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Review article

A comprehensive review of the mechanisms and efficiency of underground hydrogen storage

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ABSTRACT

Underground Hydrogen Storage can be proven very beneficial for recurring supply of clean energy throughout the world. This paper reviews different challenges like microbial growth, well integrity, and geochemical reactions faced when hydrogen is stored in subsurface. The mechanisms involved in underground hydrogen storage, monitoring techniques and optimization of injection-withdrawal techniques are also analysed by incorporating both field and laboratory research studies. If the economics of any technology is not taken into account, it will not be deemed as a common technique. Keeping that in mind, the road map for the implementation and economic aspects of hydrogen storage are discussed. Lastly, conclusions and recommendations for future research are presented.

1. Introduction

To reduce the usage of fossil fuels and the emission of greenhouse gases which adversely affects the climate, and to prepare a sustainable energy supply for the future, the transition to renewable energy seems inevitable. For decades the fossil fuels had 80% share of the global energy mix and renewable energy has the share of 15%. According to the report published by DNV energy in 2021, fossil fuel share of the energy mix will decline but still hold a 50% share of the energy mix and renewable energies will triple to 45% by 2050 [1]. According to the report by British Petroleum (BP) energy, the overall primary energy consumption in world for the year 2020 declined by 4.5%, which is the largest decline since 1945. The drop in energy consumption was mostly driven by oil, which accounted for over three-quarters of net decrease, while natural gas and coal also witnessed large drops. Despite a decline in overall energy consumption, wind, solar and hydroelectricity all increased because of the low operating costs [2]. Fig. 1 shows the overall energy consumption of different energy sources for 2020. The renewable energy consumption of U.S. in the year 2020 has reached a high of 12% of the total U.S. energy demand [3]. This increase in consumption of the renewable energy for the past year shows that renewable energies are gaining attention around the world. Renewable energies such as solar, wind and tidal energies can produce electricity without the emissions of CO_2 [4–6]. The above-mentioned energy sources are dependent on seasonally varying atmospheric drawbacks (e.g., sunlight level and intensity, wind force) and geographical constraints which when combined with annually varying energy demand results in renewable energy excesses or deficits. Because of the unpredictable nature of renewables (wind, solar and tidal), hydrogen, not seasonally-dependent, can be used as an alternative source of renewable energy which can be stored for a longer time and used when demand arises [7–9]. Hydrogen, the lightest most abundant element with high energy content, can be produced from both renewable and non-renewable sources. Hydrogen can be stored and used as a fuel in transportation, in power generation systems and in internal combustion engines [10]. Hydrogen is also the main decarbonization alternative for hard-to-abate sectors (aviation, shipping, iron and steel production) because it can be stored, combusted and combined in chemical reactions in a ways that are similar to natural gas, oil and coal [11].

The seamless operation of large-scale intercontinental hydrogen value chains will be dependent on the availability of suitable storage capacity and functionality. Geological storage is considered as the best option for large-scale and long-term storage whereas tanks are considered as the best option for short-term and small-scale storage [11]. Hydrogen can be stored in various geological structures such as salt caverns, depleted oil/gas fields, aquifers, and hard-rock caverns.

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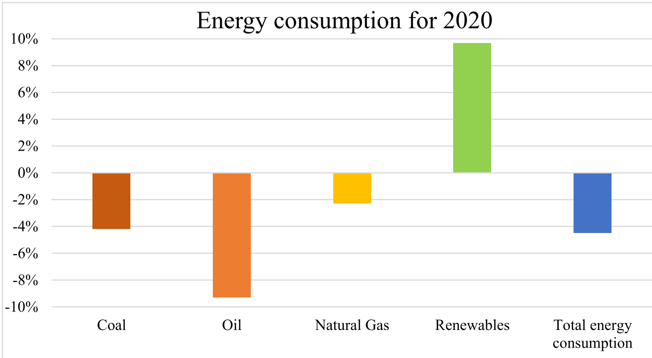

Fig. 1. Overall energy consumption of different energy sources for 2020 [2].

Subsurface storage of hydrogen lacks experience compared to other types of storage. Numerous research projects have been initiated on underground hydrogen storage in the last decade, such as Hychico (2010), ANGUS+ (2013), H2STORE (2012), HyInteger (2016), HyUnder (2012), Underground Sun Storage (2016), Hystories (2020), HyUSPRe (2021) and HYPSTER (2021) [12–15].

These studies aim to investigate the feasibility of the production, storage and utilization of hydrogen and commercialization of Underground Hydrogen Storage (UHS). UHS has many advantages over other storage systems and some of the major benefits are:

(i) Security since the storage facilities are mostly unaffected by fire, harsh weather, military, or terrorist attacks [16].
(ii) Variety in the volume of accessible space, from a few Mm^3 to hundreds of Mm^3,
(iii) Low cost, as operational and investment expenses are lower than those of large-scale surface tanks, and electrolyze costs are minimal [17]
(iv) Existence of ample appropriate geological storage locations, which allows for decentralized storage [18].

Additionally, when it comes to the storage of electricity, present storage methods are restricted in terms of both capacity and discharge time [15]. Fluctuations in energy use and output can be balanced by using large-scale energy storage. Fig. 2 clearly shows that energy storage using hydrogen can be done on a far larger scale than many other current storage approaches. UHS is akin to natural gas storage in many ways. Geological structures for underground hydrogen storage should be chosen based on a detailed geological study that considers both geological and engineering criteria. For over three decades, scientists and engineers have been working to store hydrogen in geological formations for long and seasonal periods. However, there are many lab-scale and large scale studies needs to be done before hydrogen storage can be incorporated into a real-world system [19].

2. Analysis of the literature

The operational process of UHS is same as UGS but the uncertain behavior of hydrogen in subsurface needs further investigation for the efficient UHS. Numerous studies on the feasibility of hydrogen storage in subsurface have been conducted by researchers in the past decade. The selection of suitable storage site as the first step plays an important role in UHS process. The multi criteria decision-making (MCDM) methods are proved to be useful for subsurface site selection. This technique is based on modelling and analyzing the decision process in terms of criteria and alternatives based on decision makers' (DMs') judgements. Decision makers are experts in the field of subsurface hydrogen storage [19]. The criteria used in this method include technical, environmental, and economical parameters. The MCDM has various methods of which Fuzzy set (FS) proposed by Zadeh [20] is the most common one later improved by various researchers. The methods proposed by various researchers for subsurface site selection is presented in Table 1. These studies considered four main parameters including technical, economical, health, safety and environment (HSE), and social criteria that influence sustainable UHS sites [21,22].

After selecting a suitable site, its subsurface properties must be evaluated using the core samples obtained from it. The reactivity of hydrogen with subsurface samples was studied by various researchers

Table 1
: Site selection methods for subsurface storage.

References	Storage Component	Methods used
[23]	Carbon dioxide	• Fuzzy TOPSIS • Fuzzy ELECTRE I • Fuzzy VIKOR
[24]	Natural Gas	• Choquet integral
[21]	Hydrogen	• Analytic Hierarchy Process (AHP)
[19]	Hydrogen	• Additive Ratio Assessment (ARAS) • Interval Type-2 Hesitant Fuzzy Sets (IT2HFSs)
[25]	Hydrogen	• Normal Wiggly Dual Hesitant Fuzzy Set (NWDHFS)
[26]	Hydrogen	• Multi-Attributive Ideal-Real Comparative Analysis (MAIRCA) • Trapezoidal Fuzzy Neutrosophic Numbers (TrFNN)
[27]	Hydrogen	• Fuzz-Delphi

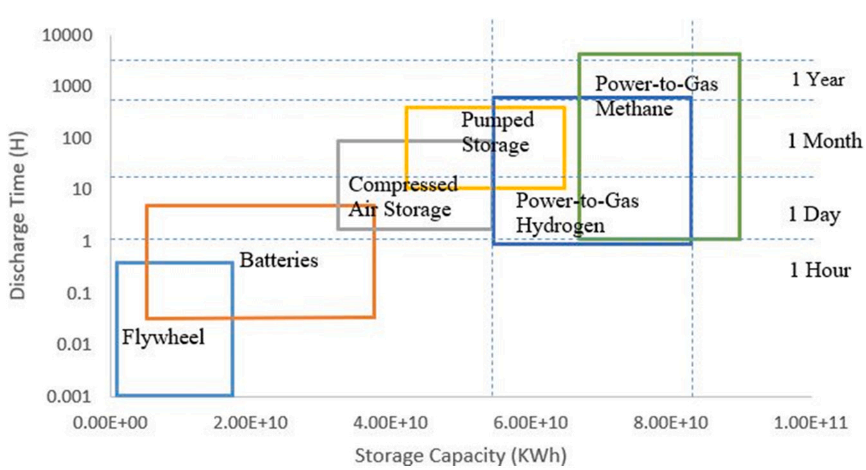

Fig. 2. Discharge time vs storage capacity for different storage techniques Moore & Shabani [138].

presented in Table 2. Geochemical reactions of hydrogen with subsurface formation and storage capacity of the storage system were simulated by some researchers (Table 3). Alireza E. Yekta et al. [28] conducted both experimental and simulation studies to analyze the geochemical reactions of sandstone in presence of hydrogen. The experimental results demonstrated that there was no change in mineralogy of sandstone in presence of hydrogen. However, the results of the simulation study demonstrated minor changes in the mineralogy.

Four available subsurface storage structures are: i) depleted gas/oil reservoirs, ii) aquifers, iii) salt caverns, iv) lined-hard rock caverns. The cyclic nature of both production and consumption on hourly, daily, and seasonal scales necessitates adaptable storage solutions on all these timescales. Storing energy underground may provide large storage capacities for a large amount of energy on the daily, weekly, and seasonal scale [29]. The usage of subsurface storage for large quantities induces secondary effects such as an increase in pressure, migration of reservoir fluids, geochemical changes, biological changes, changes in geomechanical stresses, and leakage of injected fluids or gases into drinking water aquifers. To prevent these effects, a proper understanding of the system and process is required [17]. This paper gives us an understanding of UHS in various geological structures, challenges involved in them, and their mechanisms. Also, monitoring techniques with the road map for the implementation of underground hydrogen storage are analysed by incorporating both field and laboratory research studies. Finally, some recommendations for future laboratory research are made.

3. Hydrogen properties in the context of UHS

Hydrogen is the most prevalent and lightest element on Earth. H_2 gas is a colorless, odorless, and non-toxic gas. At atmospheric pressure, the melting point is 14 K (−259.14 °C), whereas the boiling point is 20 K (−252.87 °C) [39]. Moreover, the density of hydrogen changes when the temperature alters (Fig. 3: Storage density of hydrogen under certain pressures and temperature conditions [43]Fig. 3). Considering these properties, storing hydrogen at regular atmospheric temperature and pressure is exceedingly challenging [44]. Unlike CO_2, H_2 cannot be liquefied around the normal temperature of 293 K (19.85 °C). Since liquid storage and cryo-compressed storage needs extremely low storage temperatures (−253°C), only compressed hydrogen may be used at underground storage locations [45]. H_2 may be securely held as a gas at pressures ranging from 50 to 300 bar (5×10^6–3×10^7 Pa) and temperatures ranging from 300 to 400 K (26.85–126.85 °C).

The energy density per mass of hydrogen is higher than the hydrocarbons [7,8]. The increase of pressure will lead to an increase in the density of hydrogen which results in an increase in the efficiency of hydrogen storage relative to depth. Hydrogen particles may exist in several states depending on temperature and pressure, as demonstrated in Fig. 4. When comparing the viscosity of CH_4 and CO_2 to hydrogen, the latter has a lower viscosity, with less fluctuation in viscosity with pressure and temperature, and is within the range of normal subterranean storage conditions [46]. Hydrogen has an extremely high thermal conductivity, which rises with pressure and temperature. It has a threefold higher heat conductivity than other gases. Like other gases, hydrogen solubility in water increases with rising pressure and decreases with increasing temperature and salinity. Because of its low viscosity, hydrogen has high mobility, allowing it to fill or empty the reservoir more quickly, however displacing the in-situ fluids is more challenging [7,8]. Due to the presence of contaminants, the thermal-physical characteristics of hydrogen rich stream, that is injected, differ from pure hydrogen streams, resulting in more complex engineering and operational concerns such as toxicity, safety, and compression or dehydration needs. Hydrogen storage requires compressing a massive amount of hydrogen gas present [47]. At ambient temperature and atmospheric pressure, one kilogram of hydrogen has a volume of 11 m^3 [48]. To enhance the hydrogen density for storage, compression or chilling below the critical temperature is necessary. Otherwise, the

Table 2
Experimental studies on underground hydrogen storage.

References	Objective	Tests conducted	Summary of results
[30]	H_2 solubility in saline solutions under reservoir conditions was studied.	• Solubility	The solubility data obtained from the test disagrees with the theoretical models. More data and more analysis are needed to deepen the understanding of the system.
[31]	The petrographic and petrophysical variations in reservoir sandstones from laboratory experiments under simulated reservoir conditions were studied. The core samples from two different locations were used for this study. The samples were measured before and after experiments	• Porosity and permeability • Specific surface area • Scanning Electron Microscope (SEM) • μ-CT scan	The samples which have carbonate and anhydrite has experienced significant petrographic changes after the experiments compared to other samples.
[32]	The capillary pressures and relative permeabilities of sandstones have been measured for hydrogen-water system at two different conditions were studied. Capillary pressure and relative permeability are one of the parameters governing the fluid migration in subsurface.	• Porosity and permeability • Capillary pressure • Relative permeability • Surface tension • Contact angle	The capillary pressure and relative permeability were similar in both experimental conditions which is not the case in CO_2-water system. The results from this study are applicable to a wide range of pressure and temperature conditions.
[33]	The hydrogen wettability of clays (Kaolinite, Illite, and Montmorillonite) at various pressures were studied. Clays are present in the subsurface which affects the injection/ withdrawal rates, storage capacity, and leakage of H_2 in UHS.	• Contact angle measurement	All the clays used in this study were found to be water-wet which implies that water-wet clays are efficient in trapping hydrogen gas.
[34]	All the formations of have minute concentrations of organic acids which affect the wettability of the rocks. The objective of this study was to observe the change in wettability of sandstone rock aged in organic acid in the presence of hydrogen.	• Elemental composition and organic surface coverage • Surface roughness measurement • Contact angle measurement	The wettability of the sample aged with organic acid changes from water-wet to intermediate water-wet which will affect the underground hydrogen storage.
[34]	To study the potential of a saline aquifer in a sandstone formation to store hydrogen. Micro-computed tomography was used to visualize the pore network of core in 3D.	• Micro-CT • Core flooding • Avizo 2021 (Image analysis)	The result of the study shows that 65% of the pore volume is occupied by H_2 and 35% brine after hydrogen initial saturation, further injection of brine results in 41% H_2 saturation which

(continued on next page)

Table 2 (*continued*)

References	Objective	Tests conducted	Summary of results
			is trapped residually. The H_2 occupied all the large pores of the sample.
[35]	To study the contact angle of hydrogen in contact with sandstones at various pressure, temperature, and brine salinity using captive-bubble method.	• Contact angle measurement	Contact angle is not affected by temperature, pressure, and salinity. The wettability of the sandstone is not affected due to the presence of hydrogen.

interaction of hydrogen with another substance must be employed to minimize repulsion [25]. Another key concern for hydrogen storage systems is the reversibility of hydrogen injection and withdrawal. Six techniques for high volumetric and weight density reversible hydrogen storage have been proposed: as a gas, a liquid, a gas adsorbed at a material surface, metal hydrides, complex hydrides, and chemical processes [49]. The hydrogen tightness of argillaceous rocks has been calculated to be 2% throughout one storage cycle, defined as the loss of hydrogen via diffusion in these rocks and its solubility in reservoir water. Hydrogen diffusion through the salt walls may be a concern since salt caves are inherently dry structures. Chemical reactions are another concern that might develop when hydrogen comes into touch with ambient rocks [40]. Reactions in the mineral matrix appear to occur very slowly in the absence of catalysts and at temperatures below 100 °C. The responses may substantially speed up if the pressure increases. When a metal pipeline is exposed to hydrogen over a lengthy period, especially at high concentrations and pressures, its durability substantially decreases. The effects of hydrogen on the characteristics of steel alloys are referred to as hydrogen blistering, hydrogen-induced cracking, and hydrogen embrittlement. Other problems might arise because of hydrogen leakage through pipe walls. The permeability index for hydrogen is four to five times greater than that for methane in a typical polymer pipe used in natural gas distribution systems. In contrast, the loss of natural gas and hydrogen mix 60:40 estimated for US pipe installations would equal just 0.0002% of the entire quantity of gas utilized in the US. As a result, such leaks are considered minor from a financial standpoint [48]. Table 4 shows the comparison of properties of various gases.

At different time scales, beginning with medium and long-term storage, underground hydrogen storage may be a viable alternative. However, there are several obstacles in this sector. Specific characteristics of gaseous hydrogen will need to be carefully controlled to avoid leakage during subterranean storage, transit, and extraction. Because of their huge storage capacity, large-scale UHS is conceivable in depleted natural gas reserves [50]. Although gas reservoirs have been proved to be sealed, there is limited understanding regarding hydrogen-induced fluid-rock interactions for such reservoirs due to a paucity of field pilots. Additionally, economic and environmental evaluations are necessary before commercializing UHS projects [51].

Table 3
: Simulation studies on underground hydrogen storage.

References	Objective	Software Used	Storage medium	Injected gas	Summary of results
[36]	Based on an actual geological structure, a hypothetical storage site is developed to study the system behavior of subsurface porous media hydrogen storage.	• Eclipse (E300)	Aquifer	N_2 and H_2	Subsurface porous media has potential for storing large quantities of hydrogen. The storage performance of the subsurface increases from the first to fourth cycle.
[37]	A model was developed to compare and study the hydrodynamic effects of injecting hydrogen and injecting methane to subsurface.	• DuMux	Aquifer	H_2 and CH_4	The gravitational forces were dominant in low injection rates and viscous forces become dominant in high injection rates. It has been observed that at higher injection rates lateral spreading of hydrogen was faster than methane.
[38]	A reservoir was modelled based on a Rough Gas Storage Facility located on UK to study the reactions between H_2 and the mineral components present in the reservoir.	• PHREEQC	Depleted gas reservoir	H_2	Under reservoir conditions, the clay-bearing sandstone and iron oxides was stable. Around 3.7% of H_2 could be lost due to biological activity for the modelled scenario.
[39]	A 3D model was developed to study the different extraction well configurations in a saline aquifer for three annual injection-production cycles.	• COMSOL	Aquifer	H_2	Cushion gas is not required when H_2 is stored in steeply dipping structures. The major limitation of storing H_2 in saline aquifers is up coning. Hydrogen recovery is efficient if several shallow extraction wells are located beneath the caprock.
[40]	The evaluating the possibilities of storing hydrogen in deep aquifer. A mathematical model of aquifer was modelled for one injection and withdrawal well on the assumption of not exceeding the fracturing pressure and capillary entry pressure.	• PetraSim TOUGH2 software	Aquifer	H_2	The capacity and maximum amount of hydrogen which can be stored and injected into the aquifer was found from the simulation. Recovery percentage of hydrogen increases with more withdrawal cycles. Hydrogen storage in deep aquifer is possible but more detailed understanding is required.
[41]	A 3D heterogeneous model was developed to study the effect of caprock and hydrogen injection rate in a sandstone reservoir.	• TOUGH2 • EWASG (Equation-of-State for Water, Salt and Gas)	–	H_2	High injection rates of H_2 leads to the leakage of H_2 and reduces the amount of hydrogen recovered.
[42]	To study the hydrogen loss caused by geochemical processes as function of temperature and pressure by geochemical modelling.	• PHREEQC	Depleted oil and gas reservoir	H_2	Temperature and pressure have a limited effect on hydrogen loss. The presence of calcite in subsurface results in the loss of hydrogen.
[7]	The injection, and storage of hydrogen in an open saline aquifer was simulated and studied. The role of cushion gas in an aquifer is also studied.	• Petrel • GEM	Depleted gas reservoir	H_2	Open saline aquifers have a potential for storing hydrogen. Injectivity and productivity of hydrogen is controlled by cushion gas. Cushion gas also influences the storage capacity of hydrogen. The geological parameters such as, reservoir permeability, and reservoir depth has to be considered for cushion gas to working gas capacity.

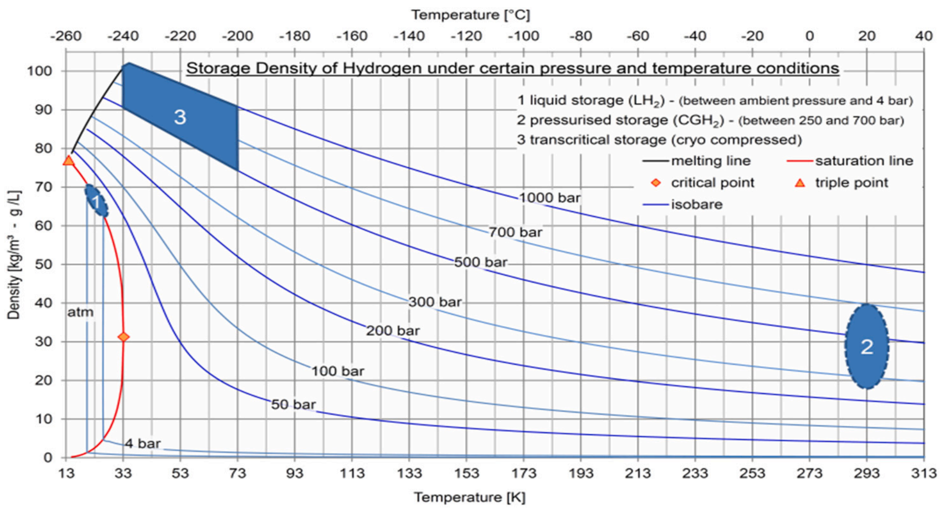

Fig. 3. Storage density of hydrogen under certain pressures and temperature conditions [43].

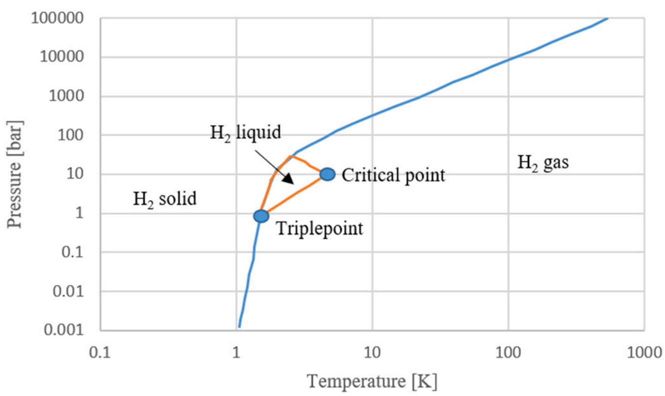

Fig. 4. Hydrogen phase diagram [48].

Table 4
Comparison of properties of various gases [48].

Properties	Hydrogen	Methane	Gasoline
Molar mass	2.016	16.043	~107
Density at NTP (kg/m^3)	0.08375	0.6682	751
Heating value (kJ/g)	120–142	50–55.5	44–47.3
Flammability limits (vol% in air)	4–75	5.3–15	1–7.6
Minimum ignition energy (mJ)	0.02	0.29	0.24
Autoignition temperature (°C)	585	540	228–471
Detonability limits (vol% in air)	11–59	6.3–13.5	1.1–3.3
Diffusion coefficient in air at NTPa (cm^2/s)	0.61	0.16	0.005

4. Hydrogen storage in geological structures

Underground energy storage is best for long-term and large-scale usage. Compressed Air Energy Storage (CAES) is a storage method that may be used for short-term (hourly) storage [17]. Porous media, in which the gas is stored in the pore space of sandstones or carbonate formations, and cavern storage, in which the gas is contained in excavated or solution-mined caverns in the thick rock, are the two types of geological locations used to store gases underground [52]. The prerequisites and standards for selecting a deposit or structure as a UHS location must be based on comprehensive geological investigations utilizing deposit engineering techniques. Structure depth, thickness, tightness, reservoir pressure, reservoir characteristics (porosity and permeability), geo-mechanical properties, and appropriate features of insulating caprocks are the essential criteria. There are also additional technological, environmental, legal, economic, and other considerations. It is also crucial to select the right exploitation parameters, taking unique reservoir features into account, as well as injection and withdrawal pressures that do not exceed the fracture pressure of the subsurface formation. Most of the literature on underground hydrogen storage in geological formations is based on natural gas and carbon dioxide storage analogues [48]. This is due to a wealth of practical expertise in storing these gases in geological structures. To inject and recover gas from subsurface geologic storage, sufficient permeability is necessary, as is high porosity for storing the requisite volume of gas, and permeability should not be too high to prevent hydrogen from leaking [52]. In porous depleted oil/gas reservoirs and aquifers, impermeable caprock is required. The major rationale for storing hydrogen gas is to fulfil both base and peak load needs. Base load storage facilities withdraw once or twice a year, whereas peak load storage facilities withdraw many times over a short time-period to fulfil short-term demand. Peak load demands are met by salt caverns, mining caverns, and aquifers, while base load demands are met by depleting oil/gas reservoirs [53]. Like natural gas, H_2 may be stored in geological structures such as salt caverns, aquifers, and depleted gas reservoirs that will be explained in the upcoming sections.

4.1. Salt caverns

Salt caverns are artificial caverns created in deep subterranean salt deposits constructed from the surface by injecting water into a well in the salt rock in a controlled manner [52]. Salt domes or bedded salt formations are the ideal locations for creating salt caverns. It can be created by solution mining such as putting fresh water into the big holes to flush them out [49,54]. Because of the homogenous nature of salt domes, they can be used for constructing the caverns within them. Bedded salt formations can be found in shallower depths and, they are thinner than salt domes. Formations present in bedded salt formations include salt (halite) and non-soluble beds such as dolomite, anhydride, and shale [55]. The lithologies of the caverns formed in a bedded salt formation vary, and each layer has its own set of characteristics that impact creep rates, deformation, and slip between bedding planes ([56]). Because of the slip, gas can move laterally [53]. There are three basic conditions for selecting the location for the construction of an underground storage cavern: the presence of salt layers in appropriate depths and thickness, presence of small quantity of insoluble parts, availability of ample supply of freshwater and ecological and environmentally safe disposal methods of brine [48]. The maximum depth of salt caverns is 2000 m, they can hold up to a volume of 1,000,000 m^3, and their height and diameter range from 300 to 500 m and 50–100 m, respectively [57]. The operational pressure in the salt cavern is 30–80% of the lithostatic pressure at the depth of cavern roof [58]. When hydrogen is injected at a constant pressure, no cushion gas is required because hydrogen is injected with saturated brine whereas hydrogen can also be injected at variable pressure in which the cavern is occupied by one third volume of cushion gas [55]. Variable or constant pressures for injection can be used in caverns [53]. When the working gas is removed at different pressures, the reservoir pressure drops, causing salt creep and compromising cavern integrity. To avoid creep, the reservoir pressure must be maintained, which necessitates the use of a specific quantity of cushion gas. The frequency of injection and withdrawal cycles in salt caverns is large, up to 10–12 per year [58]. Salt caverns with smaller diameters are built with the knowledge that changing leaching conditions may make achieving the intended cavern form difficult [59]. The leak rate in salt caverns is approximately 1% which only transpires through leaky wells [53]. Salt caverns are feasible alternatives for subterranean hydrogen storage due to their tightness and mechanical characteristics, as well as their resistance to chemical reactions. Additionally, the same well can be used for injection and withdrawal and storage facilities can be easily managed [52]. Low storage capacity is one of the disadvantages of salt caverns. It cannot be operated at higher depths than 2000 m as it requires substantial volumes of compressed gas which leads to high pressure buildup in the cavern. Whereas for lower depths (500 m) the requirement of cushion gas is less which leads to less operational cost. Large amount of water is required for leaching the cavern. Another disadvantage of salt caverns is corrosion of the casing equipment, which is caused by brine, air, microbes, effect of hydrogen on steels, and gas injection velocity [60,61]. Effects of corrosion are discussed in detail in the challenges section.

4.1.1. Salt cavern shape and volume

The volume of a salt cavern is determined by the form and size of different rock salt layer thicknesses. Lowering the operating pressure and therefore the volume of cushion gas while boosting the highest operating pressure improves the operational economics of an underground hydrogen storage cavern [62]. Fig. 5 shows the simplified shape of a salt cavern. The volume of a salt cavern will shrink if the minimum operating pressure is reduced too low due to the creep of the surrounding salt [63]. The factors such as depth of the cavern, location of the cavern, geological and mining conditions, strata lithology and the solubility of salt need be considered for cavern shape selection [64,65].

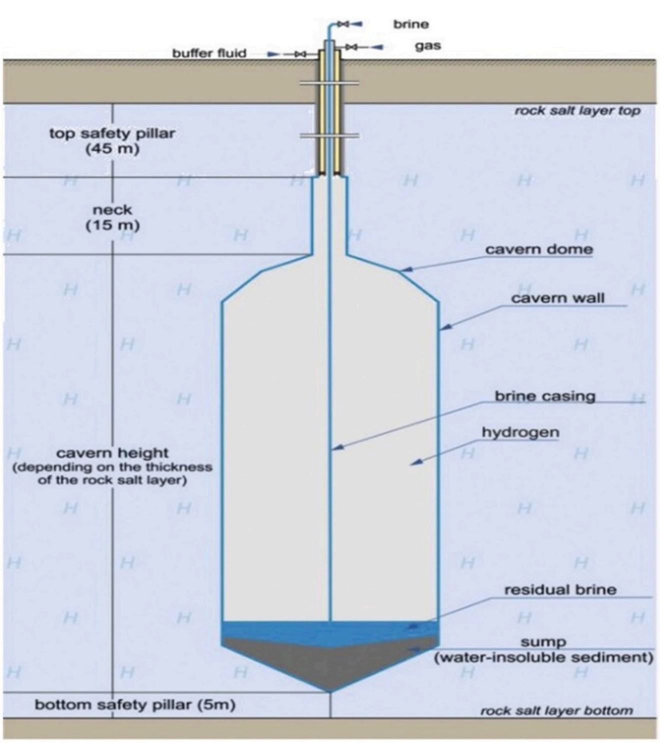

Fig. 5. Simplified shape of analysed cavern [59].

4.1.2. Salt cavern design and placement

The properties of the salt deposit heavily influence the design of a salt cavern; for example, the depth of the cavern (Fig. 6). Hence, to ensure the cavern's long-term viability and safety, several issues must be addressed. To ensure the cavern's geo-mechanical safety, the thicknesses of salt layers in the hanging-wall and footwall (above and below) must be defined and assessed. These layers' minimum thicknesses for safe operation are calculated as a function of cavern diameter. Furthermore, a minimum height-to-diameter ratio of 0.5 is specified for salt caverns in bedded salt deposits. At 1200 m depth and 270 bar overburden pressure, capsule-shaped salt caverns are more stable and have a reduced stress risk than elliptical or cylindrical salt caverns [46,66].

4.2. Depleted oil and gas reservoirs

Due to their substantial pore space, tightness, high permeability, lack of gas admixtures such as hydrogen sulphide, and ability to drill fresh wells, depleted natural gas reservoirs are widely used for underground storage [67]. This is the most frequent and popular natural gas storage option. Depleted gas reservoirs are simple to develop, manage and operate due to existing infrastructure [55]. Because of their well-identified geological features, the stability of their caprock, and the pre-existence of the requisite surface and subsurface equipment, they are ideally suited for UHS. Due to the prior use of such areas for oil and gas production, storing hydrogen in exhausted oil and gas reserves reduces geological exploration efforts. On the other hand, cushion gas is required in exhausted oil and gas fields to prevent reservoir rock disintegration [66]. Because of contamination caused by earlier hydrocarbon extraction, contamination control and gas upgrading equipment for purification may be required.

When the working pressure is raised, there will be a loss of stored natural gas. The reservoir must seal the gas within the reservoir to function as a good storage container. Successful traps (structural, such as an anticline, or stratigraphic, such as an impermeable layer, e.g., caprock), high permeability for easy gas movement, and high porosity (combined with reservoir volume) for high storage requirements can all help achieve this [55]. The cushion gas volume, which is necessary to sustain reservoir pressure and withdrawal rates, is equal to half of the reservoir volume. There are still some gas/oil reserves in abandoned reservoirs that might be utilized to satisfy the cushion gas requirement [48]. Only gas fields that satisfied the following criteria had their potential storage capacity calculated: At the time of evaluation, they were

i) Developed and accessible through production wells
ii) Had a minimum depth of 1000 m
iii) Did not contain significant amounts of H_2S (<10,000 ppm)
iv) Had a permeability >0.1 mD (i.e., no stimulation required)
v) Was not being used for storage

The following criteria were added to the working volume criteria:

i) Transmissivity of gas fields >100 mD.m
ii) Gas Initial in Place (GIIP) volume of <30 billion m^3 (because of the anticipated necessary large cushion gas and geological complexity of big fields), and
iii) Initial well productivity of >1 million m^3/day [68].

Because the amount of the present gas in some depleted reservoirs is insufficient to fulfil the cushion gas requirement, extra gas must be injected to maintain reservoir pressure. Wells which are not properly

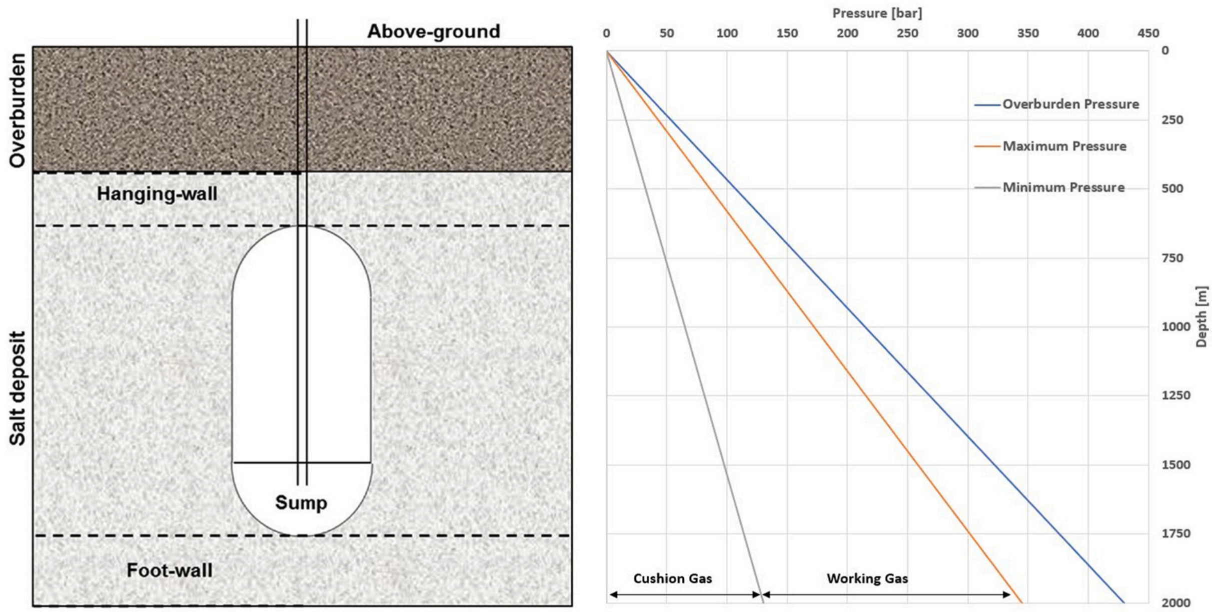

Fig. 6. A simplified representation of an exemplary cavern and estimated pressure limits as a function of depth [66].

sealed are responsible for the loss of cushion gas in depleted reservoirs [37]. Lack of caprock integrity, dissolving into connate water, diffusion into the surrounding groundwater, fingering with surrounding reservoir water, and contamination with pre-existing hydrocarbons are responsible for gas loss in a depleted reservoir [53].

4.3. Aquifers

In areas where depleted reservoirs are unavailable, aquifers can be utilized for gas storage. Aquifers, in general, are porous rocks, like sandstone, located thousands of feet underground which hold water [69]. For efficient storage, the aquifer must have the same geology as depleted hydrocarbon reservoirs. It must have high porosity, permeability, and reservoir capacity [53]. Because of the lack of infrastructure and information about geology they are more expensive than depleted oil/gas reservoirs [69]. To produce effective gas traps, geological structures such as anticlines with impermeable caprock and adequate surrounding hydrostatic and threshold pressures are necessary [70]. To transform an aquifer into a subsurface hydrogen storage site, additional expenses such as installation of above-ground infrastructures such as wells, pipelines, and injection systems are required [71]. Additionally, a system to dehydrate gas must be emplaced [55]. Aquifers without any existing gas require about 80% of the total reservoir volume of cushion gas [69]. The loss of hydrogen and loss of cushion gas is inevitable in aquifers. Sulfate reducing bacteria can pollute stored gas in deep aquifers [72]. The amount of cushion gas necessary for aquifers might be more than that required for depleted oil and gas reserves [66]. Cushion gas extraction may impede reservoir development, but it is more likely to be trapped if reservoir heterogeneity is high and gas cannot escape capillary pressure at pores. Because of the fingering between gas and water in aquifers, gases become unrecoverable, causing the gas to flow down the structure [53]. Gas is also lost through leaking wells, as it is in other storage systems, and some minor losses occur through caprock, dissolving into connate water, and diffusion into the surrounding groundwater.

4.4. Hard rock cavern

Hard rock caverns are a recently created storage option that may be employed in areas where there is no salt or porous sandstone. This technology involves excavation of hard rock caverns and encasing them with steel or plastic liners. The lining functions as an impermeable covering that keeps the gas contained [73]. Normal hard rock caverns, which are not as impermeable as lined caverns, may function at higher pressures than lined caverns. They are made by excavating igneous or metamorphic rocks and placing a concrete layer between the rock and the lining that functions as an impermeable layer. The pressure load must be borne by the host rock in which the liner is installed [73]. The cavern's pressure vessel is the rock mass that surrounds it and functions as a pressure-absorbing medium [55]. For the most common rocks investigated so far, pressures >4 MPa cause the surrounding rock mass to deform both elastically and plastically. The magnitude of these early deformations is crucial since it contributes to the eventual strain on the lining [73]. The purpose of the concrete layer is to transfer pressure loads from the liner to the surrounding rock while also creating a smooth surface for the lining to adhere to. The liner needs to be both gastight and chemically resistant. Although liner is not meant to hold any weight, it should be able to endure modest elastic and plastic deformation caused by the gas pressure exerting to the rock face by the concrete backing. The most common types of liners commercially available are Stainless steel or polypropylene liners [74]. The cushion gas demand is less in hard rock caverns since they are more stable than other storage choices. The hard rock cavern employs a groundwater drainage system around the outside of the cavern to decrease the hydrostatic pressure force on the lining that occurs during the depressurization of the cavern [53].

5. Underground gas storage in the world

Over the last century, underground gas storage facilities have gradually expanded in quantity and capacity, notably on the continents of the Northern Hemisphere. As of January 2010, 642 UGSs were being used globally. Most of them were found in depleted hydrocarbon reserves (476), aquifers (82), and salt caverns (76). Figs. 7 and 8 show the share of worldwide UGS by storage type and the share of UGS by regions. Depleted hydrocarbon deposits are the mostly used structures to store gas. The importance of salt caverns as UGSs has recently increased. According to CEDIGAZ, an International Association Dedicated to Natural Gas Information, 82 of the 202 newly planned and expanded existing UGSs around the world in 2010 were in salt caverns. Germany has extensive experience in underground natural gas storage in Europe. According to CEDIGAZ, there were 680 UGSs facilities in operation

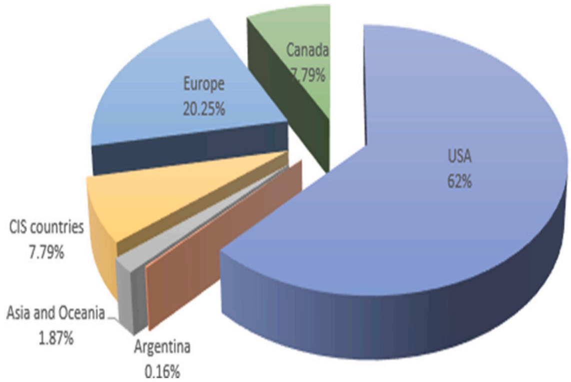

Fig. 7. Share of UGS by regions [48].

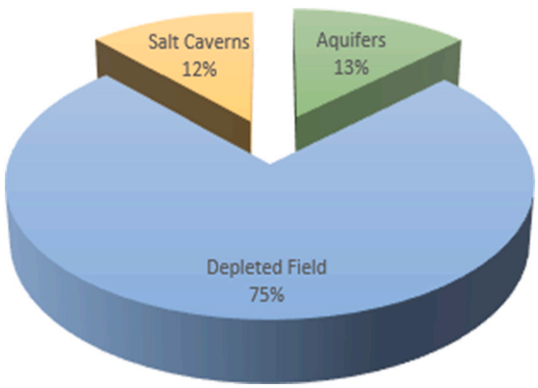

Fig. 8. Share of worldwide UGS by storage type in 2010 [48].

around the world at the end of 2015, with a total working gas capacity of 413 bcm (billion cubic meters). Most of this capacity is concentrated in the three developed gas markets: North America – 38%, the Commonwealth of Independent States (CIS)-29%, Europe-27%. Whereas Asia/ Oceania and the Middle East account for 4% and 2% of the global UGS capacity, respectively. Working gas capacity by the type of UGS facility is distributed as follows: depleted gas fields-80%, aquifers-12%, salt caverns-8% as shown in Fig. 9 in given below [48]. The figures are described in detail in Table 5 given below.

Hydrogen has been stored in three salt caverns on Teesside in the United Kingdom since 1972, and two on the Texas Gulf Coast since 1983. Germany has a huge number of underground natural gas storage facilities. In Russia, many clean hydrogen storage facilities were built

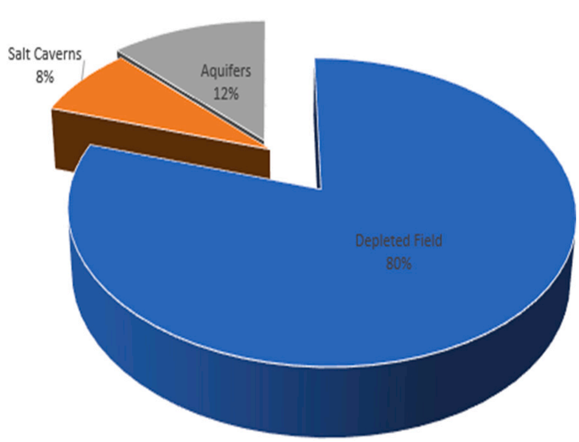

Fig. 9. Working gas capacity by storage type in 2015 [48].

Table 5
Description of figures.

Fig. 7	With 399 UGSs in the United States and 50 in Canada, North America accounted for the bulk of UGSs. Europe ranked second with 130 UGSs, followed by the Commonwealth of Independent States (CIS) countries (50), Asia and Oceania (12), and South America and Argentina each with one facility.
Fig. 8	Shows the share of storage type globally. Depleted field has the largest share of all the storage types because of the number of fields created by oil and gas industry. Salt caverns and Aquifers has almost same percentage of shares.
Fig. 9	Shows the distribution of working gas capacity from various storage types. Depleted field has the highest share in working gas from salt caverns is the least percentage of all.

underground to meet the demands of the aviation sector. The experience with hydrogen storage in subsurface caverns has demonstrated that hydrogen can be securely kept for lengthy periods of time [62,63,75]. Table 6 displays the locations of existing hydrogen storage caverns in the United States and the United Kingdom. A British company in Teesside, Yorkshire, stores one million m^3 of pure hydrogen (95% H_2 and 3–4% CO_2) in three salt caverns at a depth of around 400 m with a pressure of five MPa. Conoco Phillips has been storing 95% hydrogen in the Clemens salt dome in Texas since the 1980s [36]. The ceiling of the cavern is roughly 850 m below ground level. The cavern is formed like a cylinder, with a diameter of 49 m and a height of 300 m. The capacity of useable hydrogen is 30 million m^3, or 2520 metric tons. For some years, Praxair has been using UHS in a salt cavern in Texas to enable peak shaving of their hydrogen production [76]. Underground hydrogen storage is part of the energetic cycle, which includes energy generation from renewable sources, hydrogen conversion, hydrogen storage, hydrogen reconversion, and energy consumption. The component of the cycle relevant to subsurface hydrogen storage involves conveying it via a specialized conduit from the site of production (electrolysis) to the point of injection. The surface installation at the storage location includes compression, decompression, purification, and dehydration sections [77]. A subsurface injection/extraction well and a cavern produced by dissolution, in the case of salt storage, or a tight structure in porous rocks – aquifer or depleted hydrocarbon deposit – make up the subterranean portion. For storage and recovery, hydrogen must be compressed and decompressed between the minimum and maximum working pressures, which vary depending on the depth of the cavern [54]. Up to 10 injection-withdrawal cycles may be done per year [48].

6. Challenges faced in underground hydrogen storage

Some of the challenges faced in underground hydrogen storage are, site selection, geochemical reactions, microbial growth in reservoir, well integrity, and geological integrity of caprock. Fig. 10 depicts some of the issues to consider while storing hydrogen in porous medium [7,8].

Table 6
Existing hydrogen storage caverns in the USA and UK [48,52,78].

	Clemens (USA)	Moss Bluff (USA)	Spindletop (USA)	Teesside (UK)
Geology	Domal salt	Domal salt	Domal salt	Bedded salt
Operator	Conoco Phillips	Praxair	Air Liquide	Sabic Petroleum
Stored fluid	Hydrogen	Hydrogen	Hydrogen	Hydrogen
Commissioned (year)	1983	2007	Information not available	~1972
Volume (m^3)	580,000	566,000	906,000	3 × 70,000
Reference depth (m)	930	> 822	1340	350
Pressure range (bar)	70–135	55–152	68–202	~45
Possible working gas capacity H_2 Mio (kg)	2.56×10^6	3.72×10^6	8.230×10^6	0.83×10^6

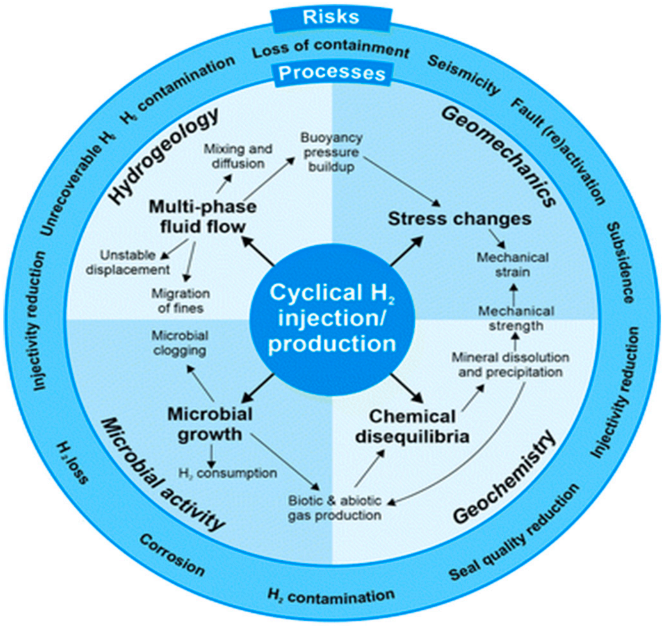

Fig. 10. Scientific challenges in subsurface storage of hydrogen [7,8].

6.1. Site selection

For successful operation of large-scale underground hydrogen storage facilities, site selection is seen as a critical component [27]. The depth and storage capacity of a potential site are critical since they can affect the entire UHS process. Therefore, more information must be collected regarding the depth and storage capacity of a storage site for efficient storage [52]. Information on the seismic activity of a potential storage site is an important safety concern in the context of UHS, because they might affect the integrity of storage facilities (e.g., the fracturing of caprock or caverns). As a result, seismic hazard maps of potential sites must be created and studied to ensure that the site is free of seismic activity [9,49].

Bauer et al. [17] created a workflow for safe and efficient use of geological structures for the UHS. To estimate the number of feasible storage sites and their environmental consequences, in addition to planning, a proper evaluation of common effects when hydrogen is injected on the subsurface is required. The main obstacle for proper planning is lack of subsurface information and monitoring. The virtual subsurface energy storage sites are suggested in this study. The virtual sites are made of data from existing site investigations from typical sites for subterranean applications, including energy storage. Since they are synthetic sites, all the characteristics are known, and any missing data may be supplied with the data available. This approach can be used to simulate storage activities or to investigate the impact of permeability heterogeneity on the consequences of operations. They may also be used to investigate the effects of nearby storage operations as well as the effects of protected compartments like groundwater aquifers on storage sites. The workflow for virtual storage site assessment is shown in Fig. 11. This workflow may be used to assess storage capabilities for hydrogen, methane, or compressed air in caves or aquifers. This virtual methodology allows for the measurement of the impacts of storing hydrogen on the subsurface, other uses, and protected commodities, as well as the assessment of monitoring methods for these subsurface usages. Deep subsurface storage of renewable energy in the form of gases or heat in caverns and porous rocks, as well as shallow heat storage via near-surface geothermal systems, both are possible using this approach. Furthermore, principles for subsurface usage planning may be established using the results of these evaluations and extrapolated to similar geological situations [17].

6.2. Microbial growth in reservoir

A diverse microbial world is present in the earth crust and the subsurface [79]. The presence of microbes is one of the most important issues in hydrocarbon reservoirs that must be considered for the viability of hydrogen storage. Microbes can arise in the subsurface because of natural processes or human operations such as drilling, pumping, or mining. Short- and long-term consequences of subsurface operations on the microbial world have not been thoroughly investigated [79]. Permanent loss of hydrogen owing to the conversion of hydrogen into products like CH_4 and H_2S is irreversible harm that microbial development may bring to a reservoir. The continuous development of

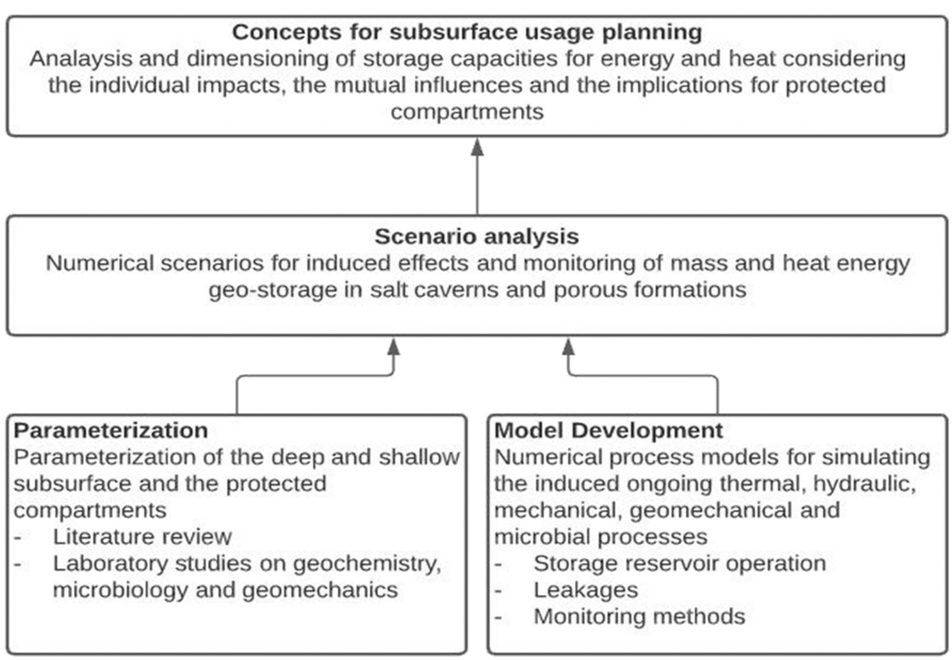

Fig. 11. Workflow for virtual storage site assessment [17].

microorganisms in the reservoir might cause pore blockage resulting in a decrease in hydrogen injectivity. Bacteria and Archaea are the two-primary single-cell types of microorganisms that generate numerous biochemical and geological processes [80]. They were discovered at a depth of 2–3 km [81], with 10^4 and 10^8 cells per gram of rock, and an estimated total number of 2–6 $\times$ 10^{29} cells within the continental subsurface [82]. The two mechanisms that contribute to the consumption and production of hydrogen in the subsurface are the chemical reactions involving abiotic and biotic components. Abiotic components are nonliving, such as water, rock minerals, and gases, whereas biotic components are living, such as bacteria. Methanogenesis, acetogenesis, sulfate reduction, and iron (III) reduction are some of the typical biotic processes that can transpire in underground gas storage wells. At these temperatures, the archaea bacterium, a methanogenic bacteria family, thrives. This species, unlike others, can live underground at extreme depths where there is no oxygen [60]. The temperature range for microbes life is −15 °C up to 121 °C, pH range is zero to 11, with significant variety and cell number between pH 6–7, and there are no apparent pressure or brine salinity limits [31]. Sulfate-reducing bacteria have also been discovered in underground gas storage, causing corrosion in subsurface well completions and surface facilities. The presence of manmade materials like steel components in gas wells enhances abiotic reactions. Because of casing corrosion and pyrite or Fe (II) oxidation, the cement sealing capacity will be decreased [83]. The most common concentration of hydrogen that can be added to natural gas without the occurrence of above-mentioned effects is around 5–10% [60]. Other variables that can limit the existence of microorganisms include high levels of harmful chemicals, poor water activity, high radiation, and low rock permeability. Microbes can acquire energy by oxidizing electron donors while simultaneously reducing an electron acceptor [79]. The reduction of hydrogen injectivity because of the growth of microbes is a common problem encountered in the subsurface.

The microbial activity in artificial salt caverns is less compared to other storage sites. The cavern's small surface area prevents the development of biofilm and clogging. The saline and sump present in the high salinity salt cavern induce osmotic stress in cells, resulting in greatly reduced variety of microbes [32]. Halobacterium is a particularly evolved organism that can survive in salty surroundings, even at concentrations of 100–150 g/L. When sulphate is present, the results reveal that there is a substantial danger of microbiological H_2S production in salt caves at the brine-gas-interphase [79].

The storage site Lobodice in the Czech Republic porous media storage was investigated and found that at the comparatively low temperature of 35$^\circ C$, about half of the H_2 (45–60%) of the stored town gas was microbially transformed into methane or hydrogen sulfide. Šmigáň et al. [84] observed that decreasing reservoir pressure results in increasing the methanogenic microbial cell numbers. The transformation of H_2 into CH_4 is found in methanogens cultivated in the lab. Another example of porous media storage is the former town gas storage site in Ketzin, Germany, where total gas volume loss, corrosion, permeability changes, and gas composition changes with a loss of CO and a gain of H_2, CH_4, and CO_2 were observed [85]. Contrary findings were reported for the town gas storage in the saline aquifer at Beynes, France, while no operational difficulties or H_2 losses were recorded [85,86]. Other researchers, on the other hand, found significant microbial activity in response to changes in gas composition [62,87]. In the Underground Sun Storage project in Lehen, Austria, 10% H_2 from renewable sources was blended with natural gas for a four-month testing period and only 18% of the injected H_2 was retrieved, although there was a concomitant rise in CH_4 [88]. The percentage of CO_2 in the subsurface has dropped from 0.2% to 0.05%. The fluids produced from the reservoir shows acetate production up to 100 mg/L over the withdrawal period and a decrease in sulphate from 20 mg/L to 0 mg/L. The DNA and RNA results showed that microbes changed from bacteria to Archaea. Different metabolisms were activated because of the foregoing results, including the activation of microbial methanogenesis, which resulted in CO_2 and H_2 consumption, demonstrating that methanogens may live even with extremely low levels of CO_2. Acetogenesis and sulphate reduction appear to have been boosted as well [89]. The Underground Sun Conversion Initiative, a follow-up project, is concentrating on using the observed methanogenesis and converting H_2 and industrial CO_2 into methane inside the reservoir [79].

The existence of diverse microbial activities can result in a variety of UHS side effects. Microbial activity observed in UHS can also be seen in the oil and gas and geothermal sectors. The following are some of the negative consequences of subterranean H_2 storage.

- Gas mixture changes: Metabolism of microbes can lead to decreasing H_2 content, increasing other gases such as methane or H_2S, and increasing CO_2 as it is a by-product of the decomposition of microbes. Small concentrations (10–20 ppm) of H_2S can affect gas quality, material integrity, safety, and health making gas treatment a necessity [90]. The loss of H_2 due to microbial activity increases steadily over H_2 injection/withdrawal cycles whereas the loss of H_2 by diffusion increases after the initial cycle. The reduction in CO_2 in the gas mixture indicates microbial activity, which is also indicated by an increase in the concentration of methane in the gas mixture [79].
- Souring and H_2S formation: The toxic and corrosive H_2S gas can be formed due to the presence of active microbial sulphate reduction. Sulphate must be present in the subsurface, either dissolved in water or in the form of sulphidic minerals, for microbiological processes to take place. H_2S will precipitate with the available Fe_2^+ if dissolved iron or ferrous iron minerals are present. Any reduction in dissolved iron and sulfide contents in the re-produced liquids can be used to identify this process [47].
- Steel corrosion by microbes and H_2S: Microbially Influenced Corrosion (MIC) is an issue which is caused mainly due to the presence of microorganisms that affects steel infrastructure in subsurface [91]. The intricate interaction of abiotic and biotic corrosion processes can result in largely localized corrosion in steel infrastructure, as well as eventual equipment failure. MIC is difficult to detect because it is very confusing to distinguish from chemical corrosion and exhibits a wide variety of corrosion rates and occurrences. A combination of corrosion properties, corrosion products, and microbiological activity can be used to determine the existence of MICs. Furthermore, H_2S produced by microbes can accelerate corrosion and cause H_2S-induced stress cracking [92].
- Microbial-induced plugging: The decrease in pore space occurs at subsurface storage sites due to microbial biomass plugging/clogging, exopolymers, or microbially induced mineral precipitation, which results in a reduction in permeability and, as a result, in injectivity as well. This plugging/clogging transpires because of the presence of iron-oxidizing microbes which can act as a catalyst to redox reactions. Microbial Induced Carbonate Precipitation (MICP) is another kind of plugging potential in underground storage sites. Plugging can be identified by a decrease in injectivity and/or an increase in injection pressure. Biomass and mineral plugging can both be severe and long-term, however, mineral precipitation is thought to stay longer than biomass plugging [93].
- Dissolution of minerals and change in reservoir properties: The activity of acid-producing microorganisms, acetogens or heterotrophic microbes, as well as the dissolution of carbonate and other easily dissolvable minerals induced by the generated acids, can lower the pH of the reservoir fluid. Mineral dissolution in carbonate-containing reservoirs can influence permeability, porosity, and fluid flow behavior of the reservoir. A small increase in temperature in a reservoir could be an indicator of strong microbial activity. This behavior, however, has not been observed in an H_2-storage facility [94].
- Possible effects of hydrogen leakage: Hydrogen leakage in the subsurface can transpire by operational problems, well integrity issues,

cap rock diffusion, or geological fractures. Aside from the dangers of hydrogen explosions, it has an impact on microbial populations in soil and groundwater, as well as related nutrient cycles. The produced products of the sulphate reduction and acetate generation process may cause the leakage. Changes in the microbiology of groundwater and topsoil caused by hydrogen might lead to long-term chemical changes in the water and soil, which must be monitored [79,95].

The key takeaway from all industries is that preventing microbial activity is always better and more cost-effective than treating it. To avoid failures induced by microbial activities, an appropriate monitoring strategy must be created [13,96].

6.3. Cementing

For safe and dependable storage of H_2 and Natural gas mixtures, the integrity of injection and withdrawal wells must be addressed in addition to storage reservoir integrity Shi, Jessen, & Tsotsis [139]. Well cementing plays a major role in ensuring the integrity of a well. Mechanical and chemical degradation occurs in the cement during H_2 storage. Mechanical degradation of the cement occurs due to frequent injection and withdrawal cycles [91]. Chemical degradation occurs due to the presence of hydrogen sulfide, and carbon dioxide in the subsurface [97]. If proper attention is not given to the effect of underground storage on the cement sheath and the casing string, this might lead to long term wellbore integrity issues [98,99].

The addition of hydrogen with natural gas affects well integrity and well completions owing to the high diffusivity of hydrogen. Mixing hydrogen with methane results in lower viscosity compared with methane. If the viscosity of a gas mixture reduces, the gas will flow through the cracks present in a cement seal. The leakage is low if the concentration of hydrogen is also low, but if the concentration of the hydrogen increases the leakage will increase as well [100]. Utilization of finest completion cements with possible low concentrations of SiO_3 in cement binders can reduce the leakages in UGS wells that operate in presence of hydrogen with natural gas. The formation of drying voids and low fissures can be avoided by the usage of low water-cement ratios. Because the redox reaction (H_2/H_2O) is not particularly strong at low temperatures, the effects of hydrogen on cement barrier sealing characteristics and solubility are thought to be minor [76], but, at higher temperature and depth, hydrogen can severely affect the cement sheath. The above-mentioned processes are highly dependent on water saturation, formation rock salinity, temperature, pH value, and oxygen presence and concentration. Additionally, microbiological activity can enhance these processes; however, if siderite, dolomite, or calcite are present, these reactions can be hindered [60].

6.4. Effects on casing string

This section gives an overview on the effects of hydrogen on casing. As previously mentioned, hydrogen will have effect on casing string which is made of steel [101,102]. In a high-temperature geothermal Icelandic well, hydrogen embrittlement was observed in conditions comparable to UGS [103]. As a result, it is worth looking at the interactions between hydrogen and steel casing in UGS wells, as well as the possible difficulties that may arise. In a research study, steel was subjected to hydrogen at the pressure of 12 bar and the results demonstrated that molecular hydrogen had no significant effect on the steel, and no type of embrittlement or loss in ductility was detected. In acidic conditions, dissolved carbon dioxide in conjunction with hydrogen was found to result in embrittlement and ductility loss in steel, which encourages hydrogen sulfide attack. A similar research study, conducted in Iceland, revealed that fluid compositions are composed of small levels of hydrogen sulfide.

Since hydrogen embrittlement in steel is difficult to detect visually, tensile strength tests are implemented to detect it. Fig. 12 shows a stress/strain curve of a steel sample that demonstrates hydrogen embrittlement. The orange curve depicts brittle failure caused by hydrogen embrittlement, whereas the blue curve depicts ductile failure in a steel not exposed to hydrogen. The primary indication of hydrogen embrittlement in metals is brittle failure.

Fig. 13 shows the failure process of the samples tested to failure to identify probable hydrogen damage. Visual inspection of the fractured sample reveals no brittle break, as seen by the plastic deformation in the steel samples. As a result, there is no evidence of hydrogen embrittlement upon ocular inspection.

Figs. 14 and 15 depicts the stress-strain diagrams of two steel samples, N80 and J55, before and after hydrogen exposure under UGS conditions. Furthermore, as previously observed in the research, none of the samples exhibited hydrogen embrittlement. The final yield strength is not lowered in general, but the samples exhibit an increase in elasticity, which indicates strain hardening.

6.5. Material durability

Packers in general consists of elastomer materials made of rubber, and the body consist of steel materials. The rubber parts of a packer, as well as tubing and casing beneath it, are all subjected to the corrosive effects of reservoir fluids. The failure of packer will affect well integrity, leakage, and it leads to environmental consequences [91]. Elastomers will experience a change in tensile strength, and hardness in the presence of gases like hydrogen sulfide, carbon dioxide, and methane [105]. The molecular structure of elastomers is affected by leaching of chemicals [106]. The concentration of hydrogen can have an impact on the durability of rubber seal components [60]. Blisters do not damage the materials themselves, but the edges function as notches, producing an increase in triaxial stress and the buildup of lattice hydrogen, which can lead to fractures. Furthermore, an increase in micro-void development or tensile strains can result in HIC, which causes abrupt collapse with no indications of weakness. Hydrogen concentration, stress level, metal composition, metal tensile strength, grain size, microstructure, kind of impurity in the metal structure and heat treatment history of the tube are the elements that impact the packers. High-strength steels, tensile strength above 900 MPa, are susceptible to hydrogen embrittlement. The protection of a casing against hydrogen-induced embrittlement is the most important consideration for underground storage of natural gas with additional hydrogen. The corrosive resistant alloys must be used for steel body of the packer to prevent it from corrosion. To prevent the failure of elastomer, hydrogenated nitrile butadiene rubber (HNBR) is recommended because of its resistance to corrosive environments, and suitability for high temperature and pressure conditions [91].

An experimental study was done by Boersheim et al. [104] to investigate the integrity issues of UGS containing hydrogen. In this

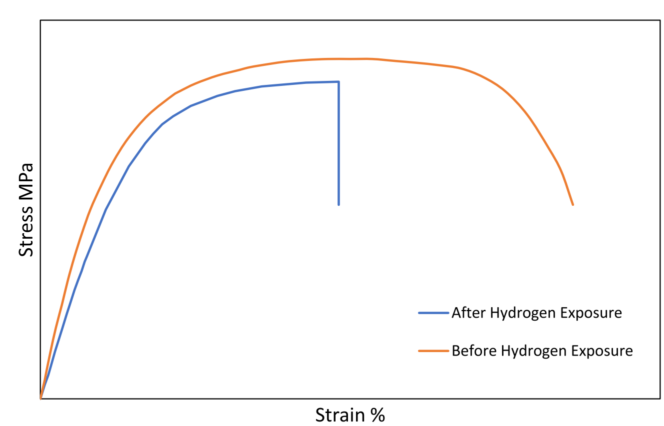

Fig. 12. Deformity of steel when it is exposed to hydrogen [104].

Fig. 13. Ductile failure of N80 steel when exposed to hydrogen.

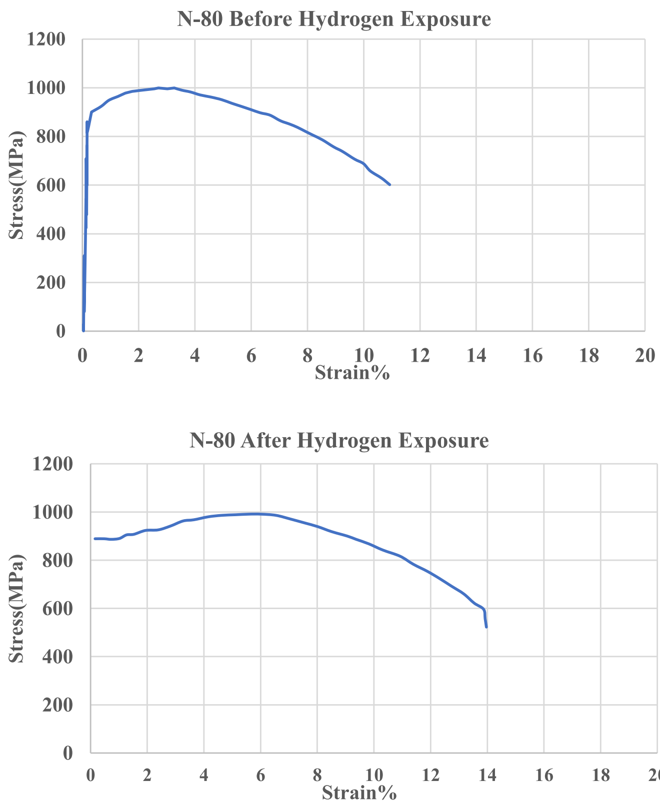

Fig. 14. Effect of hydrogen on tensile strength properties of N-80.

study, a novel approach to study the integrity issues of UGS, autoclaves, which can recreate the conditions in UGS, were used to investigate the integrity issues when hydrogen is injected. The potential changes occurring in the subsurface can also be analysed because autoclaves can simulate the reservoir conditions and have the flexibility of testing parameters as it can recreate any UGS with any gas mixtures. In this experiment, API grade steels and API class G cement were used. The core samples used in this experiment are specially designed and manufactured to recreate the subsurface components in the general UGS facility. The components used in the experiment and their material composition is given in the Table 7. The autoclave used in this experiment is shown in Fig. 16. To prevent explosion, the whole set-up is sealed in a cabin. The samples were saturated in high saline synthetic brine, placed in autoclaves and exposed to hydrogen at 80 °C and 50 Bar for a period of 4 weeks [104].

The following observations were made:

- Reductions in permeability and porosity of the composite cores
- The cements samples were affected minimally by the working gas
- The pH of the synthetic brine increased from 7 to 10
- The steel samples show no signs for hydrogen embrittlement both visually and empirically.

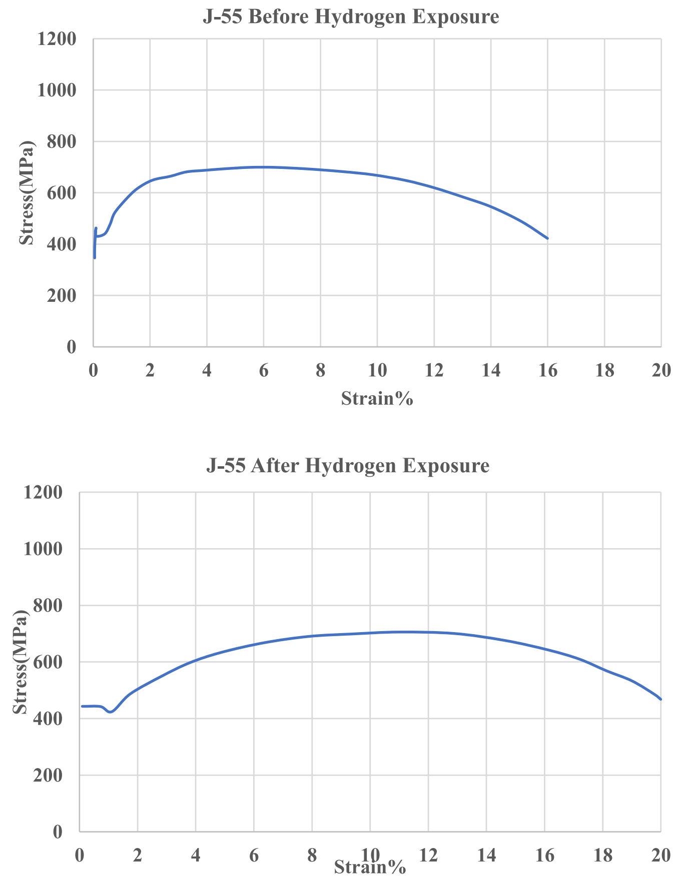

Fig. 15. Effect of hydrogen on tensile strength properties of J-55.

Table 7
Components and their material composition [104].

Component	Material
Casing	N80, P110, K55, and J55
Cement	Cement Type G (clean)
Reservoir Rock	Sandstones sourced from UGS facilities in Germany
Brine	High salinity Brine (TDS 352.23), Low salinity Brine (TDS 18.09), Distilled water
Working Gas	Hydrogen, Nitrogen

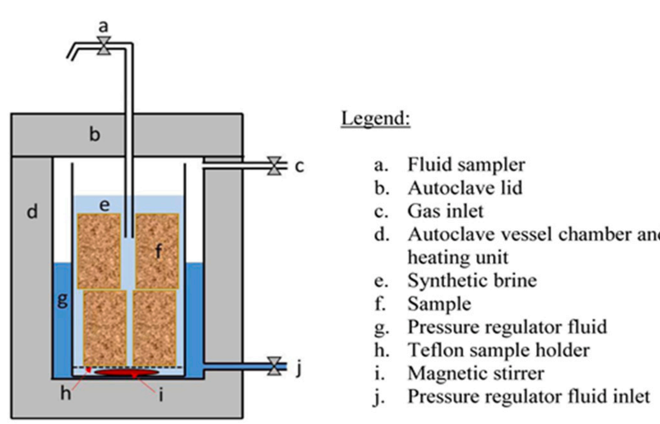

Fig. 16. Autoclave set-up [104].

Therefore, the autoclave experiment under applied conditions shows that the addition of hydrogen in UGS results in small integrity failures.

6.6. Geochemical reactions in reservoir

Injecting hydrogen into a reservoir changes the chemical equilibrium between the formation pore water, dissolved gases, and the rock matrix, resulting in several geochemical reactions, including:

i) High hydrogen loss,
ii) Production of other gases that contaminate stored hydrogen,
iii) Reduction or increase in injectivity due to mineral dissolution/ precipitation, and
iv) Changes in mechanical properties.

Mineral dissolution causes hydrogen sulfide (H_2S) production lowering the quality of stored hydrogen [7,8]. More experimental and simulation studies are required to understand the geochemical reactions in the subsurface. A modelling case study was done by Hassannayebi et al. [107] to study the geochemical reactions in the subsurface.

In this study, Molasse Basin in Upper Austria a small, depleted gas field was selected as a storage site and a target for the integrated pilot project "Underground Sun Storage." This is the first-time storage of hydrogen produced from a surplus of renewable resources that has been tested in a depleted gas reservoir. A methodology for assessing gas-brine-mineral interactions was established in this study. The recommended methodology starts with an equilibrium batch modelling examination of the geochemical system [108].

In this study, the geochemist's workbench was employed as a geochemical modelling tool. Throughout the study, the internal LLNL (Lawrence Livermore National Laboratory) thermodynamic database was employed. The thermodynamic characteristics of the missing minerals were looked up and entered in the LLNL database as part of the modelling method. For minerals for which thermodynamic characteristics could not be obtained, proxy minerals were used.

As a part of the UHS feasibility evaluation, a multistep technique for evaluating the behavior of geochemical systems in the presence of hydrogen is presented. The process is depicted in Fig. 17. Because there was no field data available to compare simulation findings against at the time of the study, numerous scenarios based on various assumptions were simulated. Using this model, the short and long-term impacts of hydrogen on the reservoir can be evaluated.

By enforcing an equilibrium assumption for hydrogen-brine-mineral interactions, the first stage involves identifying all potential reactions within different phases. The kinetic rate parameters for primary and secondary minerals were incorporated in the primary kinetic model in the second stage. The findings reveal that the kinetic speeds of H_2 geochemical interactions with minerals are usually slower than previously thought. Finally, hydrogen dissolution kinetics were included in the model. The dissolution kinetic rate was calculated using a normal storage cycle of 12 months and an operating hydrogen partial pressure of 7.5 bars. The risk of hydrogen loss and reservoir integrity disturbance associated with geochemical interactions with hydrogen cannot be ruled out given the whole spectrum of uncertainty, which is largely owing to a lack of accurate kinetic data. The risk of hydrogen loss increases when redox pair reactions are considered to be in equilibrium [109]. The reaction rates used in this study are largely taken from laboratory studies and published data. It should also be emphasized that the current simulation findings are site-specific; hence, extending the results and conclusions to other storage sites should be done with caution [107].

6.7. Geological integrity of the reservoir and caprock

Caprock in the subsurface can be affected by various parameters including but not limited to: capillary pressure, contact angle, wettability, and minerals precipitation and/or dissolution. Since caprock is one of the most important aspect of UHS, the parameters that affects its integrity must be studied [91]. Capillary pressure is influenced by the contact angle and interfacial tension. If the pressure exceeds the threshold pressure, the caprock drains all the water present in it and the rock becomes permeable which leads to leakage and loss of gas [110]. The loss of gas in UGS is mainly because of the high diffusivity of hydrogen. In UGS sites, there are high chances for the stored gas to get dissolved in the formation water of the caprock which leads to the diffusion of gas into the formation rock. Solubility controls the amount of gas that can be dissolved into water. Hydrogen has lower solubility than methane, but it has higher diffusivity than methane [60]. Due to the high concentration gradient of hydrogen, its diffusion into caprock is higher than methane. The loss of hydrogen due to diffusion is high in the initial stages, however later it reduces because of the gradual decrease in the concentration gradient of hydrogen that restricts further diffusion. The overall loss of stored hydrogen because of diffusion into caprock is 1.5–2.2% [111]. Wettability is an another important factor which affects the sealing ability of a caprock [112,113]. According to Carden &

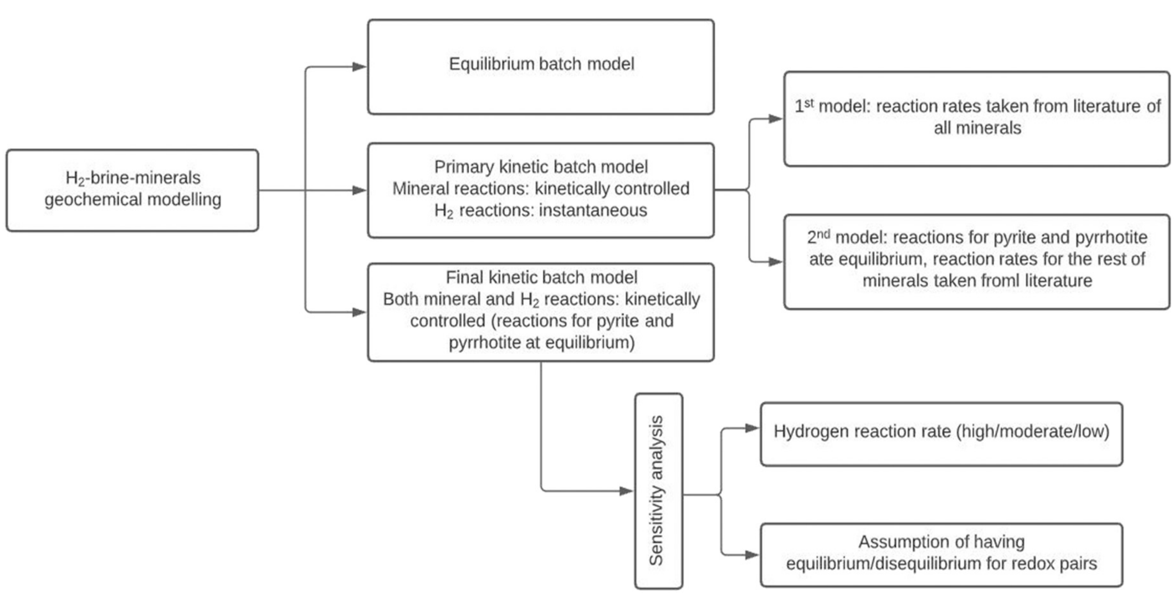

Fig. 17. The geochemical modelling workflow [107].

Paterson [110], the total quantity of stored hydrogen that can be lost because of diffusion and transport into the aquifer during their whole storage period is 2%.The contact angle, interfacial tension, and wettability of caprock must be studied for efficient hydrogen injection and withdrawal. In the literature some studies were conducted to analyze the contact angle, interfacial tension, and wettability [32,35,42,114]. The information on these parameters is limited, more experimental studies are needed.

6.8. Mechanisms and monitoring in UHS

Studying the mechanisms and monitoring them throughout the lifetime of hydrogen storage cycles is required for optimum efficiency. Depending on the energy demand required by the market and the type of field used for storage, a UHS project generally follows a cyclic operation with an alternate period of injection and withdrawal. The flow behavior of fluids must be studied from each withdrawal cycle because of changes in the concentration and state of fluids for each cycle [52]. Gas fragmentation occurs at low injection rates, whereas gas rising and lateral spreading occur at high injection rates [37]. Fingering in subsurface may contaminate the hydrogen which requires additional effort and equipment to mitigate the contamination. As a result, the flow rate must be optimized by ensuring that bottom-hole pressure, capillary entry pressure, and fracture pressure are all met to prevent fingering. To prevent lateral gas spreading, a technique known as selective technology is utilized, in which two distinct injection and withdrawal wells are used [115]. A correctly constructed withdrawal well may enhance withdrawal efficiency while also preventing problems like water coning (especially in aquifers) [52]. The microbial effects must also be considered along with geological, physical, and economical aspects for planning an underground gas storage site [71,79,107,116,117]. The mixing of cushion gases is one of the reasons for hydrogen loss in subsurface storage [118]. Yet, there is no comprehensive monitoring technique available for underground hydrogen injection and storage. Other geological storage techniques like CO_2 sequestration and natural gas storage have influenced the monitoring systems employed for UHS [52]. The monitoring techniques such as neutron log, cement bond log, sonic detection, spinner survey, camera inspection and radioactive tracer survey can also be considered for UHS monitoring [55]. Other techniques such as chemical and electrochemical tools, gravity methods, electrical resistivity tomography (ERT), muon tomography, and seismic full waveform inversion (FWI) can also be considered for effective monitoring of microbial, geochemical, and geo-hydraulic effects in subsurface storage [62,70,119,120].

7. Economic aspects of underground hydrogen storage

Regardless of hydrogen purity, the main advantage of subsurface hydrogen storage is its lowest storage cost as compared to other modes of storage [57]. The economic feasibility of underground hydrogen storage is determined by the expenses of capture, transportation, storage, extraction, and monitoring of hydrogen [52]. The geological features of the storage location, as well as the need to cover any potential leakage spots, affect the cost of storage. Compressor and gas holder cost, transformer and primary breaker installation cost, cost of building, cooling, heating, ventilation, lighting, alarm system and gas monitoring, cost of interconnected pipe network to the compressor, and new wells and wellhead equipment are the major capital costs associated with UHS. The largest cost contributor among the capital costs stated above is gas compressor machinery [57,121]. In terms of operating costs, aquifers are more costly than caverns and depleted hydrocarbon reservoirs; however, cavern systems are more expensive than depleted reservoirs in terms of storage costs [52]. Additional expenses for salt caverns exist; the most important ones are capital expenditures connected to cavern building and the cost of the cushion gas used to maintain the minimum storage pressure. The size of the caverns, the geological features of the reservoir, and the length of the brine-disposal pipeline are the factors that influence cavern costs. The cost of gas compressors, whose number and power are determined by the depth and volume of a cavern, is included in surface installation capital costs [121]. Gas dehydration, measurements, and service buildings are the less expensive installations. The proportions of individual capital expenditures may be calculated using recognized technical and economic data as follows: 60% - gas compression machinery, 6% - borehole, 5% - cushion gas and 29%- cavern construction. The outcomes of the research done for each of the various nations revealed that the expenses of investing in and running electrolyzers are the most important components of the costs, aside from future power bills [48]. The findings also revealed that electrolysis costs dominate the combined costs of production and underground hydrogen storage, >80% of the cost of investment and maintenance, accounting for the majority of electric power expenses [12]. Although most of economic studies found in the literature focused on hydrogen production from renewable sources and using them for commercial purposes [122–124], few studies focused on underground storage of produced hydrogen and their economic feasibility. The study by Michalski et al. [54] focused on hydrogen generation by electrolysis and storage in salt caverns. The results show that hydrogen generation and storage in salt caverns will have a positive impact on the power system in terms of reduced curtailments of wind power plants and lower residual peak demands. The two economic case studies by Lord et al. [53], and Le Duigou et al. [125] are presented in the following sections.

7.1. Economic analysis of UHS – a case study

H2GSM (Hydrogen Geologic Storage Model) is the model utilized in this investigation. This model is an analytical framework that depicts the components of a large-scale hydrogen storage facility that is 'gate-to-gate' (i.e., solely the storage infrastructure). This study was conducted to compare the costs of four different types of underground storage facilities: salt caverns, depleted oil and gas reservoirs, aquifers, and hard rock caverns. The parameters in this study were taken from the literature and other well-known case studies. The geologic storage module and the economic systems module are the two overarching basic components of the H2GSM model. These modules are then used to generate city-specific demands that rely exclusively on salt cavern storage.

The methodology comprised of modelling reservoir characteristics, infrastructural facilities, cost components, geologic site cost and comparison, geologic formation site preparation costs, compressor costs, cushion gas costs, well and surface pipeline costs, total capital cost, and levelized cost of hydrogen, which is the cost of hydrogen production per unit of hydrogen. Fig. 18 illustrates the cost comparison for each of the four storage alternatives. Depleted oil/gas reservoirs and aquifers, with costs ranging from $1.23 to $1.29 per kilogram of hydrogen stored, are the most cost-effective storage option. While salt caverns and hard rock have the greatest levelized storage costs, ranging from $1.61 to $2.77 per kilogram. Because of hydrogen's migration potential, economic

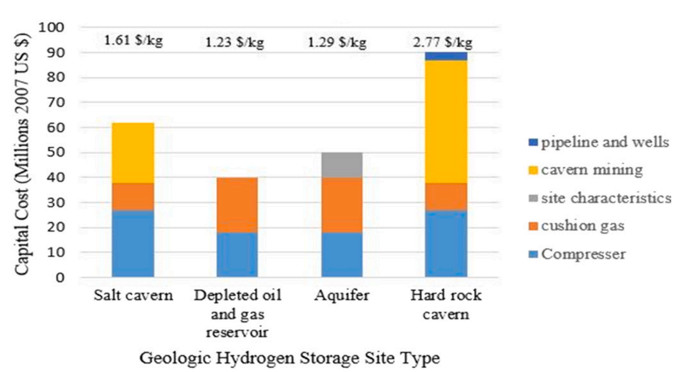

Fig. 18. Cost analysis for all four storages [53].

concerns in depleted oil and gas reservoirs and aquifers exist. Because of their poor permeability, walled hard rock caverns and salt caverns can be utilized to minimize H_2 migration. It is worth noting that the complete supply chain of hydrogen production (extraction), delivery to the storage location, and other expenditures associated with sitting studies, as well as possible risk reduction through measurement, monitoring, and verification (MMV) were not considered in the price evaluation [53].

The following study investigated four large cities in the U.S (Houston, Detroit, Pittsburgh, and Los Angeles) to determine the overall size of geologic hydrogen storage to meet the cities demand. These cities were chosen based on their geographical variations and closeness to salt deposits. The hydrogen storage demands of each city were determined by considering different relative amounts of future demand and the summer surge in transportation. Salt cavern is the only storage option considered for this method because of their low permeability. The Sandra National Laboratories' H2GSM model was updated to include city demand scenarios for hydrogen storage in salt caverns. The city-specific geologic storage solutions in the H2GSM framework were designed based on the population, the summer increase in hydrogen consumption, and various market penetration levels, all while staying within their area geological limits. The predicted demand, which determines the number of required caverns, is determined by the population of each city [53].

Fig. 19 shows the capital expenditures, levelized costs ($/kg), and the number of caverns necessary at 10%, 25%, and 100% market penetration rates for Houston, Detroit, Pittsburgh, and Los Angeles. Due to availability of thinner salt formations in Detroit and Pittsburgh, the costs of storing hydrogen and levelized costs ($/kg) are higher, whereas Houston and Los Angeles have access to thicker and more suitable salt formations so the cost of storing hydrogen and levelized costs ($/kg) are lower. Table 8 shows the caverns developed for each city scenario. Out of all the cities, Houston which is located near the Gulf Coast salt dome has the lowest cost and requires fewer number of salt caverns to meet the demand [53].

7.2. Relevance and costs of large-scale underground hydrogen storage in France

The goal of this study is to examine the technological and economic viability of large-scale subsurface hydrogen storage in France [125]. The methodology consists of two distinct but compatible modules: a static model aimed to simulate the operation of a storage facility in a simplistic manner using aggregate yearly values and a dynamic model aimed to simulate the storage facility hourly operation over a one-year period. The calculations were carried out using HOMER (Hybrid Optimization Model for Electric Renewables) software developed by the National Renewable Energy Laboratory (NREL) in the United States and is currently offered by HOMER energy. HOMER is a model frequently used to estimate, among other things, the optimal integration of vector resources like wind and sun. The initial stage of the project involved compiling a comprehensive inventory and the assessment of the country salt areas. The geology of salt formation, green energy potential in the surrounding environment, and anticipated demand for hydrogen in the region were all used to evaluate each region. The economic modelling was centered on assumptions that were either specific to France (e.g., hydrogen demand, energy rates, etc.) or that could be applied to all case studies (e.g., technology costs, cavern costs, etc.) [125].

Table 8
Number of salt formation caverns required to meet city demand scenarios [53].

City	Number of caverns (Corresponding percentage market penetration level)		
	10%	25%	100%
Houston	1	1	4
Detroit	3	7	26
Pittsburgh	2	5	20
Los Angeles	1	3	10

In France, the techno-economic feasibility and commercial case for large-scale hydrogen subterranean storage were examined. First and foremost, geological hydrogen storage in subterranean salt caverns were demonstrated to be theoretically possible in France, perhaps in six different areas. Second, in terms of both quantity and economic value, the transportation sector is clearly the major trigger. If the mobility industry takes off, the projected quantity may well surpass future demand from other industries. A dynamic study for the year 2050 was performed on an hourly basis, with electrolysers supplied by either wind energy or a mix of wind and grid power [126]. Because hydrogen may be produced and delivered on demand to the mobility sector by a sufficient combination of grid electricity and wind power, low shares of variable renewables do not require hydrogen storage. Based on these patterns, it is reasonable to expect that storage requirements will emerge around 30% renewable penetration rate (The rate is a measure of how much of energy produced being used by consumers compared to the total estimated production of energy) and become substantial between 50% and 75% renewable penetration rate. Bottleneck would necessitate storage for practical reasons. With high renewables share, it becomes more cost effective to invest in storage rather to create more renewable power to serve demand. Underground hydrogen storage in salt caverns is a potentially possible option for large-scale energy storage in general, but it requires appropriate geology, which is not present in all nations and everywhere. While a cavern requires a considerable upfront investment, it only contributes a small fraction of overall actual hydrogen costs: a few percent, and this ratio is unaffected by energy pricing [125].

To conclude, demand for underground hydrogen storage will be driven by three key stakeholders: the power industry, automobile transportation, and hydrogen consumers [54,125,127]. Hydrogen will soon become increasingly essential in addressing the technical and environmental demands of the power and transportation sector in the future. The major stakeholder, the decarbonization of the power industry, needs a further expansion of hydrogen energetics in conjunction

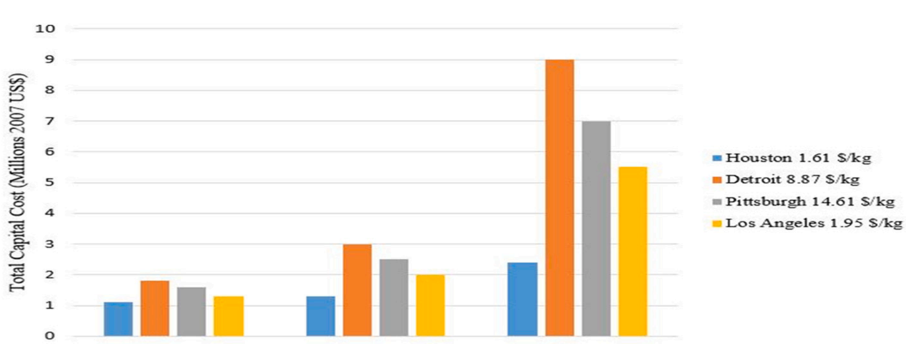

Fig. 19. Total capital costs for four cities [53].

with the use of surplus energy from intermittent renewable sources such as wind, solar, and hydrogen. This will make the initiative financially feasible, with nations having a significant share of wind and solar energy in their energy mix being the initial beneficiaries [48].

8. Road map for the implementation of underground hydrogen storage

The selection of sites for underground hydrogen storage will require adhering to a timetable, a road map in which governmental and non-governmental groups, universities, research institutions, and industry will all play specific roles. These operations typically take place simultaneously and in a coordinated time period [12]. During the early phase of the enterprise, government authorities should consider an inventory of available renewable sources, hydrogen production sites, and the initial selection of hydrogen storage places. Geological, economic, political, environmental, social, and other variables must be considered in a preliminary assessment of the value of geological formations for underground hydrogen storage. The other criteria are flexible, and they might alter in a short amount of time [128]. Geological studies, assessing the feasibility of potential underground hydrogen storage sites, developing calculation and prediction methods for the future capability of underground hydrogen storage, and methodologies for predicting how confined hydrogen would behave in an underground storage facility are all tasks that research organizations excel at [129]. Industry can lead the way in selecting acceptable locations and providing detailed geological information based on geophysical surveys, boreholes, and modelling [110]. In addition, the industry will oversee designing and constructing installations and storage facilities. Government authorities should monitor the subterranean hydrogen storage operation, while research institutes should participate in these operations and regularly update capability predictions and potential perils. Starting with the earliest phases of construction on the subterranean hydrogen storage, the local people should be engaged on a regular basis, keeping in mind that these operations do not hinder their daily schedule. Government departments should ensure judicial remedies for various situations that could possibly arise [48].

9. Conclusions and recommendations

Hydrogen among other renewable energy sources is a top candidate for the most cost-effective, clean, and sustainable energy carrier for future energy and global economic stability. The storage of hydrogen is a barrier to its integration into the global economy [10]. For large-scale H_2 storage, UHS is considered as the best option due to its safety, economic, and technical factors [41,115]. The experience and knowledge of UHS are very limited which makes it difficult for the implementation in large-scale. This paper provides an overview over the different geological structures used for UHS. It also provides a brief understanding of advantages, disadvantages, challenges involved in different storage types, and economic aspects of UHS discussed with some case studies which will help with research development in the future. UHS requires more experimental and simulation studies to achieve effective large-scale implementation. Some of the future work recommendations are:

- More studies are required on modelling of viscous fingering by using different model geometries and parameters [130].
- The solubility and diffusivity of hydrogen were published mainly for bulk liquid phases, ignoring the fact that gas dissolution and diffusion can occur in the reservoir pore network's micro and nanopores [131–133]. Future research should focus on the quantity of solubility and diffusivity of hydrogen in such pore networks for different porous media.
- Wettability of rock/brine/H_2 was examined for sandstones, clay minerals, and basalt [33,35,113,134]. However, further research on chemical active surfaces such as carbonate and their proxy minerals are needed to adequately understand the behavior of H_2-wettabilty on a large scale.
- In general, the quantity of gas flow in porous media varies with the size and connectivity of the pores, and although Darcy's rule holds true for macropores, it is strongly recommended that hydrogen transport in micro and mesopores be investigated further using different pore geometries [9].
- Multiple injection/withdrawal cycles may cause in-situ fluctuations of temperature and pressure in the reservoir, altering reservoir permeability and sealing capacity [3]. The effect of physical and thermal stresses on the reservoir rock requires further investigation.
- The biogeochemical reactions in subsurface such as reaction of hydrogen with iron/sulphur will lead to loss of hydrogen [135,136]. Therefore, the relationship between microbiological activity and geochemical parameters must be studied to reduce the loss of hydrogen.
- The microbial ecology in the subsurface changes during the injection and withdrawal periods because of the change in concentration of hydrogen. This change in concentration and microbial ecology affects the long-term storage [79,137]. The effects of hydrogen concentration need to be studied for better understanding of the ecology of the subsurface.

However, studies on UHS have been performed in countries such as Europe, US, and Canada more studies on UHS must be performed worldwide to reduce the usage of fossil fuel and the emission of greenhouse gases. More economic studies focused on UHS needs to be performed. Attention must be given to site selection studies because the selection of optimum site helps in reducing the transportation cost of hydrogen, cost of surface equipment, and the overall cost of the project. Studies on carbon capture and utilization, and underground storage of natural gas can be replicated for UHS to study the subsurface in the presence of hydrogen. As a concluding remark, if all the concerns are carefully addressed and the projects are carried out meticulously, UHS has a tremendous potential to meet the growing energy demands of the world.

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.

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