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Invited review

Review of underground hydrogen storage: Concepts and challenges

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Abstract:
The energy transition is the pathway to transform the global economy away from its current dependence on fossil fuels towards net zero carbon emissions. This requires the rapid and large-scale deployment of renewable energy. However, most renewables, such as wind and solar, are intermittent and hence generation and demand do not necessarily match. One way to overcome this problem is to use excess renewable power to generate hydrogen by electrolysis, which is used as an energy store, and then consumed in fuel cells, or burnt in generators and boilers on demand, much as is presently done with natural gas, but with zero emissions. Using hydrogen in this way necessitates large-scale storage: the most practical manner to do this is deep underground in salt caverns, or porous rock, as currently implemented for natural gas and carbon dioxide. This paper reviews the concepts, and challenges of underground hydrogen storage. As well as summarizing the state-of-the-art, with reference to current and proposed storage projects, suggestions are made for future work and gaps in our current understanding are highlighted. The role of hydrogen in the energy transition and storage methods are described in detail. Hydrogen flow and its fate in the subsurface are reviewed, emphasizing the unique challenges compared to other types of gas storage. In addition, site selection criteria are considered in the light of current field experience.

1. Introduction

Fossil fuels currently provide more than 80% of the world's energy (Iordache et al., 2014). A growing world population and economic development will inevitably lead to increases in energy consumption while reserves of fossil fuels are depleted. This would be a major challenge without the existentialist threat of dangerous climate change: the world needs to provide more energy while also, very rapidly, moving away from fossil fuels (Rusman and Dahari, 2016; Elsaid et al., 2020). The principal zero-carbon energy sources are nuclear (Zhan et al., 2021) solar thermal (Rezk et al., 2019), solar photovoltaics (Kamel et al., 2021), geothermal (Olabi et al., 2020), wind (Mahmoud et al., 2020), hydro (Soudan, 2019), and biomass energy (Inayat et al., 2019; Hussain et al., 2021), as well as energy generation using fossil fuels, but with carbon capture and storage (Boot-Handford et al., 2014). The supply of renewable energy, notably solar and wind, are variable, largely uncontrollable, and hard to predict with variations over time-scales from minutes to years (Lehtola and Zahedi, 2019). Therefore, energy must be stored to equalize generation and consumption in both the short and long term. Small decentralized sites can provide short-term energy shortage. However, mid to long-term electricity storage is still a challenge. Large-scale energy storage allows renewables to displace fossil-fuel generation without the costs of huge excess capacity to ensure supply during still, cloudy periods. Energy storage can balance supply and demand, increases energy security, and provides better management of the grid, allowing a more rapid and

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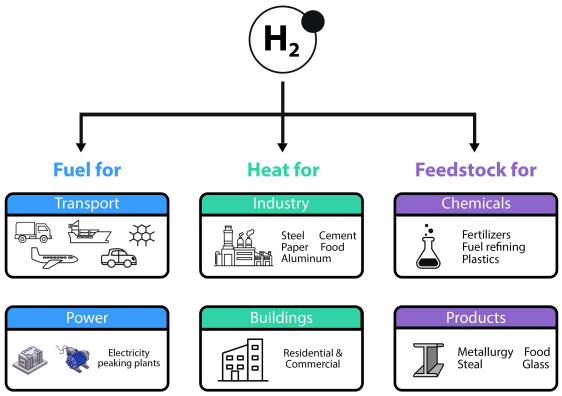

Fig. 1. The main uses of hydrogen as a clean fuel (Kobina and Gil, 2022).

effective transition to a low carbon economy (Al Shaqsi et al., 2020; Rahman et al., 2020).

Hydrogen is a colorless, odorless, tasteless gas and the most abundant element in the universe. Excess electrical power can be used to generate hydrogen by electrolysis; this hydrogen can then be used as an energy store making it a key component of the transition to a net zero greenhouse gas emissions energy system. Total global hydrogen use is currently already substantial, around 115 Mt (115×10^9 kg; Mt is a Megatonne) per year (Abe et al., 2019), but will have to increase by over one order of magnitude to meet the challenges associated with long-term global-scale energy storage. The main potential uses of hydrogen are shown in Fig. 1. At present, however, hydrogen is confined to rather specialized applications, mainly the removal of sulfur from petroleum products in refining, fertilizer production, treating metals, and as a rocket fuel (Abe et al., 2019).

Hydrogen can be produced through various processes, often categorized into a color group: green, blue, gray, white, pink, yellow, turquoise, brown and black. The color codes are used to differentiate how the hydrogen is produced and the emissions associated with production. The three main types are gray, blue and green hydrogen, illustrated in Fig. 2.

Gray hydrogen is produced from fossil fuels, with steam methane reforming the most common production process. Blue hydrogen follows the same process but with the addition of CO_2 capture and storage, preventing CO_2 from reaching the atmosphere. Lastly, green hydrogen is the production of hydrogen by zero-emissions processes such as gasifying biomass, or through the electrolysis of water, where the electricity is generated from renewable sources (Giovannini, 2020). However, at present, the vast majority of hydrogen is produced from natural gas, gray hydrogen, with resultant carbon dioxide emissions; in the future hydrogen has to be generated either from renewable energy (green hydrogen), or with carbon capture and storage (blue hydrogen).

There are six ways in which hydrogen can be stored: (1) compressed gas (surface tanks, depleted reservoirs, aquifers, salt caverns); (2) liquid hydrogen (also known as slush hydrogen, which requires cryogenic storage); (3) adsorbed hydrogen on materials with a large specific surface area; (4) absorbed

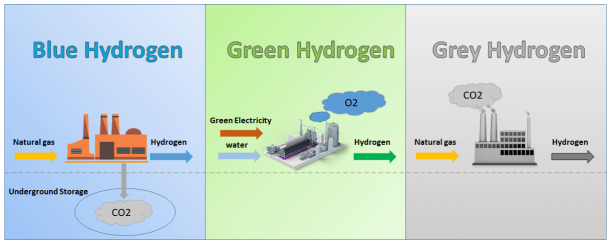

Fig. 2. The three main types of hydrogen production.

on interstitial sites in a host metal; (5) chemically bonded in covalent and ionic compounds; or (6) through oxidation of reactive metals, e.g., Li, Na, Mg, Al, Zn with water.

The only viable method for large-scale, long-term storage for national-scale electricity and hydrogen grids involves storing compressed gaseous hydrogen in large, underground geological structures. These structures are comparatively cost effective and have the capability to store the massive volumes required (Niaz et al., 2015). The same considerations pertain to large-scale carbon dioxide storage; however, as we highlight in this review there are unique challenges with hydrogen: we need to both inject and withdraw the hydrogen, not just dispose of it, while mixing with other gases, microbial activity, and hysteresis in flow properties are all potentially significant as discussed in detail later.

The four major underground hydrogen storage types are depleted hydrocarbon reservoirs, aquifers, salt caverns, and (with a small share) hard rock caverns: these will be reviewed in detail. Each of these storage sites is, to a certain extent, a unique geological structure that has been designed to operate within its functional parameters (Matos et al., 2019).

Underground hydrogen storage (UHS) can provide storage in the 100 GWh range (up to 1 EJ = 10^{18} J) (Tarkowski, 2019). To place this in context, world energy consumption in 2021 was approximately 600 EJ (Enerdata, 2022). This represents the combustion energy of 2×10^{15} moles of hydrogen with a mass of 4 gigatonnes (Gt) (4×10^{12} kg) or a volume of around 6×10^{11} m^3 at a typical underground storage pressure of 10^7 Pa (100 bar) and a temperature of 50 °C; this is 5×10^{13} m^3 at surface conditions. While, obviously a whole year's worth of energy will not be stored at any one time as hydrogen, and no account has been taken of the efficiency of converting hydrogen to electricity and vice versa, it does emphasize the scale of the problem: it is only in large underground structures, at the scale of many km in lateral extent, that sufficient storage volume will be found.

To stimulate the hydrogen economy, the Hydrogen Valley Platform (www.h2v.eu) is a global collaboration to share information on large-scale hydrogen projects: there are 36 projects in 19 countries which consider the whole chain of hydrogen production, use and storage. Approximately 37 billion Euro has been invested in this area. These projects involve production up to a few kt/day and storage at the Mt scale, several orders of magnitude below the gigatonnes required by the calculation in the previous paragraph if hydrogen is to make a significant, global, contribution to the energy system. Table 1 provides data about the name of each project, developer company, level of investment, location, and amount

Table 1. A listing of Hydrogen Valley production and storage projects (www.H_2v.eu).

Name of project	Developer	Investment (Million Euro)	Location	Production (ton/day)
CEOG	Hydrogène de France	121	French Guiana	2
ACESP	Mitsubishi power and magnum development	1000	United states	100
Hy-Fi	ORFO	No information	Chile	650
CBHS	Neoen Australia	370	Australia	25
FNXLHVT	Foshan and Nanhai government	No information	China	No information
RHET	Rugao city government	No information	China	No information
FH_2R	NEDO	No information	Japan	0.5
GHCO	ACME group	2065	Oman	390
GHBD	Verbund AG	No information	Romania	220
HVST	IIT -	55	Italy	1
Hy-balance	Air-liquide	15	Denmark	No information
Normandy hydrogen	Normandy region	No information	France	No information
Hydrogen delta	Smart delta resources	No information	Netherlands	140
H_2 proposition zuid-holland	Port of rotterdam	1000	Netherlands	3180
HVPAR	Port of amsterdam	No information	Netherlands	No information

of hydrogen production.

This study highlights recent research on UHS, modeling of hydrogen flow and reaction, and challenges in controlling and predicting these processes. Finally, site selection algorithms and case studies are discussed. The uniqueness of this paper is the application of concepts in reservoir engineering and geology to assess UHS. This paper also reviews the most recent storage projects.

2. Underground hydrogen storage

Methods for storing hydrogen are chosen based on the stored volume, the duration of storage, the required speed of discharge, and the geographic availability and cost of the various alternatives. Currently, at the small scale, hydrogen is stored as a gas or liquid in tanks for stationary or mobile applications. When handling significant amounts of hydrogen, at the Gt scale, necessary for continuous operations at a national or international scale, pressurized tanks or liquid storage vessels do not suffice: subsurface storage is essential. Table 2 provides a comparison of the different storage types. Natural gas (methane) storage in has been applied for decades. The knowledge gained by this can be easily transferred to hydrogen (Ozarslan, 2012). The materials required in access wells, the well head and transmission infrastructure are the main differences between hydrogen and natural gas storage (Ozarslan, 2012). In the case of hydrogen, embrittlement due to long-term diffusion can cause fracturing, followed by leakage, especially in the steel components, which reduces the strength and stresses that can be safely applied to metal components (Caglayan et al., 2020).

2.1 Salt caverns

Salt caverns have been used to store pure hydrogen in the United Kingdom since the 1970s and in the United States since the 1980s (Cihlar et al., 2021). They also require a cushion gas to maintain pressure (this is another inert gas, such as nitrogen or CO_2, that is initially placed in the cavern). Typically about 30% of the total capacity is comprised of the cushion gas. Salt caverns have sufficient injection and withdrawal rates to perform up to 10 injection and withdrawal cycles per year, but are often lower in capacity than natural gas reservoirs, making them ideal for peaking storage facilities.

The surrounding salt of the cavern has low porosity and permeability, which prevents leakage, while the salt itself is ductile which prevents the formation of fractures as possible escape paths for the hydrogen (Peng et al., 2020; Zhang et al., 2020). Other favorable properties include chemical inertia towards hydrogen (Cihlar et al., 2021), good heat conduction, and the suppression of hydrogen consumption by microbes due to the high salt content and limited water available (Sainz-Garcia et al., 2017; Zivar et al., 2021). These properties guarantee the long-term stability and security of hydrogen storage. To recap: the walls of salt caverns are essentially impermeable to this gas (Liu et al., 2015).

The main phases of salt cavern generation are leaching, debrining, and filling. The leaching process makes the cavern in the first place and starts by pumping water (direct or indirect circulation) into the salt formation through an access well.

Table 2. Comparison of underground storage types (Lord et al., 2014; Cihlar et al., 2021).

Storage type	Depleted gas field	Aquifer	Salt cavern	Lined rock cavern
General suitability for hydrogen	Site-specific	Site-specific	High	High
Typical type of operation	Seasonal	Seasonal	Peaking	Peaking
Typical number of cycles per year	1-2	1-2	10	10
Working/Total gas capacity	50%-60%	20%-50%	70%	70+%
Depth	300-2,700 m	400-2,300 m	300-1,800 m	1,000 m
Operating pressure	1.5-30 MPa	3-30 MPa	3.5-20 MPa	2-20 MPa
Cost of development (relative)	Low	Low	Low	High
Cost of operation (relative)	Low	Low	Medium	Medium
Largest expenses (new development)	Well infrastructure, cushion gas, compression	Exploration and determination of geology, well infrastructure, cushion gas, compression	Formation of the cavern, disposal of brine, cushion gas, compression	Blasting of the cavern, steel lining, cushion gas, compression
Geographic availability	Most countries	Most countries	Limited	Anywhere with igneous or metamorphic rock
Suitability for hydrogen	Hydrogen-methane blending proven; pure hydrogen storage under study	Under study, but experience from depleted fields can be used	Proven	First hydrogen storage in development (2022)
Suitability factors	Operational conditions, fluid and rock composition, bacteriological activity	Operational conditions, fluid and rock composition, bacteriological activity; tightness (for new storage development only)	Salt domes are superior to bedded salt structures	Metamorphic or igneous rock; low steel price
R&D needed	Effects of residual natural gas, bacterial reactions	Bacterial reactions, tightness of rocks	Accuracy of the timing of injection and withdrawal	Compatibility of lining materials with hydrogen

The salt will slowly be dissolved, and the brine produced is extracted and then used or disposed of. Transport and disposal of brine is the most difficult step in salt cavern development. Two-well construction methods have also been proposed (Wan et al., 2019). After this the de-brining process starts. The brine is displaced by injecting the cushion gas into the cavern. The gas is injected through the outer pipe, while the brine is extracted through the inner leaching pipe. Fig. 3 provides a schematic of UHS in salt caverns illustrating two possible configurations for their formation.

The design of a salt cavern depends on the properties of the salt deposit (Lux, 2009). There are a number of factors that must be taken into account to maintain its sustainable and safe operation. The thicknesses of the salt layers in the hanging wall (the salt layer above the cavern) and foot wall (the salt layer below the cavern) should be considered in order to guarantee that a cavern is geomechanically safe (see Fig. 4). Usually, the minimum (critical) thickness of these layers is considered as a function of cavern diameter and overburden pressure for a safe operation (Caglayan et al., 2020).

Due to concerns about geotechnical safety, maximum and minimum gas operation pressures are limited to between 24% and 80% of the overburden pressure (the pressure in the rock at the depth of the cavern); these pressures are used based on experience with natural gas storage (Caglayan et al., 2020).

While salt caverns are a promising storage type for hydrogen due to their decades of success and storage security, their availability is limited geographically. In addition, brine accumulating at the bottom of the cavern increases the water vapor content in the stored hydrogen (Luboń and Tarkowski, 2020), which requires the gas to be dried on production in a surface drying system.

Currently, only a few sites for hydrogen storage in salt caverns exist in the United Kingdom and the United States, see Table 3. Hydrogen storage in elliptically-shaped salt caverns at a depth of 350-450 m and with a total volume of up to 210,000 m^3 has been operation in Teeside in the United Kingdom since the 1970s. The salt caverns at Clemens Dome and Moss Bluff in the United States are built in salt domes at a depth of 800 m (top of the cavern), with volumes of approximately 580,000 m^3. Clemens Dome and Moss Bluff have operated since 1983 and 2007, respectively and demonstrate that underground hydrogen storage is a technically-feasible option (Crotogino et al., 2010).

More recently, different studies have investigated the potential of hydrogen storage in salt caverns from different per-

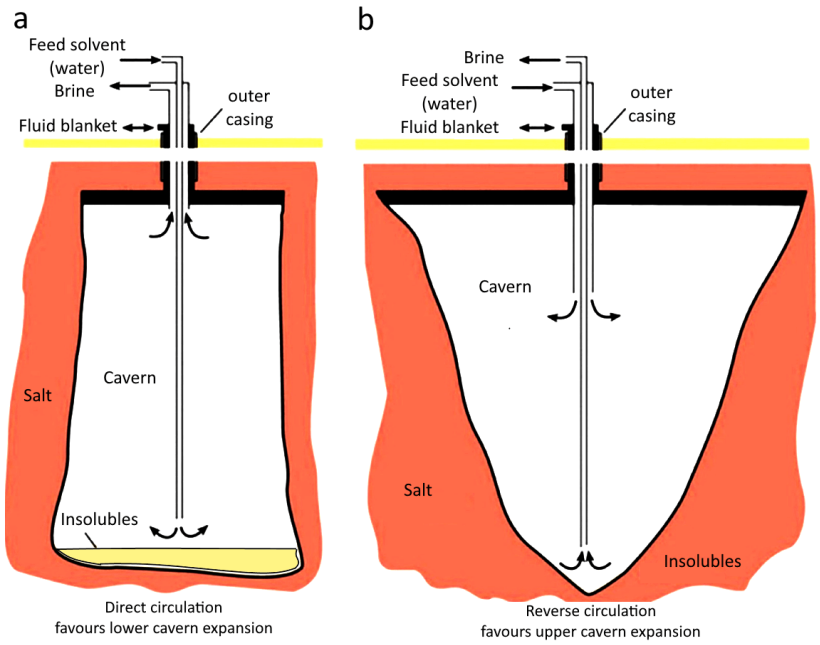

Fig. 3. A schematic of UHS in salt caverns with two configurations for creating and enlarging the cavern (Muhammed et al., 2022).

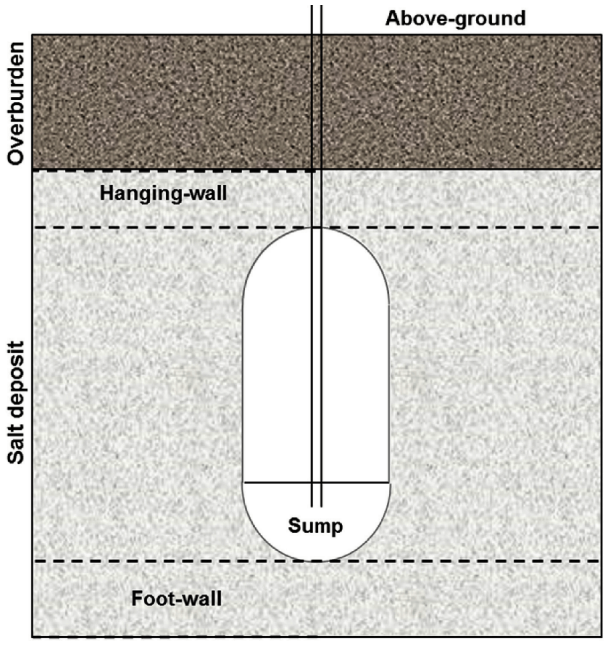

Fig. 4. Simplified schematic of a salt cavern (Caglayan et al., 2020).

spectives, ranging from the analysis of the thermo-mechanical properties of the cavern (Böttcher et al., 2017), to the identification of optimal regional sites (Ozarslan, 2012; Le Duigou et al., 2017; Juez-Larré et al., 2019; Tarkowski, 2019), and to the assessment of the financial and environmental performance of underground storage of hydrogen and natural gas (Peng et al., 2020). However, in all cases the scale of storage, as emphasized previously, with volumes at most 10^8 m^3 at surface conditions (masses of the order of 10 kt) are well below the Gt scale required for global operations: either many thousands of caverns are required, which may be challenging to achieve around the world, or sites with more storage volume, namely subsurface porous formations, are required.

2.2 Depleted gas fields

To date, depleted reservoirs are the most common storage sites for natural gas. Depleted natural gas reservoirs consist of porous, permeable sedimentary rocks located underneath an impermeable cap rock. There may be multiple wells located throughout the reservoir, often remaining from when it was operational for gas production and sometimes drilled strategically to improve the storage operation (Matos et al., 2019). Fig. 5 is a schematic of a depleted hydrocarbon reservoir, also showing the pore structure of the rock, which could be a good candidate for hydrogen storage. It is also possible to consider storage in a depleted oil field, where the same considerations discussed below pertain.

Natural gas reservoirs should be able to operate for hydrogen storage because they have demonstrated their ability to store gas for millions of years (Tarkowski, 2019). An important characteristic of hydrogen that differs from natural gas is its reactivity, both chemically and through bacterial action (Toleukhanov et al., 2015). Blends of natural gas and hydrogen are less likely than pure hydrogen to react with minerals, and can be used with existing infrastructure (Ganzer et al., 2013). The amount of hydrogen that can be added before new higher-grade steel components are required needs to be assessed on a case-by-case basis (Cihlar et al., 2021). The main consideration, mentioned before, is possible hydrogen embrittlement of steel piping and other equipment.

Hydrogen has a higher compressibility and diffusivity, and lower viscosity than natural gas, meaning it may be more difficult to contain and mixes more rapidly with other

Table 3. List of underground hydrogen storage projects.

Country	Project name	Storage fluid	Storage volume (1,000 m^3)	Type of storage	Status	References
Argentina	Hychico-diadema	CO_2 & H_2	49,500	DGR	P	www.hychico.com.ar
Austria	RAG-Sun Storage	NGH	6,000,000	DGR	P	www.rag-austria.at
Czech Republic	RWE-Haje	NGH	100,000	Aquifer	P	www.rwe-gasstorage.cz
	RWE-Lobodice	NGH	100,000	Aquifer	S	www.rwe-gasstorage.cz
Denmark	GHH	H_2	66	Salt cavern	P	www.greenhydrogenhub.dk
	ANGUS+	H_2	-	Geological formations	P	www.angusplus.de
EU countries	HyStorIES	H_2	-	DGR and aquifer	P	Londe (2021)
	HyUnder	H_2	4,000	Salt cavern	P	www.hyunder.eu
France	HyPster	H_2	484	Salt cavern	P	HyPSTER... (2021)
	TEREGA	H_2	3,300	Salt cavern	P	www.terega.fr
	Beynes	NGH	330,000	Aquifer	S	Liebscher et al. (2016)
Germany	HyCAVmobil	H_2	500	Salt cavern	S	Thaysen et al. (2021)
	HYPOS	H_2	-	Salt cavern	P	HYPOS... (2019)
	InSpEE	H_2	-	Salt cavern	P	Zapf et al. (2015)
	HyINTEGER	H_2	-	DGR	P	Boersheim et al. (2019)
	Ketzin	NGH	130,000	Aquifer	S	Liebscher et al. (2016)
	Kiel	NGH	32	Salt cavern	S	Liebscher et al. (2016)
Germany and Austria	H_2STORE	H_2	-	DGR	P	Henkel et al. (2014)
Ireland	Green Hydrogen @ Kinsale	H_2	990	DGR	P	www.energy-storage.news
Netherlands	HyStock	H_2	66	Salt cavern	S	www.hystock.nl
	LSES	H_2	14,000	Salt cavern	P	Groenenberg et al. (2020)
	LSES	H_2	75,000	DGR	P	Groenenberg et al. (2020)
Sweden and Finland	HYBRIT	H_2	120	Lined rock cavern	P	www.hybritdevelopment.se
UK	Aldbrough	NGH	330,000	Salt cavern	P	www.ssethermal.com
	HyStorPor	H_2	-	Geological formations	P	www.gtr.ukri.org
	Teesside	H_2	210	Salt cavern	S	Liebscher et al. (2016)
USA	SHASTA	NGH	-	DGR	P	www.edx.netl.doe.gov
	Spindletop	H_2	906	Salt cavern	S	Liebscher et al. (2016)
	Clemens Dome	H_2	580	Salt cavern	S	Liebscher et al. (2016)
	Moss Bluff	H_2	566	Salt cavern	S	Liebscher et al. (2016)

Note: P denotes prospective, S denotes successful, GHH denotes green hydrogen hub Denmark, DGR denotes depleted gas reservoir, NGH denotes natural gas & H_2.

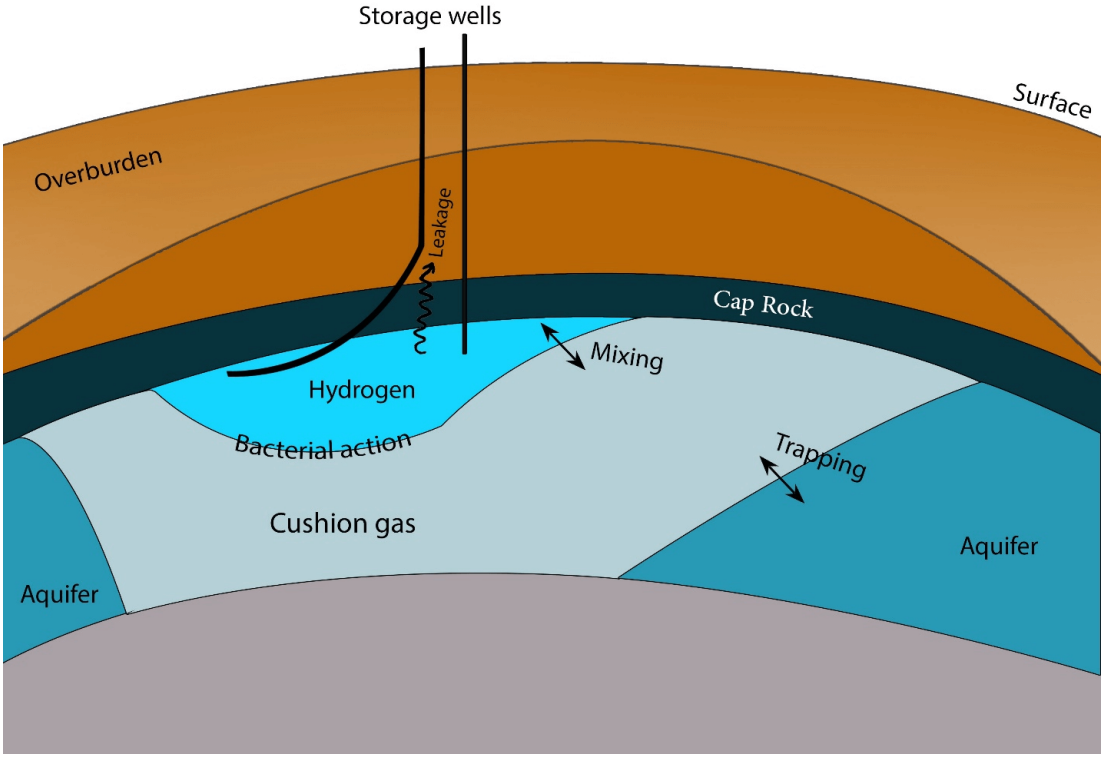

Fig. 5. A depleted hydrocarbon reservoir which could be a good candidate for hydrogen storage. The cushion gas is most likely natural gas originally present in the reservoir. Also indicated are some of the processes, including mixing, bacterial action and leakage, that may occur. These processes are discussed in more detail later in the review. In the diagram the vertical scale is exaggerated at the storage site. The total depth is typically 1-3 km, while the lateral extent of the hydrogen plume is several km. The height of the storage formation itself is typically a few tens of m.

gases in the subsurface. Computer simulations have shown that hydrogen diffusion through the cap rock is, however, negligible, and the most likely method of escape, if any, would be through the wells, as is the case with all types of underground gas storage (Cihlar et al., 2021). However mixing and bacterial action, described later, are significant concerns.

In comparison to aquifer UHS options, depleted gas reservoirs are more advantageous as the remaining gas can be used as a cushion gas. Between 50% to 60% cushion gas is required maintain pressure and prevent trapping of hydrogen by water encroachment from the aquifer; the exact percentage varies depending on the structure and desired injection and withdrawal rates (Lord et al., 2014), but is generally higher than the 30% required in salt caverns.

Operating pressures and depths vary considerably depending on the structure, with pressures in the range of 1.5 to 30 MPa and depths of 300-2,700 m, see Table 2. It typically takes 3-10 years to develop a depleted gas field into storage depending on the characteristics of the field and the extent of the tests required to determine its suitability (Cihlar et al., 2021). Unlike salt caverns, the injection and withdrawal rates of porous rock structures are limited by the permeability of the rock (typically one cycle per year). They are used most commonly for large volume seasonal natural gas storage, though there are examples of them being used for more short-term flexibility (Lord et al., 2014).

The advantages of depleted gas fields for hydrogen storage are that they are larger in volume than salt caverns, do not need to be artificially constructed, and their geology is already well understood from being operated for natural gas recovery. Compared to the development of new salt caverns, they already have a well infrastructure for natural gas, some of which can be potentially retrofitted or repurposed for hydrogen (Lord et al., 2014). Gas fields are also more widespread than salt caverns (Zivar et al., 2021).

Pure hydrogen has not yet been stored in depleted gas fields; however, there is some experience in storing of blend of hydrogen and natural gas as listed in Table 3. The Underground Sun Storage project, a pilot conducted by RAG Austria from 2014 to 2021 in the Molasse Basin, tested storage of a blend of 10% hydrogen and 90% methane in a depleted gas field. This field exhibits unique characteristics as it is homogenous, optimally sealed by shale layers and without connection to an aquifer, which makes it a promising site for a pilot project. The reservoir layer is characterized by a thickness of about 800 m, 22% porosity, 22% irreducible water saturation and a temperature of 40 °C (Pichler, 2019). Several storage operators are planning to examine a broad range of hydrogen and methane admixtures, up to storing pure hydrogen within a series of field tests (Cihlar et al., 2021). The first operational pure hydrogen storage in a depleted gas field is expected in 2030, operated by RAG Austria (Hemme and

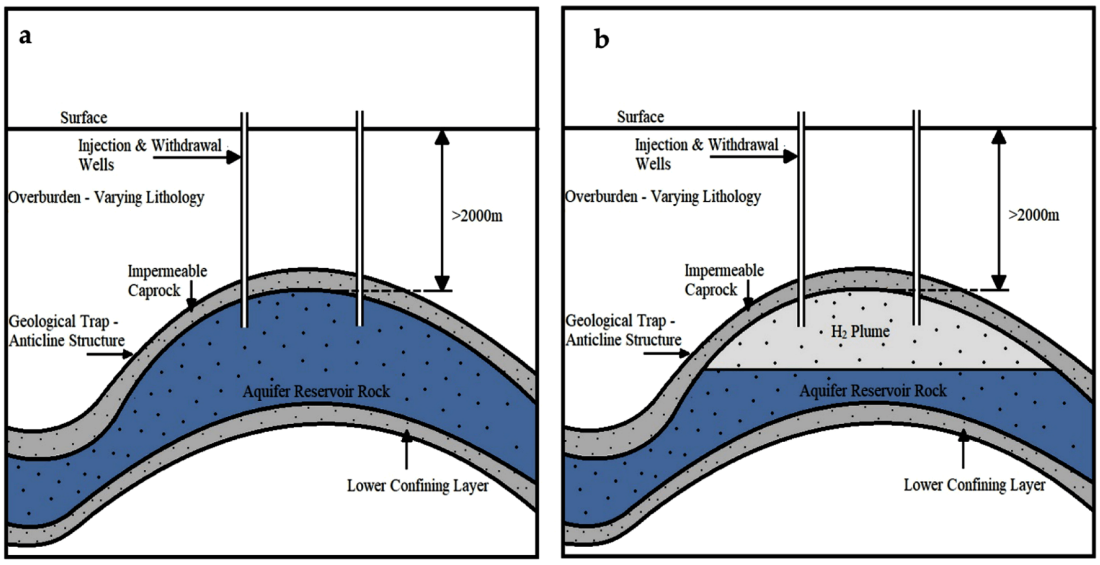

Fig. 6. A schematic of aquifer structure before (a) and after (b) hydrogen storage (Muhammed et al., 2022).

van Berk, 2018).

2.3 Aquifer storage

Aquifers are the most common type of storage site for natural gas after depleted reservoirs. Similar to gas reservoirs, they are porous and permeable; however, they contain water instead of natural gas. Unlike depleted gas fields, which are known to be reliable traps because they were originally filled with gas, geological surveys are required to guarantee the integrity of the caprock. Aquifers typically take a similar length of time to develop as depleted gas fields, plus added time for geological studies when creating new storage (Sainz-Garcia et al., 2017; Zivar et al., 2021).

Aquifers are developed by injecting cushion gas followed by hydrogen through one or more wells strategically placed, displacing water. Depending on the structure of the aquifer and the positioning of the wells, the displaced water can sometimes be used in place of cushion gas, refilling the pores as the gas is depleted and maintaining pressure, but using cushion gas instead is often more desirable as it contributes to pressure maintenance and now the cushion gas, and not the gas stored, is trapped by water (Tarkowski, 2019; Cihlar et al., 2021). Fig. 6 depicts an aquifer structure before and after conducting UHS.

In comparison to depleted reservoirs, normally more cushion gas, up to 80% of the storage volume, is required to prevent gas trapping, although the precise amount required depends on the geological structure, placement of wells, and operational needs. Operational pressures of aquifers range from 3 to 30 MPa, and depths range from 400 to 2,300 m, see Table 2.

In comparison to salt caverns, they are larger in volume (commonly for large volume seasonal storage) and the injection and withdrawal rates of aquifers are limited by the permeability of the rock (typically one cycle per year), as in depleted hydrocarbon fields (Scafidi et al., 2021).

The successful demonstration of hydrogen storage in depleted hydrocarbon fields suggests it is possible in aquifers as well, since in both cases storage is in porous rock. As in depleted gas fields, geochemical and microbial reactions must be studied for hydrogen storage development. Water is a common impurity in gas stored in aquifers, so gas drying infrastructure is an important component of the gas treatment process. Repurposing aquifers used to store natural gas for hydrogen storage is similar to using depleted gas fields due to the similarities in structure and initial conditions (Cihlar et al., 2021; Zivar et al., 2021).

Undeveloped aquifers do not have an existing well infrastructure, so all surface and subsurface components would have to be purchased and installed. The well infrastructure, geological studies, and the cushion gas are additional capital costs that make aquifers more costly to develop than depleted gas fields. However, because the storage space itself does not need to be constructed, they are still often cheaper to develop than salt or lined rock caverns (Lord et al., 2014). Porous aquifers are quite widespread and are available all around the world. However, it is uncertain what portion of the aquifer is suitable for hydrogen storage (Tarkowski, 2019).

Currently, no pure hydrogen storage has been successfully achieved in aquifers. In France, at Beyens, Gaz de France stored gas containing 50% hydrogen in a saline aquifer with a capacity of 3.85×10^8 sm^3 between 1956 and 1972. The gas was produced in a coal and steel coking process in Eastern France. The objective was to regulate fluctuations in gas production/demand. However, intense bacterial activity and consequent transformation of the gas was observed (Panfilov, 2016). At Lobodice, Czech Republic, coal gas containing 50% hydrogen and 25% of methane was stored in an aquifer (Sørensen, 2007; Reitenbach et al., 2015; Wang et al., 2015); see Table 3.

As for depleted gas fields, there are plans to study the storage hydrogen and methane admixtures, including storing pure hydrogen (Liebscher et al., 2016). The Franco-Spanish Lacq Hydrogen Project is designed to use an aquifer for

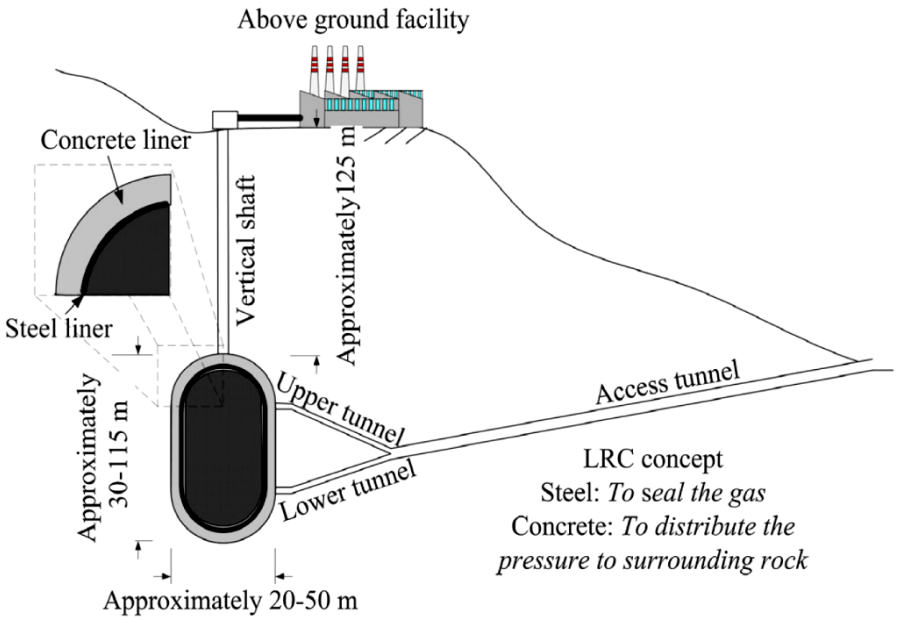

Fig. 7. A schematic of the use of lined rock caverns for hydrogen storage (Lalanne and Byrne, 2019).

hydrogen storage and should be operational by 2026.

2.4 Lined rock caverns

Lined rock caverns are the newest of the four main underground storage technologies, with only one facility in operation for natural gas storage in Sweden. Lined rock caverns (Fig. 7), like salt caverns, are artificial structures created in metamorphic or igneous rock. The caverns are covered with a layer of concrete to create smooth walls, which are then lined with steel or plastic. Because they are carefully crafted and lined, hard rock caverns have no risk of impurities and can be operated at higher pressures than other structures. They can also experience several injection and withdrawal cycles per year, making them well suited for rapid supply of peak demand. They also require relatively little cushion gas.

Hydrogen has not yet been stored in a rock cavern; however, SSAB, LKAB, and Vatten are preparing a site as part of the HYBRIT green steel project. Rock caverns will likely be reserved for peaking facilities in geographies with no other storage options because they are costly to develop. One potential concern with steel-lined caverns is that long-term exposure of steel to hydrogen can cause embrittlement. This implies that a higher grade of steel or another kind of liner, such as plastic, may need to be used (Cihlar et al., 2021). Lined rock caverns have limited storage capacity and so cannot alone supply the Gt scale of storage necessary in a global hydrogen energy economy.

There are many research and industrial projects which have focused on underground hydrogen storage. Table 3 lists the current underground hydrogen storage projects.

3. Rock-fluid properties in hydrogen storage

Hydrogen was first recognized as a distinct element in 1766 by the English scientist Henry Cavendish, when he reacted hydrochloric acid with zinc. He described hydrogen as "inflammable air from metals" and established that it was the same material (by its reactions and its density) regardless of which metal and which acid he used to produce it. The French scientist Antoine Lavoisier later named the element hydrogen (1783). The name comes from the Greek 'hydro' meaning water and 'gene' meaning forming; hydrogen is one of the two elements that comprise water (Hoffmann, 2019).

At low pressures, the behavior of hydrogen can be predicted with the ideal gas law, but at high pressures, more complex equations of state are required. Hydrogen has good thermal conductivity for a gas which increases with pressure and temperature. The solubility of hydrogen and its effect on the pH of brine is low due to its non-polar nature. Hydrogen also has a low viscosity compared to other gases. Fig. 8 shows the density of hydrogen as a function of temperature at different pressures. Underground storage is likely to be performed in a pressure range of 5-30 MPa and a temperature between 30 and 130 °C. For reference, Fig. 9 shows the conditions for different types of storage proposed in the United Kingdom.

For comparative purposes, Table 4 indicates some properties of hydrogen, methane, and carbon dioxide, CO_2, relevant to subsurface storage. The most important feature is that on a mass basis hydrogen has a higher heat of combustion than methane. While many properties of hydrogen are known, unlike methane and CO_2, it has not been so widely studied at subsurface storage conditions, and the interactions between hydrogen and other gases, reservoir brines and host rock are not known.

Many factors such as physical, geo-hydraulic, geochemical, biochemical, and mineralogical processes must be considered for a successful UHS study. In the following the physics of fluid flow relevant for hydrogen storage is described. The emphasis in this section will be on aquifer and depleted gas reservoir storage where we need to consider multiphase flow

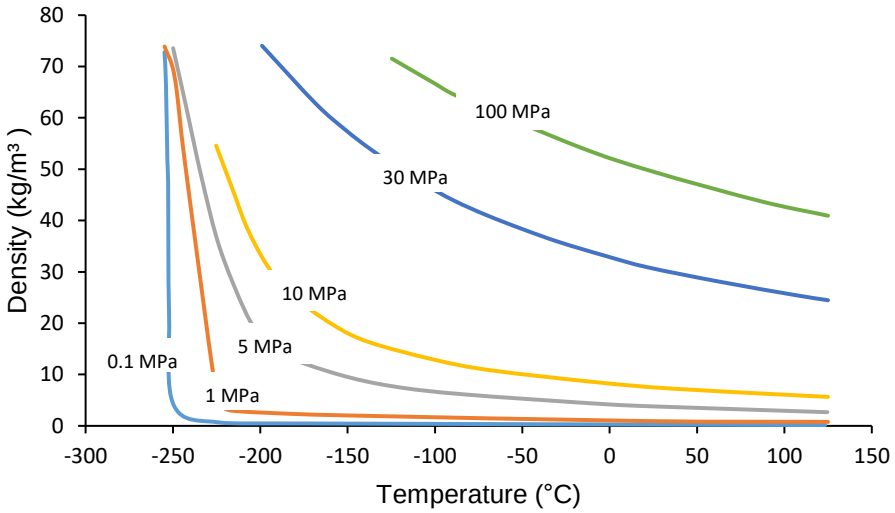

Fig. 8. Hydrogen density at different temperatures and pressures. Data from Aziz (2021).

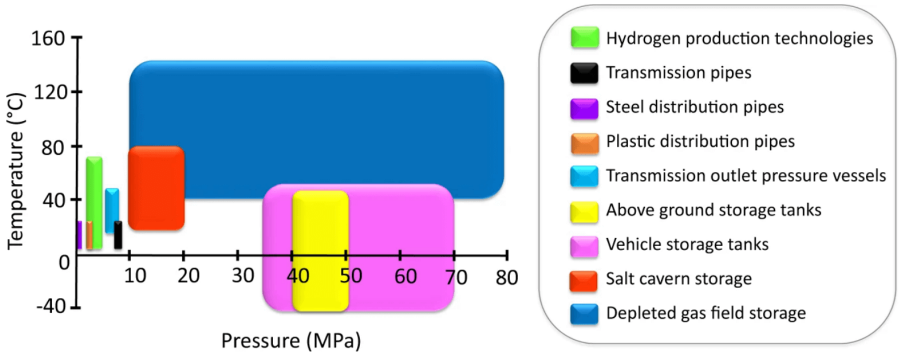

Fig. 9. The average allowed pressures and temperatures for hydrogen for safe and effective operation in the United Kingdom (Hassanpouryouzband et al., 2020).

in porous media and characterize interfacial tension, contact angle, capillary pressure, relative permeability, hysteresis and reaction (Zivar et al., 2021).

3.1 Interfacial tension

Several studies have measured interfacial tension (IFT) for CO_2 and CH_4 in contact with brine at conditions relevant for gas storage projects (Georgiadis et al., 2011; Sarmadivaleh et al., 2015; Kashefi et al., 2016). In contrast, there are fewer measurements for IFT for hydrogen-brine systems.

The first measurement of IFT between hydrogen and brine at 25 °C and up to a pressure of 10 MPa was made using the capillary-rise method (Slowinski et al., 1957; Massoudi and King, 1974). It was found that for pressures up to 10 MPa, the IFT for the H_2-water system is almost constant with less change compared to other denser gases such as CO_2 and CH_4. The pendent-drop method has also been used to measure the IFT for hydrogen-water and hydrogen-water-CO_2 systems at pressures from 0.5 to 45 MPa, and temperatures from 25 to 175 °C (Chow et al., 2018). It was found that the IFT decreases with both pressure and temperature. An empirical correlation for the IFT of the H_2-water system was developed with an average deviation from the measurements of only 0.0016 N/m (Chow et al., 2018).

The IFT can be estimated indirectly using the approach of Yekta et al. (2018). They found the IFT that would scale the measured H_2-water capillary pressure to match the standard mercury invasion capillary pressure. For two conditions (5.5 MPa, 20 °C; 10 MPa, 45 °C), the interfacial tensions were 0.051 and 0.046 N/m, respectively which are lower values than obtained from direct measurement.

Table 5 provides a summary of measured IFT values for H_2-water and CO_2-water at different pressure and temperature conditions. With CO_2, particularly at supercritical conditions (pressures greater than approximately 8 MPa and temperatures exceeding 40 °C), where it forms a dense phase, the interfacial tension is much lower than between H_2 and water. The H_2-water interfacial tension is close to the surface tension of water in contact only with its vapor.

3.2 Contact angle

The first attempt to derive contact angle for hydrogen in contact with brine was made indirectly from capillary pressure, mentioned above (Yekta et al., 2018). The contact angles measured through water in the presence of hydrogen in Vosges (France) sandstone were 22° and 35° for the two conditions

Table 4. Properties of pure hydrogen, CO_2 and methane relevant to subsurface storage.

Property	Unit	Hydrogen	Methane	CO_2
Molecular weight	g/mol	2.016	16.043	44.0095
Density @ 25 °C and 0.1 MPa	kg/m^3	0.082	0.657	1.795
Heat of combustion	MJ/kg	141.8	55.5	-
Heat of combustion	kJ/mol	286	890	-
Viscosity @ 25 °C and 0.1 MPa	Pa·s	0.89×10^{-5}	1.1×10^{-5}	8.36×10^{-6}
Specific heat capacity	kJ/(kg·K)	14.05	2.165	0.658
Ignition point	°C	560	600	-
Dynamic viscosity @ 20 MPa/50 °C	mPa·S	0.00935	0.018437	0.01614
Critical temperature	°C	-239.97	-82.3	30.98
Critical pressure	MPa	1.2.8	4.579	7.208
Solubility in pure water @ 65 °C and 20 MPa	mol^{-1}	0.14	0.1929	0.33
Flammability limits	vol% in air	4-75	5.3-15	2.5-13
Auto ignition temperature	°C	585	540	-
Diffusion coefficient in air @ normal temperature & pressure	m^2/s	0.61×10^{-5}	1.6×10^{-5}	1.39×10^{-5}
Diffusion coefficient in pure water @ 25 °C	m^2/s	5.13×10^{-9}	1.85×10^{-9}	1.91×10^{-9}
Melting point @ atmospheric pressure	K	532.25	90	216.85
Boiling point @ atmospheric pressure	K	20	111	194

References: Diamond and Akinfiev (2003); Bai et al. (2014); Laban (2020); Zivar et al. (2021)

Table 5. Summary of measured interfacial tensions (IFT) for H_2-water and CO_2-water at different temperatures and pressures.

Fluid system	Method	IFT (N/m) *	IFT (N/m) **	References
H_2-water	Capillary-rise	0.071	-	Slowinski et al. (1957)
H_2-water	Capillary-rise	0.071	-	Massoudi and King (1974)
H_2-water	Capillary pressure	0.051	0.046	Yekta et al. (2018)
H_2-water	Pendant-drop	0.072	0.068	Chow et al. (2018)
CO_2-water	Capillary-rise	0.038	0.033	Chun and Wilkinson (1995)

Note: * denotes at 5.5 MPa, 25 °C, ** denotes at 10 MPa, 45 °C.

studied.

Iglauer et al. (2021) performed an experimental study to measure the contact angle for a hydrogen-brine system for pressures between 0.1 and 25 MPa and temperature from 23 to 70 °C. Both pure quartz surfaces and surfaces rendered oil-wet with stearic acid were studied in contact with 10 weight% NaCl brine. An increase in pressure or temperatures increased the contact angles from 0° to a maximum of around 50° for pure quartz, while intermediate-wet conditions, indicating greater hydrogen wettability, were seen in the presence of stearic acid. This result has been used in a simulation study of hydrogen storage in a sandstone reservoir (Mahdi et al., 2021).

Hashemi et al. (2021b) have also characterized the contact angles of hydrogen-water/brine systems, on Bentheimer and Berea sandstones at pressures of 2 to 10 MPa, and temperatures of 20 to 50 °C using the captive-bubble method. Intrinsic contact angles of 25° to 45° were estimated, but no meaningful correlation with temperature and pressure was found.

Table 6 summarizes the contact angle values of hydrogen-water-sandstone and CO_2-water-sandstone systems at different temperatures and pressures. In all cases the contact angle is much less than 90°, indicating strongly water-wet conditions, in the absence of organic material. This makes hydrogen the non-wetting phase, which will tend to occupy the larger pore spaces in the rock, where it can also be trapped by water, discussed below.

3.3 Capillary pressure and relative permeability

Capillary pressure controls the equilibrium distribution of gas in the pore space. The threshold, or entry pressure, needs to be overcome to allow hydrogen to enter the porous medium.

Table 6. Summary of the contact angle values in degrees, θ, for hydrogen-water-sandstone and CO_2-water-sandstone systems at different temperatures and pressures.

Fluid system	Rock	Method	θ *	θ **	References
H_2-pure water	Sandstone	Capillary pressure	22	35	Yekta et al. (2018)
H_2-brine (10,000 ppm)	Pure quartz	Tilted plate method	~ 6	~ 20	Iglauer et al. (2021)
H_2-pure water	Sandstone	Captive-bubble	~ 32	~ 32	Hashemi et al. (2021b)
H_2-brine (5,000 ppm)	Sandstone	Captive-bubble	~ 29	~ 37	Hashemi et al. (2021b)
CO_2-pure water	Pure quartz	Tilted plate method	~ 10	~ 32	Sarmadivaleh et al. (2015)

Note: * denotes at 5.5 MPa, 20 °C, ** denotes at 10 MPa, 45 °C.

It is important that this entry pressure is sufficiently high in the cap rock, higher than the capillary pressure due to a buoyant column of gas in the storage formations so that the gas cannot escape.

The relative permeability quantifies how easily the fluid phases flow as a function of saturation. The end-points also indicate how much hydrogen can be trapped in the pore space by capillary forces: this trapped hydrogen cannot be produced which represents a loss of gas and lowered efficiency.

The simplest approach to assess multiphase flow for hydrogen storage is to use data obtained for natural gas (methane) reservoirs in the literature, under the assumption that the properties are similar (Hassannayebi et al., 2019). This may be justified by the fact that, as shown above, hydrogen, like natural gas, is the non-wetting phase in the presence of water.

An extension of this approach is to fit experimental data to closed-form empirical correlations such as the Brooks & Corey and van Genuchten models, but again the experiments on which the matches were based were derived from hydrocarbon-water or CO_2-water systems (Pfeiffer and Bauer, 2015; Tarkowski, 2019; Luboń and Tarkowski, 2020).

The final indirect approach is to use pore-scale modeling to predict the hydrogen-brine relative permeabilities. For instance, a pore-network model has been used to predict capillary pressure and relative permeability for drainage (injection) and imbibition (withdrawal) cycles, making assumptions about the wettability consistent with the contact angle measurements mentioned in the previous section (Hashemi et al., 2021a).

The lack of robust experimental work to directly quantify relative permeability and capillary pressure for hydrogen-brine systems is obvious in the literature. Although there are various studies for CO_2-brine, methane-brine, and nitrogen-brine systems (Burton et al., 2009; Ham and Kantzas, 2013; Benson et al., 2015; Manceau et al., 2015; Reynolds and Krevor, 2015), only one set of experimental work has been carried with hydrogen-brine on Buntsandstein formation rock at two conditions (Yekta et al., 2018).

In the one set of hydrogen-brine data, as discussed previously, the capillary pressure was measured and compared to mercury injection results to infer the contact angle and interfacial tensions (see Fig. 10). The relative permeability for primary drainage (initial injection) measured using the steady-state technique, Fig. 11, showed a high irreducible water saturation of around 40%, explained by the low permeability of the sample (Yekta et al., 2018). The low water relative permeability was indicative of water-wet conditions, consistent with the contact angle measurements reviewed previously. There was little impact of temperature and pressure on the measured properties.

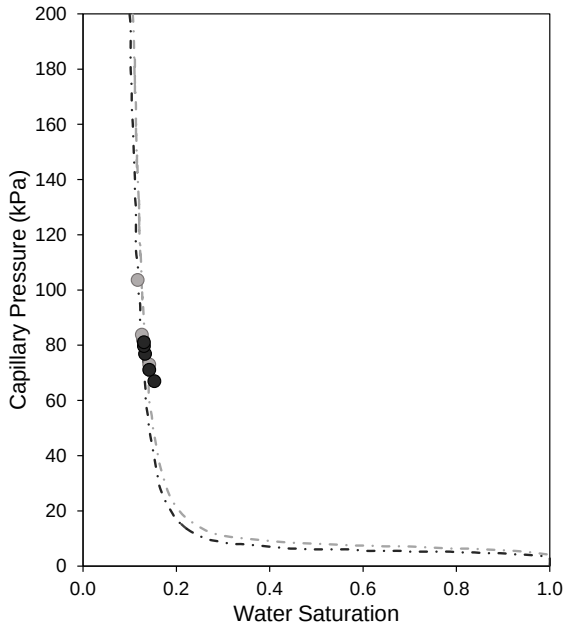

Fig. 10. Measured primary drainage capillary pressure for a hydrogen-brine system for experiments 1 (gray symbols, 20 °C, 5.5 MPa) and 2 (black symbols, 45 °C, 10 MPa) (Yekta et al., 2018).

3.4 Hysteresis

In field applications of hydrogen storage there are frequent cycles of injection and withdrawal. Therefore, primary drainage (hydrogen injection), secondary imbibition (hydrogen withdrawal), and secondary drainage (hydrogen re-injection), followed by repeated cycles of injection and withdrawal, all occur. This is unlike CO_2 storage, where only primary drainage (original injection) and secondary imbibition (where water

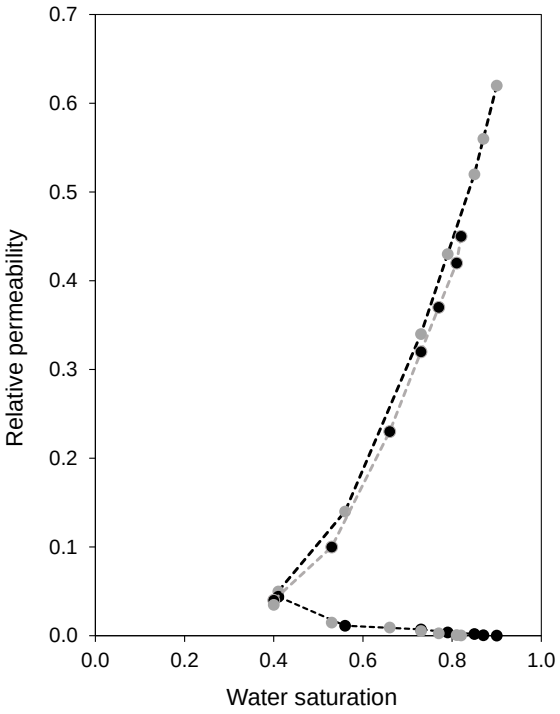

Fig. 11. Measured steady-state primary drainage hydrogen-brine relative permeabilities for experiments 3 (gray symbols, 20 °C, 5.5 MPa) and 4 (black symbols, 45 °C, 10 MPa) (Yekta et al., 2018).

displaces CO_2 as it migrates post-injection, leading to capillary trapping) need be considered. Hysteresis is therefore important in hydrogen storage applications. Hashemi et al. (2021a) investigated hysteresis in a pore-scale modeling study, mentioned in the previous section. The most important consideration is the amount of hydrogen that can be trapped during withdrawal, as the gas is displaced by water in the pore space. The amount of trapping is governed by the pore structure and wettability and, at present, it is normally assumed that the medium is strongly water-wet which leads to a large residual gas saturation.

One interesting and intriguing feature associated with hydrogen storage is that it is not correct to assume that the trapping and flow of hydrogen in its own phase is unaffected by transport of dissolved gas. Traditionally in oilfield operations it is assumed that hydrocarbon dissolution is negligible and that hysteresis is controlled by displacement between completely immiscible phases. This assumption is inherent in the use of hydrocarbon-water properties to represent hydrogen storage. With hydrogen, and CO_2, this assumption is not correct. When a phase is trapped in the pore space, it acquires a local capillary pressure governed by wettability and pore structure. Different capillary pressures result in slightly different equilibrium solubilities of the gas, thanks to Henry's law, leading to concentration gradients in the aqueous phase. Diffusion through the brine drives material from ganglia with a high local capillary pressure to others with a lower pressure. In a free fluid, this process, called Ostwald ripening, leads to the amalgamation of the gas, such that, in equilibrium, it all resides in a single large bubble. In porous media, in contrast, multiple positions of equilibrium are possible, but there is a tendency for some small ganglia to disappear and the larger ganglia to grow (Li and Fan, 2015; De Chalendar et al., 2018). This process could reconnect an originally trapped phase and certainly acts to reduce the amount of hysteresis. This is potentially significant, allowing multiple cycles of hydrogen injection and withdrawal with relatively little trapping. Ostwald ripening can lead to pore-scale rearrangement of the gas in the order of hours to days at the mm-scale, while equilibrium at the field-scale takes geological (million-year) time-scales (Blunt, 2022). Hence, for storage cycles of order a few months, we may assume local equilibrium over, say, a few cm: sufficient to change the local capillary pressures and relative permeability, but not enough to allow a large-scale change in the distribution of gas.

One result of Ostwald ripening in hydrogen storage, even if its effect is confined to fluid rearrangement at the cm-scale, is that the capillary pressures and relative permeabilities which describe macroscopic flow display less hysteresis and trapping than those used for hydrocarbon production. This means that injection and withdrawal may be more efficient than would be assumed ignoring this effect, meaning, for instance, that less cushion gas could be used. This is evidently a topic for future work to find the macroscopic flow properties which correctly encapsulate both flow, capillary forces and Ostwald ripening over the time and length scales pertinent to hydrogen storage.

3.5 Hydrogen reactions

When hydrogen is injected into an underground formation, the chemical equilibrium between the rock minerals, pore water, gases, ions and bacteria will be changed. Generally, underground hydrogen consumption or production includes two processes: 1) abiotic (chemical), and 2) biotic (bacterial). These reactions could lead to significant hydrogen loss, stored hydrogen contamination by the production of other gases (such as hydrogen sulfide), and mineral dissolution/precipitation which can increase/reduce injectivity, allow leakage and change the geo-mechanical properties of the rock. Any of these reactions can compromise secure and efficient UHS, although their impact is still poorly constrained (Lassin et al., 2011).

Hydrogen-driven redox reactions can occur with iron-bearing minerals such as hematite, goethite, or Fe^{3+} bearing clays and micas. Such reactions could change the mechanical strength of the rock matrix if hematite-containing cements or clay at grain-grain contacts in sandstone reservoirs are removed. The dissolution of minerals within the caprock could create new leakage paths, but research has indicated that the extent of such reactions is limited (Kampman et al., 2016).

In addition to redox reactions, reactions of hydrogen with dissolved sulfur species or sulfur-bearing minerals (e.g., pyrite) are expected to occur (Reitenbach et al., 2015). Besides the direct impact of mineral dissolution on porosity, permeability and mechanical properties, these reactions lead to the formation of hydrogen sulfide (H_2S), decreasing the quality of the stored hydrogen gas. Additionally, H_2S can modify the redox potential and the pH of pore waters (Truche et al., 2013) triggering further fluid-rock reactions. H_2S can also compromise the infrastructure due to its corrosive, flammable and toxic nature (Wei et al., 2017). In the case of town gas

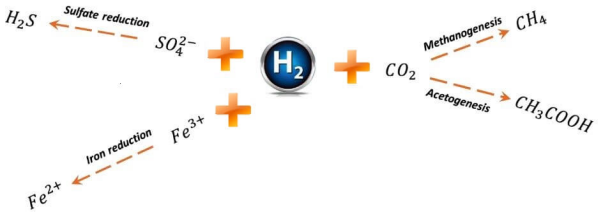

Fig. 12. Biotic reactions in underground hydrogen storage (Ebrahimiyekta, 2017).

storage in Beynes (France), it has been argued that abiotic pyrite reduction resulted in H_2S production (Reitenbach et al., 2015). As the hydrocarbon industry has decades of experience of safely producing H_2S-rich natural gas (Boschee, 2014; Bihua et al., 2018), this would be a surmountable, though costly, side-effect of hydrogen storage.

Experimental studies on reservoir sandstones under subsurface conditions (40-100 °C, 10-20 MPa) show dissolution of carbonate and sulfate cements, leading to an increase in porosity during hydrogen exposure (Flesch et al., 2018). Similar experiments on reservoir and caprock material of a natural gas storage site show an overall decrease in permeability in both rock types, due to the alteration of clay minerals (Shi et al., 2020). However, in both studies, framework minerals, such as quartz and feldspar, appeared to be unaffected by hydrogen exposure. Some potential hydrogen storage reservoirs are located in carbonate formations (Heinemann et al., 2021). Therefore, the dissolution of carbonate and sulfate minerals are of importance, as it may lead to mechanical weakening of the reservoir rock or carbonate/sulfate-cemented faults in the caprock, depending on the distribution of these cements and the local fluid to rock ratio (Hangx et al., 2015).

To predict the impact of chemical reactions over the lifetime of a hydrogen storage site, geochemical modeling is needed. To quantify the extent of reactions in the reservoir and caprock, and to assess the probability and magnitude of the expected processes, the development of a geochemical database is needed, analogous to those made for CO_2 storage (Heinemann et al., 2021), containing the reactions of hydrogen with dissolved ions and mineral surfaces including their kinetics, as well as possible catalysis. In addition, complementary flow-through experiments at realistic in situ conditions, using site-specific rock from potential storage sites, as well as studies from natural hydrogen fields (Prinzhofer et al., 2018), need to be benchmarked against reactive transport models.

Biotic reactions are known to be important in hydrocarbon reservoirs and may compromise the feasibility of storage at some sites (Gregory et al., 2019). Although several studies have looked at hydrogen utilization under natural concentrations little is known about the effects that high hydrogen pressures expected in UHS will have on the subsurface microbial system. The main microbial H_2-consuming terminal electron-accepting processes likely to occur in UGS sites are methanogenesis, sulfate reduction, iron reduction, and acetogenesis, shown in Fig. 12. These processes have been observed to occur at temperatures as high as 90 °C and at high salinities (Basso et al., 2009). Thus, H_2 can be consumed to generate methane or acetate in the presence of CO_2/HCO_3, or hydrogen sulfide in the presence of sulfate (Heinemann et al., 2021). The potential impact of microorganisms is controlled by temperature, salt concentration, pH, and substrate supply, with optimal and critical values, as summarized in Table 7.

Conditions where microbes can thrive range between 15 °C up to ~ 120 °C without any clear thresholds for brine salinity. The possible pH range for microbes lies between 0-11 with the highest diversity between 6-7. Other factors such as high concentrations of toxic chemicals, low water activity (Payler et al., 2019), radiation (Jagger, 1983) and low rock permeability (Heinemann et al., 2021) can also have a significant influence and reduce the activity of microbes (Thaysen et al., 2021).

The main impact of microbes on hydrogen storage is the permanent loss of hydrogen due to the conversion of hydrogen into products including CH_4 and H_2S. This H_2 loss will continue over the whole H_2 injection/production cycles in contrast to the initial H_2 loss due to diffusion, which will be strongest in the first cycle and decrease over time (Dopffel et al., 2021).

As the microbial population density increases, microbially formed biofilms or mineral precipitation could lead to pore-clogging, and therefore to a reduction of hydrogen injectivity and productivity. This is a common problem encountered in geothermal applications (Würdemann et al., 2016) and CO_2 storage operations (Zettlitzer et al., 2010). Because microbes catalyze redox reactions, a variety of different mineral precipitations can be triggered, leading to plugging and declining injectivity (Kryachko, 2018; Dopffel et al., 2021). Experiments on microbial enhanced oil recovery recorded an overall change of the absolute permeability by a factor of 0.56 to 0.86 accompanied by an increasing microbial density (Heinemann et al., 2021). Modeling of pore-clogging effects in the near well-bore area during hydrogen injection provide evidence that lateral gas flow near the wellbore improves, while vertical flow rates decrease (Gaol et al., 2019). Field data from the Sun Conversion and the HyChico projects (Table 3) however did not show indications of pore-clogging effects after one storage operation cycle. Overall, pore-clogging due to microbes has not been investigated in detail and further study is required to assess the probability and severity of the process during the long-term operation of hydrogen storage.

Experience from storage operations of hydrogen-rich town gas shows ranges from zero hydrogen consumption in Beynes (France), to a significant loss of hydrogen, with a concurrent reduction of CO_2 and increasing CH_4, over a seven-month cycle in Lobodice, Czech Republic. Approximately half of the H_2 (45%-60%) in the stored town gas was microbially transformed into methane or hydrogen sulfide at relatively low temperatures (35 °C) (Šmigáň et al., 1990). In Ketzin, 61% of the H_2 volume has been lost as well as important modifications to gas composition and H_2S generation with pressure losses and temperature changes (Stolten and Emonts, 2016). However, it is unclear whether or what kind of microbial processes had been active at the site. Microbial hydrogen consumption was also reported during combined storage of natural gas with additions of hydrogen and CO_2 (e.g., the Underground Sun Storage and Sun Conversion projects, Austria;

Table 7. Main storage impact, hydrogen consumption, and growth conditions for cultivated hydrogenotrophic methanogens, hydrogenotrophic sulfate reducers, homoacetogens and hydrogenotrophic iron(III)-reducing bacteria (Thaysen et al., 2021).

Class of microorganism	Storage impact	Reaction	Temperature (°C)	Salinity (g/L)	pH
Methanogens	Hydrogen loss by methane production & clogging	$4H_2 + CO_2 = CH_4 + 2H_2O\downarrow$ or$\downarrow$ $3H_2 + CO = CH_4 + H_2O$	Optimum: 31-41 Critical: 120	Optimum :61 Critical: 200	Optimum: 6.1-7.6 Critical: 4.4-9.1
Sulfate reducers	Hydrogen loss by methane production, corrosion, clogging	$5H_2 + SO_4^{2-} = H_2S + 4H_2O$	Optimum: 21-31 Critical: 115	Optimum: <100 Critical: 245	Optimum: 6.1-7.4 Critical 0.7-11.6
Homoacetogens	Hydrogen loss by CH_3COOH production, clogging	$4H_2 + 2CO_2 = CH_3COOH + 2H_2O$	Optimum: 21-31 Critical: 73	Optimum: <41 Critical: 302	Optimum: 6.1-7.6 Critical 3.5-10.8
Reducing bacteria	Hydrogen loss by Fe(n) production, clogging	$H_2 + Fe_2O_3 = 2FeO + H_2O$	Optimum: 0-31 Critical: 91	Optimum: <41 Critical: 200	Optimum: 6.1-7.6 Critical: 1.5-9

Optimum conditions is where the growth peaks; critical is the maximum conditions beyond which no growth is possible

HyChico project, Argentina (Perez et al., 2016)). During the Underground Sun Storage Project in Lehen, Austria, 10% H_2 from green sources was mixed with natural gas and stored for a test period of four months (Pichler, 2019). After this period 18% of the injected H_2 could not be recovered and a concurrent increase in CH_4 was observed. The same route was taken by the Argentinian HyChicon project, where the initial tests in 2010 were planned to store H_2 generated form electricity from a nearby wind farm in a depleted gas reservoir. Available information is limited, but during the storage cycle 10% microbially triggered H_2 loss was observed (Perez et al., 2016).

Overall, this review demonstrates that microbial activity can lead to significant loss and contamination of the injected hydrogen (Ebigbo and Gregory, 2021). This is potentially a serious problem that needs to be understood on a site specific basis before large-scale investments in storage can be made.

4. Challenges for hydrogen underground storage

In this section we synthesize the previous discussion to describe the different challenges associated with underground hydrogen storage.

4.1 Reservoir engineering

In the context of CO_2 storage, the principal mechanisms by which the gas can be retained in the pore space are stratigraphic trapping beneath caprock, capillary trapping as a residual phase surrounded by water, dissolution into the formation brine, and reaction (Boot-Handford et al., 2014). The problem here is that unlike CO_2 storage, where these mechanisms are desirable for long-term secure storage, preventing the CO_2 escaping, in the context of hydrogen, we need the gas to be mobile so that it can be withdrawn. The accumulation of hydrogen below a caprock at a high saturation is desirable, as the hydrogen can then be produced, but the other mechanisms result in a loss of useable gas, and may compromise a long-term project. As discussed in sections 2 and 3, two other concerns are the mixing of hydrogen with cushion gas, driven by hydrogen's large diffusion coefficient, and bacterial activity. The mixing of gases in the subsurface is difficult to control, with a combination of spreading and diffusion (mixing) causing the produced gas to be a mixture of the gases resident in the formation.

4.2 Geology

The geological properties of a storage formation, particularly for aquifer storage, are both important yet uncertain: these include depth, pressure, storage capacity, permeability and possibility of leakage through the cap rock, faults or fractures (Zivar et al., 2021). Commonly, the depth of salt caverns is 400-1,000 m and depleted reservoirs or aquifers are more than 800 m. The deeper the formation, the higher pressure that the site can be operated at, but this comes with additional costs from wells and site preparation. In depleted hydrocarbon reservoirs, the hydrogen could be affected by residual fluid in the reservoir which would lower the operation performance. Since the amount of required cushion gas in aquifers is large, these storage types have lower priority compared to salt caverns for current developments, but the larger capacity and geographical availability of aquifers makes them important for global-scale storage, as emphasized already (Bai et al., 2014).

4.3 Economics

Storing hydrogen is one significant challenge for a future hydrogen economy. Fig. 13 compares the storage costs of various hydrogen storage technologies in different cycles, although aquifer storage was not considered. The key finding is that underground hydrogen storage may unlock hydrogen's competitiveness sooner than previously anticipated (BloombergNEF, 2020).

While there is considerable experience of subsurface projects in the oil and gas industry, and growing application of CO_2 storage, hydrogen storage introduces additional challenges. Its unique subsurface behavior, specifically microbial

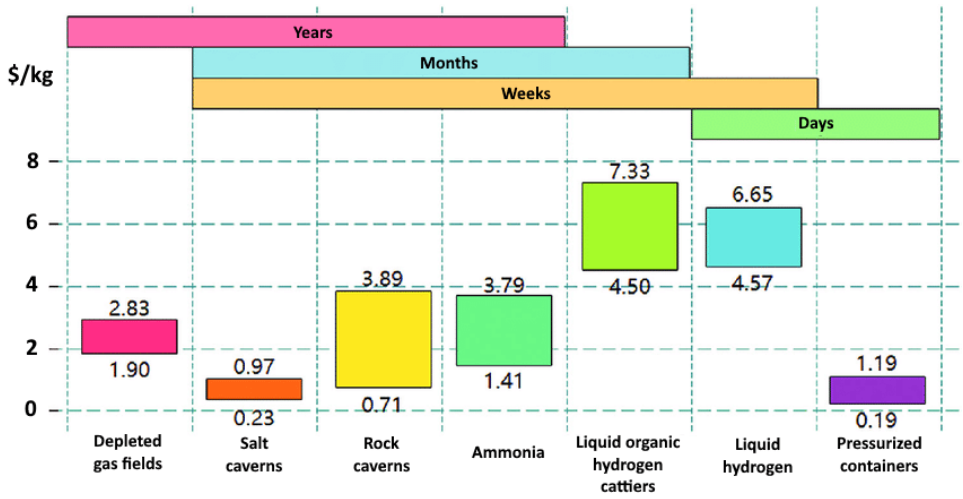

Fig. 13. The storage costs of various hydrogen storage technologies (The Hydrogen Economy Outlook, 2020).

reaction, mixing with other gases, and hysteresis, needs to be understood for sites of interest. Since hydrogen is a commodity, not a waste, any loss of gas comes at a cost. On the other hand, the value of hydrogen does enable proper investment in site characterization, infrastructure and innovative engineering, something that is difficult for CO_2 storage where investment in a site is viewed as simply a cost with little or no financial return.

5. Site selection for underground hydrogen storage projects

Storing hydrogen in underground geological formations is potentially a costly process which requires careful appraisal, decision-making and evaluation. From primary levels of UHS appraisal which include site detection and exploration, to the final stages of providing and installing hydrogen production facilities and crew costs, all of these procedures need investment and time, and if an appropriate site for UHS projects is not selected, much, if not all, of the associated costs will be wasted. Furthermore, ensuring long-term and safe storage of hydrogen requires a wise selection of the storage site.

For a successful UHS project, along with considering technical challenges for site selection, environmental issues must be equally taken into account (Zapf et al., 2015; Deveci, 2018). Below are listed the criteria used for site selection (Lewandowska-Śmierzchalska et al., 2018; Nemati et al., 2020; Zivar et al., 2021).

Technical criteria are divided into eight sub-categories.

1) **Geology:** the ability of the underground formation to store hydrogen dependent on cap rock and host rock properties. Also can UHS facilities be developed at the site?
2) **Depth:** defined as the required depth assuring safe and economic UHS.
3) **Area:** one of the main factors that determines the capacity of underground formations for UHS.
4) **Thickness:** the effective thickness of reservoir (or layers) for UHS.
5) **Cap Rock:** a suitable cap rock for UHS must have sufficient thickness in conjunction with low permeability.
6) **Reservoir Permeability and Porosity:** the more porous and permeable the reservoir, the greater the capacity and injectivity.
7) **Storage Capacity:** is the total capacity of the underground formation for storing hydrogen.
8) **Reservoir Pressure:** reservoir pressure must be high enough to assure that all injected hydrogen will be produced. This factor determines the amount of cushion gas required.

Economics is divided into four sub-categories.

1) **Labor:** human resources and their cost associated with the UHS project.
2) **The Distance between Supply and Demand:** determines the amount (cost) of pipeline or transportation which is required for transporting the produced hydrogen to the area of demand.
3) **Infrastructure Availability:** is defined as the availability and cost of infrastructure for storage.
4) **Initial Investment**: in order to establish the facility, primary investments are vital in all UHS projects.

Health, safety, and environment is divided into three sub-categories.

1) **Regional Risks:** the probability of occurrence of natural disasters such as earthquakes at the location of UHS projects which can lead to additional expenses and leaks.
2) **Environment and Public:** UHS projects must be based on accurate predictions and estimations to assure that any risks are minimized with little impact on the environment.
3) **Legal Restrictions:** before starting a project, any legal hurdles need to be addressed.

Social criteria are divided into two sub-categories.

1) **Social Acceptance:** before starting a project, all local residents and communities at or near the location of the project must be surveyed, and UHS projects can only

Table 8. Criteria considered in the literature for site selection of hydrogen underground storage in six countries.

Location	Criteria	Storage type	References
Romania	Operator interest, geology, brine demand, infrastructure Good geological conditions	Salt cavern	Iordache et al. (2014)
Poland	Geology (geothermal gradient), volume, depth	Salt cavern	Lewandowska-Śmierzchalska et al. (2018)
	Geology (tectonic activity, overburden lithology), volume, depth	Aquifer	
Poland	Geology (shape and complexity of the structure), volume, recognition stage, depth	Salt cavern	Tarkowski and Czapowski (2018)
Turkey	Investment cost, storage cost, gas pressure, reservoir porosity and permeability, geology	Salt cavern	Deveci (2018)
US (Oregon)	Infrastructure, stakeholder access, cost, environment	Pilot scale project	Taie et al. (2021)
Canada	Geology, production capacity, cost, government support	All types	Lemieux et al. (2020)
UK	Depth, reservoir quality, volume, number of entries	Saline aquifer	Scafidi et al. (2021)

proceed with local acceptance.

2) **Job Creation:** the number of jobs created by implementing a new UHS project and facility and its effect on the economy of the local area.

In summary, Table 8 lists some of the considerations used to select project sites worldwide.

6. Conclusions

Large-scale underground hydrogen storage is a necessary component of efforts to move toward a sustainable zero-emissions economy. This review has provided a comprehensive reference outlining current projects and plans. The properties of hydrogen relevant to storage and the types of underground storage have been discussed in detail.

At present only salt caverns have been used successfully for underground hydrogen storage. While they prevent leakage of the stored gas, thanks to the impermeable and ductile nature of the salt, and can allow rapid injection and withdrawal several times a year, they are expensive to construct, limited in capacity, and can only be formed in a few areas where there are natural salt deposits. Lined rock caverns can be constructed at more sites and also allow rapid injection and withdrawal. However, they are expensive to build and also limited in capacity.

Storage needs to be implemented a Gt scale to power a global renewables economy. To achieve this, depleted hydrocarbon reservoirs and aquifers are needed, which have greater capacity than salt caverns, and are more widespread geographically. Depleted oil and gas fields have the advantage of an existing injection and production infrastructure and are bounded by known geological traps. On the other hand, the use of hydrogen is likely to require the replacement of most of the steel used in wells and pipelines.

Two significant sets of challenge remain to be overcome before hydrogen storage, and the use of hydrogen as an energy vector, can largely or completely displace our current dependence on fossil fuels. The first is technical. There is limited experience of the use of hydrocarbon fields and aquifers for long-term hydrogen storage. Particular problems that need to be considered, that go beyond our current experience with carbon dioxide storage, are the possibility of bacterial degradation, mixing of hydrogen with cushion gas, loss of hydrogen due to capillary trapping with water, and the impact of Ostwald ripening on reducing hysteresis, at least at the small scale. Careful analysis of putative sites over a range of temperatures and pressures is needed before the overall efficiency of storage can be quantified with confidence.

The second challenge is economic. Today most hydrogen is produced from fossil fuels, gray hydrogen, and the cost of combining this with carbon capture and storage (blue hydrogen) or direct generation by electrolysis using renewable energy is, at present, a barrier to large-scale implementation. This review has suggested that these costs may decrease over time, allowing an economically viable energy transition, but this does require efficiencies of scale and improvements in technology. Furthermore, loss of hydrogen during underground storage, or expenses associated with new injection and production infrastructure, combined with separating hydrogen from cushion gas or water, all add to the cost of storage. However, hydrogen is a valuable commodity, which may help drive innovation in storage design.

Conflict of interest

The authors declare no competing interest.

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