



Introduction

Due to access to large volumes and extended storage periods, underground hydrogen storage (UHS) provides seasonal adjustment and meets peak demand to stabilize the power grid. Storage in salt caverns, depleted oil or gas reservoirs, and aquifers is seen as a potential option for short to long-term, large-scale, hydrogen storage project development. Furthermore, existing underground gas storage reservoirs (UGS) can be converted into hydrogen storage reservoirs, enhancing the availability of UHS near to existing natural gas supply systems.

However, the safe and efficient storage of hydrogen in depleted oil, gas reservoirs or aquifers is still unproven on a commercial scale. Only a small number of hydrogen operational pilot projects, most experiences and information on UHS and the behaviour of hydrogen in the subsurface come from so-called Town Gas Storage (Stolten and Emonts, 2016), which was an industrially produced fuel gas historically used and stored in limited locations. Town gas is still in use today in some locations in the Far East.

The usage of natural gas in each hemisphere is cyclic due mainly to the demand for heating during the winter months. In the future, hydrogen is expected to partially replace natural gas as an energy source, thus the demand for hydrogen would most likely also see some form of seasonal cycle as natural gas.

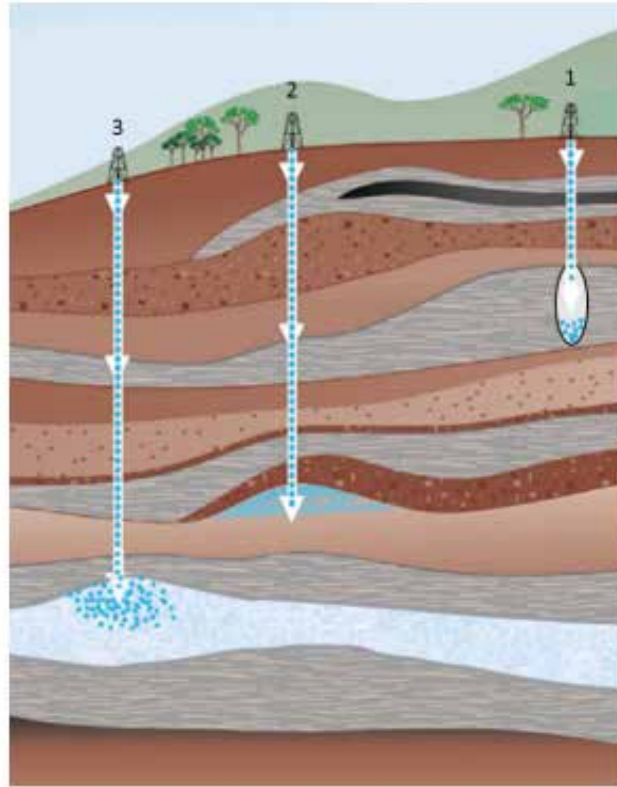
The future energy mix is also expected to have an ever-increasing blend of renewable energy sources, with obvious periods of energy excess and shortfalls. The fluctuations in renewable power, such as wind power and solar energy, could be part used to produce green hydrogen (or stored in battery devices), also providing for some short-term fluctuations in hydrogen supply, which would also require some form of storage facilities.

Underground Hydrogen Storage (UHS)

The three most common possible UHS systems include salt caverns, depleted oil or gas reservoirs, and aquifers (Figure 1).

Table 1 also shows a list of UHS facilities worldwide. Current state of play for each storage type will be briefly discussed in the following.

Figure 1: Three different UHS: 1- Salt Caverns, 2- Depleted Hydrocarbon Reservoirs, 3- Saline Aquifers



Source: Amid et al, 2016

Table 1: Underground Hydrogen Storage Examples Worldwide (Panfilov, 2016 & Ebrahimiyehta, 2017)

	Type of Storage	Depth (m)	Since	Electricity Generation	Hydrogen Percentage	Pressure (bar)	Capacity (m ³)
Teeside, UK	Salt Caverns	370	Since 30 years	30 GWh	95%	45	3 x 70,000
Moss Bluff, Praxair, US	Salt Caverns	850-1,400	2007	80 GWh		70-135	566,000
Spindletop, (Air Liquid), US	Salt Caverns	850-1,400			95%	Up to 150	600,000 m ³
Clemens Dome, (ConocoPhillips), Texas, US	Salt caverns	850	Since 1986	892 GWh	95%	150	580,000
Kiel, Germany	Salt caverns	1,335	Since 1971		62%	80-100	32,000
Ketzin, Germany	Aquifer	200-250	Since 1964		62%		
Beynes (GDF), France	Aquifer	430	Approx 20 years		50-60%		1,185 MMSm ³
Lobodice, Czech	Aquifer	400-500	Since 1960		45-50%	45-59	400 MMSm ³
Kasimovskoie, imovskoie, Russia	Aquifer						1,800 MMSm ³
Hychico Argentina	Depleted Gas Reservoir	600-800	2015	24.6 GWh	100%	25	

Salt Caverns

Salt Caverns have been used for high purity hydrogen storage by the chemical sector in the UK since the 1970's (Teesside) and in the US since the 1980's (US Gulf Coast and The Chevron Phillips Clemens Terminal in Texas) as well as Yakshunovskoe in Russia (Crotofino et al., 2010), (Panfilov et al., 2006), (Pichler, 2013) and (Stolten and Emonts, 2016).

Salt caverns are characterised by minimum leakage due to tightness of the salt rock, low operational cost and have a high recovery efficiency (produced H₂/ injected H₂).

The Teesside storage facility has three shallow elliptic salt caverns enabling large-scale hydrogen storage. The depths of the salt caverns are between 350 and 400 m, in the upper Permian salts and each cavern has a volume of approximately 70,000 m³; the stored gas is 95% hydrogen and 3-4% CO₂ with an energy storage capacity of 30 GWh for the working gas (Stolten and Emonts, 2016). There are two much larger caverns in Texas (Crotofino et al., 2010). The ConocoPhillips Clemens Terminal in Texas has stored hydrogen since 1980. The cavern roof is about 850 m underground. The cavern is a cylinder with a diameter of 49 m, a height of 300 m, and a usable hydrogen capacity of 30 MMm³, or 2,520 metric tons. This storage facility is directly connected to the Old Ocean refinery. The stored gas is 95% hydrogen with an energy capacity of 892 GWh. Also in Texas is Air Liquid, a major producer of hydrogen in North America uses a large salt cavern on the Gulf Coast to store 95% hydrogen. The cavern allow hydrogen storage with the aim of enhancing flexibility to meet increasing customer demand for hydrogen. The hydrogen network is expected be extended by 90 miles to southeast Texas.

Moss Bluff, Texas (US), Praxair has been operating UHS in a salt cavern for several years, to enable "peak shaving" of its hydrogen production. This facility is connected to the Praxair Gulf Coast hydrogen pipeline network, which serves the petrochemical needs of Texas and Louisiana (Panfilov, 2016).

Porous Media

Another form of potential UHS is represented by depleted hydrocarbon reservoirs or existing gas storage reservoirs that are no longer in operation such as Rough gas storage in the UK (Blanco et al., 2018).

The risks associated with UHS in porous media (such as depleted reservoirs) include contamination due to hydrogen contacting solids and fluids in the reservoir. Likewise, the formation of hydrogen sulphide (H₂S), which is a corrosive and poisonous gas, occurs due to activity of microbes, resulting in loss of hydrogen inventory. Another possible challenge is leakage, due to the low density and high diffusivity of hydrogen. Therefore, it is possible for hydrogen to leak out of the reservoir through the caprock which would be significantly reduced in the case of natural gas. The recovery efficiency of these storage facilities is considered a challenge, relating to the volume of injection and production of hydrogen in and from these storage facilities.

There is very limited experience of UHS in depleted hydrocarbon reservoirs and aquifers on a commercial scale. Two pilot projects that are associated with the UHS in subsurface porous media and investigate the feasibility of UHS in geological formations include the German Hydrogen to Store (H2STORE) project (Pudlo et al, 2013) and the Austrian SUN.STORAGE project (Sun Project Final Report, 2017). The H2STORE Project is a collaborative research project which assesses the potential of hydrogen produced from wind and solar power stored in depleted gas reservoirs sealed by mud rock layers (H2Store Project, 2013). The H2STORE project also investigates the geochemical, microbiological and mineralogical interactions produced by the injection of hydrogen into depleted gas reservoirs.

Similarly the SUN.STORE project included research into an array of subjects related to the storage of hydrogen in subsurface porous media from geochemistry and reactive transport modelling, to economic and legal assessment of hydrogen production, storage and transportation; the project was concluded in 2017 (SUN Project final report, 2017). The project's main theme included an actual field test of injection and the production of natural gas and hydrogen mixture under actual reservoir conditions (SUN Project final report, 2017).

The Hychico project in Argentina (Pérez et al., 2016), is an example of the use of a depleted gas reservoir as an UHS facility. The hydrogen is produced from wind powered electrolysis. The first stage of the project is a hydrogen plant with 120 Mm³/h (99.99% purity), a Wind Turbine (6.3 MW) with an average capacity of 50%. The second stage includes connecting the hydrogen plant to the wellhead of an oil and gas field in Diadema, Patagonia (Pérez et al., 2016) via a 2.3 km hydrogen pipeline constructed in 2014. The hydrogen is reported to be injected into a sandstone gas reservoir at a depth of 600-800 m, under a pressure of 10 barg and temperature of 50°C (Pérez et al., 2016).

The storage of hydrogen in aquifers is not proven, and studies to date use experience in town gas (manufactured gas) in aquifer storage facilities. Town gas is a mixture of gases, typically 25-60% hydrogen, and smaller amounts of CH₄ (10-33%), CO / CO₂ (12-20%) and N₂ < 30% which is generated by coal gasification (Buzek et al., 1994), (Panfilov 2016), (Pichler, 2013) & (Heinemann et al., 2021), (Stolten et al., 2016). Town gas storage has been used in France (Beynes, Ile de France), Czechoslovakia (Lobodice) and Germany (Ketzin, Bad Lauchstradt, Kiel, Burggraf-Bernsdorf) (Panfilov 2016), (Ebrahimiyejta, 2017), (Buzek et al., 1994), (Heinemann et al., 2021) and (Stolten et al., 2016).

Hydrogen Fluid Properties

Hydrogen is the third most abundant and the lightest element found on Earth and is a non-toxic, odourless and colourless gas. Hydrogen has the second lowest melting and boiling points (after Helium) at 14 K and 20 K respectively at atmospheric pressure which makes it difficult to store under standard conditions. Other gases like Carbon Dioxide (CO₂) and Methane (CH₄) can be liquefied at temperatures of around 293 K (Ebrahimiyehta, 2017) & (Visser, 2020). As seen in Figure 2 it is not possible to store hydrogen in the liquid state under standard conditions (273 K, 1 atm).

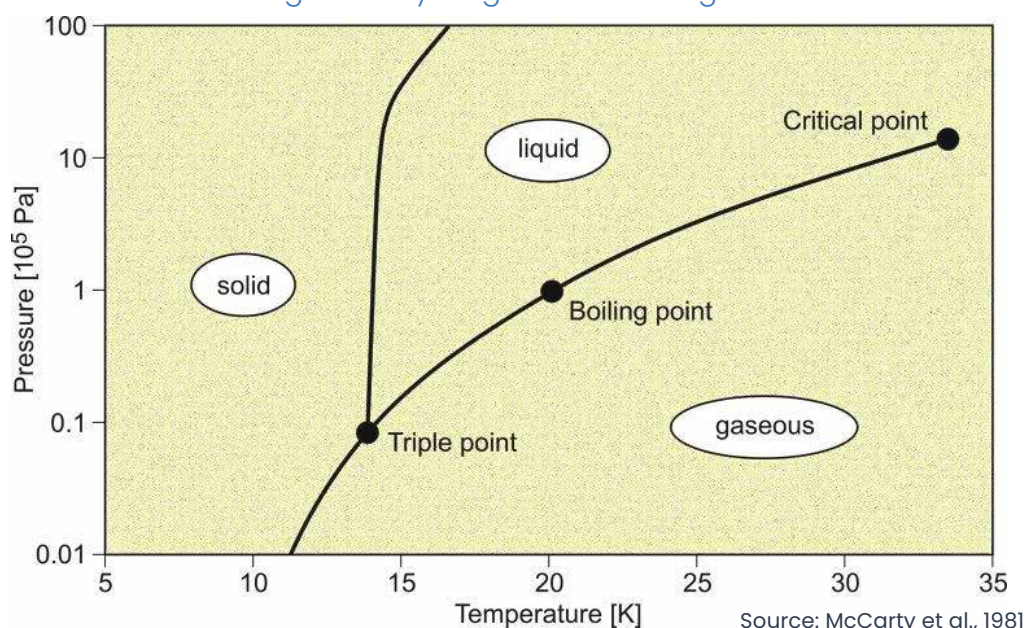
Hydrogen has a very low density (0.084 kg/m³) at standard conditions (Lanz et al, 2001). Hydrogen is about 8 times less dense than CH₄ and 22 times less dense than CO₂ in which more space and pressure will be required for hydrogen to store the same mass amount of gas (methane) for the same energy content (Table 2).

The critical point of hydrogen is at a temperature and pressure of 33 K (-239.97°C) and 13x10⁵ Pa (188 psia) respectively (Figure 2). Therefore, hydrogen needs to be stored in the gaseous phase (Heinemann et al., 2021).

Table 2: Physicochemical Properties of H₂, CH₄ and CO₂ (H2tools, 2021)

Properties	H ₂	CH ₄	CO ₂
Molecular weight	2.016	16.043	44.09
Density @25C & 1 atm	0.089 Kg/M3	0.657 Kg/M3	1.98 Kg/M3
Viscosity @ 25 C & 1 atm	0.89 x 10 ⁻⁵ Pa s	1.1 x 10 ⁻⁵ Pa s	1.49 x 10 ⁻⁵ Pa s
Solubility in pure water @25°C & 1 atm	16 x 10 ⁻⁴ g/L	22.7 x 10 ⁻³ g/L	1.45 x 10 ⁻³ g/L
Boiling point	-253°C	-162°C	-78.4°C
Critical Temperature	-239.9°C	-82.3°C	-31°C
Critical Pressure	12.8 atm	45.8 atm	72.8 atm
Heating Value Range	120-142 kJ/kg	50-55.5 kJ/kg	-
Diffusion in Pure Water@25°C & 1 atm	5.13 x 10 ⁻⁹ m ² /s	1.85 x 10 ⁻⁹ m ² /s	1.60 x 10 ⁻³ m ² /s
Flash Point	-253°C	-188°C	-
Flammability Range	4-75°C	5-15°C	-
Research Octane Number (RON)	>130	125	-
Auto Ignition Number	585°C	540°C	-

Figure 2: Hydrogen Phase Diagram

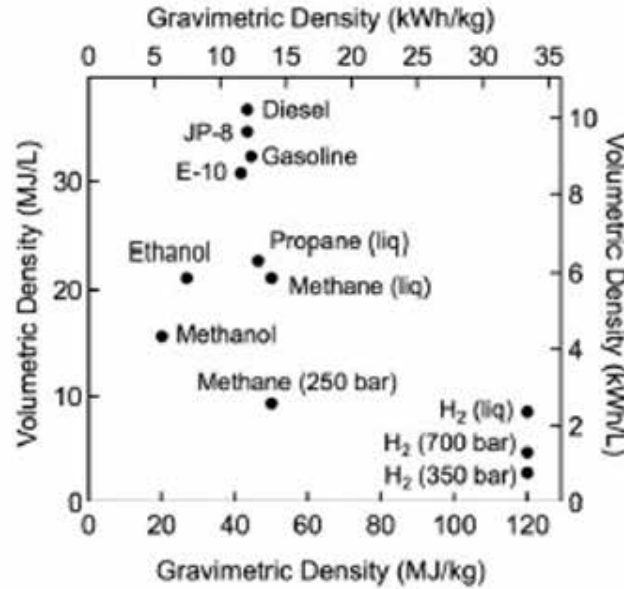


Similarly to the density, hydrogen viscosity is low compared to CH₄ and CO₂.

Hydrogen has a high gravimetric energy density of 120 MJ/kg which offers a high storage potential of energy compared to natural gas (55 MJ/kg), but it has a low volumetric energy density of 10.8 MJ/m³ at standard conditions compared to natural gas (32.5 MJ/m³).

Figure 3 shows the volumetric and gravimetric density of a number of energy carriers.

Figure 3: Volumetric and Gravimetric Density of a few Energy Carriers



Source: Fisher et al., 2009

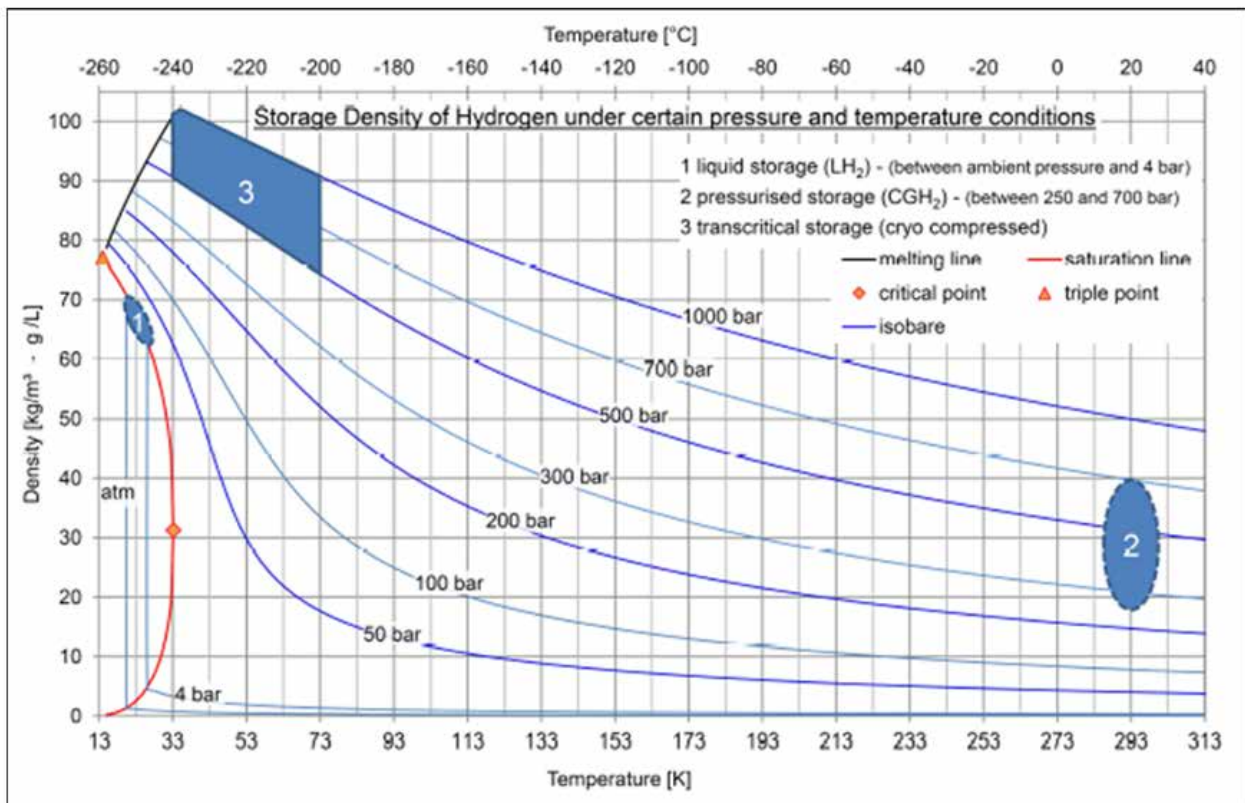
Hydrogen Fluid Properties

Figure 4 shows three different storage technologies which have differing storage densities for hydrogen under certain pressure and temperature conditions.

1. Liquid storage;
2. Compressed gas storage;
3. Cryogenic-compressed storage.

It can be seen from Figure 4 that liquid storage and cryo-compressed storage require extremely low temperatures, therefore, hydrogen is generally stored as a gas at temperatures ranging between 300-400 K and pressures ranging between 50-300 bara.

Figure 4: Storage Density and Hydrogen under Certain Pressure and Temperature Conditions



Source: ILK Dresden. "Storage Density of hydrogen under certain pressure and temperature conditions"

Main Risks and Challenges of UHS

Costs and Economics Aspects

The cost for using salt caverns for UHS is higher than other UHS options as they are expensive to construct as well as requiring infrastructure development, but their operational costs are lower than depleted hydrocarbon reservoirs and aquifers. It is also seen that there is a wide range of costs associated with UHS.

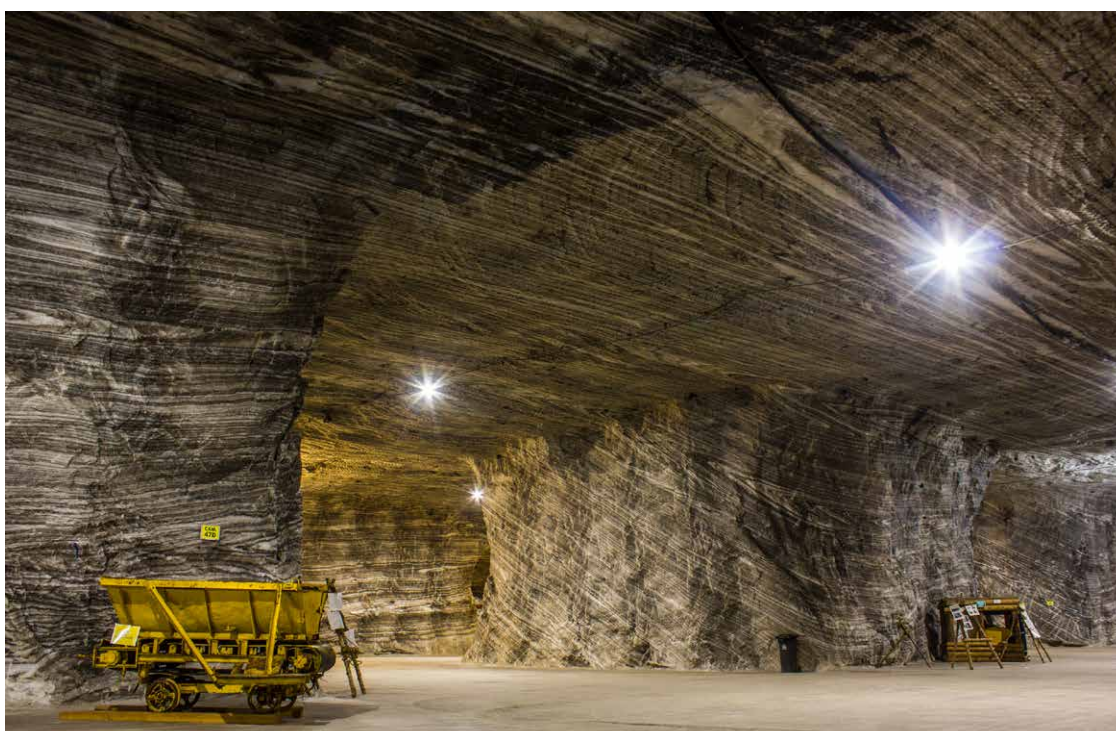
Table 3 shows the Levelised Cost of Storage (LCOS) and Capital Costs (CAPEX) associated with storing hydrogen in Salt Caverns, Depleted Reservoirs and Aquifers.

Table 3: LCOS and CAPEX Costs for Hydrogen Storage

Type of Storage	LCOS (US\$/kgH ₂)	LOCS (US\$/kWh)	CAPEX (US\$/kgH ₂)	CAPEX (US\$/kWh)	Working Volume (tH ₂)
Salt Caverns	0.28	0.01	27.2	0.82	500
	1.40	0.04	51.5	1.55	1,912
Depleted Gas Reservoirs	0.88	0.03	18.4	0.55	1,912
Aquifer	0.89	0.03	19.3	0.58	1,912

Notes:

1. It is difficult to generalise storage costs because of the wide variety in sizes, operating conditions of storage, and the number of injection and withdrawal cycles.
2. These estimates are based on literature with a set of assumptions about the storage specifics and the way the storage would be operated (e.g. the number of cycles), which all influence the calculated levelised cost of storage (LCOS).
3. Levelised cost of storage (LCOS) - The cost of kWh or MWh electricity discharged from a storage device accounting for all cost incurred and energy produced throughout the lifetime of the device.
4. It should be noted that it is likely for the larger working volumes >750 tH₂ more than one salt cavern may be required.
5. Data in the table is based on the following references, adjusted to 2021 basis: 2020 Grid Energy Storage Technology Cost and Performance Assessment, Ahluwalia et al, 2019 and Lord et al, 2014, 2020 Grid Energy Storage Technology Cost and Performance Assessment, 2020.

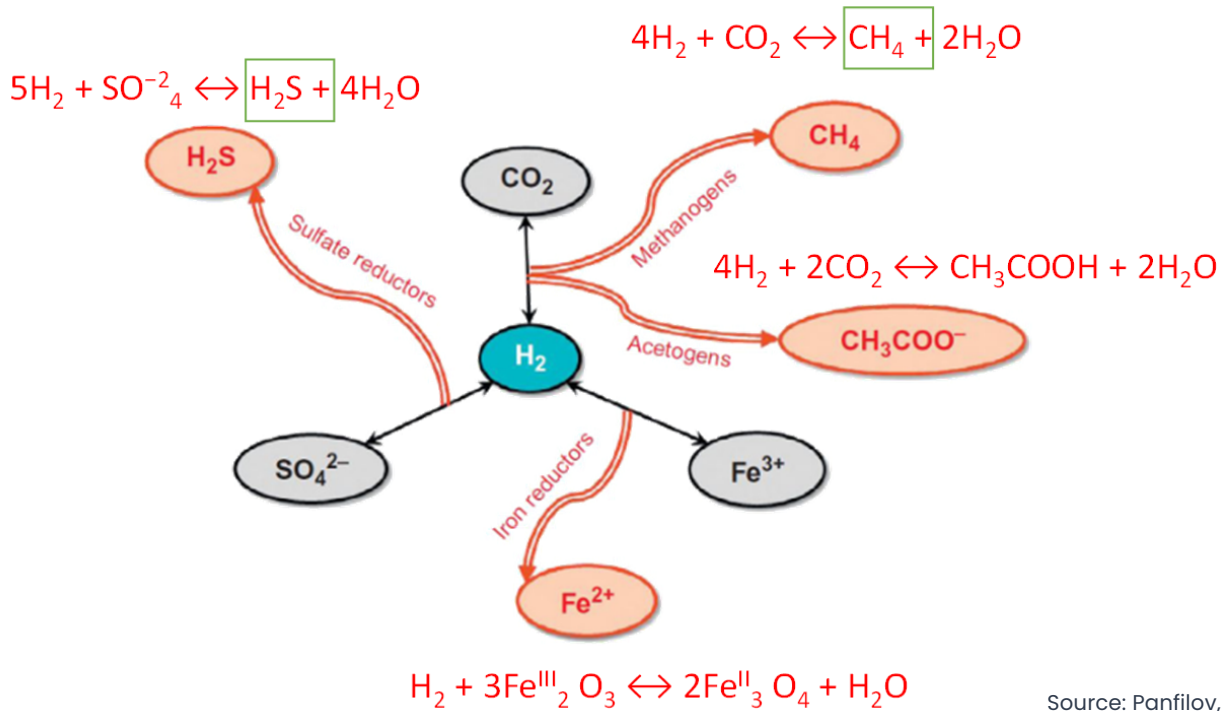


Main Risks and Challenges of UHS

Hydrogen Reactions in the Subsurface Environment

Hydrogen reactions in the subsurface can either be Geochemical, Biochemical or microbial growth as show in Figure 5.

Figure 5: Hydrogen Reaction with the Subsurface Environment



Geochemical Reaction

The storage of hydrogen in reservoirs exhibits risks due to geochemical reactions with rock minerals and reservoir fluids. This could lead to fluid losses in addition to damage to the rock, such as changes to the porosity and permeability of the reservoir rock and dissolution of minerals. Hydrogen is a highly reactive element and will change the chemical equilibrium between the formation pore water, dissolved gases and rock matrix. Such reactions can cause the dissolution or precipitation of minerals in the reservoir rock or fluid, which can alter the porosity and permeability of the rock, impacting the rock strength and productivity of fluids from the rock.

Other geochemical reactions expected to take place during hydrogen injection in UHS include redox reactions with iron minerals such as iron bearing clays, micas, Hematite and Goethite impacting rock strength as well as formation of leakage pathways in the caprock (Figure 5). However, such reactions could also lead to some deterioration in the caprock porosity and permeability, presenting a better seal and preventing leakage (Visser, 2020) and (Heinemann, et al., 2021).

Experiment studies show that typical sandstone reservoirs are not highly reactive with hydrogen (Hemme et al., 2018).

It is prudent to know the chemical composition of the rock and fluids during the screening of a reservoir for UHS. Performing such studies on depleted gas reservoirs could include laboratory experiments using cores from the reservoir rock in addition to geochemical studies.

Main Risks and Challenges of UHS

Biochemical Reaction and Microbial Growth

Micro-organisms are both naturally occurring in reservoirs and introduced during drilling or injection of gas or water.

The main risk related to the presence of microbes for UHS is the conversion (and loss) of hydrogen into products such as CH_4 or H_2S . The hydrogen reactions in the subsurface are shown in Figure 5.

Studies show a reduction in hydrogen ranging between 3–17% (Panfilov 2016). This process usually referred to as methanogenesis, which is the loss of hydrogen and the associated energy loss.

Further evidence of hydrogen consumption due to microbial growth can be found in town gas storage sites in Czech Republic France, Germany, Poland and Belgium.

Another type of hydrogen loss is due to Acetogenesis, in which microorganisms cause the pH value to decrease and usually occurs when micro-organisms are in contact of H_2 and CO_2 .

Other risks include hydrogen loss, and hydrogen sulphide or hydrogen sulphate formation with associated corrosion and acidification (Figure 5).

In France (Beynes) hydrogen (50%) has been stored from 1956 to 1974 in a saline aquifer of 385 Msm^3 by Gaz de France (Carden et al., 1979). Foh et al 1979, predicted that hydrogen is unlikely to react with microbes present in the reservoir at temperatures $< 80^\circ\text{C}$, which has been supported by the fact that no hydrogen losses were reported during 18 years of operation.

However, town gas was stored in a 200–250 m deep sandstone aquifer in Ketzin (40 km west of Berlin, Germany) in which total gas losses were circa $2 \times 10^8 \text{ m}^3$ with a working gas volume of circa $1.3 \times 10^8 \text{ m}^3$ (between 1964 and 1985); the losses have been attributed to chemical and microbiological processes in the reservoir (Stolten et al, 2016). At Lobodice in the Czech Republic, a 54% hydrogen town gas was stored in a saline aquifer with depths of 400–500 m. Following seven months of storage and a decrease in the reservoir pressure, losses of hydrogen (from 54% to 37%) were also attributed to methanogenic microorganisms present in the reservoir (Stolten et al, 2016) coincidental with an increase in CH_4 and N_2 . The carbon isotope analyses of the increased CH_4 indicated microbial origin.

In the Underground Sun.Storage project (Sun.Storage Final Report, 2017), a significant shift in the microbial consortium was identified and it was concluded that 3% of the injected hydrogen was converted to CH_4 by methanogens.

Hydrogen Fingering

Fingering is a condition whereby the interface of two fluids, such as gas and water, bypasses sections of reservoir as it moves along, creating an uneven, or fingered, profile.

Similar to natural gas, injecting hydrogen into a depleted reservoir or aquifer causes a gas-water displacement and the difference in viscosity between the gas and water can lead to the gas fingering.

Diffusivity

Hydrogen has high diffusivity and can therefore exhibit enhanced migration through fractures and across faults in the caprock, potentially leading to leakage. The low solubility of hydrogen in water may minimize the losses of hydrogen due to diffusion as the water saturated caprock will act as permeability barrier to hydrogen (Panfilov, 2016).

From numerical simulation studies, hydrogen losses by diffusion through the caprock are estimated between 2–6% (Carden and Paterson, 1979), (Pichler, 2013) and (Panfilov, 2016). Results from the SUN.PROJECT Report, 2017 suggest a total loss of 18%, but which include losses due to diffusion and solubility.

Main Risks and Challenges of UHS

Leakage

The sealing potential of a caprock to hydrogen gas depends on the caprock ability to withstand mechanical and hydraulic gas infiltration. Leakage is prevented by the presence of a caprock with low permeability and a high capillary entry pressure above the reservoir, and a trap structure that will prevent the hydrogen from migrating laterally. For salt caverns the risk of hydrogen leakage is low as the hydrogen gas is contained by salt that is effectively impermeable. In the case of depleted gas reservoirs, the caprock and trap mechanism keeping the natural gas in place may not necessary work for hydrogen trapping under the same conditions. Similarly for aquifers, a caprock that seals water in place is not necessarily guaranteed to seal for hydrogen.

To date, pure hydrogen has not been stored in porous rocks and therefore, it is not fully known if this is technically possible. There will dependency on the type of UHS reservoir, for example, if it is fractured or not, the degree of faulting, and fault-sealing properties, as well as porosity and permeability properties of the reservoir and caprock. Possible “micro fractures” in the caprock could provide a means for the hydrogen to escape from the storage reservoir. Experience from town gas shows no evidence of leakage from the reservoir via the caprock but rather losses due to microbial growth (Stolten et al, 2016).

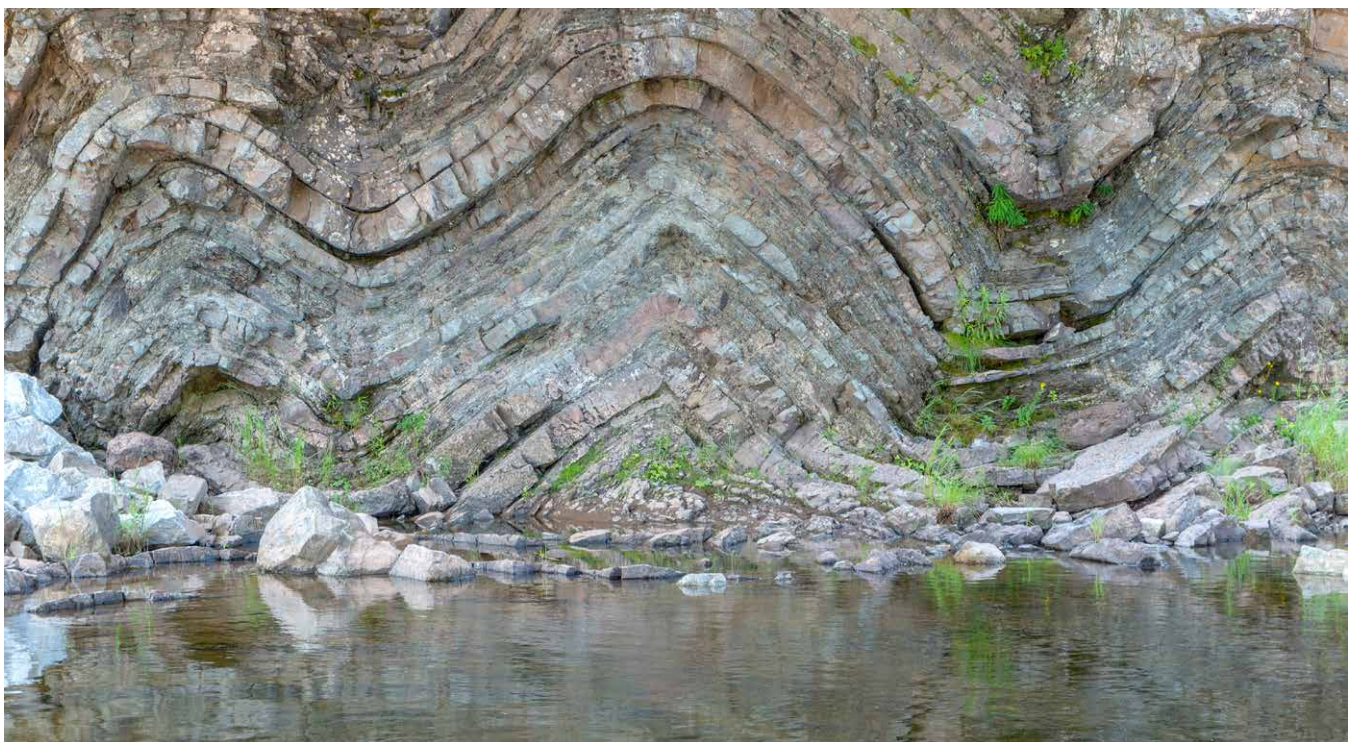
Use of Cushion Gas

A minimum pressure needs to be maintained in salt caverns, depleted gas reservoirs and aquifers if used as a UHS reservoir. This is necessary to prevent and limit any geomechanical issues including induced seismic activity and the risk of fracturing the formation, specifically if cyclic UHS is being considered. The geotechnical properties of the storage reservoir and fracture pressure gradients need to be assessed prior to the selection of storage reservoirs.

Cushion gas is also necessary to prevent brine incursion into the reservoir and to maintain the required reservoir pressure ensuring deliverability of the hydrogen. Cushion gas is usually between 30-70% of the total storage volume and could be any appropriate gas or hydrogen, although the use of hydrogen would be more costly. The mixing of cushion gas and hydrogen leads to the gas composition changing the gas properties of each gas, which introduces uncertainty to the injection and production behaviour and contaminates the hydrogen which may require processing prior to use.

Some recommendations for cushion gas include nitrogen, methane or CO₂ as well as hydrogen. Nitrogen is seen to be a more efficient cushion gas as it is denser than methane, which would allow a more efficient displacement of water relative to methane but would constrain hydrogen volumes and production to 50-80% (Pfeiffer et al., 2015). CO₂ could also be used as a cushion gas due its relative high density (Oldenburg et al., 2003), but it is also reactive with hydrogen, which would cause hydrogen loss. The cushion gas is considered as another cost which is part to the CAPEX costs (see Table 3).

Salt caverns are expected to need less cushion gas than depleted reservoirs and aquifers.

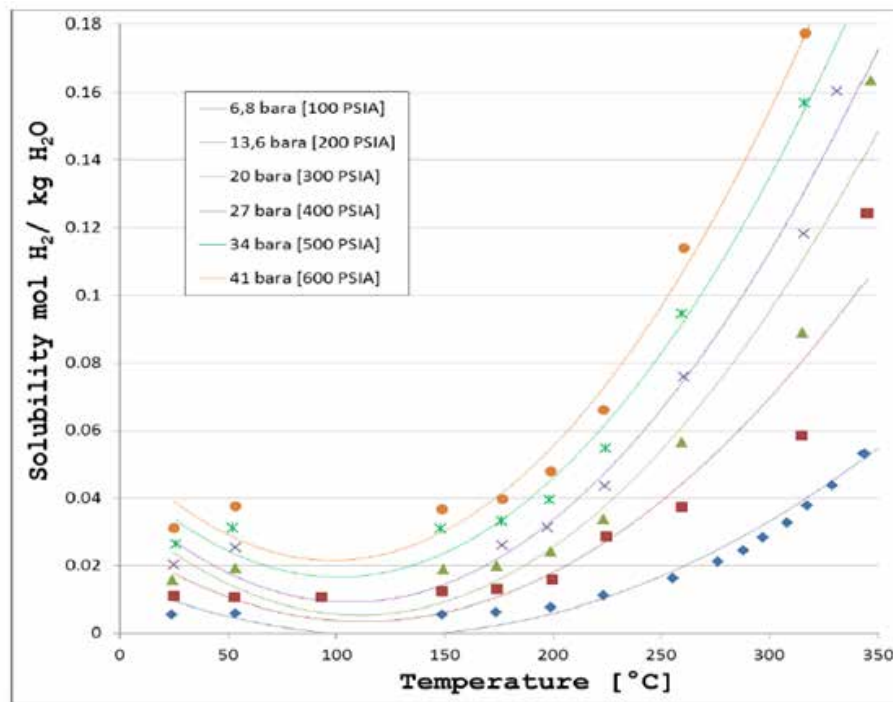


Main Risks and Challenges of UHS

Solubility in Water

During injection and storage of hydrogen in porous media, hydrogen will be in contact with formation brine, which leads to the loss of hydrogen, reduction in the pH value and reduces the RedOx potential of the system (Lassin et al., 2011). The solubility of hydrogen in water varies as a function of temperature (Figure 6). It can be seen that solubility of hydrogen in water decreases with the increase of temperature, until an inflection point where increasing temperature increases the solubility of gases in water. For temperatures ranging between -6°C to 100°C, the solubility of hydrogen in water decreases with increasing temperature (Pray et al., 1950).

Figure 6: The Solubility of Hydrogen in Water as a Function of Temperature



Source: Pray et al., 1950

The solubility of hydrogen in water is proportionate to the pressure as predicted by Henry's law (Pray et al., 1950). However Pray et al., did not include a correction factor for salinity. Lassin et al., 2011, suggested that any dissolved species such as salts reduces the gas solubility in fluids and also predicted a very small solubility of hydrogen in water.

Crozier et al., 1996, stated that the hydrogen solubility in water is about 37 mol/m³ at 30°C and 50 bar and 80 mol/m³ at 25°C and 100 bar. From a successful field study test in Austria using 10% hydrogen in the injected gas, the hydrogen loss due to dissolution was estimated at 0.88-2% (Carden and Paterson 1979) & (Pichler, 2013). Results from the Sun project Report, 2017 suggest a total loss of 18% by diffusion and solubility.

More work needs to be done in order to understand if the solubility of hydrogen in water can be ignored.

Recovery Production Efficiency

The production efficiency is the ratio of produced hydrogen over the injected hydrogen in a reservoir or salt cavern, which indicates the amount of hydrogen return. The production efficiency is a function of hydrogen diffusion leakage and solubility in water in addition to geochemical and biochemical reactions.

Experiments were carried out on a real reservoir as part of the SUN Project in which a final volume balance showed, that 82% of the injected hydrogen could be recovered. The other 18% are accounted for by, diffusion, solubility and conversion (Sun.Project Final Report 2017).

Conclusion

Hydrogen is seen as an energy carrier that can be stored readily in large quantities and can address the problem of energy supply fluctuations associated with renewable energies such as solar, wind and hydro-power by conversion of excess/curtailed electricity into hydrogen. Hydrogen could provide large energy availability balancing, in the range of GWh or more and could provide diurnal or seasonal balancing, to provide additional energy supplies when demand is high.

There are three types of hydrogen storage facilities currently used or in consideration, these include; salt caverns, depleted hydrocarbon reservoirs as well as aquifers. Salt caverns have been successfully used for hydrogen storage since the 1970's but are costly to create and usually have limited volumes. There are pilot projects related to hydrogen storage in depleted gas reservoirs, but no commercial development as such, hence such developments are more long-term expectations.

The main challenges of hydrogen storage include costs and hydrogen losses when re-produced from the storage facility. Hydrogen losses are mainly attributed to geochemical and biochemical reactions as well as leakage and diffusivity, hydrogen fingering and the need for cushion gas. Experience from historical town gas (mix of CH₄, CO₂ and hydrogen) usage and storage in aquifers show the main challenges are losses due microbial biochemical losses.

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