room temperature (40 °C) and atmospheric pressure (10 MPa) conditions without paying much attention to the formation fluid composition and microbial interaction effects [151]. Through the collaborative research effort of the H2STORE project, factors such as geo-hydraulic, petrophysical, mineralogical, microbiological, and geochemical interactions induced by the injection of H_2 and H_2-CO_2 mixtures in a depleted gas reservoir have been studied [103,152].

Specifically, Henkel et al. [103] investigated the lithological variation for H_2 storage in a depleted gas reservoir on a siliciclastic rock sample selected from 5 locations (Bravaria, Brandenburg, Lower Saxony, Thuringia, and Saxony Anhalt). The chemical and mineralogical composition of the selected samples were analyzed by conducting laboratory analyses, such as XRF, ICP-OES, SEM, XRD, and PLM. Special attention was given to the pore-exposed grain surface as they have great potential for an in-situ reaction after long-term storage. Based on the microscopic analysis, the selected samples were composed of subarkoses to litharenites, which implies a high variability in the pore-filling cement and porosity. Each sample was found to have unique rock compaction, which therefore suggests different pore spaces for H_2 storage [103]. Further, carbonate minerals, anhydrite, quartz, and feldspar were highlighted as the major pore-filling cementing materials. Results from the petrophysical study gave a variation in samples based on location. It confirms that authigenic clay minerals in Saxony-Anhalt minimize porosity and permeability, thus, higher H_2 storage containment. In conclusion, the authors reveal that rock-fluid interaction is almost absent in locations where the pore spaces are filled with blocky cementing material (anhydrite, quartz), and it entirely depends on the permeability, clay minerals, as well as their surface morphology [103].

In a comprehensive study, Henkel et al. [152] investigated 670 siliciclastic reservoirs (sandstones and clay/silt caprock) from 25 well sites in Germany and Austria using different analytical methods. In this case, mineral reactions were studied systematically before and after 3 % H_2 and 97 % CO_2 injection in the depleted gas reservoir at UHS conditions (T = 120 °C, P = 20 MPa, and formation salinity = 35 %). The extent of mineral dissolution and precipitation, their impact on the reservoir properties, and microbial metabolism triggered by H_2/CO_2 exposure

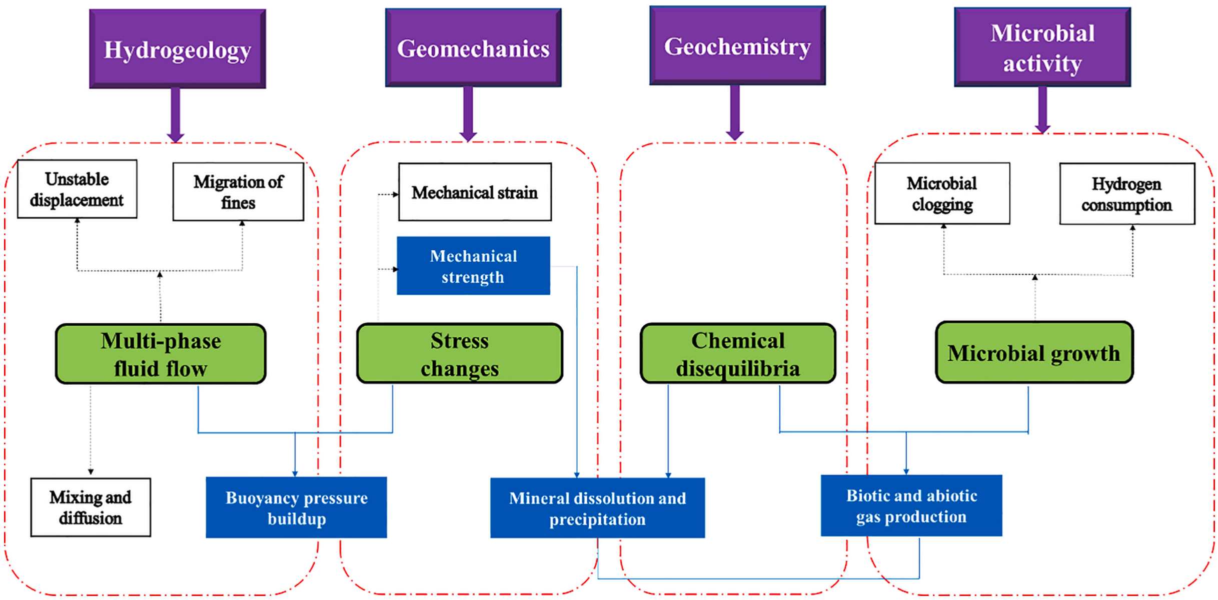


Fig. 9. Factors affecting the efficiency of H_2 storage in porous media. Modified after [8].

were highlighted. Based on geochemical, petrography, µCT, and FE-SEM analysis, it was found that CO_2 exposure led to the dissolution of the pore-filling calcite cement, which led to the formation of pores within the sample for underground storage, thus increasing permeability. The result also corresponds with the BET analysis, as the specific surface area in the Permian samples increased after the CO_2 autoclave batch experiment. Although the batch experiment was conducted using CO_2, the authors suggest that the same outcome will be achieved for H_2 injection [152]. The recent study by Shi et al. [153] also validates the findings of Henkel et al. [152] regarding mineral dissolution, who conducted similar tests using pure CO_2 injection in Mount Simon sandstone.

Based on the availability of depleted natural gas reservoirs and the low capital investment associated with them during conversion to H_2 storage sites, the effects of blending natural gas and H_2 in a depleted oil and gas reservoir were investigated again by Shi et al. [112]. Emphasis on rock types and relevant gas mixtures and their adverse impact on reservoir integrity and storage were investigated, as the selected core and cement samples (from California natural gas storage formation) were characterized based on permeability, porosity, surface area, mineralogy, and other structural characteristics prior and after incubation (3-month durations). Specifically, 11 cylindrical core plugs (7 extracted from the storage reservoir zone and four from caprock) with a cement sample (which served as a representative cementing material used for production casing at the storage site) at different depths were used for this experiment. The authors discovered that before sample exposure to H_2 and natural gas mixture, there was an increase in permeability of the caprock (e.g., sample #2) and reservoir rock (e.g., sample #4) from the first set of experiments, based on permeability measurements. This was attributed to the pre-existence of micro-fractures within the samples [112]. However, when exposed to H_2 and natural gas mixtures, the permeability of the caprock samples decreased, and the reservoir rock moderately increased. These changes in permeability (before and after exposure) were found to be controlled by convection (due to macropores or fractures) rather than Knudsen diffusion (due to micro and mesopores). In the second batch of the experiment, a similar finding was made, though to a lesser amount [112]. Because the presence of native fluids (as determined by the technique employed in the second batch) prevents the loss in permeability as the pore throat was blocked; thus, the adverse effects of the rock/gas interaction are lessened. Porosity measurements in caprocks and reservoir rocks were also reported to be similar (before and after incubation). The influence on permeability, on the other hand, was more significant. Furthermore, the rock mineralogy and the pore size distribution (based on BET analysis) of the samples (from the second batch) changed due to the interaction between the rock and fluids [112].

In the case of the polymeric samples used as wellhead seal material representatives, an increase in diameter after incubation was reported (especially on the elastomer materials compared to the Teflon). This observation was attributed to gas absorption in the elastomer samples, making them swell. The authors further recommended that the impact of swelling (mechanical strength of the different polymeric cementing materials) due to the H_2/natural gas mixture needs further study [112]. This is because the swelling of gasket materials due to H_2 exposure was previously reported [154] and it was hypothesized that the H_2 molecule is likely to penetrate via diffusion through the gasket materials (and could dissolve at higher pressure). This swelling process is explained as explosive decompression failure (XDF) or blister fracture [154]. It was found that when the H_2 is decompressed quickly enough before the sample is removed from the incubator, diffusion in the gaskets cannot keep up, and the gaskets become oversaturated with H_2. As a result, the rapid release of H_2 from the materials could result in void defects inside the material, which could contribute to swelling. Moreover, since Shi et al. [112] also observed substantial changes when the elastomer materials were immersed in natural gas, XDF might be possible, especially during long storage periods and fast rates of gas depletion.

Flesch et al. also evaluated the effect of exposing rock samples (obtained from Tertiary, Triassic, and Permian strata; containing siltstones [massive and laminated] and sandstones [massive, laminated, and fine layered]) to a mixture of H_2 and brine from three different probable H_2 storage sites within the stratigraphic units in Germany and Austria. The samples were immersed in 100 percent H_2 and aged for up to six weeks in a static batch autoclave experimental setup under various relevant reservoir conditions for petrophysical assessment [151]. The characterization (i.e., microscopic, petrophysical, petrographic, and computer tomography analyses) before and after exposure to the H_2 and brine mixture revealed that the Tertiary sandstones showed no chemical reactions. In contrast, the Permian and Triassic sandstones showed notable changes in permeability (ranging from 60.5 % to 38.5 %) and porosity (ranging from 56 % to 107.8 %). These changes were linked to pore-filling anhydrite, carbonate cement, and scattered carbonate ooids in the Permian and Triassic sandstone samples, Fig. 10 [151]. In conclusion, the preliminary results obtained by [151] from the different

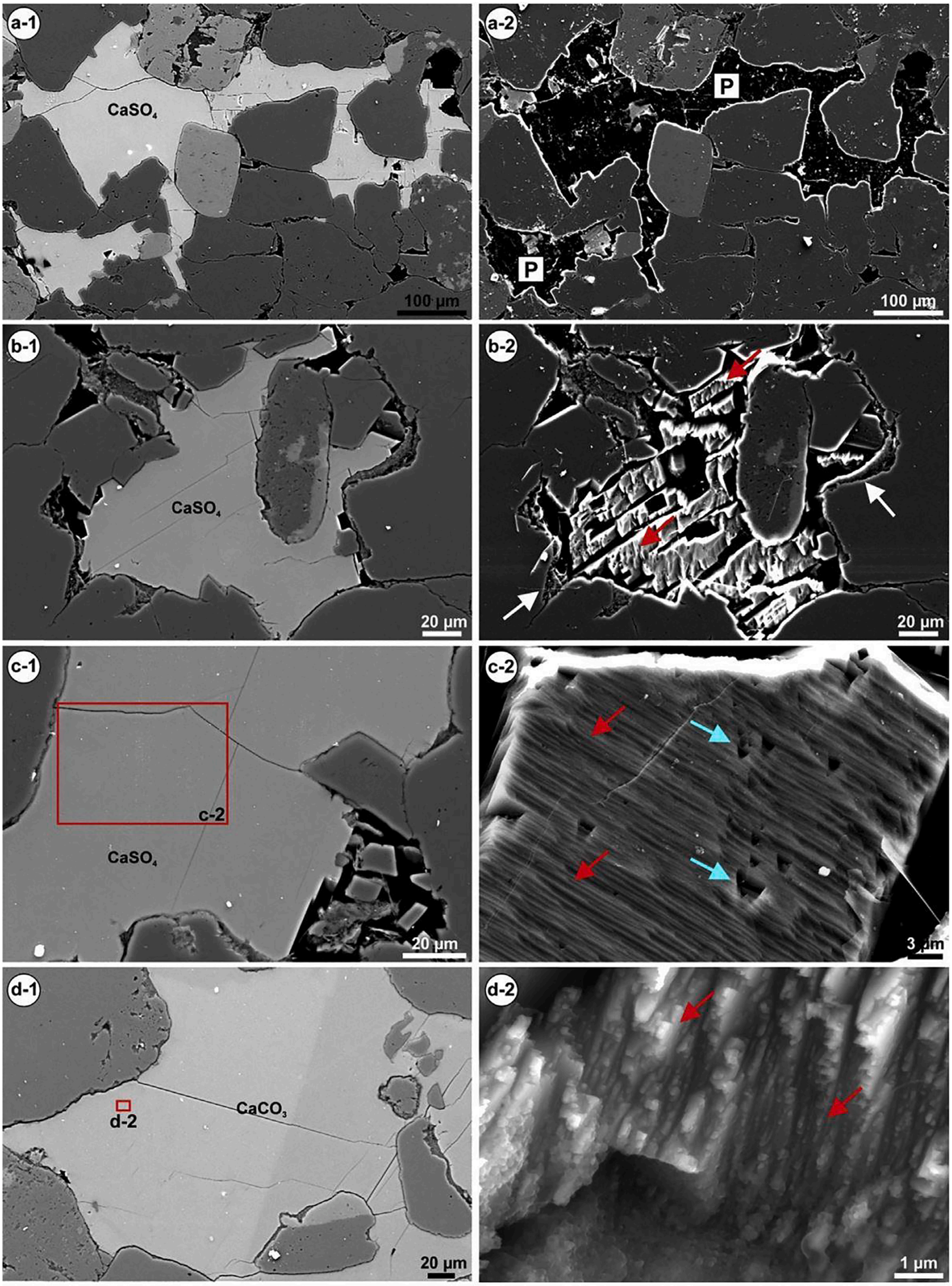


Fig. 10. SEM images of the Permian sandstone thin section displaying the effect of pore-filling anhydrite ($CaSO_4$) and carbonate ($CaCO_3$) cement. Pictures on the left (a-1, b-1, c-1, and d-1) represent before H_2 injection, while a-2, b-2, c-2, and d-2 on the right denote after injection. As can be seen from the high-resolution images, a total $CaSO_4$ dissolution was observed, leading to the creation of open-pore space for storage (a-1 vs a-2) and (b-1 vs b-2). In the case of c-1 vs c-2, the effect of cement alteration led to the formation of triangular (pyramidal) edge pit shapes and staircase structures. In contrast, the dissolution of carbonate cement (d-1 vs d-2) altered the formed staircase structures. All these points toward the potential storage capacity of the selected reservoir formations and the effect of H_2 injection as more open pore structures were formed, enabling H_2 storage [151].

dimensions (2D and 3D) and the measurement scales (mm to cm) of the 21 samples show the potential for H_2 storage.

The primary observation of the changes in permeability by Flesch et al. [151] and Shi et al. [112] was attributed to both mineral dissolution and precipitation. For instance, the decrease in permeability (from caprock samples) observed in the second batch of the experiment by Shi et al. [112] where the core sample was allowed to maintain its native fluid or, in some cases, was saturated with oil/brine was attributed to the precipitation of new minerals that led to pore throat blockage during incubation. On the other hand, the increase in permeability was caused mainly by mineral dissolution because of the presence of additional brine and oil.

Pilot studies from the underground sun storage project in Austria [105] and the Hychico project in Argentina [106] also reported some analysis of depleted oil and gas reservoirs. The Sun project of Austria, led by RAG Austria (the most prominent gas operator and thus energy storage company in Austria), has assessed the blending of 10 to 20 % H_2 (green H_2) and 80 % CH_4 within a small, depleted gas reservoir in the Molasse Basin. While the outcome successfully highlights the possibility of H_2 storage to a certain degree, no possible evidence of migration (H_2 loss) within the reservoir was documented, and there was no detrimental impact from the current natural gas storage facility [105]. This again supports the clear coexistence and suggestion for using natural gas storage facilities for H_2. In conclusion, the authors reported that the sun storage project is intended to serve as a fundamental for a further 'underground sun conversion' to achieve large-scale storage of solar energy in the form of H_2 in underground gas reservoirs.

Alternatively, the Diadema-led project by Hychico proposes to produce green methane by tapping on any methanogenesis that may occur within the reservoir [114]. Though H_2 storage is not the primary focus, kinetic rates and H_2 displacement could provide valuable insight into other parameters, such as reservoir temperature, pH, salinity, and organic substance, which were investigated [114]. Findings reveal that the Glauconitic sandstone gas reservoir overlayed by clays is a good candidate for H_2 storage and green methane production. Even though Hychico recorded chemical, microbial, as well as alterations in the mechanical properties due to in-situ fluid interaction with H_2 as the potential challenge [106,114], the long-term goal is aimed at providing a future regional and international market with green H_2 production from renewable sources, as well as green methane, by using H_2 as raw material.

4.2. Modelling works

Several methods, including laboratory measurements and numerical simulations, can forecast multiphase flow parameters in reservoirs. Experimentation is costly and time-consuming and can only be utilized on limited sample numbers and conditions. On the other hand, numerical modelling and simulations are required to augment laboratory tests for various studies and sensitivity analyses. A wide choice of numerical tools is employed for modelling the transport processes in UHS. Even though the application is based on process types, these tools represent commercial petroleum software packages and combined commercial and scientific packages to open-source codes. In addition, the application of these tools depends on the affecting properties that are to be studied during UHS projects, as each tool has certain limitations, as presented in Table 4.

4.3. 1 Hydrodynamics of hydrogen storage

Many factors and mechanisms, particularly hydrodynamics, affect the ultimate storage of H_2. Often, the mass conservation equation for two-phase fluid: liquid (l) and gas (g), is used with the help of the tools and codes to study the potential of H_2 storage, **Eq.** (4).

$$q^k = \phi \frac{\partial(\rho_w c_w^k S_w + \rho_g c_g^k S_g)}{\partial t} - \nabla\cdot\left(\rho_w c_w^k \nu_g + J_g^k + \rho_g c_g^k \nu_w + J_w^k\right) \quad (4)$$

where ϕ is the porosity, ρ is the molar density in (mol/m^3), c is the molar concentration, S is the saturation, ν is the advective volumetric velocity in (m/s), and J is the diffusive/dispersive flux in (mol/(sm^2)), q^k the source of the sink term (mol/(sm^3)) and the subscripts g and w denote the gas and water phase, respectively, and the superscript k represents the chemical component.

As previously discussed in **section 2**, H_2 physicochemical properties influence the dynamics of H_2 storage (e.g. Mahdi et al. [160]). For instance, the low density and viscosity of H_2 often result in high mobility, leading to H_2 losses due to unstable displacement. Controlling parameters such as viscous fingering and gravity override are pertinent to most underground gas storage operations as they affect storage efficiency. Paterson observed that structural geometry could influence the nature of these controlling parameters [161]. They also observed that the viscous fingering could spread within the lateral geometry of the reservoir beyond the spill point, which can only be controlled by reducing the injection rate. Between both controlling parameters, it was further observed that the gravitational forces (due to density difference, low injection rate, and large vertical permeabilities) are more dominant at a slow injection rate. Thus, a stable front can be achieved. In contrast, a fast injection rate promotes viscous fingering as the less viscous fluid (H_2 gas) moves ahead, the more viscous (brine/native gas) fluid; thus, a high unstable displacement was recorded [161]. As an extension, Hagemann et al. [162] studied the effect of unstable displacement reported by [161]. Through selective technology, Fig. 11, the authors addressed the lateral spread of injected gas beyond the spill point. Even though such a problem can be addressed by injecting H_2 at the bottom of the structure, it was observed that such an injection strategy could only

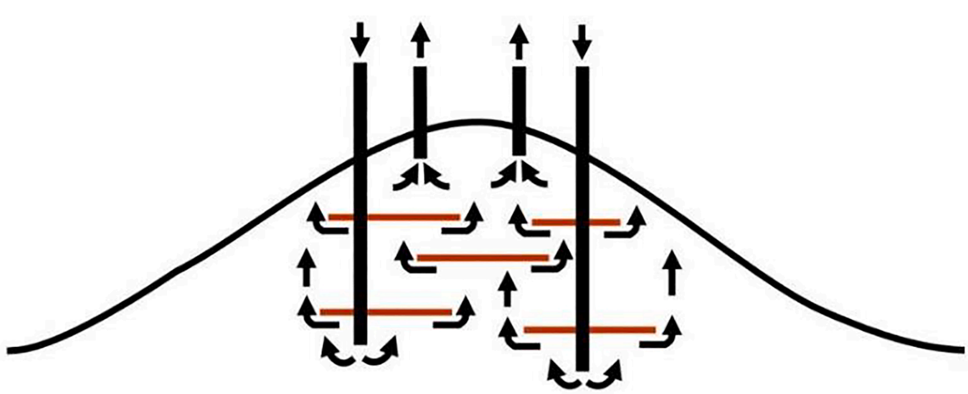


Fig. 11. Selective technology schematics. The red lines denote low permeable barriers, whereas arrows indicate gas flow direction [162]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

delay the lateral spreading until the gas gets to the cap rock. However, the proposed selective technology in prospect can be used to reduce the effect of gas rising during UHS projects.

As shown in Fig. 11, this technology is a two-well system (vertically heterogeneous) that adopts a conceptual highly permeable and porous sandstone reservoir interrupted by low-permeable formations such as shale and mudstone intercalations. During gas injection, H_2 is pumped by one vertical well system down to the bottom, which eventually rises due to buoyancy force. However, as the H_2 gas encounters the low permeable layers (shale and mudstone), which serve as a barrier, the migration is delayed until the gas penetrates or flows across the obstacles. Consequently, on reaching the reservoir top (caprock), the other well systems are used to withdraw the H_2. In comparison, this technology looks promising as it can minimize the lateral spreading of H_2 [162]. Implementation of this system still requires detailed planning in terms of (i) injection/withdrawal rate, (ii) gas rise time, and (iii) availability of a suitable storage medium with such properties. Though analytical simulation from their work was used to infer the exact gas rising velocity and its effect, numerical modelling via COMSOL showed the impact of barrier length (as longer length slows the penetration rate). This technology will require more understanding before implementation [162]. This technology was further assessed by [96], and it was found that despite the extra perforation in the uppermost layer, H_2 accumulation in the uppermost perforation was due to horizontal propagation and not vertical migration along the well as such, the selective technology by [162] to address unstable displacement is quite complex.

In a similar vein, an extension to Hagemann et al.'s work based on isotropic and homogenous (structured) conditions in a 2D reservoir was conducted by Feldmann et al. [48]. For better realization, the authors used a 3D domain to study the hydrodynamic properties of H_2 and gas mixing in heterogeneous (unstructured) depleted gas reservoir conditions. Interestingly, no noticeable viscous fingering as compared to the aquifer study by [162] was observed. This was due to the less displacement effect between gas–gas and gas–water mixture as it was reported that the impact of gravity override is a function of density contrast. Notably, at an initial reservoir condition (P = 170 bar; and T = 125 °C), H_2 was less dense than methane and nitrogen by 75 kg/m^3 and 125 kg/m^3, respectively. This phenomenon is illustrated in Fig. 12, as H_2 in a gas–gas system had less gravity effect; Fig. 12**(a)** as compared to the gas–water system in Fig. 12**(b)**, where clear separation due to gravity can be observed. Also, it was found that the injection rate contributes significantly to the effect of mixing as low injection/extraction rate limits H_2 mixing in depleted gas conditions. In addition, they found that H_2 injection resulted in a highly saturated and homogenous H_2 plume in the near well area, whereas H_2 purity in the extracted gas mixture increased with an increasing number of withdrawal cycles [48].

In another study, Pfeiffer and Bauer [47] chose N_2 as a cushion gas because it is reasonably inert to chemical reactions and relatively inexpensive to test the feasibility of H_2 storage in Schleswig-Holstein, Germany, for a week. Simulation studies reveal that such formation can store a large amount of energy in the form of H_2. The simulated storage could deliver 20 % more of the total energy demand at the final storage cycles. However, a major limitation was observed with the injection scheme. It was reported that well 3, which had the worst amount of H_2 in the first cycle, improved much more towards the final storage cycle. This was attributed to the optimized injection scheme, as they noted that including shut-in periods during the initial filling of the reservoir to decrease the pressure level may improve the well injectivity subsequently [47]. Sainz-Garcia et al. [99] also conducted a 3D numerical simulation from a sandstone formation (aquifer condition) in the San Pedro Belt of Spain to further understand the influence of different well configurations. Their finding corroborated that of [48] where maximum H_2 saturation appeared near the well area close to the reservoir top and the absence of viscous fingering, which they attributed to the steepness of the reservoir structure.

While depleted gas conditions (e.g., [48]) observed no fingering, the work by [99] yet, reported the absence of viscous fingers in an aquifer setting. Thus, the structural geometry of the reservoir significantly impacts the effect of fingering. About 78 % maximum recovery (in the best case) was observed even though it was at the expense of water up-coning [99]. The overall conclusion was that steeply dipping structures will make a suitable geometry for H_2 storage and using H_2 as a cushion gas (compared to nitrogen) can limit the effect of gas mixing during injection, even though it is prone to up coning. Pfeiffer et al. also corroborated the steeply dipping structure reported by [99], as their sensitivity analysis shows that depth, low permeability, dipping reservoir structure, and low pressure ultimately favour H_2 extraction [163] since, after four years of operation, it was found that the H_2 volume in place and the H_2 volume produced rose steadily with each storage cycle, with just 1.75 % of the H_2 lost by dissolution into the resident brine.

In a different study, N_2 gas was employed as a cushion gas to examine the possibilities of H_2 storage in a saline aquifer in the North German Basin [46]. Although the authors observed an influx of H_2 accumulation near the well area due to gravitational effects, nitrogen gas remained laterally spread within the reservoir for pressure maintenance. Further investigation showed that while H_2 recovery increases with the withdrawal cycle, as with many studies [46,47,163,164], the cushion gas injection pattern may improve the storage efficiency. The importance of injection rate in a depleted gas was investigated using Dumu^x open source code to compare the hydrodynamics between natural gas and H_2 [165]. Despite the different scenarios (low injection, medium injection, and high injection rates), it was concluded that H_2 spread laterally faster due to its unfavourable mobility ratio due to the low dynamic viscosity of H_2, which limits the withdrawal rate. In a recent study, seasonal H_2

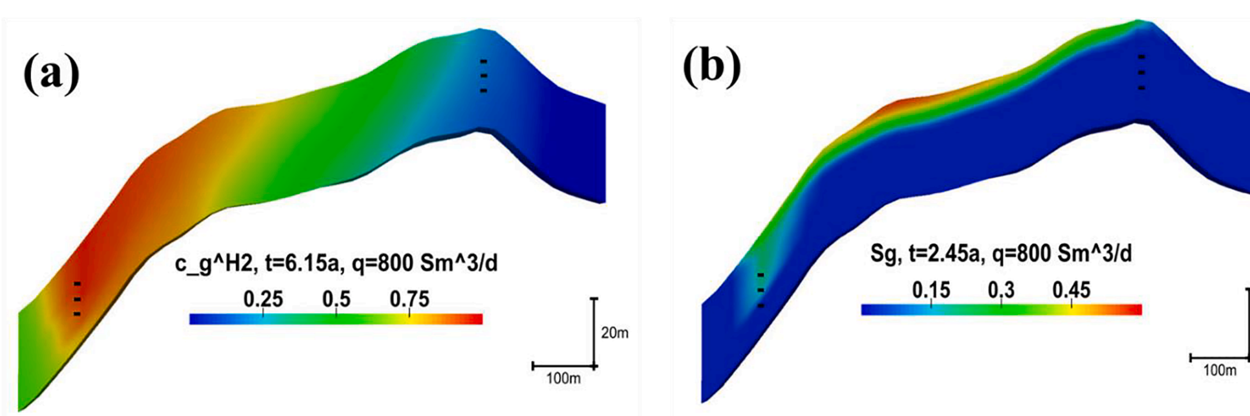


Fig. 12. Effect of gravity override between (a) gas–gas and (b) gas–water systems. **Note:** (a) represents H_2 concentration in an H_2-gas displacement while (b) denotes gas saturation in an H_2-water displacement [48].

storage in an aquifer was assessed using an existing anticline structure in Poland [100]. Findings suggest the presence of water up-coning after H_2 injection as the H_2 accumulated along the reservoir top. More so, it was observed that a high permeable layered zone favoured H_2 injection. Additionally, the authors found the absence of viscous fingering even though the aquifer structure was steeply dipping.

Lysyy et al. [96] also studied the seasonal storage of H_2 in a depleted oil and gas field in Norne, Norway. Through a 3D numerical simulation using Eclipse 100, a systematic investigation involving storage site initialization, four annual cyclicities, and the prolonged withdrawal period was performed. The first case study investigated the possibility of 100 % H_2 storage in well C-3H. In contrast, the second case study investigated the effect of various parameters, such as cushion gas, injected gas composition, and structural geometry. Based on the storage scheme examined (gas, oil, and water zones) for pure H_2 storage, the thin gas zone (the best) gave a recovery factor of 87 %. Furthermore, the distribution of H_2 in the subsurface was consistent with the geological structure of the field but was restricted by fluid saturation and displacement efficiency [96]. The other case studies showed that pre-injection of natural gas would increase the ultimate recovery rate for H_2 though at the expense of its purity [96]. Lastly, the authors recommended further investigation on the percentage injection of gas mixtures with lower H_2 content because their study (70 % natural gas and 30 % H_2) found a varying H_2 fraction during H_2 delivery [96].

4.3.1. Hydrogeochemical and microbial effects of hydrogen storage

As discussed in the previous section, H_2 loss has been attributed to several parameters, such as diffusivity, mineral dissolution, precipitation, gas mixing (blending), fingering, and gravity segregation. However, the effect of microbial activities in the reservoir can also affect the overall storage capacity of the injected gas, especially when considering a depleted reservoir meant for seasonal storage.

Hemme and van Berk [110] used a 1D reactive mass transport (1DRMT) model to investigate H_2 loss due to microbial effect, a chemical reaction between the reservoir and over/underlying seals, and diffusion. The authors reveal that the loss of H_2 by bacterial conversion to CH_4 (methanogens) and H_2S (sulfate-reducing bacteria) depends not only on the active amount of electron receptors (CO_2 and $SO4^{2-}$) available for a reaction but also on the amount of co-injected gas, duration of H_2 storage as well as the kinetic rate constant. The authors [110] further reveal that the contacting region between the reservoir rock and caprock (top) and that between the reservoir rock and underlying rock (bottom) were the primary hotspot for H_2 loss due to diffusion, as they served as a permeable medium for CO_2 and SO_4^{2-} which promotes (H_2S and CH_4 formation) further loss of H_2. Although, dissolution within the carbonate in the caprock and anhydrite in the bottom seal can also increase the loss of H_2. They also reported that H_2 loss in a depleted gas field could increase reservoir porosity, which can cause losses via leakage, especially in a fractured reservoir. For instance, an increase in the cap rock porosity would decrease the sealing capacity and increase the risk of pathways for the stored H_2 to reach the overlying aquifers. In contrast, a decrease in the cap rock porosity would increase its sealing capacity [110]. This finding thus indicates that an increase in dissolution rate (which can occur both at the reservoir and cap rock level) over precipitation rate can compromise the caprock integrity. Thus, H_2 loss by leaking via increased porosity and permeability, whereas when the precipitation rate is more significant than the dissolution rate, the caprock integrity may still be intact. Still, the porosity and permeability near the wellbore region may be jeopardized, which could further affect the ultimate deliverability (injection/withdrawal) of the UHS operation [24].

A geochemical study by Hassannayebi et al. investigated the mineral interaction in a gas brine system to assess the extent of H_2 loss in the Molasse Basin, Austria [159]. The equilibrium approach was used to estimate the long-term effect of H_2 storage, whereas kinetic modelling investigated the interaction between H_2 and formation over a typical storage cycle condition. They discovered that the pyrite reduction into pyrrhotite resulted in a considerable increase in reservoir pH, as well as H_2 loss. In another study, Amid et al. [12] investigated several mineral assemblages using PHREEQC on the effect of the chemical stability of H_2 in the reservoir. Specifically, three different scenario assemblages were investigated, i.e., sandstone and iron mixture, sulfur, and carbonates. The first scenario was chosen since North Sea reservoirs are predominantly clay-bearing sandstone with iron oxide cement [166,167]. The second scenario was examined based on the usual presence of H_2S in natural gas. The third scenario was based on the Sabatier reaction, where CO_2 reacts with H_2 to produce methane and water. Regardless of the overall investigations, the clay-bearing sandstone and iron mixtures were found to be stable under the reservoir conditions (temperature: 365 K; and pressure: 5 – 10 MPa), while sulfur and carbonate assemblages were not suggesting that H_2 conversion is a potential challenge in the rough gas storage facility (RGSF). Furthermore, they found that leakage within the reservoir aquifer and overlaying caprock for the modelled RGSF amounted to a total of 0.035 % (0.029 % in the aquifer and 0.006 % in caprock) after 12 months based on the reservoir temperature and pressure [168].

Generally, it is expected that the solubility of gas is highly dependent on the reservoir temperature, pressure, and salinity [12,169]. However, since the estimated H_2 solubility assumed the water as pure (no effect of salinity), such percentage of H_2 loss from the aquifer and caprock is expected to be overestimated; only 0.1 % of H_2 was lost due to dissolution and diffusion [12]. A general observation confirms that the leakage (or loss of H_2) via microbial archaea is more likely to put the reservoir at risk as its percentage loss is higher. A maximum H_2 loss of not more than 3.7 % of the entire H_2 injected due to conversion to methane and biomass throughout the storage was observed, which was considerably higher than 0.1 % [12]. This is expected as the amount of CO_2 present in the liquid and gas phase has phased out, whereas the concentration of Ca^{2+} ions (0.26 mol/L) has already reached equilibrium; as a result, no more H_2 consumption can be achieved. While methanogens and Sabatier bacteria are less sensitive to the reservoir, sulfate-reducing bacteria can grow within the residual hydrocarbon, potentially contaminating the gas with H_2S. Moreover, many of the Southern North Sea reservoirs are at higher temperatures, and as such, hyperthermophilic sulfate-reducing bacteria (SRB) cannot be underestimated as they accommodate temperatures well beyond 100 °C [170]. Furthermore, because Southern North Sea reservoirs are predominantly gypsum or anhydrite-based mineral cement, which encourages microbe habitation, caution should be taken during reservoir selection, as lithologies with little sulfur or that are too hot to sustain sulfate-reducing bacteria will serve as a better storage option within the RGSF.

Microbes could be in the form of methanogens, acetogens, sulfur-reducing, or iron-reducing organisms [37]. However, particular microbes such as temperature-loving (thermophilic) and salt-loving (halophilic) found in naturally occurring high-salt environments, such as salt-lakes or salt-ponds, are likely to withstand harsh temperature, pressure, and salinity in the reservoir during the metabolism of H_2 [37,171]. Hence, investigating the potential damages of each possible microbe is of great importance before H_2 injection. This effect is reported by [172] using the microbial growth rate and microbial population density. It was found that some microbes can metabolize the injected H_2, which could affect the H_2 content, thus increasing energy loss. Again, they found that the extent of the metabolism is a factor of the H_2 percentage in the gas mixture and is likely to increase further after a few cycles. A similar observation was also reported by [173] when the impact of methanotrophs on hydrogenotrophs in UHS was analyzed numerically. Recent work by [38] reviewed three H_2-consuming processes (methanogenesis, homoacetogenesis, and sulfate reduction) in the subsurface to enable the extent of microbial growth during H_2 storage. Based on the 42 depleted oil and gas fields and the data on the H_2 storage tests sites, the authors documented that five fields may be considered sterile to H_2-consuming bacteria at $> 122\ ^oC$ [38]. In

Table 4
Summary of key attributes of popular numerical tools for modelling UHS transports. The data presented were modified from [155].

Tools/codes	Spatial discretization*	Time discretization*	Remarks
Eclipse 100	FDM	Implicit	• This is a commercial software package used for the black oil model. • Uses an empirical model. • Fluid properties estimation. • Capable of modelling flow in oil and gas reservoirs with up to three phases. • Excellent history matching potential [96]. • Cannot be used for physical and biological processes such as mechanical dispersion, microbial growth, and a few bio-chemical reactions.
Eclipse 300	FDM	Implicit	• It is a commercial software package capable of conducting compositional modelling and can be adjusted for UHS studies. • Uses EOS and feed-forward calculation for modelling. • A similar limitation with E100 • Despite its usage for H_2 storage [46], it has poor performance for history matching [96,156].
CMG (IMEX, GEM and STARS)	FDM	Implicit	• Robust commercial software with an excellent graphical user interface. • Can model multiphase flow molecular diffusion, mechanical dispersion, chemical reaction, solid mechanics, and non-isothermal flow.
COMSOL	FEM	Implicit	• Commercial Multiphysics solver with an efficient physics-based graphical user interface that simulates physical and chemical processes for different disciplines. • The limitation is primarily attributable to the suitability of the discretization method implemented for fluid flow problems. • Its physical models include compositional multi-phase flow, molecular diffusion, chemical reactions, non-isothermal flow, and solid mechanics.
OPM	FVM	BE	• OPM, i.e., (Open Porous Media) is an open-source code developed for transport process simulation, especially problems related to petroleum reservoirs. • It is based on the C++ programming language. • It can model multi-phase flow, molecular diffusion, and non-isothermal conditions.
DuMu^x [157]	FVM	BE	• Open-source code written in C++ program language for flow and transport processes in porous media. • This FVM-based software can simulate multi-phase flow, molecular diffusion, mechanical dispersion, non-isothermal flow, and solid mechanics. • It is often used for ground water and hydrology studies, just like OpenGeoSys.
OpenGeoSys [158]	FEM	BE	• This open-source tool is written in C++ programming language. It allows the simulation of complex flows involving molecular diffusion, mechanical dispersion, chemical reactions, biomass growth and decay, non-isothermal flow, and solid mechanics. • Unlike OPM, OpenGeoSys and DuMU^x are often used for ground water and hydrological applications. • It differs from DuMu^x based on its method of discretization. • All these open-source codes (OPM, DuMux, and OpenGeoSys) can be adjusted for bio-reactive modelling in UHS.
TOUGHREACT	FVM	BE/CD	• This commercial tool is based on the Fortran programming language used for multiphase process modelling in porous and fracture media. • It can be used for multi-phase flow simulations, molecular diffusion, chemical reactions, non-isothermal flow, and biomass growth and decay modelling. • Physical and biological process modelling is often integrated into the available source code for UHS studies.
Phreeqc [12]	FDM	**n/a**	• Aqueous geochemical modelling. It can be used to simulate (i) speciation and saturation index calculations, (ii) inverse modelling, and (iii) batch reaction and 1DS-transport calculation [110].
GWB [159]	FDM	Implicit	• Often used for kinetic rate law studies for mineral dissolution and precipitation reactions, redox transformation, 1 and 2D reactive transport, and bio-reactive and colloidal transport.

*__FDM__ - Finite Difference Method, **FEM** - Finite Element Method, **FVM** - Finite Volume Method, **BE** - Backward Euler, **CD** - Central Difference, **IMPES** - Implicit Pressure and Explicit Saturation.
Note: *The remarks presented herein are solely based on the present author's surveys of the user experiences in the versions of the tools mentioned above (as of the time of this review) in the scientific literature and authors' experiences. These remarks should neither be considered an endorsement of any product nor encourage/discourage its usage.*

contrast, six fields were found to have the capacity to sustain all the H_2-consuming processes based on either temperature, salinity, or pressure constraints [38]. The outcome of this work shows that H_2 properties have a significant impact during storage and the type of facility also determines the potential development of microbial activities since each facility has a different characteristic.

A summary overview of the presented result from the numerical studies discussed herein (and more) involving hydrodynamics, geochemical, and microbial effects in the porous medium is shown in Table 5**.**

4.3.1.1. Effect of kinetic rate constant. The kinetic rate constants of reactions also affect H_2 geo-storage. As reported by [110]**,** who used an initial kinetic rate constant of $9.26 \times 10^{-8}\,\mathrm{mol\,kgw^{-1}s^{-1}}$ at 30 °C [175] for SRB and $2.30 \times 10^{-9}\,\mathrm{mol\,kgw^{-1}s^{-1}}$ at 37 °C [176] for methanogens observed that, by increasing the rate constant slightly to $2.40 \times 10^{-7}\,\mathrm{mol\,kgw^{-1}s^{-1}}$ for SRB, the total stored H_2 is lost within 20 years of the reference simulation condition, Fig. 13**(a).** Albeit generated H_2S amount remained constant as the reactive anhydrite within the reservoir rock was entirely consumed, and the only sulfate source will be the region (hotspot) between the reservoir rock and the underlying rock via diffusion, which is considered as a very slow process. Consequently, if the kinetic rate ($2.40 \times 10^{-7}\,\mathrm{mol\,kgw^{-1}s^{-1}}$) is maintained, and the storage period is increased from 30 to 300 years. The effect of H_2 loss due to diffusion becomes significant as more sulfate will diffuse through the region. In contrast to H_2S, which was not generated, the amount of CH_4 increased from 0.2 to 0.35 mol/L (averagely) after 30 years. This was so as methanogenesis immediately commenced after SRB became fully consumed. This was further supported by the fact that since sulfate was consumed faster due to the increased kinetic rate constant (from 9.26×10^{-8} to $2.40 \times 10^{-7}\,\mathrm{mol\,kgw^{-1}s^{-1}}$), the timeframe for methanogenesis becomes larger. Consequently, when the SRB rate constant was decreased to $9.26 \times 10^{-9}\,\mathrm{mol\,kgw^{-1}s^{-1}}$, it remained constant as no H_2S was generated within the reservoir rock, Fig. 13**(b).** However, CH_4 was generated, even though it was less than when the rate constant was increased.

Table 5
Summary of selected simulation studies on H_2 storage in porous media.

Research focus	Software tools/ codes	Model type	Cushion gas	Storage medium	Storage duration	Approach	References
Technical feasibility for H_2 storage.	• Eclipse 300	2D	N_2	Saline aquifer	Short term	• K_r and P_c from Brooks and Corey's relations • Peng-Robinson EOS for property modelling	[47]
Lateral displacement and injectivity effect of H_2 in an isotropic and homogenous (structured) condition.	• DuMu^x	2D	H_2 and CH_4	Aquifer	Short term	• K_r and P_c from Brooks and Corey's relations • Darcy's law • Thermodynamics assumptions • Ideal gas law • Wilke correlations • Box method spatial discretization	[162]
Hydrodynamics and gas mixing in heterogeneous unstructured) reservoir condition.	• DuMu^x • COMSOL • Petrel	3D	CH_4 and N_2	Depleted gas	Long term	• Peaceman's model (for automatic adjustment of injection/production rate) • Molecular diffusion • Mechanical dispersion	[48]
Geochemical investigation and storage potential for Rough Gas Storage Facility.	• PHREEQC	-	H_2 and CH_4	Depleted gas	Short term	• Volumetric analysis • Chemical stability study • Biological consumption of hydrogen in the reservoir • Leakage study	[12]
Storage and geophysical study of a synthetic heterogeneous reservoir.	• Eclipse 300	2D	N_2	Saline aquifer	Short term	• K_r and P_c from Brooks and Corey's relations • Peng-Robinson EOS for property modelling • Geophysical modelling (EWI, ERT, and Gravity technique) for site monitoring	[163]
Coupled hydrodynamic and biochemical investigation	• DuMu^x	2D	CH_4 and CO_2	Depleted gas	-	• Darcy's law • K_r and P_c from Brooks and Corey's relations • Molecular diffusion • Mechanical dispersion • Microbial modelling • Spatial discretization (Box method)	[165]
Effect of dimensioning and induced hydraulic fracturing during H_2 storage	• Eclipse 300	2D	N_2	Saline aquifer	Short term	• K_r and P_c from Brooks and Corey's relations • Penge Robinson EOS	[46]
Hydrodynamicstudy	• COMSOL	3D	H_2	Saline aquifer	Long term	• K_r and P_c obtained from Brooks and Corey relations from typical literature due to the absence of site-specific data • Gas density obtained using PR EOS • Other properties of gas and obtained via an inbuilt COMSOL state equation • Aquifer geometry obtained from the ALGECO$_2$ project	[99]
Comparative study between homogeneous and heterogenous reservoir during H_2 integration in energy network using different spatial variation methods	Eclipse 300	2D	N_2	Saline aquifer	Short term	• K_r and P_c from Brooks and Corey's relations • Penge Robinson EOS • Analytical averaging methods	[164]
Hydrogeochemical investigation	• PHREEQC	1D	CO_2	Depleted gas	Long term	• Molecular diffusion • Microbial growth modelling • Stored gas composition • Storage time modelling	[110]
Geochemical exploration in Molasse Basin	• Geochemist workbench (GWB)	-	CH_4	Depleted gas	Short and long term	• Equilibrium modelling • Primary kinetic batch modelling • Final kinetic batch modelling	[159]
Maximum storage potential, capacity, degree of saturation, and time effect during injection/withdrawal cycles	• PetraSim-TOUGH2	3D	H_2	Aquifer	Long term	• Isothermal assumption • Multi-phase flow modelling • Geothermal modelling	[100]
Hydrodynamics investigation	• Eclipse 100	3D	CH_4	Depleted oil and gas field	Long term	• History matching • Storage initialization • Cyclic operation • Prolonged withdrawal investigation and case studies	[96]
Investigated the effect of H_2 injection and withdrawal from a 3D heterogenous sandstone reservoir in different geological settings	• TOUGH2	3D	-	Aquifer	Long term	• K_r and P_c from Brooks and Corey's relations • EOS for water, salt, and Gas (EWASG) for property modelling • Dirichlet boundary conditions	[160]

(continued on next page)

Table 5 (*continued*)

Research focus	Software tools/ codes	Model type	Cushion gas	Storage medium	Storage duration	Approach	References
						• Reservoir heterogeneity was simulated using the porosity and permeability data from the 10th SPE comparative solution project • The influence of porosity and permeability heterogeneity on Pc was obtained via Leverett J-function • Pc and Kr were imported into TOUGH2 via Van Genuchten–Mualem model.	
Role of cushion gas on H_2 recovery, heating value, injection/ withdrawal, storage time, and incremental condensate recovery	• Eclipse 300	3D	CH_4, N_2, and CO_2	Partially depleted gas condensate reservoir	Long term	• K_r from SCAL data • Molecular diffusion by Ficks law and Fuller method • Three parameter Penge Robinson EOS • Different injection and withdrawal cases	[174]
Investigated the effect of cushion gas injection, H_2 recovery, losses, and solubility in the oil and water	• CMG/GEM	3D	CH_4, N_2, and CO_2	A depleted heterogeneous oil reservoir	Long term	• Different injection scenarios for cushion gas • Oil-water and Gas-oil Kr measured via unsteady-state methods • Thermodynamic properties were obtained via a compositional model simulator	[29]

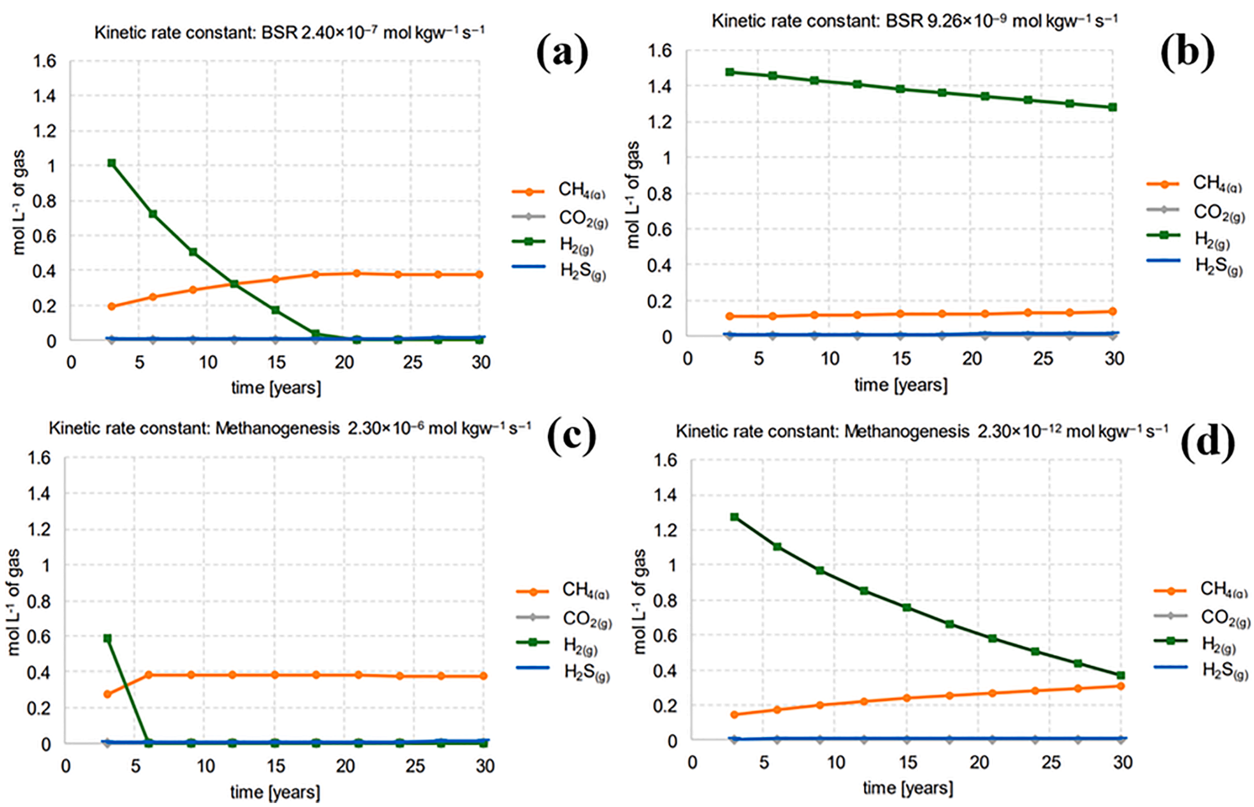


Fig. 13. Kinetic rate constant for SRB at (a) higher (2.40×10^{-7} mol kgw^{-1}s^{-1}) and (b) lower (9.26×10^{-9} mol kgw^{-1}s^{-1}) and for methanogenesis at (c) higher (2.30×10^{-6} mol kgw^{-1}s^{-1}) and (d) lower (2.30×10^{-12} mol kgw^{-1}s^{-1}) [110].

In the case of increasing the kinetic constant to 2.30×10^{-6} mol kgw^{-1}s^{-1}, H_2 was lost in less than six years for methanogenesis, Fig. 13**(c)**. CH_4 concentration in the reservoir increased to about 0.30 mol/L after 30 years. However, when the kinetic rate constant decreased to 2.30×10^{-12} mol kgw^{-1}s^{-1}, H_2 loss was slightly lower, whereas only a minute amount of CH_4 was generated, Fig. 13**(d)**. The authors concluded that, while the kinetic rate constant is a vital component controlling H_2 loss during geo-storage, the maximum amount of H_2S generated (if all reactive anhydrite is consumed) is limited by diffusion only, whereas the maximal amount of CH_4 depends not only on the kinetic rate constant but storage time [110].

4.3.1.2. Pressure and temperature conditions in gas reservoirs. Reservoir pressure and temperature have a significant influence on H_2 geo-storage.

(e.g. there is an optimal thermo-physical H_2 storage depth for structural H_2 storage [177]). The simulation work of Hemme and Berk [110] was conducted at 40 atm (pressure) and 40 °C (temperature). However, when increased pressure (161 atm) and temperature (80 °C) were assessed for H_2 storage (at a constant kinetic rate), the effect of SRB and methanogens were no longer active. This was ascribed to the maximum temperature for SRB and methanogenic bacteria being 80 °C [125,178]. The authors also found that, at the set operating conditions for the depleted gas reservoir, only 75 % of H_2 was consumed as compared to 76 % based on the simulation conditions (40 °C and 40 atm). This could be due to the increased pressure resulting from the decreased gas volume at a higher gas concentration.

5. Economic analysis of depleted reservoirs, aquifers, and salt caverns

Economics analysis is key to determining any storage site's success as successful storage infrastructure investments must provide attractive financial outcomes [179]. Several investment metrics are employed by the industry to assess the commercial viability of projects, including, but not limited to, net present value and internal rate of return as it measures a project's viability, and cash flow against capital (CAPEX) and operating (OPEX) expenditures [179]. The physical properties and capacities of a given storage field, the services to be offered, and, to a lesser extent, regulatory regimes all impact CAPEX and cash flow, and thus the economic attractiveness of storage sites. In contrast, the development of OPEX varies greatly depending on the type of gas storage and its performance characteristics during design. Often, costs (either CAPEX or OPEX) are site-specific based on (i) the quality and variability of the geologic structure of the proposed site, (ii) the required amount of compressive horsepower, (iii) the type of surface facilities (i.e., see [50]), (iv) the proximity to pipeline infrastructure for the ease of transport and (v) legal and environmental issues [85,93,179].

As previously stated, underground storage is unique. It can store vast amounts of H_2 and is less expensive (a few hundred dollars or less) than other modes of storage (a few thousand dollars or more) independent of H_2 purity [50]. However, geological issues are the most difficult to deal with; thus, they must be explored first when deciding on a prospective UHS location [31,34]. Taylor et al. classified CAPEX and OPEX for various storage options, with compressor costs accounting for most of CAPEX and H_2 generation costs accounting for most of OPEX. The overall economic viability of the whole H_2 storage project was summarised to depend upon the capture cost, transportation cost, storage cost, withdrawal cost, and monitoring cost [50].

Despite the lack of economic data, the work of Lord et al. [28] is frequently cited to interpret the many sources of CAPEX for H_2 geostorage technologies. Table 6 shows the city-specific geological storage alternatives based on population, summer spike demand for H_2, and various market penetration levels (e.g., market penetration to adopt H_2 over other fuels in the transportation sector) using the Hydrogen Geological Storage Model (H2GSM) framework.

As with petroleum economics, CAPEX and OPEX are the major determinants of a financial asset's profitability [50]. However, the geological site hugely affects these indexes, as seen in Table 6, depending on whether it is surface or subsurface [28].

For instance, the data presented for salt cavern shows that CAPEX at the surface level includes gas compression equipment and building construction cost. Whereas, well and cavity development cost – i.e., leaching - is under subsurface [28]. Generally, as this cost is often unrecoverable once incurred during storage, cushion gas can therefore be considered as CAPEX [28]. Moreover, cushion gas percentage and the interplay of other factors are depth-dependent on the geological site. Thus, lower depth implies lower cost since it leads to lower injection pressure and reduced equipment compression load, leading to low H_2 storage capacity. However, an increase in storage depth will lead to larger H_2 storage capacity and lower cushion gas requirement [180],

Table 6
The typical assumption for a geological site design-specific cost analysis [28].

Assumed cost analysis for a specific storage site	Salt cavern	Aquifer	Depleted oil and gas
Cushion gas capital cost (USD)	**11,227,540**	**21,492,278**	**21,492,278**
Cost of H_2 Gas (USD/kg)	6	6	6
Geologic site preparation total cavern site development (USD)	**23,340,000**	**n/a**	**n/a**
Mining costs (USD/m^3)	23	n/a	n/a
Leaching Plant Costs (MUSD)	10	n/a	n/a
Site Characterization (MUSD)	n/a	10.3	n/a
Compressor capital costs (USD)	**27,539,480**	**18,359,654**	**18,359,654**
Injection rate (kg/hr)	2960	2487	2487
Withdrawal rate (kg/hr)	4920	2487	2487
Compressor power (kWh/kg)	2.2	2.2	2.2
Compressor (kWh/yr)	988,819	499,836	499,836
Operating (days/yr)	350	350	350
Compressor capacity factor (%)	96	96	96
Cost of electricity (cents/kWh)	5	5	5
Levelized electricity cost per compressor (USD/kg)	0.002	0.001	0.001
Water and cooling cost/ compressor (USD/100 L)	0.02	0.02	0.02
Water requirements per compressor (l/kg H_2)	50	50	50
Water and cooling costs (USD/kg H_2)	0.012	0.012	0.012
Compressor O&M (USD/kg H_2)	0.014	0.014	0.014
Number of compressors	3	2	2
Compressor size (kg/hr)	2000	2000	2000
Compressor size (kW)	3700	3700	3700
Cost per compressor (USD/kW)	2481	2481	2481
Pipelines and wells capital cost, Full pipeline costs (USD/ tonne)	**4.39**	**6.26**	**6.26**
Pipeline fixed costs (USD/tonne)	4.03	4.03	4.03
System flow rate (tonne/hr)	4.78	4.78	2.42
Pipeline maximum flow rate (tonne/hr)	445.9	445.9	445.9
Transport distance of H_2 (km)	16	16	16
Base transport Distance (km)	100	100	100
Well O&M multiplier (%)	4	4	4
Number of injection/withdrawal wells	1	1	1
Capital cost per well (MUSD/well)	1.15	1.15	0.26
Well variable cost (USD/km)	1,434,409	1,434,409	318,757
Well depth (ft)	3800	3800	4604
Well variable cost (MUSD)	1.66	1.66	0.45
Equipment lifetime (years)	30	30	30
Discount rate (%)	10	10	10
Capital recovery factor	0.11	0.11	0.11
Full H_2 well costs (USD/tonne)	46.27	46.27	10.55
Mass flow rate/day/well	2500	2500	2500
Injection rate (kg H_2/hr)	283,836	283,836	283,479
Full H_2 surface piping (USD/ tonne)	0	0	0
H_2 pipeline and well costs (USD/ tonne)	50.66	53.71	16.80
Total capital costs	**63,253,547**	**40,999,458**	**40,106,938**
Levelized total capital costs (USD/ kg)	1.54	1.21	1.19
Levelized cost of H_2 storage (USD/ kg)	1.61	1.29	1.23

though at a higher cost. Despite the low cushion gas content, which is depth-dependent, the number of boreholes and material cost will also increase the CAPEX requirement for salt caverns [28]. In the case of OPEX, which depends on H_2 production sources and others [50], cavern's maintenance and post-storage processing as the leached brine often contains impurities that must undergo proper separation before final disposal is considered as the major OPEX. Additionally, the mining of salt caverns takes several years. It requires huge investment (see Table 6), and if there is no industrial use for the mined brine, the remediation of its effect after disposal can be detrimental.

Table 7
Different utility-scale subsurface storage and aspects considered during decision-making [31].

	Salt cavern	Aquifer	Depleted oil and gas
Point in development	Commercial	Laboratory	Laboratory
No of injection/withdrawal cycles	Up to 10	1 to 2	1 to 2
Storage capacity (tonnes of H_2)	Small to Medium(1000 – 3500)	Large to Very Large(7200 – 53,000)	Medium to Large(2000 – 23,000)
Cushion gas percentage	20 to 33	45 to 80	50 to 60
Operating pressure (bar)	45 to 202	30 to 137.8	100 to 400
Rate of discharge (GW/day)	0.467 to 10.128	1.09 to 8.55	2.66 to 100

In the case of the aquifer, a Levelized cost between the salt cavern and the depleted deposit was observed (1.21 – 1.29 USD/kg of H_2). This is because CAPEX due to leaching and mining relevant to the salt cavern is not present in the aquifer. Contrary to the study by [28] presented in Table 6, some authors reported that the CAPEX and OPEX involved in aquifer site characterization make this subsurface storage the most expensive. Specifically, Tarkowski highlighted that the construction and operation costs for H_2 storage in aquifers are higher than those in caverns and depleted hydrocarbon reservoirs [23]. A typical CAPEX for an aquifer involves (i) surface infrastructure, (ii) pipeline and well cost, (iii) cushion gas, (iv) compressors, and (v) survey cost, whereas OPEX plus maintenance cost for H_2 storage in an aquifer includes (i) viscous fingering, (ii) well leakage, (iii) pipelines and wells, (iv) compressor maintenance, (v) H_2 compression and (vi) post storage processing [31]. All highlighted CAPEX besides cushion gas cost are inevitable when drilling an exploratory well to study the geological characteristics of the intended aquifer for storage assessment as it involves a detailed survey of the site porosity, intrinsic permeability, and the capillary entry pressure to allow for reservoir properties, such as deliverability and capacity to be determined [23,28]. Notwithstanding, the significance and type of cushion gas are also essential as aquifers often contain in-situ reservoir fluids, which contribute to the high percentage of cushion gas requirement for pressure maintenance. Even when working gas and cushion gas capacity is uniform (1:1), cushion gas is reported to account for approximately 42 % of the total CAPEX [28]; thus, cushion gas cost cannot be overlooked. Alternatively, different cushion gas types can be used (e.g., N_2 and CO_2), but the stored H_2 gas is at risk because of the possible microbial reaction (i.e., methanogenesis) that could occur in the presence of CO_2; thus, H_2 withdrawal will be reduced. Furthermore, adopting a multiple extraction strategy and well configuration when using H_2 as cushion gas could further increase the withdrawal rates [99] as it also promotes low OPEX associated with post-separation of the produced gas as both are identical.

As for depleted oil and gas reservoirs, site characterization cost (10.3 million USD) found in the aquifer is absent as they already have identified reservoir characteristics [28]; thus, the total capital cost is usually cheap (1.23 USD/kg of H_2). Albeit, this is not always the case, as the hydrodynamics of H_2 in terms of its higher mobility than oil and natural gas in the reservoir could incur more cost if a secondary site characterization is required [28,45] to ensure adequate storage containment. CAPEX under this category usually involves the cost of repositioning the current petrochemical infrastructure initially used for natural gas storage and whether the site is onshore or offshore. In the case of OPEX, similar factors could influence the economic performance as both aquifer and depleted reservoirs are porous mediums. Thus, they often exhibit the same characteristics. Though additional OPEX could arise from the more intensive separation processing due to the native oil/gas/brine and the management of this by-product [31].

Notwithstanding, storage cost is higher for storage in depleted oil reservoirs than in depleted natural gas reservoirs [22] as it requires more complex multiphase fluid flow interactions with H_2, which will result in pore space reduction and greater storage costs due to separation after withdrawal. Again, oil reservoirs do not have the added benefit of a cushion gas already in place as compared to depleted gas reservoirs. Another factor that makes depleted gas reservoirs cheaper is the fact that, rather than using the native gas (CH_4) to reduce the percent of cushion gas required, an endpoint injection strategy can be adopted for injecting the cushion gas for an efficient sweep, which thus generates value from what would initially be considered a loss [48] and such strategy are only applicable to gas–gas reservoirs displacement.

In summary, Table 6, in retrospect, highlights a typical economic study for H_2 storage using the analytical framework of the H2GSM model as it illustrates the analysis from a different perspective. H_2 storage in depleted natural gas reservoirs turned out to be the cheapest (even though the study was conducted using previous petrochemical equipment with no site characterization cost) compared to aquifers and salt caverns. In the review of Wallace et al. [31], it was reported that the cost per kg of H_2 for salt cavern, aquifer, and depleted reservoir ranges between 1.60 and 1.61, 1.29, and 1.23–1.48 USD, respectively, suggesting that the depleted reservoir is often considered as the least expensive option. The extent to which it is less costly depends on how the field was initially developed. Some fields may have suffered from poor drilling, operating, and abandonment procedures, which could affect the cost of their conversion and adaptation for storage purposes. Nonetheless, as seen in Table 7, other factors influence the different utility-scale during storage, especially the geographical location (based on the UK). Hence, further studies will be required to analyze each mode's life cycle cost, especially in depleted gas reservoirs where pure H_2 and a mixture of H_2 and CH_4, CO_2, and N_2 gases can coexist for a realistic reservoir condition.

6. Challenges and risks associated with H_2 storage

It is difficult to list all the challenges peculiar to the depleted gas reservoir as it affects the entire potential UHS mediums (salt and hard rock caverns, aquifer, and depleted hydrocarbon). Clearly, well integrity, plays a significant role in the geo-storage of gases. Although depleted reservoirs are known for impeccable and tested characteristics, the wells must be able to withstand extreme conditions to prevent potential leakage and corrosion in the presence of formation fluids (gas, oil, and brine) [55,181]. H_2 geo-storage is novel and is currently not well understood. The design of new gas storage wells should meet or exceed relevant design manuals and standards to ensure storage safety and long-term efficiency during injection and withdrawal cycles.

As defined by the International Organization for Standardization (ISO), well integrity is the capacity of the drilled hole to maintain its shape and remain intact throughout the well's life cycle [182]. Based on the NORSOK D-010 standard, it has also been defined as the application of technical, operational, and organizational solutions to reduce the risk of uncontrolled release of formation fluids to the surface throughout the well's life cycle [183]. Loss of well integrity will lead to project failure, resulting in financial losses and possibly jeopardizing people's health and lives. To control potential leakage problems during storage, it is vital to address some factors that can potentially inhibit the well integrity, such as microbial actions, embrittlement, cementing problems, sulfidation, elastomer failures, caprock sealing failures, and purity of the injected gas. A schematic showing the possible leakage channels along the wellbore during injection and withdrawal is presented in

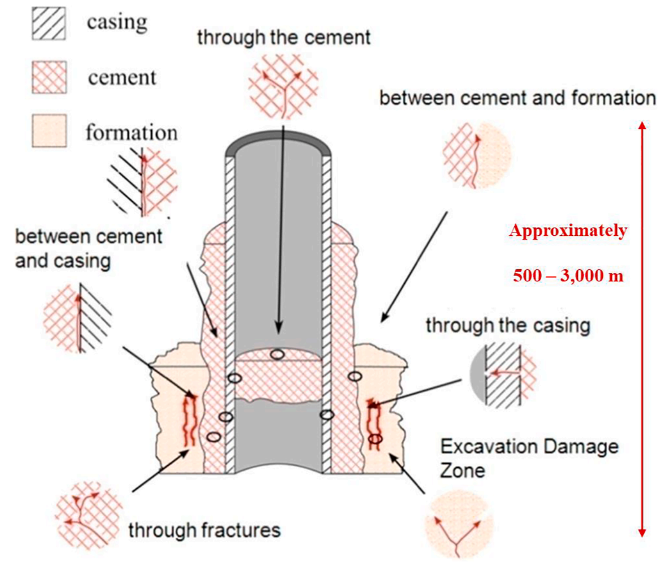


Fig. 14. Potential pathways of H_2 leakage along the wellbore. Modified after [184].

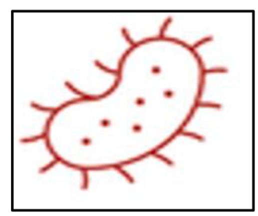
Fig. 14. On a broad scale, the following highlight sums up the key challenges associated with gas geo-storage as it affects UHS, UGS and CCS.

6.1. *Microbial induced corrosion (MIC)*

The activities of microbes are considered the foremost challenge associated with H_2 geo-storage irrespective of the storage medium, and it is the most common problem for steel infrastructure in various environments (water, oil, and subsurface installations). In MIC, H_2 is an electron donor for microorganisms in the subsurface [34]. This is evident as many reported cases have observed changes in gas composition during storage cycles after H_2 injection. The permanent conversion of H_2 to CH_4 and H_2S by microorganisms, the chemical reactivity of H_2 with the minerals of the reservoir-rock or cap-rock, the resulting potential porosity changes, the effect of ferric ions on metal surfaces in the presence of H_2, as well as the H_2 loss through diffusion through the caprock, are all potential risks and concerns due to microorganisms [36,91,110]. Although, corrosion like this mainly depends on the type of microbes and the inhabiting sites (caverns or porous media) [34,37]. It is reported that sulfate and iron reductions and, methanogenesis (see also Table 8) are the processes that cause MIC in UHS [43]. Moreover, corrosion induced by these living organisms can weaken several metallic downhole components, thus, compromising the containment integrity of the stored gas [43].

Sulfate-reducing bacteria, SRB (obtained via anhydrite [$CaSO_{4(s)}$] mineral dissolution) consumes H_2 to produce H_2S [38,173], and the gained energy subsequently serves as input by SRB for cell growth [185]. The major risk caused by SRB comes with its high toxicity (as it contributes to developing inflammatory bowel diseases [186] for humans when present in an environment) and its potential corrosion effect on the facility [37]. It can inhabit any gas storage type, especially natural gas reservoirs [43]. SRB can corrode the metallic casing surface in two ways: chemically by producing H_2S and electrically by the withdrawal [187]. The survival of this type of microbes depends on the reservoir temperature, salinity, brine complexity, pH, and pressure of the underground environment [38]. Notwithstanding, pressure, amongst other environmental factors, has a lesser effect on microbial cellular activity, particularly in thermophiles and hyper-thermophiles [38]. Optimally, SRB occurs at around 38 °C [188] under near-neutral pH (6 to 7.5) conditions [185]. Extreme temperature conditions include (0 to 60–80 °C) [110,189], 110 °C [178], and even 122 °C [38] as reported in some

Table 8
Biotic transformation and implications for subsurface storage. Modified after [36].

Hydrogenotrophic microbes	Reaction and potential mineralization	Potential impact on UHS
Sulfate Reducing Bacteria (SRB) 	$SO_4^{2-} + 5H_2 \leftrightarrow H_2S + 4H_2O$ • H_2 reacts with anhydrite and other inorganic sulfate sources and reduces sulfate to sulfide.	• H_2 sulfide release. • Gas mixing effect. • Iron corrosion. • pH reduction. • Hydrogen embrittlement. • Mineral precipitation.
Iron Reducing Bacteria (IRB) 	$H_2 + 3Fe_2O_3 \leftrightarrow 2Fe_3O_4 + H_2O$ • Microbes can reduce passive film, e.g., ferric components on metal surfaces. • Water can release and occupy the sandstone's interstitial pore space by-product, causing excess water saturation and mineral dissolution.	• Low sulfide reduction. • Metal corrosion. • Carbon steel corrosion. • Mineral dissolution.
Methanogens Archaea 	$4H_2 + CO_2 \leftrightarrow CH_4 + 2H_2O$ • Microbes attach to the edges of clay surfaces, e.g., kaolinite, to prevent CH_4 flow. • The presence of Aluminium ions in the kaolinite can be toxic to the growth of methanogens.	• Gas mixing effect. • Reduces injection and withdrawal capacity due to plugging of pores. • Permeability and porosity alteration.

depleted oil and gas fields. Since most reservoir fluids in geo-storage predominately contain NaCl, a kosmotropic solvent (as they promote stability and habitability), they are thus more permissive to microbial growth than methanogens [38].

Iron-reducing bacteria (IRB) uses a metabolic reaction where iron (III) oxide (ferric oxide) consumes H_2. Iron-reducers could be heterotrophs (that use organic carbon as food) or autotrophic (those bacteria that can synthesize their food) [190]. Besides SRB, IRB is another form of bacterial group that can generate corrosion to the casing, but to a smaller degree compared to SRB. IRB relies on the availability of iron oxides, organic carbon, and minerals like smectite and chlorite to promote metal oxidation processes and reduce iron (III) phases. IRB and SRB can survive in the same environment and work together in the corrosion process as IRB uses acetate, a typical product of SRB [43,191]. However, IRB can thrive over SRB, particularly in an environment rich in iron oxides and organic carbon, due to their high affinity for H_2 [192].

In the case of methanogens, H_2 is consumed as a donor, while CO_2 is used as a receptor to produce CH_4 [39]. Methane is not highly corrosive for metals; however, the presence of CO_2 and H_2S within the ambient natural gas (initially present before H_2 injection) may further facilitate its corrosiveness. It is also dependent on temperature, pH, and salinity [38]. Preferred temperatures for growth are 30 to 40 °C [193]. Though, higher temperature conditions (i.e., 80 °C [125] and 97 °C [193–195]) have been reported in the subsurface. The review of Thaysen et al. also reported 122 °C in some depleted oil and gas studies [38]. The effect of methanogens in field cases has been reported for gas storage (e.g., town gas) technologies. Although other storage facilities (Beynes in France; and Kiel in Germany) reported no mal-functions due to methanogens [151], the storage facility at Lobodice in the Czech Republic noted a change in the stored town gas composition as 54 vol% of injected H_2 gas reduced to 37 vol% H_2 gas. In contrast, the CH_4 increased from 21.9 % to 40.0 vol% within seven months of storage [176,195].

In the CCS context, CO_2 can lead to a crevice, galvanic, and pitting type of corrosion in downstream equipment, which is mainly enhanced by the presence of chloride (in the case of crevice corrosion) [43]. Generally, material selection is the best mitigation for corrosion (especially the non-microbial). However, these materials are expensive and must be ensured that proper handling techniques are used in running casing and tubing not to create surface defects that will reduce the fatigue life [43]. Another option is chemical inhibition, where specific chemicals are used in either batch or continuous injection [196]. The batch method involves undesirable, healthy downtime, while the continuous injection may be complicated logistically due to gas storage wells' sometimes-unpredictable injection/production pattern. Other corrosion mitigation measures include annulus protection fluid, coatings, and cathodic protection [197]. Also, corrosion can be detected and measured using multi-fingered caliper logs even though they can only detect tiny changes in the internal surface tubing condition to identify corrosion and mechanical damage. However, when combined with magnetic thickness tools, the internal and external conditions of the tubing can be examined [147,196].

Additionally, since CO_2 forms carbonic acid when dissolved in water [82,198], the produced carbonic acid can generate an electrochemical reaction with the steel to form iron carbonate scale precipitate [199], which are inorganic precipitates that can plug the reservoir pores during injection cycles, thus reducing deliverability [200]. Due to SRB and methanogens in the subsurface, both corrosion scenarios can cause metal thinning, resulting in metal fatigue if not addressed. As a result, it is critical to understand the reservoir rock's geological structure and mineral composition, as this outcome in prospect suggests that a depleted gas field rich in sulfate, and carbonate-bearing mineral, is generally not recommended for UHS. The work of [110] also made a similar recommendation for reservoirs with a low amount of sulfate and carbonate minerals but with a high amount of reactive iron-bearing minerals as the best option for geo-storage.

6.2. H_2 embrittlement

H_2 embrittlement (H_2 blistering or H_2-induced cracking) of downstream equipment is another crucial challenge found in UHS storage [201–203]. H_2 blistering occurs when an atomic H_2 is formed on the metal surface by dissociative chemical sorption, whereas H_2-induced cracking occurs after H_2 entry into the steel surface [43]. Though pipelines are often used for UGS and UHS transportation, their selection and application depend on the potential corrosion severity and the project type. Suppose metal is exposed to H_2 for an extended period, especially at greater concentrations and pressures such as those found in underground storage, its durability may be compromised even at a more significant strength [204], leading to H_2 embrittlement [205]. This process is tied to the growth of microbial biofilms on steel surfaces [37]. Fig. 15 shows the H_2-induced twins in the form of needles and deformation bands on the steel surface due to embrittlement effects. High-strength steel is often conducive for UGS and CCS. However, they are severely affected by UHS. This indicates that low to medium steel is considered a better option for UHS [43]. If by chance, a higher steel strength is required based on the properties of the potential location, the low-strength steels should be employed in softened conditions since they exhibit better toughness [39].

Steel selection requires specific material types, such as corrosion-resistant alloys (CRA), as recommended by the oil & gas industry [43]. These material types have a martensitic structure that can resist moderate acid environments (resistance against H_2S or CO_2), yet it is susceptible to H_2 permeation as diffusion can still occur [39]. While the diffusion rate for H_2 depends on the type of crystal lattice [122], the susceptibility of this material to H_2 depends on the steel composition, processing history, and deoxidation practice [207], as these parameters affect non-metallic inclusions (type, size, and morphology) and their ability to hold H_2. Some studies through experimental analysis have found a way of reducing H_2-induced cracking. San Marchi et al. [208] documented that austenitic steel samples with high nickel content (e.g., greater than 12 %) and the presence of nitrogen gas could effectively prevent H_2 permeation and diffusion.

In comparison with natural gas, the quality of the metallic material (used in pipeline construction in H_2 transportation) is essential for the H_2 storage process. Thus, the evaluation of well integrity requires critical analysis. For instance, if the tensile stress of the casing or the micro-porosity of the interior of the casing increases gradually, casing failure could occur without any indication [184]. In pre-treated steel materials, induced H_2 cracks are more complex, with a generation rate almost three times that of the original crack. This transformation can lead to a more rapid H_2 diffusion than CH_4 (UGS) and CO_2 (CCS) gases in the crack tips and even the entire material due to their different physico-chemical properties.

Aside from the possible harm that H_2 could cause to steel pipes, valves, and other hardware, the cement that seals the well-bore from the formation may not be as impervious to H_2 as it is to other gas molecules [209,210] thus, experimental mass transfer studies on the rock

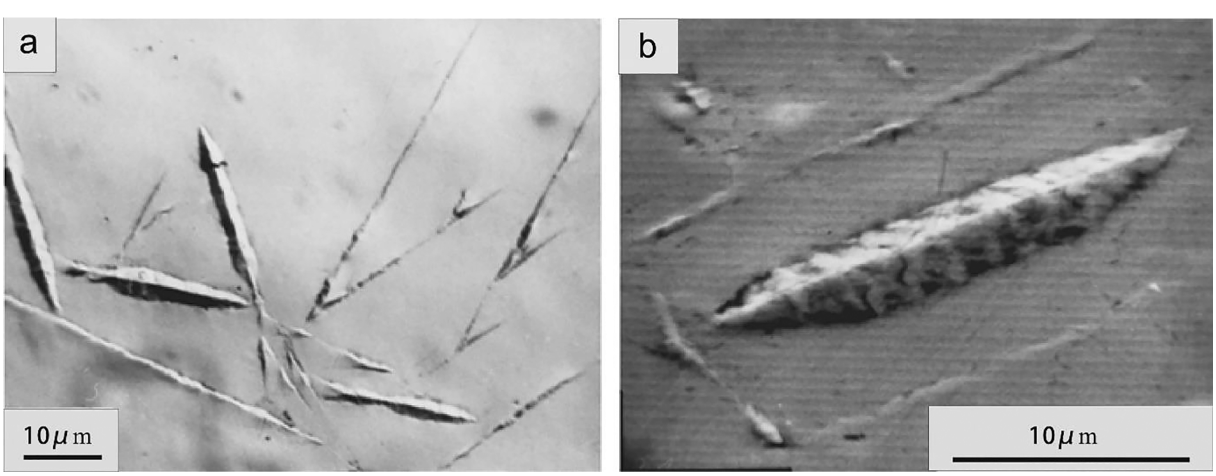


Fig. 15. H_2 - induced twins in the form of needles and deformation bands on the steel surface [206].

formation, caprock, and cement samples may provide answers to some of these concerns. It is vital to use representative H_2 and natural gas combinations and that laboratory circumstances properly imitate the depleted gas reservoir pressure and temperature conditions to understand the effect of the gas on the steel pipes.

6.3. Cement carbonation

Cement is an essential component that ensures the integrity of a well during storage life [211,212]. As previously discussed, depleted gas reservoirs are already characterized since they have undergone production (compared to aquifers and caverns). Yet, the quality of the cement over a long period can be compromised. Cement can undergo both mechanical and chemical degradation in the subsurface. Mechanical degradation occurs due to exposure to extreme loading conditions due to pressure, thermal expansion, and volume change during hydration while chemical degradation of cement (carbonation) occurs due to CO_2 presence in an aqueous environment [213] or H_2S attack after H_2 oxidization with sulfate, as shown in Fig. 16 [43].

It is also vital to note that the swelling potential for clay is influenced by the water activity of the fluids and the active clay minerals [214–217]. The ability of active clay minerals in clay containing reservoirs, caprocks, and faults to absorb H_2 could produce swelling and stress changes. While the H_2 sorption capacity for common clay minerals (such as montmorillonite and laponite) is two to four times less than that of CO_2, the associated stress–strain sorption behavior yet provides a significant mechanical and transportation issue to the storage complexity [8].

In the underground environment context, the carbonation of the cement sheath is considered the major setback associated with the CCS because a CO_2 attack can reduce the well compressive strength, which may affect the wellbore's integrity by increasing its porosity and permeability, thus, promoting leakage [218]. For UGS and UHS, carbonation depends on the CO_2 percentage found in the stored gas or cushion gas. Hence, cushion gas selection is considered one of the major factors when considering carbonation effects. This supports the findings of Hemme and van Berk, who stated that depleted gas fields with low residual CO_2 concentrations for H_2 storage are highly recommended

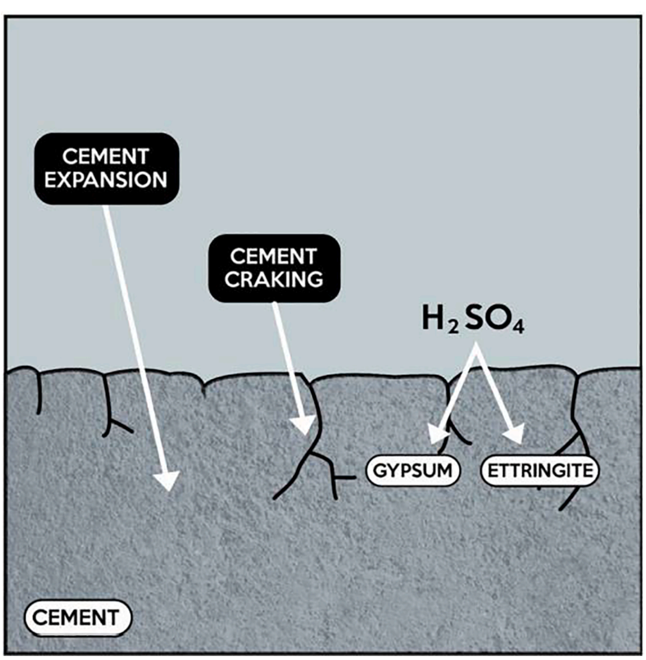


Fig. 16. Chemical degradation of cement due to H_2S attack in an aqueous environment [43].

since they can reduce H_2 loss. This conclusion was reached because, whereas longer storage duration results in more H_2 loss and shorter storage periods result in less H_2 loss, the overall H_2 loss in a short storage time is higher since additional CO_2 is available for methanogenesis after each gas injection cycle [110].

Class G and H cement types are commonly used in oil and gas applications [211,212] for UGS [43]. In CCS, Portland cement is modified to slow the reaction of CO_2 by blending Pozzolanic material to reduce the chemical attack of CO_2. In a recent development, geopolymer is currently tested in place of typical cement systems for CCS due to the absence of calcium hydroxide ($Ca[OH]_2$), making it more effective and environment-friendly [219]. For UHS, no field test proves that class G and H cement types will be suitable for UHS due to the overall capacity of H_2 to permeate through the cement sheath [43] as they are more mobile and diffusive. Hence, more investigation on this aspect will be required. Other associated problems of UHS include cement deterioration due to H_2S formation caused by H_2 reaction with SRB archaea that may be present on the surface of the cement sheath or within its interstitial spaces, primarily when active clay minerals exist within the rock's lattices.

6.4. Sulfidation

Sulfidation (a process of installing sulfide ions in a material or molecule which is used to convert oxides to sulfides) can also impact the integrity of a well during injection and withdrawal as it can lead to corrosion. As for UGS and CCS, the risk of sulfidation depends on the H_2S amount present in the respective formations [43]. Ettringite or hydrous calcium aluminium sulfate mineral formation within the cement sheath is a major concern in UHS as it causes expansion. Primary ettringite that occurs before cement hardening is often easier to address than secondary ettringite, which leads to expansion and internal stresses within the cement sheath [220,221]. Given the strong diffusion rate of H_2 to CH_4 and CO_2, the permeability and porosity of the cementing material must be carefully examined [184], especially in caverns (where new excavations are drilled), to ensure better impenetrability.

6.5. Elastomer degradation

An elastomer is a polymer with viscoelasticity and weak intermolecular forces with low Young's modulus and high failure strain compared to other materials. In underground facility design, packer assemblies isolate fluid within the casing, tubing, and annuli. They also serve as a secondary barrier after the drilling fluid (primary barrier). Material-wise, packers are composed of rubber-like polymers rounded with steel. However, it is susceptible to chemical degradation (corrosion) when exposed to H_2S, CO_2, or CH_4 environments [43]. Usually, when an elastomeric material contacts drilling fluid, completion fluid, production fluid containing various solvents [222], formation brine, or corrosive chemical generated due to gas intervention, it may facilitate chemical degradation of such elastomer, which subsequently affects the packer durability [43]. For instance, in SRB and methanogens cases where H_2 reacts with sulfate and CO_2 to produce H_2S and CH_4, respectively, the elastomer performance may be inhibited. Specifically, the exposure of an elastomer to H_2S is reported to cause hemolysis and heterolysis as it increases the reaction activity in the subsurface [43]. This may cause a reduction in the tensile strength of the material, ultimate elongation, and even result in hardness [222,223].

In the context of UGS, CH_4 is chemically stable and does not react with an elastomer, though permeation within the material may be observed [222]. In the case of the UHS environment, which is mainly affected by H_2S, Nitrile Butadiene Rubber (NBR) elastomer is most preferred, as it is more responsive to corrosion entrainment and with a high capacity of reducing the risk of rapid gas decompression, which may cause internal blister within the rubber seal [39,224]. As for CCS, CO_2 behaves as an inert gas (non-toxic) in the gas phase until it undergoes a carbonic acid reaction. Therefore, elastomer performance can only occur after CO_2 chemical conversion, which may reduce the sealing strength of the reservoir and caprocks [224,225].

6.6. Caprock integrity failure

H_2 reactions with reservoir rock and caprock challenges are numerous due to their physical and chemical properties [76,91]. Commonly used caprocks are salt and clay layers because they provide tightness and hydraulic integrity against H_2 [43]. However, any lithology can provide a seal. All that is required is that the seal rock's minimum displacement pressure (capillary entry pressure) is greater than the buoyancy pressure of the gas in the accumulation [226]. Parameters like solid, fluid, and solid–fluid interactions play a significant role in fluid containment as they prevent the upward migration of fluids during storage [24,60]. Specifically, caprock failures largely depend on porosity, permeability, interfacial tension, and capillary pressure, which promote the stored gas's leakage. For example, rock tightness is governed by water saturation, and because most rock-forming minerals are hydrophilic and maintained by capillary forces; as a result, the capillary entry pressure: which is the pressure required for gas to enter the pores and eject capillary water [100] on the clayey cap rock is not surpassed, the caprock will remain tight for the gaseous phase and acts as a geological barrier for the gas below [177]. However, if exceeded just once, water can be extruded from the cap rock, making it permeable to gas. This ultimately leads to gas leakage [39,100].

Leakage in porous media can occur either by (i) membrane seal failure, (ii) hydraulic seal failure, or (iii) wedging open of a fault due to tectonic activity. Details on this can be found elsewhere [226]. However, leakage risk in the subsurface is higher in UHS than in UGS or CCS. In the H_2-brine system, due to the very low H_2 density [36] and the high H_2 diffusivity [53], and also mainly due to the lack of project data, CH_4-brine systems (UGS) has a high gas–water interfacial tension, which leads to increased capillary pressure. CCS, however, has its challenges (relatively low gas–water interfacial tension and the corrosive environment created by the acidic gas - which can lead to an increase in porosity and permeability of the sealing caprocks) [43,55,227,228].

Trapping mechanisms, such as structural, residual, mineral, and dissolution (refer to **section 2.2**), play a major role in the efficiency and integrity of the sealing caprock [4,25,26]. For instance, caprock integrity may be compromised via mineral precipitation and dissolution [91,227,228]. As can be seen in Fig. 17, if the precipitation rate is higher than the rate of dissolution, the integrity of the caprock is maintained (intact) though a minute reduction both in porosity and permeability may be observed. However, at low precipitation rates, both the injection and production efficiency will be affected as this may increase the porosity and permeability, leading to leakage through the sealing caprock [22,43].

Additionally, the type of in-situ mineral and pore size distribution may change the interaction and geometry of the rock and fluid [45]. This effect can be extended to UGS and UHS as porosity deformation can either improve or deter the sealing strength. In contrast, mineral precipitation at the well equipment could result in scale formation [39]. Risk due to H_2 loss via diffusion in the depleted gas field is of major concern as H_2 dissolves in the formation water of the caprock and then diffuses through the caprock. Although, less than 0.1 % [12] and 2 % [149] are predicted to be lost via diffusion and dissolution. Some authors theorized that the high diffusivity of H_2 and low viscosity and density (as compared to CH_4) might lead to high mobility, which further increases losses due to leakages [173].

6.7. The gas purity during mixing

Regardless of the storage means previously enumerated (caverns, aquifers, and depleted hydrocarbon reservoirs), gas purity due to mixing is vital as only a part of the injected gas is often withdrawn during the peak period as 25 to 75 % (depending on the specific project, e.g. see review by [31]) needs to be retained as a cushion gas to prevent water intrusion, and also to enable maximum storage capacity and efficiency [96,184]. As discussed above, H_2 is highly mobile and diffusive (compared to methane), and mixing injected, and reservoir gas (during injection/withdrawal) could promote H_2 leakage. Experimentally, this may affect the quality of interpretation since H_2 is likely to diffuse out faster in an output facility intended for analyzing a mixture of gases (H_2 and methane).

This, however, could be mitigated as several injection schemes such as (i) cyclic injection/withdrawal using the same well or (ii) gas injection at the reservoir edge [96,99,100] have been implemented. In

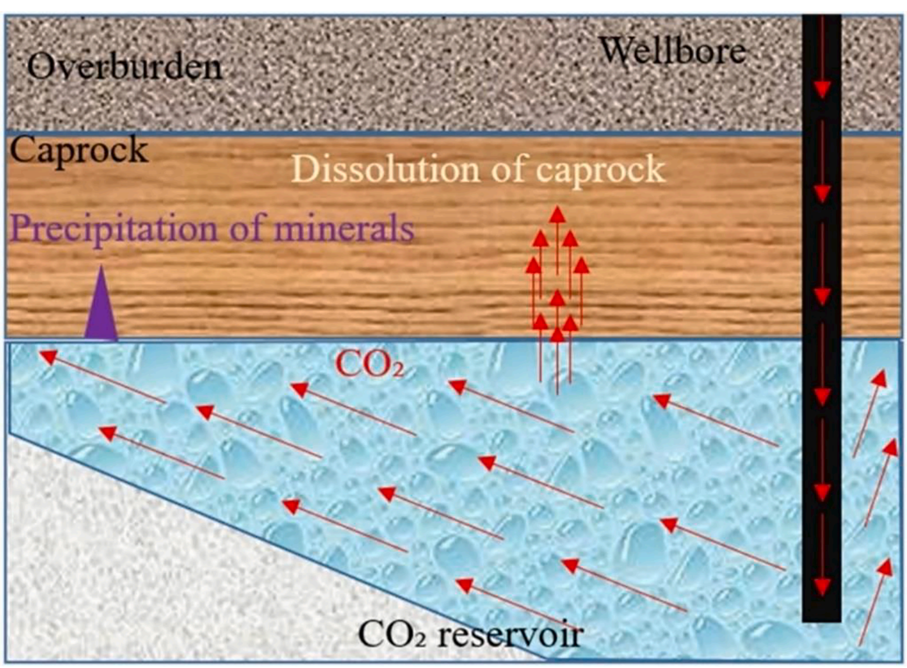


Fig. 17. Schematic representation of precipitation and dissolution effects [229].

Table 9
Comparative summary of UGS, CCS, and UHS in terms of the well integrity challenges.

	UGS	CCS	UHS	Refs.
Microbial Induced Corrosion	• CO_2 and H_2S presence responsible for related corrosion. • It varies depending on the geological formation, rock minerals, gas composition, pH, temperature, and salt content. • SRB type is more likely to occur compared to IRB and methanogens.	• MIC is absent; however, galvanic, pitting, and crevice corrosion could occur.	• All types (SRB, IRB, and methanogen) can occur since H_2 serves as an electron donor. • Microbes' survival is mainly influenced by temperature and salt content. • Pressure and pH have a low impact. • High risk of mineral precipitation/ dissolution.	[34,36–38,43,110]
H_2 embrittlement	• The moderate risk depends on the presence of H_2 near metal surfaces. • Leakage is potentially less due to its low diffusivity.	• Low risk due to H_2 absence.	• H_2 presence increases the susceptibility to cracking at lower stresses, thereby reducing material ductility and resistance to corrosion. • High leakage risk due to its high diffusivity and low molecular weight.	[37,39,43,184,204,205]
Cement carbonation	• The amount of CO_2 in the rock mineral and formation fluids will determine the reaction.	• The high risk depends on the percentage of CO_2 in the formation of carbonic acid. • Minerals dissolution and precipitation are influenced by temperature and pH	• The amount of CO_2 in the rock mineral and formation fluids will determine the reaction.	[8,43,213,218]
Sulfidation	• Depending on the amount of H_2S in the environment.	• Low danger because there's a lower chance of finding a large amount of H_2S.	• SRB-type MIC mostly found as H_2S can impact the well performance and quality.	[43,184]
Elastomer degradation	• The elastomer will not react chemically with natural gas.	• It occurs only when an elastomer contacts carbonic acid after CO_2 conversion.	• Moderate to high H_2S by-product of SRB can reduce the tensile strength and hardness of the elastomer.	[39,43,222–225]
Caprock integrity	• In a methane-water system, higher interfacial tension leads to higher capillary pressure and a lower likelihood of leakage.	• If dissolution rates in the caprock exceed precipitation rates, potential leakage is possible due to porosity and permeability enhancement.	• The high diffusion risk is due to its low interfacial tension in a hydrogen-water system, which results in low capillary pressure. • Minerals dissolution and precipitation can enhance or reduce potential leakage.	[24,55,60,85,227]
Gas mixing effect	• Neglected since working and cushion gas have the same composition.	• Moderate effect	• Mostly affected but can be used for secondary applications such as solid oxide fuel cells (SOFC) and transportation purposes.	[96,99,100,162]

another study, (iii) selective technology (as previously discussed in Fig. 11) was employed to reduce gas mixing (i.e., H_2 loss) for storage safety [162]. The cyclic injection/withdrawal via the same well is a simple transformation strategy that could result in high contaminated gas production, especially in the first year of withdrawal; thus, mixing is considered a severe problem for this strategy. As for transformation strategies where H_2 is injected at the reservoir edge, the residual gas is pushed to the other side of the reservoir or could be simultaneously produced on the opposite side. This strategy can lead to purer H_2 production during withdrawal as the effect of mixing is less and it has a better sweep efficiency.

The effect of mixing could be neglected in the case of UGS, where CH_4 is often used as both the working and cushion gas, as both gases predominantly have the same composition. However, in UHS cases where H_2 is mixed with natural gas (since natural gas facilities are easier converted to UHS), the withdrawn H_2 is likely to be less purified as both gases are miscible, thus, separation of such gases could lead to higher costs due to the application of separators. An illustration is a case of using H_2 in polymer electrolyte membrane (PEM) fuel cells (for vehicles) as it technically requires a pure form of H_2. However, solid oxide fuel cells (SOFC) can use the mixed gas of CH_4 and H_2, which is beyond the scope of this work and can be discussed in a separate article. On the contrary, if the H_2 is to be used in gas-fired turbines or fed into natural gas pipelines for other purposes beyond PEM fuel cells, blending H_2 into the natural gas grid becomes realistic, efficient, and positive for the environment.

Haven discussed the challenges and risks associated with geo storage of gases concerning well integrity, a summary of the key points discussed is presented in Table 9 for better comparison between UGS, CCS, and UHS projects.

7. Knowledge gaps and future scopes

Even though information on H_2 storage in depleted reservoir conditions is limited, the following recommendations were identified for future studies:

- Experimental and simulation reports have concentrated mainly on hydrodynamics, geochemistry/geochemical, and microbial activities regarding losses, mineral alteration, precipitation, and how it affects reservoir integrity. Future studies could focus on the geomechanics aspect of UHS in depleted gas conditions from excessive pressure increase, cyclic loading, or stresses during gas injection. Additionally, the seasonal operation's cyclic loading might induce crack/fault regeneration, leading to premature failure. As a result, more research combining hydrodynamics and geomechanics may be done to investigate aspects related to the migration, mixing, and dispersion of H_2 along fault lines inside the reservoir and across the overburden for improved recovery during injection/withdrawal.
- There is currently a scarcity of data describing the reservoir system strength variation in connection to H_2-enriched brine reactions on rock minerals (before and after incubation) [112,151]. As a result, more research is needed to focus on grain-size parameters of reservoir formations to analyze the effect of H_2 and how it affects reservoir quality, such as porosity and permeability, and migration over an extended period. Furthermore, evaluating the long-term stability of borehole seals is critical to determine H_2 leakage regardless of caprock mineral type and quality.
- While the composition of natural gas plays a significant role in depleted gas reservoirs, the effect of impurities such as NO_2, SO_2, and H_2S before H_2 injection is essential to distinguish the loss of H_2 due to microbial activities effectively. Moreover, numerical models capable of describing reservoir changes over extended periods of injection

and storage could also be utilized to understand the long-term effects of H_2 and impurities on the reservoir quality.

- Technically, geological formations chosen for the UHS project are evaluated based on (i) capacity (i.e., total usable storage volume of a site), (ii) injectivity (i.e., ease of fluids flow through the pore throats), (iii) trapping mechanisms and (iv) confinement (i.e., capability to contain H_2 in the site) [40]. As a result, rock fluid interaction through wettability studies can be used to provide insights into depleted gas reservoir conditions, particularly on trapping mechanisms. For instance, recent studies for rock/brine/H_2 systems (e.g. see refs. [25,26,61,66,76,107,230–237]) have investigated the effect of wettability using different rock minerals. While these studies mostly simulate aquifer storage conditions, more research is needed, focusing on simulating a depleted reservoir scenario [238,239], the impact of wettability changes, and the common trapping mechanism for H_2 immobilization (see **section 2.2**).
- In general, while the review technically proves the feasibility of H_2 storage in a depleted gas reservoir, low public awareness has slowed the deployment of the technology. As a result, the ethical implications of H_2 storage development need to be further evaluated, and more efficient mechanisms should be adopted to promote public acceptance of the technology.
- Lastly, pilot projects of UHS in depleted gas reservoirs are limited to Austria and Argentina, whereas potential sites, as shown in Table 2 (based on this review), are limited to Poland, Scotland, Germany, Netherlands, Norway, the UK, Canada, USA, and China. While natural gas consumption is constantly increasing, the gas reservoir is also expected to deplete accordingly. Hence, more preliminary studies on UHS must be performed worldwide to reduce the usage of fossil fuels and greenhouse gas emissions to meet the world's growing energy demands by 2050.

8. Conclusions

Hydrogen as an energy carrier is a novel technology, that is constantly improving through ongoing research, particularly on its production and storage. Porous media (aquifers and depleted reservoirs) are used for underground storage as they provide massive storage capacity. This article thus provides an overview of H_2 storage in depleted gas reservoirs. It quickly compares the properties of H_2 to those of CH_4 and CO_2, as well as generic gas trapping methods. Briefly, operating, and potential global depleted facilities and case studies were also covered. It further discussed how the thermophysical properties of H_2 (such as density, viscosity, diffusivity, and solubility) and petrophysical properties (i.e., porosity and permeability) influence storage and withdrawal capacity, injectivity, gas immobilization, and efficient gas containment. Lastly, an economic insight into depleted reservoirs in comparison to aquifers, and salt caverns was also covered, as well as the unique obstacles related to well integrity during gas storage. Based on the review, the major conclusions are summarized below:

- Although H_2 storage in the subsurface can be achieved through different means, the depleted gas reservoir is still the most viable choice due to economic factors, its wide geographical distribution, as well as its proven technical credibility based on the existing natural gas storage facilities. More so, depleted gas reservoirs have fewer uncertainties, thus generating less challenge during site evaluation as compared to aquifers.
- Many factors have been attributed to H_2 loss. However, hydrodynamics, geochemical, and microbial aspects are often considered the principal promoters of H_2 loss in the subsurface, especially in the depleted gas scenario. For instance, the loss of H_2 via hydrodynamics is majorly dependent on density, viscosity, diffusivity, and solubility whereas both geochemical and microbial can be attributed to the petrophysical properties (permeability and porosity) and the controlling environmental factors (particularly temperature, salinity, and pressure) as well as the native gas/brine compositions.
- Reservoir geometry, injection design, and pattern can effectively reduce the loss of H_2 as most depleted gas studies observed little or no presence of viscous fingering compared to some aquifer mediums.
- During withdrawal, gas mixing in depleted gas reservoirs can be reduced by correctly understanding the effect of gravity override and reservoir structure (geometry) and the interplay of injection/withdrawal schemes. In addition, cushion gas's type, and concentration, can significantly reduce the effect of H_2 mixing during storage. Furthermore, biological transformation and the in situ H_2-CH_4 cushion gas mixing can influence storage cycles, gas quality, and security.
- Experimental work often used sandstone samples to study the effect of mineral alteration, precipitation, and the overall reservoir integrity and connection within the reservoir pores of the rock by porosity and permeability measurements and through microscopy or computer tomography analysis in laboratory scales.
- Simulation studies have been based on investigating the effects of injection/production rates, operating time, cushion gas composition, storage cycle duration, and sensitivity analysis on cushion gas fractions as it affects the reservoir properties to determine the optimum design for H_2 storage in the gas reservoir.
- The well integrity plays a vital role in safeguarding the quality of gas injection and withdrawal during underground storage operations. If affected, it can result in several leakage problems due to microbial corrosion, embrittlement (in the case of H_2), cementing effect, sulfidation, elastomer failures, caprock sealing failures, and purity of the injected gas.
- Challenges of gas storage in depleted gas reservoirs are inevitable. However, proper planning during the selection of gas reservoir, cushion gas, injection strategy, duration, and practical information on the site characteristics will significantly reduce the associated risk as it could be a pragmatic solution for seasonal and large-scale H_2 storage.

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.

Data availability

No data was used for the research described in the article.

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