

A REVIEW OF HYDROGEN STORAGE IN GEOLOGICAL FORMATIONS

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DOI: 10.51865/JPGT.2025.01.22

ABSTRACT

This review presents a thorough analysis of the technical, geological, and economic aspects of Underground Hydrogen Storage (UHS), positioning it as a key enabler for large-scale deployment of clean hydrogen supply chains. As global hydrogen demand increases particularly in traditional sectors such as refining and ammonia production the development of safe, seasonal, and scalable storage systems becomes essential to support renewable-based energy infrastructures.

Geological hydrogen storage is identified as the most viable solution due to its advantages in volumetric energy density, intrinsic safety, and the availability of suitable subsurface structures such as salt caverns, depleted hydrocarbon reservoirs, and deep aquifers. Techno-economic analyses show that UHS is considerably more cost-effective and technically feasible for large volumes compared to surface-based storage methods.

The review synthesizes real-world experiences from the UK, USA, and Austria, where field-scale pilot projects demonstrated up to 82% recovery of injected hydrogen with no significant degradation in reservoir integrity. The paper also highlights challenges related to microbial activity, particularly in porous formations with residual hydrocarbons or brine, where biological methanation or hydrogen loss could occur.

A comparative analysis of hydrogen's physical properties versus methane is included, focusing on density, viscosity, interfacial tension, solubility, and diffusivity, and how these influence hydrogen behavior in porous geologic media. Additionally, the role of cushion gas (e.g., CO₂, N₂, CH₄) is evaluated for its contribution to maintaining reservoir pressure and enhancing extraction performance. The gas selection is shown to significantly impact chemical compatibility, environmental risk, and operational efficiency. In conclusion, this review underscores that the successful deployment of UHS requires a multi-scale understanding of hydrogen dynamics in subsurface formations, supported by robust experimental validation and numerical modeling, to ensure safe, efficient, and climate-resilient storage operations over the long term.

Keywords: Hydrogen, Geological formations, Underground Hydrogen Storage, Hydrogen recovery efficiency

INTRODUCTION

Global hydrogen demand reached an all-time high in 2022, yet it remains largely concentrated in traditional applications. By 2025, worldwide hydrogen consumption reached approximately 95 million tons, reflecting a year-on-year increase of nearly 3% [1]. This growth was evident across most major consumer regions, with the exception of Europe, where industrial activity was hampered by surging natural gas prices. Importantly, this expansion was not the result of deliberate policy measures to advance hydrogen use, but rather a consequence of broader global energy trends. Demand continues to be dominated by conventional sectors such as oil refining and chemical processing, while new applications in heavy industry, transportation, and power generation account for less than 0.1% of total consumption [1,2].

The adoption of low-emission hydrogen in current industrial settings remains limited, comprising only 0.7% of global hydrogen demand in 2022. This indicates that hydrogen production and use still contribute significantly to CO₂ emissions, exceeding 900 million tons annually. Nevertheless, the outlook for low-carbon hydrogen is promising – particularly in ammonia production – although progress in refining applications lags behind [3].

Concurrently, the number of announced low-emission hydrogen production projects is rising rapidly. Estimates suggest that annual clean hydrogen output could reach 38 million tons by 2030, assuming full implementation of all proposed projects. Of this total, 17 million tons stem from projects still in early developmental stages. The projected production potential through 2030 already exceeds by 50% the figures reported in the International Energy Agency's 2022 Global Hydrogen Review. However, only 4% of this projected capacity has reached Final Investment Decision (FID), corresponding to roughly 2 million tons – a notable increase from the previous year. Among the announced projects, 27 million tons are based on electrolysis powered by low-emission electricity, while 10 million tons originate from fossil fuels using carbon capture, utilization, and storage (CCUS) technologies [1].

The global hydrogen strategy is structured around three core pillars: identifying and scaling strategic high-impact applications, reducing production costs, and establishing robust regional infrastructure [4]. Within this framework, hydrogen storage plays a pivotal role in ensuring the secure and flexible deployment of hydrogen across the value chain. According to the “DOE Pathways to Commercial Liftoff: Clean Hydrogen” report, various hydrogen storage methods are critical during both the industrial scale-up phase (2027-2034) and in long-term deployment beyond 2035. These storage systems are essential to buffer supply variability associated with renewable sources and accommodate seasonal demand fluctuations. While short-term solutions – such as compressed gas tanks, pipelines, and salt caverns – offer cost-effective flexibility, large-scale geological storage is regarded as fundamental for anchoring hydrogen infrastructure and enabling widespread deployment of clean hydrogen networks over the long term [4].

Papadias and Ahluwalia conducted a techno-economic assessment of various hydrogen storage methods, including pressurized vessels, cryogenic storage, and underground storage in salt caverns and porous media. Their analysis underscored the cost-effectiveness and scalability of underground storage in geological formations [5]. Compared to surface storage, underground hydrogen storage (UHS) offers several advantages for large-scale applications [6-8]:

- Reduced surface footprint due to high volumetric energy density of hydrogen;
- Enhanced safety against sabotage and environmental risks;
- Scalability due to widespread availability of suitable underground formations.

Underground Hydrogen Storage (UHS) is conceptually analogous to the well-established practice of natural gas storage, which has been in operation for over a century using salt caverns, depleted gas and oil fields, and aquifers [9,10]. UHS utilizes either natural or engineered porous formations to provide viable capacity for large-scale, seasonal energy storage. To ensure sufficient injectivity and operational rates, the target formation must be thick, porous, and permeable – preferably a saline cavern or reservoir. Moreover, effective containment requires a continuous caprock with extremely low permeability to prevent hydrogen leakage from the storage complex [11].

The primary motivation for hydrogen storage is to meet both base load and peak demand. Base load storage facilities typically withdraw hydrogen once or twice per year, whereas peak shaving storage facilities extract multiple times over short durations to address sudden demand spikes [8,9]. Peak demand storage is best suited to salt caverns, abandoned mines, and aquifers, while base load storage is typically accommodated in depleted oil and gas reservoirs [12].

INTERNATIONAL EXPERIENCE WITH LARGE-SCALE GEOLOGICAL HYDROGEN STORAGE

Experience with geological hydrogen storage is primarily limited to salt cavern applications, with a few pilot projects exploring storage in depleted gas and oil reservoirs. Historically, hydrogen has been stored in three salt caverns at Teesside, United Kingdom, since 1972, and in two caverns along the Gulf Coast of Texas, USA, since 1983 [9,13]. In the UK, at Teesside (Yorkshire), a British company stores 1 million m³ of nearly pure hydrogen (95% H₂ and 3-4% CO₂) in three salt caverns at approximately 400 meters depth, under a pressure of 50 bar [14,15].

In Texas, ConocoPhillips has been storing hydrogen with a purity of 95% in the Clemmons salt dome since the 1980s. The cavern, located approximately 850 meters below ground, is cylindrical in shape, with a diameter of 49 meters and a height of 300 meters. The usable hydrogen storage capacity reaches 30 million m³, equivalent to roughly 2,520 metric tons. This facility is directly connected to the Old Ocean refinery. Additionally, Praxair operates a UHS facility in a salt cavern in Texas, designed to balance peak hydrogen demand. This site is integrated into the Praxair Gulf Coast hydrogen pipeline network, which supplies the petrochemical industries of Texas and Louisiana [16].

Experience with hydrogen storage in porous rock formations is limited and has mostly involved hydrogen-containing gas mixtures (e.g., town gas). In the past, town gas was stored in aquifers in France (Beynes), the Czech Republic (Lobodice), and Germany (Engelborstel, Bad Lauchstädt, and Kiel). At Beynes, France, Gaz de France (GDF) stored manufactured gas containing 50% hydrogen in a saline aquifer with a capacity of 385 million standard m³ between 1956 and 1972 [17].

Hydrogen-containing gas mixtures have also been injected into depleted gas fields in pilot projects in Argentina (Diadema field) and Austria, aimed at managing supply-demand fluctuations. However, intense microbial activity and associated biochemical transformations were observed. Methanogenic bacteria initiated the Sabatier reaction ($4\text{H}_2 + \text{CO}_2 \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}$, $\Delta H = -134 \text{ kJ/mol}$), effectively converting hydrogen into methane. This behavior implies that geological formations used for hydrogen storage may function as underground methanation reactors (UMRs) under certain conditions [15].

The Czech pilot project at Lobodice revealed that after several months of hydrogen injection into coal gas reservoirs, approximately 50% of the stored hydrogen was biologically converted into methane via microbial degradation in the presence of CO and CO₂ [9,13].

One of the most advanced demonstrations of underground hydrogen storage is the Austrian project “Underground Sun Storage,” which involved injecting 20% hydrogen into a depleted gas reservoir [18]. The injected mixture comprised 90% natural gas and 10% green hydrogen. This pilot was the first real-world test of storing hydrogen in a subsurface sandstone formation at a depth of 1027 meters and an average temperature of 40 °C [18,19]. The project confirmed the technical feasibility of storing renewable energy in the form of hydrogen within porous underground media. No negative effects on reservoir rock integrity were observed. The complete life cycle – hydrogen injection, storage, and withdrawal – was analyzed. In the final stage, mass balance analysis showed that approximately 82% of the injected hydrogen could be recovered, while the remainder was lost due to diffusion, dissolution, or microbial transformation. The field test also confirmed that blending up to 10% hydrogen into natural gas reservoirs does not compromise the integrity or operability of existing gas infrastructure [18,19].

Projects specifically dedicated to storing 100% pure hydrogen are relatively rare but growing in number. Notable examples include [20]: (I) HyPSTER (France): storing 100% hydrogen in a salt cavern at Etrez; (II) H2CAST (Germany): storage in a salt cavern in Etzel; (III) HyStock (Netherlands): storage in a new salt cavern in Zuidwending; (IV) HyBRIT (Sweden): storage in a hard rock cavern near Luleå.

GEOLOGICAL FORMATIONS FOR HYDROGEN STORAGE

Storage in Depleted Gas and Oil Reservoirs

Hydrocarbon reservoirs are typically composed of porous and permeable rock formations, such as sandstone or carbonates. These formations retain hydrocarbons due to the presence of impermeable caprock layers or sealing strata that act as structural traps, forming subsurface reservoirs accessible via drilling and production technologies. Once depleted, these geological structures remain intact and can be repurposed for the technical and economic feasibility of underground hydrogen storage (UHS) [13].

According to Kanaani et al. [21], depleted hydrocarbon reservoirs are considered the most suitable candidates for large-scale hydrogen storage, owing to their well-characterized geology, compact and intact seal integrity, and the availability of pre-existing surface infrastructure and well-documented historical exploration and production data [13]. These formations have successfully contained trapped hydrocarbons for millions of years, thereby proving their geological competence for long-term storage [22]. Figure 1 illustrates a conceptual representation of how excess renewable energy at the surface can be converted into hydrogen and subsequently stored within subsurface geological formations – a core principle of Underground Hydrogen Storage (UHS)[13]. The diagram highlights a critical feature of UHS technology: the typical depth ranges of various storage media. Among these, depleted hydrocarbon reservoirs – especially former gas fields – are situated at the greatest depths within the Earth's crust, necessitating technically demanding drilling operations. However, the advantage lies in the potential reuse of existing infrastructure from previous extraction activities. In contrast, shallower formations such as aquifers and salt caverns are located closer to the surface, and their depth-specific characteristics play a key role in determining their overall storage capacity and operational feasibility.

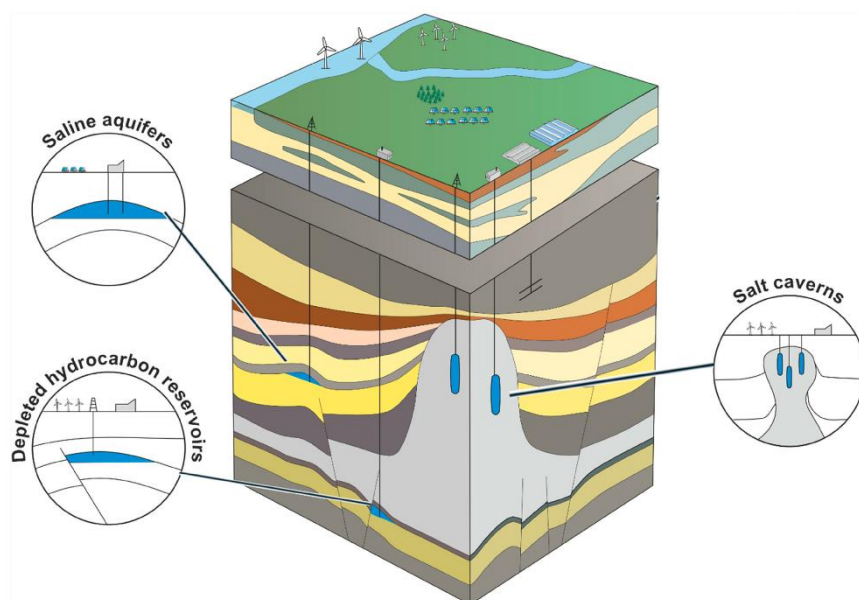


Figure 1. Overview of Geological Formations Applicable to UHS [13]

Depleted gas fields, in particular, offer well-documented formation properties, providing nearly all necessary subsurface data for safety assessments and engineering design. They also possess three critical attributes for secure UHS operations [23]: (I) A porous and permeable reservoir rock to store hydrogen; (II) An impermeable caprock to prevent vertical migration due to buoyancy; (III) A structural trap (e.g., an anticline) to inhibit lateral hydrogen movement and concentrate gas in elevated structural zones.

Nonetheless, converting a depleted reservoir into a UHS site requires comprehensive site-specific studies. For example, residual gas in the formation may act as a beneficial cushion gas, helping maintain pressure stability during injection and withdrawal.

However, it may also dilute the purity of the extracted hydrogen stream [10,13]. In the case of residual oil, chemical reactions become more probable, potentially converting hydrogen into methane or other compounds.

Operational experience from natural gas storage systems has demonstrated that UHS typically requires a base gas or cushion gas to maintain reservoir pressure during hydrogen withdrawal. Gases such as N₂, CH₄, or CO₂ are commonly used for this purpose [13,23]. This base gas not only stabilizes pressure but also co-produces with hydrogen, thereby affecting product gas purity. Consequently, additional purification steps may be required to meet hydrogen quality standards, which can significantly increase operating costs [21].

Moreover, the performance of geological formations for underground hydrogen storage (UHS) is strongly influenced by the physical properties of hydrogen. When compared to methane which serves as a reference gas due to its extensive history of underground storage applications the most notable difference lies in density. Methane is approximately eight times denser than hydrogen (as shown in Table 1), a disparity that has significant implications for the storage capacity of geological structures.

Table 1. Comparative Physical Properties of Hydrogen and Methane [13]

Properties	H ₂	CH ₄
Molecular Weight	2.016	16.043
Density (25 °C and 1 atm)	0.082 kg/m ³	0.657 kg/m ³
Viscosity (25 °C and 1 atm)	0.89×10^{-5} Pa s	1.1×10^{-5} Pa s
Solubility in pure water (25°C and 1 atm)	7.9×10^{-4} (mol kgw ⁻¹ H ₂ (g))	1.4×10^{-3} (mol kgw ⁻¹ CH ₄ (g))
Boiling point	-253 °C	-165 °C
Critical Pressure	12.8 atm	45.79 atm
Critical Temperature	-239.95 °C	-82.3 °C
Heating Value	120–142 kJ/g	205–55.5 kJ/g
Diffusion in pure water (25 °C)	5.13×10^{-9} m ² /s	1.85×10^{-9} m ² /s

Ultimately, the principal advantage of using depleted reservoirs lies in their economic viability, as many of the exploration, characterization, and infrastructure investments have already been made during prior hydrocarbon production, reducing the overall development costs over the asset's lifetime.

Storage in Aquifers

An aquifer is a subsurface layer of porous and permeable rock saturated with freshwater or saline water, often extending hundreds of meters in depth. In the absence of suitable salt caverns or depleted hydrocarbon reservoirs, aquifers are frequently considered for underground hydrogen storage (UHS) [9]. They represent a promising option due to their widespread presence across sedimentary basins globally. The storage mechanisms in

aquifers and depleted hydrocarbon reservoirs are similar, as both consist of porous and permeable formations. However, two fundamental geological criteria must be met: (i) the host rock must exhibit good reservoir properties for injection, and (ii) it must possess adequate sealing capacity to prevent migration of the stored gas [9,23,24].

When hydrogen is injected into an aquifer initially filled with water, the density contrast between the injected gas and the resident fluid leads to a pressure-induced displacement of water downward or laterally to create storage space. This process results in the formation of a gas-liquid interface, which is further intensified as injection pressure increases. Unfortunately, this interface poses a challenge for UHS in aquifers, particularly during withdrawal operations. During hydrogen production, the movement of this gas-liquid boundary often causes co-production of water, complicating gas recovery and treatment processes [24,25].

The volume of hydrogen that can be stored is primarily governed by the aquifer's total pore volume, temperature, pressure, and porosity. Compared to depleted reservoirs, aquifers typically require larger volumes of cushion gas, since they lack pre-existing gas volumes that could offset the necessary pressure buffer. As a result, the cushion gas fraction in aquifers can reach up to 80% of total pore volume, meaning that only about 20% of the reservoir capacity may be usable as working gas [8,14].

Hydrogen and cushion gas losses are generally unavoidable in aquifers. Sulfate-reducing bacteria may contaminate stored hydrogen in deep aquifers, while cushion gas recovery may be limited due to reservoir heterogeneity and capillary trapping. The extended contact between hydrogen and formation water in aquifers also increases the risk of gas dissolution and downward migration, leading to irrecoverable gas volumes [16,26].

Several additional factors affect UHS performance in aquifers, including:

- Undetected gas migration through faults and fractures;
- Microbial activity leading to gas degradation;
- Geochemical reactions between hydrogen and reservoir minerals.

Importantly, due to limited geological characterization and uncertainty surrounding aquifer-specific properties, UHS in aquifers tends to be significantly more expensive than storage in depleted gas or oil reservoirs [16]. Comprehensive site assessments often require new wells to be drilled for thorough exploration and testing [14].

As of now, no successful pure hydrogen (100%) storage has been demonstrated in aquifers. However, a town gas project (derived from coal gasification, containing notable amounts of hydrogen and other gases) has been documented as the only aquifer-based hydrogen storage trial in Europe [27].

Storage in Salt Caverns

Salt caverns are ideally suited for the storage of various gases at high pressures. These are predominantly cylindrical, man-made voids constructed within thick subsurface salt deposits by injecting water in a controlled manner to dissolve the salt – an approach known as solution mining [28]. Depending on technical and operational requirements, salt caverns can be constructed at depths of up to 2000 meters, with volumes reaching 1,000,000 cubic meters, heights between 300 and 500 meters, and diameters ranging from 50 to 100 meters, enabling the storage of large quantities of gas [14,28].

For hydrogen storage, the operational pressure inside salt caverns typically ranges between 30% and 80% of the lithostatic pressure [14,24]. Specific geological conditions – such as excellent sealing capacity, favorable mechanical properties of the salt, and its chemical inertness – make salt caverns a practical and attractive option for UHS. The surrounding salt is extremely impermeable, offering near-complete gas containment. In most cases, the only plausible leakage pathways are wellbore failures or fractures, though reported leakage rates are generally below 1% [24].

Salt caverns can be developed within salt domes or bedded salt formations. Salt domes are thick, homogeneous salt bodies that simplify the construction of structurally stable caverns under typical operating conditions. In contrast, bedded salt formations consist of interbedded salt layers (primarily halite NaCl) and insoluble strata such as dolomite, anhydrite, or shale, usually at shallower depths than domes [24,29].

At greater depths (beyond ~1800 meters), salts may undergo creep deformation due to elevated pressure and temperature, even when caverns are carefully engineered [14]. Moreover, caverns constructed in heterogeneous salt formations may exhibit reduced structural stability, making them less favorable for long-term UHS applications [14,29].

In addition to cost considerations, several constraints differentiate salt caverns from porous media storage systems (such as aquifers and depleted hydrocarbon reservoirs). These include:

- Limited geographical availability of suitable salt formations;
- Requirements for water management and disposal;
- Potential for irregular cavern geometry;
- Thermal and mechanical stability challenges at depth.

Another underexplored issue is the impact of multiphase phenomena within salt caverns. Accumulated residual water at the cavern bottom can lower injection efficiency and may necessitate sophisticated injection – withdrawal strategies. Nonetheless, caverns offer favorable cycling potential, supporting more than just seasonal storage and allowing frequent hydrogen injection/withdrawal cycles – unlike porous formations [29,30].

Salt caverns generally pose a low risk of microbial activity, especially when artificially engineered. However, in environments with high salinity, such as salt lakes or brine ponds, the presence of halotolerant or halophilic microorganisms may pose a risk to hydrogen integrity. These organisms could potentially consume hydrogen and initiate biochemical transformations [29,30]. Therefore, it is essential to assess the origin and behavior of such microbial populations, especially during cavern leaching or under long-term operational conditions. Water used during solution mining or extended injection campaigns could introduce or promote microbial growth.

Compared to porous reservoirs, salt caverns require moderate cushion gas volumes, typically ranging between 22% and 33% of total cavern capacity [29,30].

KEY PARAMETERS FOR STORAGE IN DEPLETED OIL AND GAS RESERVOIRS

The most critical parameters influencing hydrogen storage in depleted oil and gas reservoirs can be classified into three main categories: solid properties, fluid properties, and solid–fluid interaction parameters. These characteristics represent fundamental phenomena governing the simulation, design, and performance prediction of hydrogen storage systems, as well as fluid flow behavior within porous media.

To accurately model and assess storage performance, however, reliable subsurface data and reservoir-specific measurements are essential. According to recent literature, the availability of comprehensive datasets required to fully evaluate UHS flow dynamics remains limited, reflecting the early stage of research in this domain. Despite this, a few existing datasets have been published, and they serve to enhance our understanding of hydrogen behavior in such geological environments.

The parameters outlined in Figure 2 are categorized and discussed to reflect the mechanical characteristics of the solid matrix, the physicochemical behavior of the fluids, and the coupled solid–fluid interactions relevant to UHS applications [13].

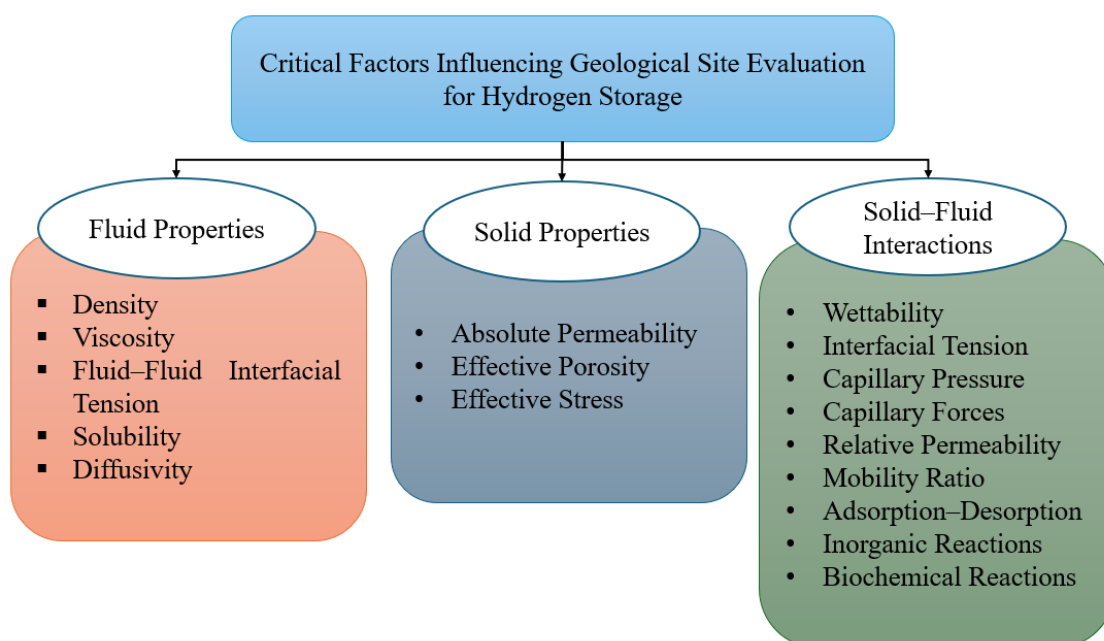


Figure 2. Key factors shaping the future of UHS site evaluation and protocol development [13].

Solid Properties

In the context of a specific UHS project, the solid properties of the storage medium play a crucial role in determining the capacity, efficiency, and operational safety of hydrogen storage. The most relevant solid-related parameters include absolute permeability (k_a), effective porosity ($\phi_{effective}$), and effective stress ($\sigma_{effectiv}$).

I. Absolute Permeability (k_a) and Effective Stress (σ_{effectiv})

Absolute permeability (k_a) defines the ability of a porous medium to transmit a fluid when it is fully saturated with that fluid. Importantly, k_a is an intrinsic property of the porous material that is, it remains independent of the fluid's properties or solid–fluid interactions [31,32]. From an engineering standpoint, higher permeability translates to more energy-efficient hydrogen injection and withdrawal processes [33].

Effective stress (σ_{effectiv}) is defined as the difference between the overburden pressure and pore pressure. Empirical studies indicate that k_a generally decreases with increasing σ_{effectiv} during loading cycles and increases again during unloading cycles [14]. This effect is particularly pronounced in fractured rocks such as limestone, coal, and shale [34]. For example, permeability in coal was shown to drop from 5.5×10^6 nD to 1.5×10^6 nD when σ' increased from 4 MPa to 7.5 MPa. Conversely, k_a increased from 1.05×10^6 nD to 2.5×10^6 nD as σ_{effectiv} decreased from 10.5 MPa to 4 MPa.

In the UHS operational cycle, hydrogen injection corresponds to stress unloading, while hydrogen withdrawal corresponds to stress loading. Thus, k_a tends to increase during injection and decrease during withdrawal. It is essential to characterize the stress–permeability relationship under in situ reservoir conditions prior to large-scale deployment in order to enable accurate modeling of hydrogen behavior.

Notably, k_a is generally higher in sandstones and limestones compared to shales and coal matrices [34], implying that UHS is likely to be more energy-efficient in conventional reservoirs composed of sandstone and carbonate rocks.

II. Effective Porosity ($\phi_{\text{effectiva}}$)

While total porosity is defined as the ratio between the pore volume and the total rock volume, only the connected pore space contributes to hydrogen flow and storage. Therefore, effective porosity ($\phi_{\text{effectiva}}$) the ratio of connected pore volume to bulk volume is the parameter of interest in determining the maximum hydrogen storage capacity at the reservoir scale [31].

This capacity is directly proportional to $\phi_{\text{effectiva}}$, which tends to be significantly higher in conventional rocks (e.g., sandstone and limestone) than in unconventional formations such as shales and coals [35,36]. Consequently, maximum storage capacity is expected to be much greater in conventional reservoirs.

It is also important to note that measuring $\phi_{\text{effectiva}}$ in unconventional rocks is more complex, often leading to increased uncertainty in performance predictions for UHS in such formations [14,34]. Moreover, like permeability, effective porosity is influenced by effective stress (σ_{effectiv}). During loading cycles, $\phi_{\text{effectiva}}$ tends to decrease, while

unloading promotes partial recovery. However, $\phi_{\text{effective}}$ is generally less sensitive to stress variation than (k_a) [35-37].

Therefore, prior to large-scale UHS deployment, it is advisable to evaluate the impact of effective stress on both permeability and porosity, particularly for stress-sensitive or fractured reservoirs.

Fluid Properties

Fluid properties are also critical parameters that significantly influence the efficiency, stability, and safety of underground hydrogen storage (UHS) operations. Key fluid-related parameters include density, viscosity, fluid–fluid interfacial tension, solubility, and diffusivity.

I. Fluid Density (ρ)

Subsurface pressure and temperature naturally increase with depth due to hydrostatic and geothermal gradients. Therefore, it is essential to assess how these variables influence the density of the storage fluid (ρ) [38]. Specifically, the density of hydrogen (ρ_{H_2}) increases substantially with pressure but decreases slightly with temperature.

For instance, at 298 K, ρ_{H_2} rises from 0.5 kg/m³ to 12 kg/m³ as pressure increases from 0.6 MPa to 16 MPa. At a constant pressure of 30 MPa, ρ_{H_2} decreases from 20 kg/m³ to 16 kg/m³ as temperature increases from 298 K to 373 K. In contrast, the density of methane (ρ_{CH_4}) under similar conditions increases from 0.6 kg/m³ to 135.67 kg/m³ over the pressure range of 0.1 MPa to 20 MPa at 323 K. Furthermore, ρ_{H_2} remains significantly lower than the density of water (ρ_{H_2O}) under equivalent thermophysical conditions.

This large density contrast between hydrogen and brine leads to strong gravitational segregation, causing upward migration of hydrogen through the formation. Consequently, this poses a serious risk of hydrogen leakage to the surface, making the mechanical integrity of the caprock a crucial factor for secure UHS an issue explored further in subsequent sections [14,34].

II. Fluid Viscosity (μ_{H_2}):

Viscosity (μ) quantifies a fluid's resistance to flow and is therefore a key operational parameter during both hydrogen injection and withdrawal. Additionally, the viscosity ratio between the displacing and displaced fluids together with relative permeability differences governs interfacial stability and the likelihood of viscous fingering, a phenomenon that can severely compromise hydrogen sweep efficiency.

During hydrogen injection, viscous fingering may result in:

- Early hydrogen breakthrough at production wells,
- Poor contact between hydrogen and resident fluids,
- Reduced storage efficiency [34].

The viscosity of hydrogen (μ_{H_2}) is moderately sensitive to both pressure and temperature. For example:

- At 20 MPa, μ_{H_2} increases slightly from 9.6×10^{-3} mPa·s to 10.7×10^{-3} mPa·s as temperature rises from 313 K to 373 K.
- At 373 K, μ_{H_2} increases from 10.4×10^{-3} mPa·s to 11.8×10^{-3} mPa·s as pressure increases from 0.1 MPa to 50 MPa [39].

In comparison, the viscosity of methane (μ_{CH_4}) increases from 13.6×10^{-3} mPa·s to 26×10^{-3} mPa·s under the same pressure range at 373 K. While the viscosities of H_2 and CH_4 are similar at low pressures, methane becomes nearly twice as viscous as hydrogen at higher pressures. Additionally, increasing temperature reduces intermolecular friction, thus decreasing viscosity for both gases.

More importantly, the viscosity of hydrogen is approximately one-half that of methane under equivalent thermophysical conditions. As a result, viscous fingering is more likely during hydrogen injection, particularly in formations with significant heterogeneity or unfavorable viscosity ratios [40].

III. Fluid–Fluid Interfacial Tension ($\gamma_{H_2-H_2O}$)

Immiscible fluids are separated by a phase boundary, the nature of which is governed by the molecular structure of each fluid and the intermolecular forces acting across the interface. A critical property of this boundary is the interfacial tension (γ_{FF}), which quantifies the energy required to create additional surface area between two immiscible phases [41].

For the H_2 – H_2O system, interfacial tension $\gamma_{H_2-H_2O}$ is relatively insensitive to pressure but strongly decreases with increasing temperature. For example: At 323 K, $\gamma_{H_2-H_2O}$ decreases slightly from 70 to 67 mN/m (~4.3%) as pressure rises from 0.1 to 40 MPa and At 10 MPa, $\gamma_{H_2-H_2O}$ decreases significantly from 72 to 44 mN/m (~38.9%) as temperature increases from 298 K to 448 K.

In contrast, $\gamma_{H_2-H_2O}$ decreases with both temperature and pressure. At 323 K, $\gamma_{H_2-H_2O}$ declines from 67 to 54 mN/m (~19.4%) as pressure increases from 0.1 to 25 MPa. At 10 MPa, it drops from 64 to 61 mN/m (~4.9%) as temperature increases from 300 to 353 K [14,34].

These variations in interfacial tension are largely governed by gas density and intermolecular forces. Interfacial tension values have also been experimentally estimated via H_2 –water–sandstone flooding tests, calibrated against mercury capillary injection pressure data. However, estimates of $\gamma_{H_2-H_2O}$ based on these methods (e.g., 51 mN/m at 5.5 MPa and 293 K, and 46 mN/m at 10 MPa and 318 K) were significantly lower than directly measured values (~72 mN/m and 70 mN/m under the same conditions, respectively) [14,34]. This discrepancy likely stems from systematic errors associated

with converting mercury-based data and raises concerns about the validity of indirect extrapolation methods [42].

IV. Hydrogen Solubility

Hydrogen solubility in formation brines or residual oil must be carefully evaluated, as dissolution represents a significant pathway for hydrogen loss in UHS systems. While H_2 solubility in water is relatively low under ambient conditions, it increases considerably under UHS-relevant pressures and temperatures. For instance: At 319 K and 0.678 MPa, H_2 solubility in water is 8.56×10^{-5} (mole fraction); and at 323 K and 7.9 MPa, it increases to 1.03×10^{-3} [14,34].

Thus, solubility data obtained under surface conditions are not reliable for UHS design. Under reservoir conditions, H_2 solubility in brines is strongly affected by pressure, temperature, and salinity: At 372 K in 3 mol/kg NaCl solution, solubility increases from 2.15×10^{-5} to 1.549×10^{-3} as pressure rises from 3.3 to 23 MPa, and at 10.1 MPa, solubility increases from 6.31×10^{-4} to 7.02×10^{-4} as temperature rises from 323 K to 372 K. At 323 K and 15.1 MPa, solubility decreases from 9.38×10^{-4} to 6.62×10^{-4} as salinity increases from 3 to 5 mol/kg NaCl [43].

Hydrogen solubility in non-aqueous liquids (e.g., crude oil, hydrocarbons, diesel, solvents) follows similar trends – increasing with pressure and temperature – but the absolute solubility is nearly an order of magnitude higher than in brines. For example: At 3.3 MPa and 373 K, H_2 solubility is 0.0269 (mole fraction) in diesel, compared to only 2.15×10^{-4} in 3 mol/kg NaCl brine [14,34].

As a result, hydrogen loss via dissolution is expected to be significantly higher in depleted oil reservoirs than in aquifers, necessitating advanced reservoir management strategies.

To quantify solubility more rigorously, Henry's law can be applied using the relation $C = k_P$, where C is the concentration of dissolved gas, k is Henry's constant, and P is the partial pressure of the gas. However, inconsistencies in reporting partial pressure data in the literature have limited the accuracy of Henry constant estimations, representing a challenge for predictive modeling of hydrogen retention.

V. Hydrogen Diffusivity (D):

Another fundamental property of relevance to UHS is the diffusivity of hydrogen (D_{H_2}), which quantifies the rate at which hydrogen molecules migrate through the surrounding rock surface or formation. This parameter is particularly important for estimating hydrogen losses over time, especially in formations where molecular diffusion dominates transport mechanisms.

Hydrogen diffusivity generally increases with temperature and decreases significantly with pressure. For instance: At 25 MPa, the diffusivity of H_2 in water increases from $14.4 \times 10^{-8} \text{ m}^2/\text{s}$ to $218.8 \times 10^{-8} \text{ m}^2/\text{s}$ as temperature rises from 650 K to 973 K, and at 323 K, the diffusivity of H_2 in butane (C_4H_{10}) decreases from $1440 \times 10^{-8} \text{ m}^2/\text{s}$ to $250 \times 10^{-8} \text{ m}^2/\text{s}$ as pressure increases from 0.35 MPa to 2.1 MPa. Additionally, hydrogen diffusivity in hydrocarbons decreases with increasing carbon number: At 323 K and 2.1 MPa: $D_{H_2} = 430 \times 10^{-8} \text{ m}^2/\text{s}$ in CH_4 , $320 \times 10^{-8} \text{ m}^2/\text{s}$ in C_2H_6 , $270 \times 10^{-8} \text{ m}^2/\text{s}$ in C_3H_8 , and $250 \times 10^{-8} \text{ m}^2/\text{s}$ in $n\text{-}C_4H_{10}$ [44,45].

These findings suggest that hydrogen diffusion may cause significant losses over time in deep aquifers and depleted hydrocarbon reservoirs. A numerical reservoir simulation showed that up to 1% of stored H_2 could be lost over 15 years in a 7-meter-thick reservoir with 20% porosity due to molecular diffusion into surrounding brine [46].

Solid–Fluid Interactions

Solid–fluid interactions govern the distribution, migration, retention, and reactive transport of fluids in porous media. In the context of UHS, such interactions include wettability, solid–fluid interfacial tension (γ_{SF}), capillary pressure (P_C), relative permeability (K_r), and the mobility ratio (M) [13]. Among these, wettability and capillary pressure are especially critical for modeling multiphase flow behavior in porous systems [13,31].

I. Wettability

Wettability describes the preferential affinity of a solid surface for one fluid in the presence of another immiscible (or partially miscible) fluid. In hydrogen storage formations, H_2 –rock wettability is heterogeneously distributed due to variations in surface chemistry, mineral composition, and pore geometry. When considered across an entire reservoir, these local-scale interactions govern macroscopic wettability [47,48].

Wettability affects several key UHS performance metrics, including: Capillary pressure (P_C), Hydrogen saturation and residual saturation (S_{H_2} , $S_{H_2,r}$), Injectivity and containment efficiency, and Overall storage capacity (C , m^3). Therefore, accurate characterization of wettability under in-situ conditions is essential [13].

Experimental assessment of H_2 wettability in sandstone is typically performed using contact angle (θ) measurements under reservoir conditions via video-based methods. For instance, at 15 MPa and 323 K, contact angles for water advancing and receding on quartz treated with 10^{-2} mol/L stearic acid (a realistic concentration in deep aquifers under reducing conditions) were 68° and 64° , respectively, indicating weak water wettability [49].

In the UHS context: The receding water contact angle corresponds to H_2 injection (gas displacing water), and the advancing angle corresponds to H_2 withdrawal, as water re-enters the pore space [49].

Moreover, θ increases with pressure, temperature, and acid concentration, as these factors alter gas density and intermolecular forces at the solid-fluid interface. However, notable discrepancies exist between measured and estimated θ values: Estimated θ values from mercury injection and sandstone imbibition (22° at 5.5 MPa/293 K; 35° at 10 MPa/318 K) were significantly higher than directly measured values for pure quartz (10° and 18° at the same conditions) [34].

This inconsistency may be attributed to differences in mineralogical composition: Sandstone cores used in imbibition tests contained ~ 81 vol% quartz, 17 vol% K-feldspar, and 2 vol% minor minerals (e.g., muscovite, hematite, illite), and whereas contact angle experiments were performed on pure quartz surfaces [34,49].

Another potential cause is the limited reliability of mercury-based extrapolation techniques. Trace amounts of organic matter present in sandstone cores may also significantly alter surface wettability.

These findings underline the need to evaluate wettability of other relevant minerals and rocks (e.g., calcite, dolomite, mica, shale, coal, carbonates) under realistic UHS conditions, to better assess the storage potential across diverse geological settings [14,34].

II. Solid–Fluid Interfacial Tension (γ_{SF})

Solid–fluid interfacial tension (γ_{SF}) plays a crucial role in governing hydrogen distribution and migration within porous media. Due to the lack of direct and reliable experimental methods for measuring this parameter, researchers often rely on semi-empirical models to estimate γ_{SF} . Recently, Pan et al. reported the interfacial tension between rock and hydrogen (γ_{rock-H_2}), showing that it decreases with both pressure and temperature reductions for minerals such as quartz and basaltic rocks [40]. Moreover, increasing the concentration of stearic acid also reduced γ_{rock-H_2} . For instance, at 323 K, increasing the stearic acid concentration from 1×10^{-9} mol/L to 1×10^{-2} mol/L reduced aged quartz–H₂ interfacial tension from 90.57 to 81.49 mN/m at 15 MPa, and from 84.91 to 72.37 mN/m at 25 MPa, respectively. Since pure quartz is not typically found in subsurface formations, stearic acid-aged quartz is considered a more representative proxy for UHS conditions.

III. Relative Permeability (k_r)

Multiphase fluid flow in porous media is strongly affected by relative permeability (k_r). Typically, the sum of the relative permeabilities of the wetting and non-wetting phases is less than one. It is well established that k_r is a function of water saturation (S_w) and the rock's wettability. Measurements of hydrogen relative permeability (k_{r-H_2}) and associated water permeability (k_{r-W}) under reservoir conditions using the steady-state method revealed that:

- At 5.5 MPa and 293 K, k_{r-H_2} increased from 0 to 0.04 as S_w decreased from 90% to 41%;
- At 10 MPa and 318 K, k_{r-H_2} increased from 0 to 0.03 as S_w decreased from 82% to 40%. In contrast, for CH₄ at 8.3 MPa and 298 K, k_{r-CH_4} increased from 0.05 to 0.8 as S_w decreased from 73% to 33%. These findings indicate that hydrogen exhibits a significantly lower flow capacity through brine-saturated sandstone than methane, likely due to the geological heterogeneity of the rock matrix [50]. However, more experimental data are needed to confirm this behavior, given the limited datasets available for H₂.

IV. Mobility Ratio (M)

The mobility ratio (M) is a critical factor influencing hydrogen injectivity, displacement efficiency, and interfacial stability between the displacing (H_2) and displaced (liquid) phases. M is defined as the ratio of hydrogen mobility to brine mobility, where mobility is the product of absolute permeability (k_a), relative permeability (k_r), and the inverse of fluid viscosity (μ) [34].

A high M can lead to viscous fingering, reducing hydrogen sweep efficiency and promoting premature breakthrough. M depends on variables such as saturation, pressure, temperature, and gas type. Pan et al. reported that M_{H_2} increased from 0.06 to 55 as water saturation decreased from 82% to 40% at 10 MPa and 318 K. Moreover, higher pressure and temperature conditions lowered M significantly. For instance, at $S_w = 73\%$, M_{H_2} was 2.3 at 5.5 MPa and 293 K, compared to 1.1 at 10 MPa and 318 K [34,40]. These results emphasize the need for further studies and suggest that viscous fingering could be mitigated by lowering injection rates or increasing H_2 viscosity, such as by generating H_2 foam.

V. Adsorption–Desorption

Gas adsorption–desorption processes become relevant in high-surface-area systems. While well studied for CO_2 , CH_4 , and N_2 , data for H_2 remain limited. For example, H_2 adsorption (A_{H_2}) on dry sodium montmorillonite at 373 K and 0.045 MPa reached up to 0.1 wt% after 30–45 days. A_{H_2} decreased with temperature, reaching 0.07 wt% at 403 K under the same pressure [51]. Molecular dynamics simulations predicted A_{H_2} on calcite up to 0.42 wt% at 0.1 MPa and 400–600 K [52].

At 363 K, A_{H_2} on raw clay (17% illite, 2.6% kaolinite, 2% chlorite, 20% carbonate, 22% quartz, 5% others; surface area = 46 m²/g) increased from 0.125 to 0.62 mmol/g as pressure rose from 0.4 MPa to 4 MPa. For purified clay (80% clay content, <2 μ m particles, 84 m²/g), A_{H_2} rose from 0.065 to 1.25 mmol/g as pressure increased from 0.4 to 5.5 MPa. Above 4–5 MPa, adsorption plateaued. These trends mirror CH_4 adsorption behavior. Temperature also slightly increased A_{H_2} : from 0.53 to 0.62 mmol/g between 293 and 363 K at 6 MPa. The anomalous increase is attributed to H_2 reactivity with Fe in clays, accelerated at higher temperatures [14,34]. Adsorption may also alter rock mechanics, permeability, and surface chemistry, necessitating in situ studies under UHS conditions.

VI. Capillary Pressure (P_C)

Capillary pressure (P_C) is central to understanding multiphase flow in porous formations. It determines phase configurations at the pore scale and is often assessed alongside relative permeability curves as a function of S_w . During hydrogen injection, brine is displaced upward, and H_2 tends to accumulate near the caprock due to buoyancy. The

pressure needed to displace brine is defined as P_C . Thus, achieving high H_2 saturation or low water saturation is favorable for maximizing storage capacity [9,23].

Only a few studies report P_C and S_w relationships for sandstone– H_2 –brine systems. For a sandstone with $k = 45$ mD and $\phi = 19\%$, P_C decreased from 110 kPa to 61 kPa as S_w increased from 17% to 31% at 5.5 MPa and 293 K; similarly, at 10 MPa and 318 K, P_C decreased from 81 to 56 kPa as S_w rose from 19% to 41%, indicating minimal pressure–temperature dependence [50].

Post-injection, spontaneous imbibition of brine can cause capillary fingering, enhancing H_2 entrapment and storage capacity. However, to enable efficient withdrawal, residual H_2 saturation (S_{H_2-r}) should be minimized. Preventing H_2 phase segregation may improve remobilization and extraction. P_C also influences structural trapping and long-term containment security [14].

CUSHION GAS IN UNDERGROUND HYDROGEN STORAGE (UHS)

Cushion gas refers to the volume of gas that remains permanently within a storage reservoir to maintain adequate pressure and enable continuous deliverability of the working gas. Its primary function is to ensure that the reservoir pressure remains sufficiently high to support consistent and efficient injection and withdrawal cycles. Throughout the operational life of a UHS system, the cushion gas undergoes alternating compression and expansion to stabilize internal reservoir conditions. Maintaining this pressure is critical for effective hydrogen recovery; otherwise, the efficiency of the withdrawal cycle may be compromised [21,53].

The required cushion gas volume varies depending on the storage technology employed [14,54]:

- In depleted hydrocarbon reservoirs, cushion gas typically constitutes ~50% of the total storage volume.
- In salt caverns, it usually accounts for ~25%.
- In aquifers, cushion gas requirements can be as high as 80% of total storage capacity.

The performance of hydrogen extraction in UHS systems is highly influenced by the cushion gas selection, which must be tailored to its physical properties. These include density, viscosity, and solubility, all of which influence gas flow behavior and ultimately affect hydrogen recovery efficiency [55].

The optimal cushion gas must also demonstrate chemical stability under reservoir conditions. Any chemical reactivity with the stored hydrogen or with reservoir rock could lead to unwanted byproducts or jeopardize formation integrity [56].

In practice, the three most commonly used gases for this purpose are: Carbon dioxide (CO_2), Nitrogen (N_2), and Natural gas (methane, CH_4) [21].

Additionally, residual hydrocarbons in depleted oil and gas fields may also be utilized as cushion gas depending on their composition [54].

These gases are typically selected based on availability, cost-efficiency, physical properties, and environmental impact. Several key trade-offs exist:

- CO₂ offers cost advantages when sourced from carbon capture and storage (CCS) technologies. Industries utilizing captured CO₂ may also benefit from tax credits. Beyond economic benefits, CO₂ use contributes to climate change mitigation [57].
- N₂ is chemically inert and reduces the risk of corrosion or undesirable reactions. Its high relative humidity facilitates easier separation from hydrogen during withdrawal, thereby improving hydrogen recovery rates and system efficiency [58].
- CH₄ provides similar recovery advantages to N₂ but additionally benefits from broad compatibility with existing natural gas infrastructure. This minimizes retrofitting costs and enhances economic feasibility.

Both CH₄ and N₂ are widely available and commercially accessible. The advantages and limitations of each cushion gas type are summarized in Table 2.

Table 2. Cushion gas comparison for UHS

Cushion gas	Advantages	Disadvantages
CO ₂	• Financial incentives for carbon capture. • Injection of CO₂ into underground formations can contribute to climate change mitigation. • Widely available as a by-product from power plants and industrial processes.	• If not properly managed, CO₂ may leak from storage sites, causing environmental harm. • The capture, transport, and injection of CO₂ into subsurface formations can be costly. • Effectiveness as a cushion gas and safe storage potential may be limited to specific geological formations.
N ₂	• Higher gas wettability. • Less corrosive than CO₂. • Inert nature implies reduced risk of chemical reactions. • Abundant in the atmosphere and easy to produce.	• Injection and withdrawal of N₂ as a cushion gas may introduce operational complexities. • High cost of separating N₂ from air. • Excessive release into the atmosphere may disrupt local nitrogen cycles and harm ecosystems.
Natural gas (CH ₄)	• Potentially high hydrogen recovery rate (up to 89.7%). • Widely available. • Existing technologies are in place for leak detection.	• Any leakage of CH₄ during storage or transport contributes to climate change. • CH₄ is highly flammable and may form explosive mixtures with air.

	• Compatible with most existing infrastructure	• Additional safety measures may increase operational costs.
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CONCLUSIONS

Underground hydrogen storage (UHS) represents a strategic technological solution to ensure flexibility and continuity in clean hydrogen supply chains. Due to its high volumetric energy density and scalability, UHS stands out as a superior alternative to surface storage methods, especially within the context of integrating renewable energy into the energy system. Geological formations such as salt caverns, depleted hydrocarbon reservoirs, and deep aquifers provide natural environments for implementing this technology, each offering specific advantages and operational challenges.

Field experience, particularly in salt caverns, has demonstrated the technical feasibility of UHS, with hydrogen recovery rates exceeding 80% under controlled conditions. Meanwhile, pilot studies in porous media have revealed potential risks, including diffusion losses, biochemical transformations, and hydrogen solubility in brine or residual oil. The physicochemical properties of hydrogen – such as low density, low viscosity, interfacial tension, high diffusivity, and specific solubility – significantly influence its behavior in geological settings.

Additionally, the characteristics of the host rock – such as effective porosity, absolute permeability, and effective stress – directly impact injectivity and retention capacity. Solid–fluid interactions, including wettability, relative permeability, capillary pressure, and mobility ratio, play a central role in evaluating system stability. The selection of cushion gas, whether CO₂, N₂, or CH₄, directly affects operational efficiency, chemical compatibility, and associated risks such as corrosion or environmental contamination. All these factors must be rigorously assessed to ensure the safe and efficient functioning of an underground hydrogen storage system.

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Received: May 2025; Revised: June 2025; Accepted: June 2025; Published: June 2025