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Gas storage in geological formations: A comparative review on carbon dioxide and hydrogen storage

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ABSTRACT

Carbon dioxide and hydrogen storage in geological formations at Gt scale are two promising strategies toward net-zero carbon emissions. To date, investigations into underground hydrogen storage (UHS) remain relatively limited in comparison to the more established knowledge body of underground carbon dioxide storage (UCS). Despite their analogous physical processes can be used for accelerating the advancements in UHS technology, the existing distinctions possibly may hinder direct applicability. This review therefore contributes to advancing our fundamental understanding on the key differences between UCS and UHS through multi-scale comparisons. These comparisons encompass key factors influencing underground gas storage, including storage media, trapping mechanisms, respective fluid properties, petrophysical properties, and injection scenarios. They provide guidance for the conversion of our existing knowledge from UCS to UHS, emphasizing the necessity of incorporating these factors relevant to their trapping and loss mechanisms. The article also outlines future directions to address the crucial knowledge gaps identified, aiming to enhance the utilisation of geological formations for hydrogen and carbon dioxide storage.

1. Introduction

The escalating global population and rapid pace of urbanisation have triggered surging energy consumption and depleted fossil fuel reserves at unsustainable rates [1–3]. The usage of fossil fuels has been linked to unprecedentedly high carbon dioxide (CO_2) emissions, recognized as the principal catalyst of climate change [4]. Current policies alone are projected to be insufficient in meeting the 2015 Paris Agreement [5]. Urgent challenges in reducing net CO_2 emissions necessitate immediate strategies for large-scale carbon capture and storage (CCS) [6,7]. Additionally, there is a need to transition to alternative, environmentally friendly energy resources to diminish our dependency on fossil fuels [8,9].

Traditional renewables such as wind and solar energy often suffer from intermittent, hindering their reliability as stable sources [10], thus requiring large-scale energy storage systems. To mitigate this issue, green hydrogen (H_2) has gained prominence as an exceptional energy carrier. It can be produced using renewable energy sources during times of excess energy generation and subsequently fed back into the electrical grid as needed [11,12]. By 2050, H_2 could potentially serve as a viable alternative to natural gas and solid fuels in the evolving energy landscape [13].

When it comes to storing large volumes of CO_2 and H_2 on a Gt scale, recent research points to geological formations as highly effective and practical options [14,15] Currently, three conventional storage media are commonly used: depleted oil and gas reservoirs, deep aquifers, and salt caverns [16]. The use of the first two types of formations is essential to meet the aforementioned storage needs owing to their broad geographical distribution and effective operational characteristics. These formations have been typically treated as porous media, whose

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petrophysical properties (*e.g.*, porosity, permeability and structural integrity) are pivotal in both design and operational stages.

Currently, investigations into underground hydrogen storage (UHS) remain relatively limited in comparison to the more established body of knowledge of underground carbon dioxide storage (UCS). Due to the analogous physical processes, specifically the complex multiphase interactions inherent to both systems, drawing on the experiences in UCS could potentially accelerate the advancements in UHS technology. However, it is crucial to acknowledge and highlight there exist distinctions encompassing several key aspects, including (1) the intended purpose of storage, whether it is permanent or intermittent; (2) the selection of geological storage media; (3) key trapping mechanisms; (4) relevant fluid and petrophysical properties; and (5) requisites for surface infrastructure and well design, as elaborated in Fig. 1. While the insights gained from UCS experiences undoubtedly could contribute valuable knowledge, their direct applicability to UHS is circumscribed by the inherent differences, which can impose constraints that warrant careful consideration.

This review offers a comparative overview of UCS and UHS, featuring the key differences in their storage media, trapping mechanisms, fluid properties and petrophysical properties, covering the past endeavours and current state-of-the-art developments. The discussion starts with addressing geological storage media and comparing their characteristics across different contexts. Based on different storage scenarios relevant to UCS and UHS, the trapping mechanisms and injection strategies are discussed highlighting the respective influences on storage and withdraw efficiency. Following this, the review delves into a detailed comparison of material features, including physical, interfacial, and geo-/bio-chemical properties, contributing to underground gas storage. Finally, the review provides concluding remarks, highlights research gaps, and outlines future directions.

2. CO$_2$ and H$_2$ storage in geological formations

2.1. Storage media

In this section, three main types of underground gas storage media are discussed: depleted oil and gas reservoirs, deep aquifers, and salt caverns [16]. It should be noted that there exist additional requirements for UHS (*i.e.*, cushion gas) to maintain reservoir pressure at a sufficient level during hydrogen extraction. Table 1 offers a comparison of characteristics between UHS and UCS across these geological formations.

2.1.1. Depleted oil and gas reservoirs

Depleted oil and gas reservoirs offer several advantages that make them well-suited. First, oil and gas initially trapped have remained contained for countless millions of years, guaranteeing storage integrity. Second, most of these reservoirs have been rigorously characterized through geological studies. Third, the existing infrastructures could be adapted for storage purposes [23]. However, the storage capacity must be carefully managed to avoid compromising the caprock due to excessive pressure [39]. Additionally, in some locations, immediate injection may not be feasible due to strategically placed cement plugs [48].

For UCS, depleted oil and gas reservoirs stand out as one of the most promising options for permanent, secure, and economically viable storage sites [49]. Beyond the previously outlined advantages, three additional benefits are notable: 1) they offer the necessary high temperature and pressure to maintain CO$_2$ in a supercritical state [29]; 2) The use of Enhanced Oil Recovery (EOR) techniques. Injecting CO$_2$ is able to extract heavy hydrocarbons from the oil phase and accelerate the oil mobility through oil swelling and reducing oil viscosity [35]. Essentially, there are two types of oil recovery techniques: miscible oil recovery (requiring reaching the minimum miscibility pressure, suitable for oil gravity more than 25° API and depth larger than 915 m) and immiscible oil recovery (suitable for depth less than 915 m or oil gravities between 17.5° and 25° API) [50]; 3) Potential for enhanced methane (CH$_4$) recovery in unconventional reservoirs (*e.g.*, Shale gas), attributed to the preferential absorption of CO$_2$ over CH$_4$ [36].

For UHS, they offer wider distribution and greater volume compared to salt caverns (discussed in Section 2.1.3), and do not necessitate artificial construction [16]. When compared to deep aquifers (discussed in Section 2.1.2), the requirement for cushion gas is minimal as the remaining gas allows for reduced cushion gas injection [26].
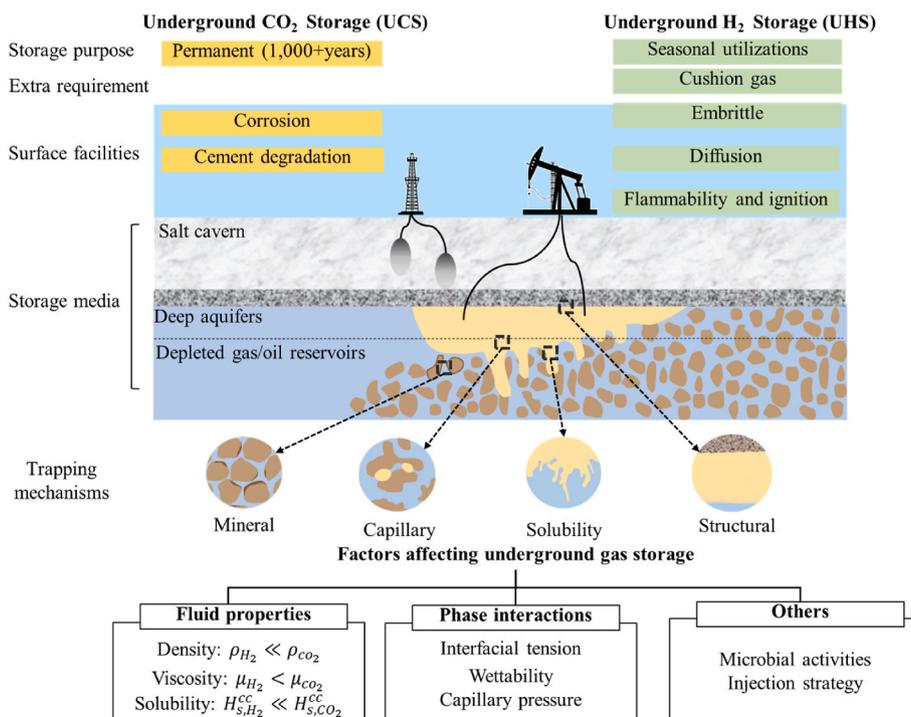

Fig. 1. Comparisons between underground carbon dioxide and hydrogen storage.

Table 1
Characteristics of UHS and UCS across different geological storage media.

		Depleted oil and gas reservoirs	Deep aquifers	Salt caverns
Depth (m)		300-2700 [17]	400-2300 [17]	300-1800 [17]
Operation pressure (MPa)		1.5–28.5 [17]	3–31.5 [17]	3.5–27 [17,18]
Flexibility of cycling		1-2/year, seasonal [19]	1-2/year, seasonal [19]	10/year, peaking [19]
Suitability	CO_2	Proven [20]	Unproven [7]	Proven [21]
	H_2	Unproven [22]	Unproven [17]	Proven [23]
Capital cost	CO_2	Low [24,25]	Medium [24,25]	High [24,25]
(relative)	H_2	Low (1.19 $/kg) [26,27]	Medium (1.21 $/kg) [26,27]	High (1.54 $/kg) [26,27]
Storage capacity for H_2		Medium (50%–60%) [28,24]	Low (20%–50%) [28,24]	High (70%) [28,24]
Advantages	Common	● Well characterized geologically [23,29] ● Proven tightness and integrity [23,29] ● Pre-existence installations [23,29]	● Large capacity [16,20] ● Widespread availability [16,20]	● Storage integrity, able to prevent leakage due to low porosity and permeability [30,31] ● Prevent the fracture formation as possible for the escape paths due to salt ductile [32,33,34]
	CO_2	● Possible to enhance residual oil or CH_4 recovery [35,36]	● Possible to extend storage duration due to stable carbonates formation [24]	/
	H_2	● Remaining gas can be used as cushion gas [26] ● Large storage capacity and widespread [16]	/	● Chemical inertia towards hydrogen [37] ● Suppress consumption of microbes [16,38]
Disadvantages	Common	● Multiphase flow complications associated with residual hydrocarbon [25] ● May not be available for immediate injection [39]	● Lack of pre-existence of surface and subsurface installation [19] ● Unknow storage integrity [40]	● Limited geographically [19] ● Limited storage scale (i.e., at most 10^8 m^3 at surface conditions) [19] ● Relatively high capital cost [27]
	CO_2	● Possibly cause corrosion of gas pipelines and cement degradation [41]		
	H_2	● Equipment needs to be reassessed due to hydrogen embrittlement [28] ● Remaining gas/oil in the reservoir can affect the purity of the H_2 [42] ● H_2 loss due to geochemical and microbial reactions [43]	● Require gas drying infrastructure [24]	● Require gas drying infrastructure due to brine accumulating induced water vapor [44]
Remarks	CO_2	● Storage capacity reduced to prevent caprock fractures due to exceeding pressures [39]	● Potential to store up to 10,000 Gt of CO_2 globally [45]	/
	H_2	● Pilot project are in the process of investigating blends, with plans to eventually store pure hydrogen by 2030 [22]	● Aquifer hydrogen storage field operation is limited to impure hydrogen (natural gas with 50–60% hydrogen) [46,47]	● Facing issues: water usage for leaching [23] and brine disposal [44]

Researchers have conducted simulation-based studies, providing positive indications regarding the feasibility [22]. Also, many countries have exhibited diligence in exploring the impacts of UHS within depleted oil and gas reservoirs, exemplified by the implementation of pilot projects such as HyStorPor [12], SUN. STORAGE [51], and Hychico [52]. However, their suitability for hydrogen storage and production is not fully evolved [22]. The residual hydrocarbons can mix with injected H_2, which may affect its purity [42]. Moreover, the reactivity of H_2 spanning both geochemical and microbial reactions possibly lead to H_2 loss [43]. Additionally, the current infrastructure might necessitate the substitution of higher-grade steel considering hydrogen-induced embrittlement [28]. To date, pure hydrogen has not yet been stored in depleted gas fields. Some mixtures of natural gas and hydrogen (up to 50%) have been found in depleted gas reservoirs [17]. RAG Austria conducted the Underground Sun Storage pilot project, which involved testing the storage of a mixture containing 10% hydrogen and 90% methane. They are now in the process of investigating a wide range of admixtures, with plans to eventually store pure hydrogen. The first operational storage of pure hydrogen in depleted reservoirs is expected in 2030 [51].

2.1.2. Deep aquifers

Aquifers are another popular choice for gas storage, primarily due to their widespread availability and their common feature of porous rock storage. However, unlike depleted oil and gas fields, which have a proven track record as secure traps, aquifers usually require geological surveys to confirm the caprock integrity [40]. Moreover, there is a limited availability of geological data pertaining to brine deposits, and these brine formations have never undergone assessment for the storage of substantial quantities of highly pressurized gas, contributing to the inherent risks for gas storage [53]. In addition, undeveloped aquifers lack pre-existing well infrastructures. The additional investment in both

geological surveys and well infrastructures render aquifers more financially demanding than depleted oil and gas fields [19].

In the context of UCS, the discovered aquifers hold the potential to globally store CO_2 up to 10,000 Gt based on the upper estimation made by IPCC [45]. Ideally, chemical interactions between absorbed CO_2 and surrounding rock could foster the formation of highly stable carbonates, potentially extending its storage durations [24]. However, on the other hand, bulk injections can acidify reservoir waters due to the formation of carbonic acid [54]. Such acidification process could result in the dissolution of carbonate minerals (e.g., dolomite) and silicate minerals (e.g., K-feldspar), which consequently can lead to an increase in permeability. This imposes a greater risk of gas leakage, drinking water pollution as well as the cement degradation and well corrosion [55,41,56].

For UHS, in comparison to depleted gas reservoirs, a larger amount of cushion gas (up to 80% of storage volume) is typically required to prevent gas trapping [17]. In contrast to salt caverns, deep aquifers offer higher geographical accessibilities and storage capacities. Gas stored in aquifers often contains water impurities, necessitating gas drying infrastructure as a key component of the gas treatment process [24]. The cushion gas and drying infrastructure are also additional capital costs. Despite these added expenses, aquifers are still generally more cost-effective to develop than salt caverns, as construction of storage space itself is not required [26]. To date, aquifer hydrogen storage field operation is limited to impure hydrogen (natural gas with 50–60% hydrogen) storage projects worldwide: Engelbostel and Bad Lauchstadt in Germany, Beynes in France, and Lobodice in the Czech Republic [46, 47].

2.1.3. Salt caverns

Salt caverns present a suitable solution for gas storage owing to their safety features and prolonged operational viability [30,21]. However,

due to their relatively high development cost, limited geographic availability and limited capacity (*e.g.*, salt cavern: 0.25 Mt/year, Crtionelle in USA; aquifer: 4.9 Mt/year, Don Valley in UK; Depleted oil/gas reservoir: 1 Mt/year, Lianyun Gang, China), they are not typical storage media for CO_2 [57]. While salt caverns are the only proven UHS technology so far [23]. Therefore, the scope of the discussion here is exclusively centred on H_2 storage. The walls of salt caverns serve as highly effective barriers, impermeable to H_2, thus preventing leakage [31]. Additionally, the ductile nature of the salt itself acts as a deterrent against the development of fractures through which gas escape [32]. These caverns possess several other advantageous qualities including chemical inertness towards hydrogen [37], and suppressing hydrogen consumption by microbes owing to the elevated salt content and limited water availability [16,38]. These inherent properties provide a robust foundation for the long-term stability and security of hydrogen storage. Despite these strengths, there are some constraints to consider. Firstly, the geographical availability is limited and the storage scale is relatively modest, with volumes reaching a maximum of around 10^8 m^3, which falls considerably short of the global operational requirement at the Gt scale [19]. Also, they require artificial construction resulting in relatively high capital cost [27]. Additionally, the accumulation of brine at base leads to an increase in water vapor content within stored H_2 [44], necessitating a surface drying system to dehydrate the gas during production. Cushion gas is also required to maintain pressure, generally comprising 30% of the total capacity [51]. Salt caverns have successfully stored pure hydrogen in the United Kingdom since the 1970s and in the United States since the 1980s [17].

2.2. Trapping mechanisms

The immobilization of gas includes four major trapping mechanisms (as shown in Fig. 1): structural/stratigraphic trapping, capillary/residual trapping, solubility/dissolution trapping, and mineral trapping, which have been extensively researched in the context of carbon dioxide storage [58–61] and, to some extent, in underground hydrogen storage. In this section, the trapping mechanisms and some of the noteworthy recent developments will be discussed. The detailed comparisons are shown in Table 2.

2.2.1. Structural/stratigraphic trapping

Structural/stratigraphic trapping can be exploited in geological sequestration, to keep gas safe below for centuries [62]. When gas is introduced into geological storage formations, it tends to move upward on account of buoyancy force [63], and laterally via preferential pathways until it reaches a caprock, fault, or other sealed discontinuity [64]. Technically, the caprock is porous but has remarkably low permeability, which helps to prohibit the upward motion of buoyant gas [65–67]. Nevertheless, capillary leakage might happen when the buoyancy force surpasses a certain threshold, which affects gas storage capacities and security [68,69]. The capillary pressure-buoyancy pressure equilibrium can be utilized to calculate the maximum gas column height h_{max} [70, 71]. The estimation h_{max} for CO_2 and H_2 at a common storage condition (*e.g.*, at 10 MPa and 323 K) is 80 m and 41 m, respectively [72]. h_{max} is highly relevant to radius of the capillary(r), interfacial tension ($\gamma_{(gas-liquild)}$) and contact angle (θ), it is necessary to determine these parameters accurately at reservoir conditions for reliable evaluations of underground gas storage projects. In a recent study, it demonstrates that h_{max} fluctuates with storage depth [58]. However, a lower depth barrier exists below which CO_2 cannot be stored permanently through structural trapping because, at approximately 15,000 m depth, density reversal (CO_2 is heavier than brine) occurs [73]. Based on this hypothesis, the maximum storage depth in UHS settings was investigated [39]. It was found that a maximum storage depth for H_2 is 1100 m.

Table 2
Summary of trapping mechanisms of UCS and UHS.

	CO_2	H_2
Structural trapping		Mechanism: capillary resistance force of seal caprock is larger than buoyancy force [55]
		🙂Maximum gas column height: 1100 m [18]
	🙂Maximum gas column height: 15,000 m [66]	🙁The injected H_2 will sweep the pore fluids and concentrate below the caprock more rapidly [50]
		🙁Fingering due to fluid mobility contrast, and gravity override due to fluid densities differences might happen [106]
Capillary trapping		Mechanism: entrapment of gas in porous medium by capillary force [67]
		🙂Capillary trapped H_2 might be recoverable by injecting cushion gas [72]
	🙂Recognized as the most efficient mechanism to storage security [70,71]	🙂Trapped hydrogen can enhance gas connectivity leading to more efficient withdrawal [73]
		🙁High pressure results in higher H_2 loss [74]
Dissolution trapping		Mechanism: gas dissolving in formation liquid [75]
		🙁Will lead to H_2 loss [82]
	🙂Solubility reduces as the temperature and salinity rise and increases with pressure [107]	😐Dissolution decreases with increase in brine salinity but increases with pressure and temperature [108]
Mineral trapping		Mechanism: the incorporation of gas into a stable mineral phase by interactions with different minerals and organic components [83]
	🙂The most reliable CO_2 trapping mechanism for long-term storage [91]	🙁Will lead to H_2 loss [65]
	😐Mainly acid-base reactions (i. e., carbonic acid formation) [192]	😐Mainly redox reactions (i. e., abiotic and biochemical reactions) [17,93]
	😐Divalent cation-containing geological formations are more suitable [88]	🙁Possible lead to the change in rock mechanical stability, which further affect storage capacity and containment security [94]

Notes: 🙂: Positive effects; 🙁: Negative effects; 😐: Explanation/No effects.

2.2.2. Capillary/residual trapping

Capillary/residual trapping encompasses the entrapment of a fluid within a porous media due to capillary forces [74]. In underground gas storage, it refers to injected gas becoming detached and immobilized [75]. The initial stage involves displacing the wetting phase (*e.g.*, brine)

with gas as it is first injected into the reservoir. Following injection cessation, the gas moves in two directions: upwards due to density variations and laterally due to capillary forces. Subsequently, the wetting phase seeks to occupy throats (*i.e.*, imbibition process). As a consequence, wetting layers within the throat expand until they completely fill the throat, forming unconnected clusters trapped within pore bodies (*i.e.*, snap-off process) [76].

In UCS, both the extent and rate of capillary trapping made it the most efficient mechanism to storage security during the first decade [77, 20]. In contrast, regarding UHS, residual trapping could result in H_2 loss during withdraw. As the injected H_2 needs to be utilized again in the future, it is crucial to minimize its residual saturation. Currently, it was found that injecting cushion gas renders the trapped H_2 recoverable during subsequent cycles [78]. Moreover, Zhang *et al.* (2023) [79] indicates that trapped hydrogen can significantly enhance gas connectivity over time, resulting in reduced hysteresis and more efficient withdrawal. Thaysen *et al.* (2022) [80] documented a residual H_2 trapping of 20% at 2 MPa, escalating to 43% at 7 MPa in Clashash sandstone. This illustrates that deeper reservoir (*i.e.*, high pressure) might be less appropriate for hydrogen storage.

2.2.3. Solubility/dissolution trapping

The phenomenon wherein gas dissolves into the formation liquid is referred to solubility/dissolution trapping [81]. Upon gas encountering formation brine in deep saline aquifers or hydrocarbons in depleted oil reservoirs, mass transfer happens as the gas dissolves into the formation liquid, ultimately reaching an equilibrium state [82]. This dissolution results in a minor density increase in the formation liquid, prompting downward flow due to gravitational instability [45,83], which enhances the mixing of the formation liquid and gas, and expediting the diffusion process [84]. The mechanism contributes to two primary effects: curbing the upward gas migration and increasing the storage capacity of the geological formation [57,85]. Although the dissolution has been recognized as a significant storage mechanism for UCS [86], it is important to recognize that gas dissolution is a gradual process due to a low molecular diffusion coefficient, taking thousands of years for complete dissolution [87]. For UHS, the amplified trapping capacity may lead to H_2 loss, necessitating careful consideration for future applications [88]. It is discussed in detail in Section 3.1.3.

2.2.4. Mineral trapping

Mineral trapping is the incorporation of gas into a stable mineral phase by interactions with different minerals and organic components, which requires a long period of geological timescale [89–91].

In the context of UCS, several geochemistry reactions lead to the entrapment of CO_2 in the form of dissolved species and the formation of carbonate minerals [63]. An illustrative instance of CO_2 mineral trapping in a H_2O-rich environment is as follows: Initially, CO_2 dissolves into the aqueous phase, resulting in the formation of four distinct "carbonate

species" [CO_2(aq), H_2CO_3(aq), HCO_3^- (aq) and CO_3^{2-} (aq)] and H^+ as a byproduct [54,92]. Subsequently, an increase in acidity triggers the dissolution of primary rock minerals containing metal ions, often encompassing Ca^{2+}, Fe^{2+}, Mg^{2+}, and Al^{2+}, leading to a pH shift towards basic conditions [93,94]. This kind of pH swing accelerates the precipitation of CO_2-derived carbonates, such as dawsonite, ankerite, and siderite [95]. These reactions typically follow a predictive sequence, as depicted in the provided Table 3. Currently, mineral trapping is regarded as the most dependable mechanism for ensuring long-term UCS [15, 96]. Nonetheless, certain processes, notably dehydration mechanisms and the dissolution of minerals like dolomite, K-feldspar, and potentially sheet silicates, could facilitate CO_2 migration and necessitate careful consideration [56].

In contrast, the mineral trapping of H_2 involves redox reactions (*i.e.*, abiotic and biotic reactions) [23,99]. Both reactions can alter changes in formation permeability, which could subsequently impact storage capacity and security [100]. Regarding abiotic reactions, recent studies highlight that quartz and K-feldspar exhibit low reactivity, whereas pyrite, hematite, and possibly other sulphide and iron-based minerals with high oxidation states manifest reactivity [101–103]. To elaborate further, the dissolution of pyrite, owing to its thermodynamically unstable nature in the presence of hydrogen, emerges as a principal abiotic reaction during hydrogen storage [104]. As of now, the data regarding abiotic reaction remains scarce, and additional minerals necessitate assessment of their chemical reactivity. As for biotic reactions, it is typically catalysed by microorganisms [105,106]. Within the context of mineral trapping, two pivotal actions come into play (others are discussed in Section 3.4): sulphate reduction and ferric reduction [97]. These reactions will lead to permanent H_2 loss, which is clearly unfavourable [72]. The corresponding reaction equations are outlined in the provided Table 3.

3. Factors affecting underground gas storage

This section will discuss factors affecting underground gas storage in detail. For comparison purposes, Table 4 indicates some properties of CO_2 and H_2, relevant to ambient and reservoir conditions.

3.1. Fluid properties

3.1.1. Density

The considerable difference in densities (ρ) can lead to pronounced gravitational segregation, potentially resulting in gas leakage [122, 123]. In the case of underground gas storage, depth-related increases in pressure and temperature contribute significantly to alterations in gas density [58]. Hence, comprehending the influence of these two parameters on fluid density becomes pivotal [124]. As indicated in Fig. 2 (a), ρ_{H_2} is less sensitive to pressure and temperature effects and ρ_{H_2} is notably lower than ρ_{CO_2} under equivalent thermo-physical conditions.

Table 3
Summary of major geochemical and microbial reactions in UCS and UHS [97,98].

Gas type	Reaction		Reaction equation	Remarks	
CO2	Dissolution of CO_2		$CO_2(g) + H_2O = H^+ + HCO_3^-$	Equilibrium constant	Reactions Order
	Dissolution of Carbonate Minerals		$CaCO_3(s) + H^+ = HCO_3^- + Ca^{2+}$	1.0×10^{-7} mol/m²/s	
	Dissolution of Silicate Minerals		$K-feldspar \rightarrow 3SiO_2(aq) + AlO_2^- + K^+$	10^{-13}-10^{-12} mol/m²/s	
			$Chlorite \rightarrow 3SiO_2(aq) + AlO_2^- + 2.5Fe^{2+} + 2.5Mg^{2+} - 8H^+ + 8H_2O$		
	Precipitation		$Dawsonite \rightarrow 3SiO_2(aq) + AlO_2^- + Na^+ + HCO_3^- - H^+$	1.0×10^{-7} mol/m²/s	
			$Siderite \rightarrow Fe^{2+} + HCO_3^- - H^+$		
			$Ankerite \rightarrow 0.3Mg^{2+} + 0.7Fe^{2+} + Ca^{2+} + 2HCO_3^- - 2H^+$		
	Precipitation of Silica		$SiO_2(aq) \rightarrow SiO_2(s)$	1.0×10^{-14} mol/m²/s	
H2	Abiotic reaction		$FeS_2 + (1-x)H_2 = FeS_1 + (1-x)H_2S$	Discussed in mineral trapping of UHS (Section 2.2.4)	
	Biotic reaction	Sulphate reduction	$SO_4^{2-} + 5H_2 = H_2S + 4H_2O$		
		Ferric reduction	$H_2 + 3Fe_2O_3 = 2Fe_3O_4 + H_2O$		
		Methanogenesis	$4H_2 + CO_2 = CH_4 + 2H_2O$	Discussed in microbial activity (Section 3.4)	
		Acidogenesis	$4H_2 + 2CO_2 = CH_3COOH + 2H_2O$		

Table 4
Properties H_2 and CO_2.

Properties		H_2	CO_2
Suitable storage conditions		5–30 MPa & 26.85–126.85 °C [107]	Above critical state and below 800 m [108]
Molecular weight (g/mol)		2.016 [107]	44.0095 [107]
Critical temperature (°C)		−239.97 [109]	31.1 [110]
Critical Pressure (MPa)		1.28 [109]	7.38 [110]
Density (Kg/m^3)	SATP	0.082 [111]	1.795 [19]
	1-km depth	4.32 [112]	397.4 [113]
Viscosity (10^{-6} Pa s)	SATP	8.90 [16]	8.36 [19]; 14.9 [110]
	50 °C and 10 MPa	9.54 [112]	29.47 [113]
Rock-gas interfacial tension (mN/m)	Clean quartz, 1 km-depth	98.93 [114]	32.30 [114]
	Basaltic rock, 1 km-depth	75.26 [114]	26.07 [114]
Contact angle (°)	Clean quartz, 1 km-depth	20.46 [115]	31.87 [116]
Solubility in pure water (g/L)	SATP	1.6×10^{-3} [110]	1.45 [110]; 1.49 [117]
	1-km depth	0.148 [118]	50.31 [117]
Diffusion coefficient in pure water (m^2/s)	SATP	5.13×10^{-9} [119]	1.9×10^{-9} [120]
	1-km depth	–	3.94×10^{-9} [121]
Diffusion coefficient in air (m^2/s)	SATP	0.61×10^{-5} [19]	1.39×10^{-5} [19]
	1-km depth	–	–

Notes: SATP represents standard ambient temperature and pressure (*i.e.*, 25 °C and 0.1 MPa); 1 km-depth represents field condition (*i.e.*, 50 °C and 10 MPa).

Consequently, the large H2-liquid density difference will lead to strong gravity segregation and accumulation beneath the caprock at a faster rate compared to CO_2 [44]. A notable risk of H_2 leakage emerges, underscoring the imperative need to ensure caprock integrity [110].

3.1.2. Viscosity

Fluid viscosity (μ), defined as the ratio of shear stress between adjacent fluid layers and the velocity gradient in the normal direction, quantifies the ability of a fluid to flow [127]. Typically, it is combined with the relative permeability ratio, known as the mobility ratio, to evaluate injection and withdraw efficiency and fluid-fluid interfacial stability. Elevated mobility will lead to the occurrence of viscous fingering [128,129], which can trigger early gas surface leakage and low gas sweep efficiency [130]. Hence, it is recommended to control gas viscosity to optimize sequestration projects. As depicted in Fig. 2(b), μ_{H_2} is slightly influenced by temperature and pressure. In contrast, μ_{CO_2} experiences a more substantial increase with pressure and it can be one order of magnitude greater at high pressure due to greater collision and friction between denser gas molecules [131]. Further, μ_{CO_2} decreases with increasing temperature at higher pressure. Notably, μ_{H_2} is approximately one to two orders of magnitude lower than μ_{H_2O} under reservoir conditions. This divergence implies higher likelihood of H_2 viscous fingering during the injection phase. To counteract this effect, it is advisable to consider reducing the injection rate or increasing μ_{H_2} (*e. g.*, H_2-foam formation) [132]. On the other hand, according to Darcy's law, the increase of viscosity is associated with greater viscous resistance, which can have an adverse effect on the inject and withdraw energy efficiency during operation. This aspect should be considered when planning gas storage projects.

When considering the storage of CO_2/H_2 in depleted reservoirs, it is common for these gases to mix with light hydrocarbons (*e.g.*, CH_4). The density of these mixtures can be calculated using a modified ideal gas law that incorporates the compressibility factor, allowing for adjustments based on varying reservoir conditions [133]. Similarly, the viscosity of the mixtures can be estimated through the Chapman-Enskog theory, enhanced by the Lennard-Jones potential to precisely account for molecular interactions and their sensitivities to environmental conditions [134,135]. The variation in temperature and pressure under reservoir conditions can be considered by incorporating these factors in the theoretical calculations, whereas the spatial heterogeneity of the gas mixture is simplified.

3.1.3. Solubility

The evaluation of gas solubility in the formation liquid is essential, as it has the potential to impact both the trapping and withdrawal efficiency. In UCS, CO_2 dissolve in formation liquid can assist its permanent storage purpose [86]. For UHS, the extended trapping capacity may lead to H_2 losses, necessitating careful consideration for future applications [88].

The solubility of both CO_2 and H_2 primarily hinges on factors such as temperature, pressure, and salinity of the formation liquid [125], as shown in Fig. 2(c). Under reservoir conditions, the solubility of CO_2 and H_2 both increase with pressure and decreasing salinity. However, the trend regarding temperature is opposite: CO_2 dissolution decreases with rising temperature while H_2 dissolution rises with temperature [136, 125]. Moreover, the absolute solubility of H_2 in non-aqueous liquids is roughly an order of magnitude greater than that in aqueous liquids. This implies that the potential loss of H_2 due to dissolution trapping is expected to be more significant in depleted oil reservoirs compared to aquifers [137].

3.2. Capillary effects

Capillary pressure (P_c) determines the fluid configuration of multiphase flow in geological porous media, strongly affecting the residual gas saturation and the mobility of gas ganglia [138]. Fig. 3 shows the comparisons of $P_{c(H_2)}$ and $P_{c(CO_2)}$ under reservoir conditions. It indicates that the influence of pressure and temperature on $P_{c(H_2)}$ was insignificant, which is opposite to $P_{c(CO_2)}$. According to Young Laplace Equation, there exists two governing parameters: interfacial tension and wettability, which will be discussed in this section.

3.2.1. Interfacial tension

Fluid-fluid interfacial tension (γ_{FF}) is a function of pressure, temperature and fluid composition The relevant experimental data presented in Fig. 4(a) highlights noticeable contrast in the behaviour of CO_2–H_2O interfacial tension ($\gamma_{CO_2-H_2O}$) and H_2–H_2O interfacial tension ($\gamma_{H_2-H_2O}$). $\gamma_{CO_2-H_2O}$ shows notable decrease with rising pressure and a slight increase with temperature. $\gamma_{H_2-H_2O}$ remains relatively constant with changes in pressure but significantly decreases as temperature increases. Generally, with interfacial tension increasing, the residual gas saturation tends to rise as the non-wetting phase is more likely to traverse pore throats and effectively migrate across the media [142]. Jiang and Tsuji (2015) [143] conducted numerical simulation employing the lattice Boltzmann method, which suggest that high interfacial tension contributes to increased residual saturation and large size distribution of residual clusters. Experimentally, Kimbrel *et al.* (2015) [77] utilized computed x-ray microtomography (microCT) to assess immobilized nonwetting phase volumes, where a positive correlation between residual saturation and interfacial tension is found. However, contrasting conclusions are presented by Wildenschild *et al.* (2011) [144], where it is found that indicate that decreasing interfacial tension results in distinctly larger areas of residual trapped gas. They explain that the decrease in interfacial tension is expected to affect the non-wetting phase blob morphology towards smaller and more numerous blobs.

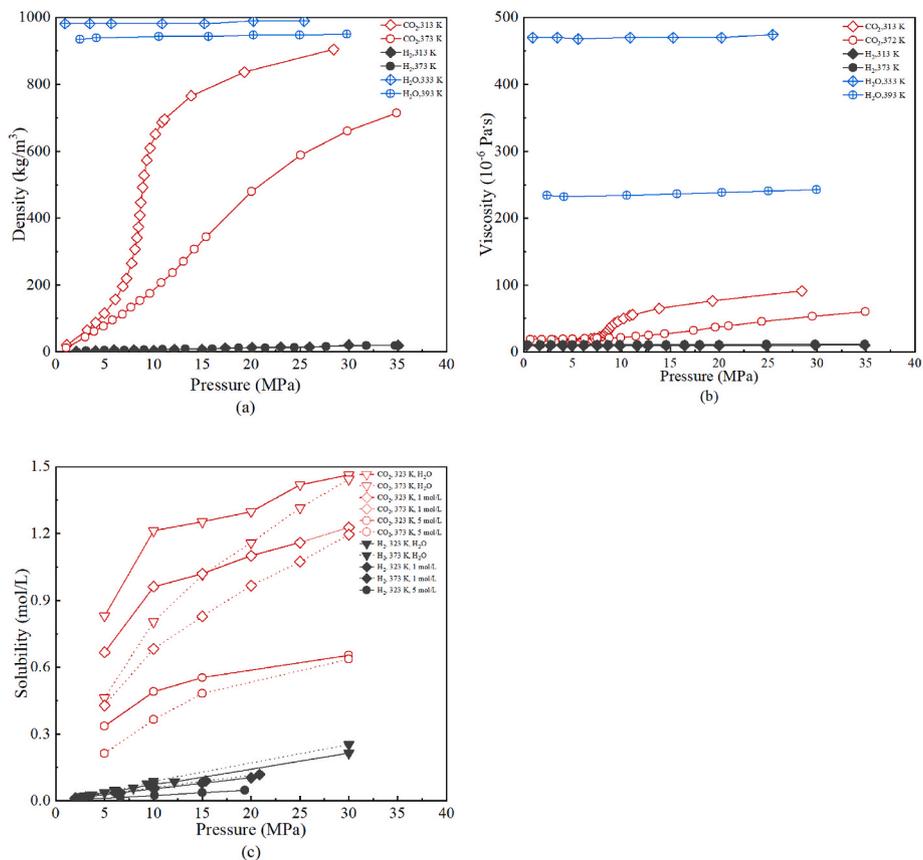As for solid-fluid interfacial tension, there is very limited data in the

Fig. 2. Comparisons of fluid properties: (a) Fluid density under different temperature and pressure (Data collected from Refs. [114,112,113]); (b) Fluid viscosity under different temperature and pressure (from Refs. [114,112,113]); (c) Solubility under different temperature, pressure and salinity (NaCl) (from Refs. [125, 118,126]).
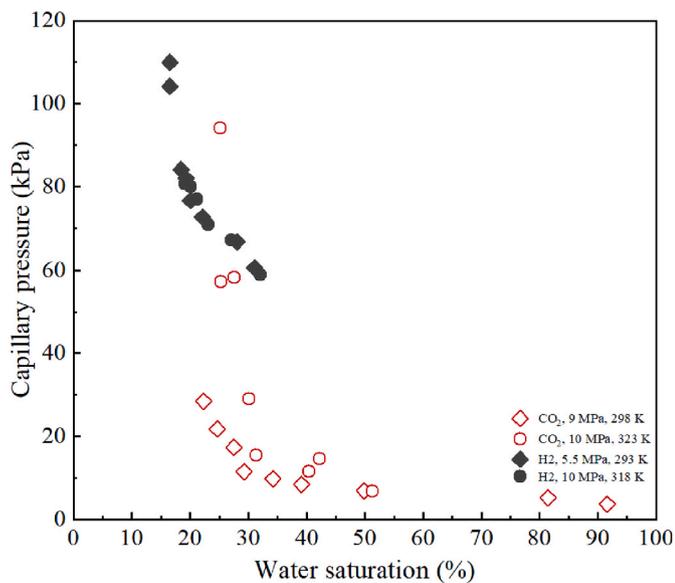

Fig. 3. Comparisons of capillary pressure under reservoir conditions (Data collected from Refs. [139–141]).

literature due to difficulty in measurement under *in situ* condition [147, 114]. Recently, technical advancements have enabled the prediction of the interfacial tension between rock-H_2 and rock-CO_2 using semi-empirical methods, as shown in Fig. 4(b).

3.2.2. Wettability

Wettability is the relative affinity of one liquid to a solid surface in the presence of another immiscible (or partially miscible) fluid, which is one of the key factors governing the distribution of fluid in a reservoir during gas flooding [148]. Though it can be quantified from interfacial tension through Young's equation, no experimental method reliably measures gas-rock interfacial tension (as discussed in Section 3.2.1). Instead, numerous approaches have been proposed to measure contact angles, including the sessile drop method [149], captive bubble method [150], tilting plate method [151], Wilhelmy plate method [152], capillary-rise method [153], and capillary penetration method [154]. The selection of a specific approach hinges on factors such as the geometry, the materials involved, and the thermodynamic state of the system [155].

Contact angle (θ) is influenced by three primary factors: pressure, temperature, and surface chemistry. Generally, θ_{CO_2} is larger than θ_{H_2} under equivalent conditions. The impacts of temperature and pressure are depicted in Fig. 5(a). The observed increase in contact angle with pressure can be attributed to the substantial rise in gas density, which leads to notably intensified gas-mineral intermolecular interactions and a significantly heightened gas-solid affinity [116,156]. As temperature increases, the contact angle also demonstrates an upward trend. This can be explained by the increased likelihood of hydrogen bonds breaking between water molecules and silanol groups on the quartz surface at elevated temperatures [157]. It should be noted that results discussed here are obtained from pure quartz. However, clean quartz does not exist in the subsurface, while stearic acid aged quartz-gas interfacial tension is more representative, as presented in Fig. 5(b). During gas cyclic injections, there exists receding contact angle corresponding to gas injection and advancing contact angle corresponding to gas
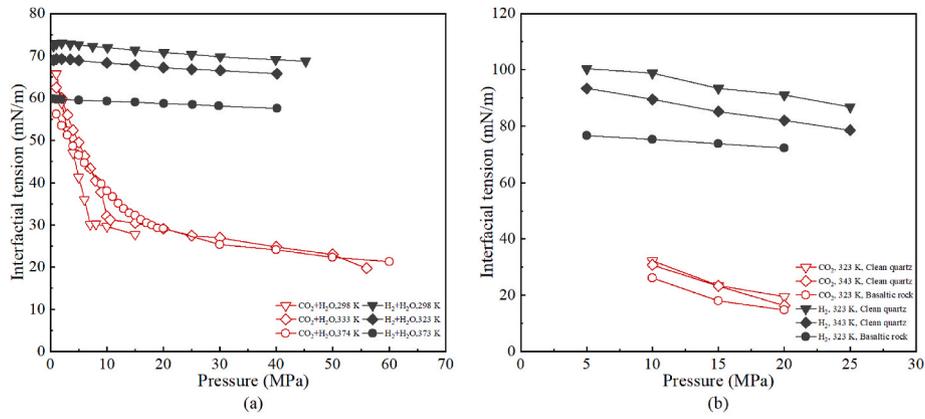
Fig. 4. Comparisons of interfacial tension (a) Fluid-fluid interfacial tension (Data collected from Refs. [145,146]) (b) Solid-fluid interfacial tension (from Ref. [114]).
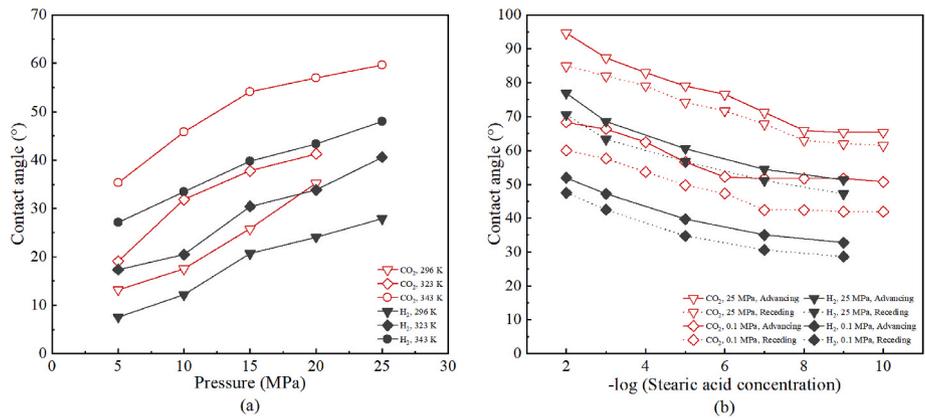

Fig. 5. Comparisons of contact angle: (a) Measured from pure quartz (Data collected from Refs. [116,115,159]); (b) Measured from stearic acid aged quartz (from Refs. [115,159]).

withdraw, which are also included in Fig. 5(b). It is evident that contact angle decreases monotonically with organic acid concentration. Remarkably, the adsorption of organic acids changes the surface into hydrophobicity even at extremely low concentrations. Consequently, this triggers the de-wetting process on the quartz surface [158].

3.3. Relative permeability

Relative permeability is the ratio of the effective permeability to the absolute permeability. As a point of note, effective permeability is the permeability of one fluid in a multi-fluid system, *i.e.*, permeability to a

fluid when its saturation is less than 100%. It is a key parameter since the construction of larger-scale gas storage model requires constitutive relationship between relative permeability and water saturation [160]. Fig. 6 presents the experimental data regarding relative permeability of both gas phase (*i.e.*, H_2 and CO_2) and liquid phase (*i.e.*, water and brine). All experiments are conducted on sandstone with steady-state technique.

As shown in Fig. 6(a), compared to CO_2-water/brine system, the relative permeability of H_2-water/brine system is less affected by temperature and pressure due to its relatively stable viscosity, as presented in Fig. 2(b). While the CO_2 relative permeability is more sensitive to
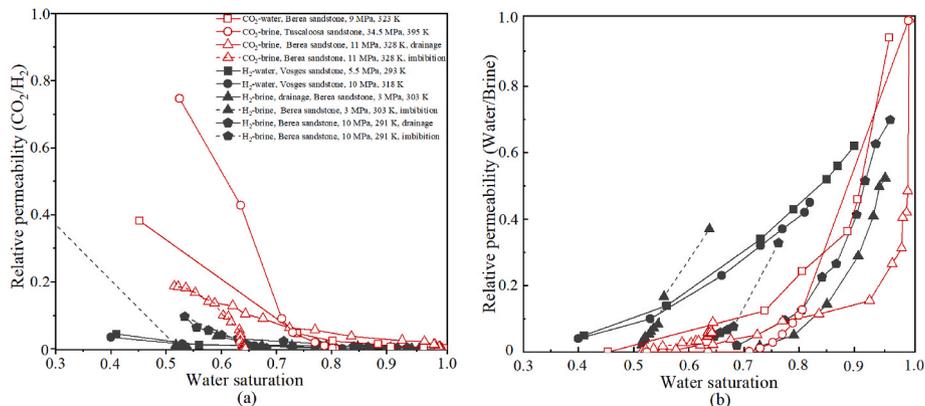

Fig. 6. Comparisons of relative permeability (Experimental data collected from Refs. [141,161–165]): (a) Gas phase; (b) Liquid phase.

changes in temperature and pressure. Notably, H_2 relative permeability is much lower than that of CO_2 under similar pressure and temperature conditions.

Hysteresis in relative permeability has a significant impact on the gas saturation distribution, gas recovery, water production, gas purity, and the reservoir pressure [166,167]. Fig. 6(b) highlights distinct wetting phase behaviours: The liquid phase relative permeability hysteresis for H_2 system is much stronger, which may be attributed to the differences in contact angle hysteresis behaviours (as shown in Fig. 5(b)). To be discussed later in Section 3.5, it is essential to accommodate intermittent injections for UHS, the strong hysteresis in liquid phase can affect its recovery efficiency. Bahrami *et al.* (2023) [167] demonstrates that hysteresis in relative permeability can result in a 16 %–25 % reduction in H_2 recovery factor. However, currently only few of UHS reservoir-scale simulation consider hysteresis, potentially neglecting the realistic multiphase flow behaviour [168,169]. Hence, the discussion here emphasizes the need for a thorough understanding regarding impacts of hysteresis in relative permeability.

3.4. Microbial activity

In addition to the biotic reactions mentioned in mineral trapping (Section 2.2.4), namely sulphate reduction and ferric reduction, there are two additional significant reactions that deserve attention: methanogenesis and acidogenesis, as outlined in Table 3. These reactions can be expedited after introducing H_2 into the reservoirs, which serves as a source of electrons for bacteria and archaea. These organic processes are impeded in highly saline conditions [170,171], while encouraged in alkaline conditions [102]. It is worth highlighting that microbial activity stands as the primary mechanism for H_2 loss in geological formations [172], thus warranting deeper investigation. Even though numerous numerical and mathematical modelling studies have explored these biotic reactions and the proliferation of microorganisms within reservoir pore fluid, the validation through experiments remains limited due to the complex nature of physical trials [97].

3.5. Injection strategy

UCS is primarily intended for permanent sequestration, yet in certain situations, its retention may require seasonal injections instead of constant ones [173]. Within the framework of UHS, where H_2 is treated as a commodity, it becomes essential to accommodate intermittent injections on shorter time scales [41]. The viability for cyclic injection processes and the hysteresis nature of multiphase flow may directly impact trapping efficiency and integrity, necessitating further investigation [174,175].

For the trapping efficiency, Land model [176] is a well-known trapping model used for estimating the relationship between maximum residual saturation to maximum initial saturation [177]. Its classic shape of the trapping shows that the residual trapping saturation increases monotonically with initial saturation [178]. However, a few investigations have demonstrated anomalous entrapment during multiple cycles. The residual gas saturation can exceed the predicted values of the land model [174]. Moreover, the residual trapping saturation does not increase monotonically, but with some hysteresis behaviour, which cannot be captured by Land model [179]. These findings imply that, at least in certain circumstances, the conventional explanation of capillary trapping may not be complicated enough to properly explain the long-term behaviour of capillary-trapped gas. As discussed above, current results regarding the trapping efficiency during cyclic injections are still inconsistent. Some demonstrated that the trapping efficiency is enhanced when multiple drainage-imbibition cycles are applied [174, 175,179–182]. In contrast, some studies indicate that there is no change over time (*i.e.*, well-predicted by Land model) [183,184]. Recently, fine migration [185] and wettability alteration [186] have been identified as the potential mechanisms responsible for the anomalous behaviour

Table 5
Summary of recent studies relevant to cyclic gas injections.

References	Experimental conditions	Rock type	Core preparation	Effects on trapping efficiency
Ruprecht *et al.* (2014) [183]	9 MPa, 50 °C	Berea sandstone	Fired	No change
Herring *et al.* (2016) [179]	8.3 MPa, 38 °C	Bentheimer sandstone	Unfired	Enhance
Garing and Benson (2019) [184]	9 MPa, 50 °C	Berea sandstone	Fired/ Unfired	No change
Edlmann *et al.* (2019) [181]	10 MPa, 40 °C	Fell sandstone	Unfired	Enhance
Dalton *et al.* (2020) [182]	12.4 MPa, 45 °C	Bentheimer and Nugget sandstones	Unfired	Enhance
Herring *et al.* (2021) [174]	8.6 MPa, 40 °C	Bentheimer sandstone	Unfired	Enhance
Herring *et al.* (2023) [175]	8.6 MPa, 45 °C	Bentheimer sandstone	Unfired	Enhance (only in the upper region)

described above. Table 5 gives a summary of recent studies that reported cyclic gas injections.

For its impact on integrity, the injection of gas into the underground storage site leads to an increase in pressure, consequently resulting in changes in the stress system in and around the underground storage site [187]. Cyclic stress fluctuations in the vicinity of the well, within the reservoir and the faults, can cause compaction of the reservoir, leading to a reduction in porosity and flow of the reservoir fluid [188], complex subsidence [189], reactivation of faults [190], or microseismic activity. In addition, reservoir compaction can lead to overburden rock subsidence [191], the formation of fractures, and thus potential gas migration pathways through the sealing rocks [112].

4. Summary and future perspectives

This review offers a comparative overview of carbon dioxide and hydrogen storage in geological formations, mainly focusing storage media, trapping mechanisms and factors affecting underground gas storage. These comparisons highlight that due to the significant differences between storage fluids, the conversion of our existing knowledge from UCS to UHS should incorporate these factors relevant to their trapping and loss mechanisms. Below we summarise the key findings through this review and offer the future research directions regarding these aspects:

1. **Storage media:** Geological formations hold highly efficient and viable solutions for underground gas sequestration and storage. For UCS, depleted oil and gas reservoirs are well-suited whereas aquifers necessitate thorough investigation due to scarce geological data. For UHS, salt caverns have been the most prominent and commonly used formations so far, however, their geographical constraints and low storage capacity pose major challenges to large-scale storage operations. The suitability of depleted oil/gas reservoirs and aquifers for hydrogen storage and production has not been fully evolved. To reach the ambitious Gt scale storage goal for hydrogen and carbon dioxide, it is imperative to prioritize future research on depleted gas/ oil reservoirs and aquifers owing to their expansive geographical presence and higher storage capacities. Moreover, cushion gas is an additional requirement for UHS with the primary aim of maintaining reservoir pressure at an adequate level. While it can enhance

withdrawal efficiency by reducing residual trapping, it may also affect H_2 purity, which requires future study on selecting types of cushion gas and optimising the injection scheme.

2. **Trapping mechanisms**: Within UCS, both the extent and rate of capillary trapping make it the most efficient mechanism for storage security. Simultaneously, supplementary mechanisms contribute to the overall storage process. However, the scenario differs in UHS. Whilst increasing storage capacity, capillary trapping could lead to H_2 loss upon withdrawal. Yet, the hydrogen entrapment by capillary mechanisms can be reduced by injecting cushion gas and controlled by enhanced gas connectivity promoted by hydrogen characteristics. Further investigations are warranted to quantify these effects on the hydrogen recovery efficiency during cyclic operations. Both solubility and mineral trapping can also result in H_2 loss, necessitating careful consideration for future applications.

3. **Fluid properties:** The substantial differences in fluid properties between CO_2 and H_2 give rise to distinctive hydrodynamic process in the storage media, further affect storage integrity and trapping behaviour. The low density of H_2 leads to pronounced gravity segregation and rapid accumulation beneath the caprock, potentially leading to early H_2-surface leakage. The low viscosity of H_2 presents significant storage challenges, including low sweep efficiency and inefficiency in displacing formation liquids. Moreover, viscous fingering induced larger interfacial area will lead to higher chance of H_2 dissolution and interactions with rock, potentially impact H_2 withdraw efficiency and purity. Measures should be taken to counter these additional effects during injection and withdraw processes.

4. **Capillary effects:** (1) Fluid-fluid interfacial tension: Most existing investigations have focused on the water-gas systems. However, data regarding brine salinity and its composition constitute, which are pivotal parameters influencing surface tension remains restricted, particularly concerning H_2. To address this, it is strongly recommended to conduct thorough assessments of interfacial tension using *in situ* formation rocks and brine. (2) Contact angle: Until now, contact angle studies have primarily focused on gas interactions with bare mineral surfaces. Nevertheless, recent research reveals that organic acids, more representative in reservoirs, can significantly affect contact angle even at low levels of presence. The actual composition can vary due to the presence of additional organic compounds in real rock formations, requiring exploration of large-scale geological formations to foster a comprehensive grasp of these complex interaction.

5. **Relative permeability:** The relative permeability hysteresis in liquid phase for H_2 systems is notably more pronounced compared to that in CO_2 systems, potentially leading to decreased recovery during intermittent injections in UHS applications. Therefore, it is imperative to conduct further studies on the effects of hysteresis on relative permeability. This will enable a more accurate representation of realistic multiphase flow behaviours.

6. **Geochemical and microbial reactions**: Geochemical reactions existing in UCS predominately relates to mineral trapping, a process that facilitates enhanced CO_2 storage efficiency. Conversely, in the context of UHS, primary geochemistry reaction centres around pyrite and microbial activities lead to H_2 loss, emerging as noteworthy challenges in feasibility of UHS. There is a need to broaden comprehension of both abiotic and biotic reactions, across a wider range of subsurface conditions. It is worthwhile to explore the potential development of additives that can efficiently inhibit microbial activities to mitigate H_2 loss.

7. **Cyclic injection and hysteretic responses:** Both UCS and UHS encounter cyclic injection schemes, and its viability and the hysteresis nature of multiphase flow may directly impact trapping efficiency and storage integrity. Current results regarding the trapping efficiency during cyclic injections are still inconsistent, indicating the conventional explanation of capillary trapping (*i.e.*, the Land model) may not be comprehensive enough to properly explain the observations. This inconsistency is possibly related to two potential mechanisms: fine migration and wettability alteration, warranting exploration of these mechanisms at pore-scale experiments and modelling. Regarding storage integrity, cyclic stress fluctuations in the vicinity of the well, within the reservoir and faults, can lead to development of fractures, and thus potential gas migration pathways through the sealing structures. Future study on large-scale experiments and reservoir modelling of cyclic storage, with a focus on exploring the cyclic and hysteretic behaviour, is required to produce information on storage efficiency and integrity.

CRediT authorship contribution statement

Haiyi Zhong: Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis, Conceptualization. **Zhongzheng Wang:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Yihuai Zhang:** Writing – review & editing, Investigation, Conceptualization. **Si Suo:** Writing – review & editing, Conceptualization. **Yi Hong:** Writing – review & editing, Conceptualization. **Lizhong Wang:** Writing – review & editing, Conceptualization. **Yixiang Gan:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

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

Data will be made available on request.

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