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Underground hydrogen storage: a review

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Abstract

Large-scale underground storage of hydrogen gas is expected to play a key role in the energy transition and in near future renewable energy systems. Despite this potential, experience in underground hydrogen storage remains limited. This work critically reviews the most important elements of this important technology, including hydrogen properties and their importance for subsurface operations, sources for hydrogen, and historical hydrogen storage operations, to set the state of the art. The cyclical nature of hydrogen storage operations will produce pressure and stress changes within the reservoir that could affect the integrity of the well, the reservoir, the caprock and the entire subsurface storage complex. To minimize geomechanical leakage risks and optimize the storage operation it is crucial to understand the pressure and stress history of the storage site, to optimize well locations to manage pressure, and to identify the reservoir-specific cushion gas-to-working gas ratio. Finally, the major scientific and operational challenges required to ensure the safe and efficient deployment of underground hydrogen storage at a large scale are here outlined.

1 Introduction

Hydrogen as a future low-carbon energy carrier is currently gaining momentum on a global scale. There is an increasing recognition of the versatile role hydrogen as a clean energy solution can play for the decarbonization of transportation, power, heating and fuel-intensive industries to enable large-scale greenhouse gas emissions reduction (Hanley et al., 2018; McPherson et al., 2018; UNIDO, 2018). Of particular interest is the role hydrogen can play in supporting the renewable energy systems such as solar, wind and hydro electrical power generation, where the supply of renewable energy is subject to daily up to seasonal fluctuating events (e.g., diurnal cycles, weather changes, seasonal changes in wind force). As an example, in the spring of 2020 California curtailed up to 300,000 MWh of excess renewable energy per month (EIA, 2021), but faced rolling blackouts in August because the grid was short on energy as there was no mechanism in place to store the excess energy for deployment during periods of high demand (California ISO, 2021). In addition, daily grid balancing involves almost always increasing output from fossil fuel power plants which increases fuel consumption and in turn emissions. The generation of hydrogen using excess or curtailed renewable energy can help alleviate the drawbacks of a renewable energy system as it can be distributed directly to the end-user or stored as required for grid balancing at all scales. For seasonal storage of renewable energy, large-scale storage of hydrogen is one strategy to help ensure that energy supply can always meet the energy demand.

Hydrogen has the highest gravimetric energy density of all known substances (120 kJ/g), but the lowest atomic mass of any substance (1.00784 u) and as such has a relatively low volumetric energy density (Tab. 1). To increase the volumetric energy density, hydrogen storage as liquid chemical molecules such as liquid organic hydrogen carriers (LHOC) or directly usable hydrogen carriers such as ammonia or methanol are being considered (Abdin et al., 2021). However, liquifying hydrogen comes with increased costs and is unlikely to be economic at the capacities required at the inter-seasonal energy storage scale(Yin and Ju, 2020). Because of its small size, low molecular weight, low viscosity, low density, and positive buoyancy over -251°C, hydrogen gas is highly diffusive. Hydrogen has a low solubility in water, however work by Chabab et al (2020) have shown this solubility increases with increasing pressure (Fig. 1). Figure 1 shows the variation of density, viscosity, solubility in water and energy density with storage depth for a hypothetical reservoir with temperature and pressure linearly increasing according to a geothermal gradient of 33°C/km and a hydrostatic gradient of 10 kPa/m.

Surface-based hydrogen storage facilities such as pipelines and tanks have limited storage and discharge capacities (MW h, hours-days), subsurface hydrogen storage in salt-caverns and porous media (such as depleted oil and gas fields, saline aquifers) has the potential to supply energy on a much larger scale (GW h/TW h; weeks-seasons (Fig. 2, Mouli-Castillo et al., (2021)).

Experience to date with hydrogen storage in geological media is limited to four salt cavern projects at Teesside (United Kingdom) and the US Gulf Coast, and to three aquifer storage projects for town gas (50% hydrogen) storage in the 1960's and 1970's (Panfilov, 2016). However, there is extensive experience in the storage of other energy and waste fluids (e.g. oil, natural gas, CO_2, compressed air or thermal water) that can provide useful learnings and workflows to ensure the safe and efficient hydrogen storage operations (Schultz et al., 2022, this publication). Different geological options have been proposed for the storage of hydrogen, including salt caverns (Böttcher et al., 2017; Caglayan et al., 2020; Ozarslan, 2012; Tarkowski and Czapowski, 2018), saline aquifers (Heinemann et al., 2018; Niklas Heinemann et al., 2021; Luboń and Tarkowski, 2020; Sainz-Garcia et al., 2017), or depleted hydrocarbon reservoirs (Amid et al., 2016; Lemieux et al., 2019) (Fig. 3). The physical and chemical characteristics of each storage type are different and so are the parameters such as working gas capacity, cushion gas requirements, maintenance cost, site preparation, monitoring, production rate and leakage risks, which will need to be considered for commercial development. Here, we briefly review the different origins of hydrogen, experiences with geological hydrogen storage, the challenges originating from the cyclic nature of hydrogen storage, and identify the major obstacles and opportunities for subsurface hydrogen storage in the future.

2 Sources of hydrogen

Hydrogen can be produced through a number of different methods, each with different efficiencies, costs and carbon intensity. The most commonly deployed methods include producing hydrogen from fossil fuels via steam methane (SMR), autothermal (ATR) reformation, partial oxidation (POX), coal gasification, or pyrolysis, either with (around 1% of global hydrogen production from fossil fuels) or without carbon capture and storage (CCS) and using electrolysis of water (Nikolaidis and Poullikkas, 2017). There are several new low carbon hydrogen production technologies in development including: microbes using light energy to produce hydrogen from water as part of their metabolic processes (Akhlaghi and Najafpour-Darzi, 2020), fermentation of biomass to produce hydrogen (Łukajtis et al., 2018), pyrolysis or gasification of biomass (Cao et al., 2020), photoelectrochemical water splitting (Shiva Kumar and Himabindu, 2019), solar thermal water splitting (Safari and Dincer, 2020), electrolysis powered by nuclear energy and methane pyrolysis to produce hydrogen and solid carbon. There have been several colour prefixes applied to hydrogen depending on the production processes (Tab. 2, (Newborough and Cooley, 2020)). A preferable differentiation for the hydrogen production processes is to consider the life cycle greenhouse gas emissions related to both the production process itself and other related processes (e.g., mining of fuel), into account (Fig. 4, (Parkinson et al., 2019)). Hydrogen production from fossil fuels results in relatively high carbon emissions, even when the emissions are captured (Fig. 4, 1.09-10.35 kg CO_2e / kg H_2, Parkinson et al. (2019)). Solar and wind sourced hydrogen generally have a low carbon footprint (0.52-2.5 kg CO_2e / kg H_2, Parkinson et al. (2019)), while negative emissions can be achieved by using biomass to produce hydrogen when emissions are captured (-11.66 - -17.50 kg CO_2e / kg H_2, Parkinson et al. (2019)).

An additional consideration in hydrogen generation is reaching cost parity with fossil fuels, which are expected to become more expensive due to carbon penalties and / or mitigation costs. The vast majority of hydrogen produced today (96%) originates from SMR without CCS, (IEA, 2021) at a cost of around \$1.8/kg (assuming 2020 natural gas prices), with some blue hydrogen projects such as Quest in Canada, with a cost of \$2-3/kg including CCS. Only 4% of hydrogen is from green hydrogen, with costs ranging from \$3 – \$6.66/kg (European Commission, 2020; Hydrogen Council, 2020). For comparison, in 2021 natural gas prices at the Henry Hub (Louisiana, US) ranged from \$0.12-0.3/kg, which translates to \$0.26-0.65 per 120 MJ, the energy content of 1 kg of hydrogen. Many climate neutral future energy scenarios such as the Hydrogen Roadmap Europe (FCH, 2019) focus on hydrogen generated from wind and solar (“green”) and from natural gas (methane) steam reforming with carbon capture and storage (“blue”). Cost trends indicate that the cost of green hydrogen production will become cheaper than natural gas-generated hydrogen over the next 10 years as the costs of wind, solar, hydro and other non-fossil energy sources and electrolysers fall with increasing deployment and that green hydrogen costs could fall below blue hydrogen costs in some locations by 2030 (BloombergNEF, 2021; Energy Transitions Commission, 2021). The IEA anticipates that with increased deployment and technology advances green hydrogen costs should fall to \$1.3/kg by 2030. If natural gas prices were to increase, green hydrogen would become competitive much earlier.

3 Historical milestones and current state of the technology

In spite of the extensive experience in natural gas storage and other sorts of subsurface storage activities, the amount of underground hydrogen experiences, particularly of high purity (>90%) hydrogen, is sparse (Tab. 3).

Storage in salt caverns

Underground salt cavern storage has been identified as one of the most promising geological storage technologies for hydrogen, due to their technological maturity, fast cycling flexibility and large volume storage capacity (Energy Technologies Institute, 2015). Salt caverns are cavities solution mined within suitable (halite-dominated) salt formations using fresh water to dissolve the salt rock (Li et al., 2019). The properties of the surrounding salt include low permeability and such high sealing capability, inert chemical behaviour with respect to hydrogen and favourable mechanical properties that provide the ability to accommodate repeated withdrawal and extraction cycles, (Evans et al., 2021) allow for the secure storage of fluids over long periods of time (Lux, 2009). Salt caverns are widely deployed for the storage of energy fluids, such as oil, natural gas, or compressed air (Crotogino et al., 2001; Le Fevre, 2013; Zhang et al., 2017). Practical experience in hydrogen storage in salt caverns is limited to three commercial storage operations, one in the UK and three in the US, that have been providing hydrogen for the chemical industry since the 1970s and one salt cavern in Kiel Germany that stored town gas with 62% hydrogen in the 1960's and 1970's (Crotogino, 2016; Panfilov, 2016). The experience from these operations in both bedded and domal salt highlights that hydrogen can be securely stored and recovered from salt caverns over many decades (Tarkowski, 2019). Cavern storage of hydrogen has seen increased interest in the last decade, with new operations being developed in the UK (SSE thermal and Equinor, Aldbrough), US (ACES, Utah), Germany (HYPOS, Bad Lauchstadt), Netherlands (Gasunie, Veendam), and France (HyGeo, Nouvelle-Aquitane and HyPSTER/Stopil_H2, Etrez) (Le Duigou et al., 2017). These projects are considering both new caverns and importantly the repurposing of existing caverns, indicating salt caverns offer rapid storage capacity through conversion of existing assets to hydrogen storage. Multiple studies have analysed the potential of salt caverns for hydrogen storage in different areas of the world in the last decade (e.g. Bai et al., 2014; Caglayan et al., 2020; Iordache et al., 2014; Lemieux et al., 2020; Liu et al., 2020; Michalski et al., 2017; Ozarslan, 2012; Tarkowski and Czapowski, 2018). The above demonstrates that this established gas storage technology will be well suited to providing a large-scale storage option for hydrogen.

Storage in porous rocks (saline aquifers and gas fields)

A combination of porous rocks overlain by impermeable mudstones or evaporites which form a sealing caprock, create conditions deep in the subsurface that are perfectly suited to trap and contain gasses such as methane, natural gas and CO_2, along with the formation brine. These formations which have been proven to securely contain gasses over millennia, could be the ideal candidate to provide inter-seasonal, TWh scale hydrogen storage, and include both saline aquifers and gas fields. Important experience of hydrogen storage in saline aquifers was gained during aquifer storage of town gas in the 1950's through to the 1970's. Town gas is produced from coal gasification, where oxygen and steam oxidize coal to produce a gaseous mixture of ~50–60% hydrogen with ~30% CH_4, and ~20% CO_2 and CO. Aquifer storage of town gas occurred in France (Beynes), Czech Republic (Lobodice) and Germany (Engelborstel, Bad Lauchstaedt). During decades of commercial operation, there were no reports of containment failures from these town gas storage sites; however, some changes in the stored gas composition are thought to have occurred as a result of bio-geo-chemical reactions within the storage reservoirs (Buzek et al., 1994; Kruck and Crotogino, 2013; Panfilov, 2016). Notable is the generation of H_2S, likely due to abiotic pyrite reduction, as well as methane generation from methanogenic bacteria within the reservoir. While hydrogen storage within porous media has recently seen growing attention, the only two pilot studies that have injected and recovered hydrogen to date are green methane projects in Austria and Argentina (Table 2, (RAG, 2019)). These storage projects inject a mixture of natural gas with 10% hydrogen produced from renewable energy into sandstone reservoirs for green methane production by coupled CO_2/H_2 injection into reservoirs hosting methanogenic bacteria.

Other subsurface storage options

Two more geological options, engineered rock caverns and abandoned mine shafts, have been proposed as options of the storage of hydrogen. Engineered rock caverns involve the excavation of cavities in extremely tight and stable hard rock formations (Crotogino, 2016). Matos et al. (2019) provide more details about the characteristics of the most suitable lithologies for hosting engineered caverns.

In certain circumstances, abandoned mine shafts could be repurposed for the storage of energy fluids, with the inclusion of engineered barriers, such as cement or resin at the rock boundary. These two options have raised much less interest than salt caverns or saline formations, mainly due to the technical challenges associated and the suitability requirements of the host rocks to accommodate the storage facility, however the Swedish HyBRIT project is developing a 100m^3 hard rock cavern for hydrogen to be used in the decarbonization of steel making.

4 Challenges of cyclical operation

To balance the discrepancy between supply and demand within the energy system, the hydrogen obtained during energy surplus periods will need to be stored until the energy demand is greater than the energy production. Storage in both salt caverns and porous rocks, can deliver the injection and withdrawal rates to provide a fast-ramping, flexible and seasonal-scale energy resource. This cyclic injection and withdrawal of hydrogen will alter the pressure and temperature around the well and far into the reservoir, resulting in changes to the stress equilibrium that could impact the integrity of the well, the reservoir, the caprock and the entire storage complex. Potential failure mechanisms include induced failure of the caprock/overburden, fault reactivation, and well sealing failure. These geomechanical aspects and their response to hydrogen storage must be evaluated in order to minimize leakage risks and assure the integrity of hydrogen storage.

For salt caverns, that are likely to experience faster cycling rates than porous stores, the integrity and stability of a cavern is related to geomechanical and geological factors of the salt and the cavern shape as well as the pressure of the stored gas (Ozarslan, 2012). During the operational lifespan of the salt cavern, it will experience complex mechanical, thermal, and hydraulic processes. The stress state of the cavern is dependent on depth, geological stress state, internal gas pressure, and injection/ withdrawal rates (Ramesh Kumar et al., 2021). It is also controlled by the creep behaviour of salt and cavern geometry (Spiers et al., 1990), which are controlled by heterogeneity which in turn influences stress redistribution during loading and unloading cycles (Asgari et al., 2020). Salt has a number or attributes that equip it to deal with these cycles, it has a near-isotropic stress state that provides resistance to hydrofracturing, and it is ductile, behaving in a viscoplastic manner when subject to stresses so has the ability to heal any induced cracks and faults (Urai et al., 2019). However, geological heterogeneities such as non-salt (halite) interbeds can compromise permeability and alter steady-state creep creating strain partitioning (Taheri et al., 2020; Tarkowski, 2019). These must be kept to a minimum during site selection. It is imperative that the stress changes during injection and withdrawal cycles do not cross the dilatancy boundary, which separates dilatancy behaviour from compressibility behaviour resulting in increased permeability, reduced rock strength and potential failure leading to loss of cavern integrity (Hunsche and Hampel, 1999). The temperature of the gas in the cavern fluctuates in response to thermodynamic and heat exchange processes which are generally transmitted to the immediate vicinity of the cavern wall. As salt has a relatively low coefficient of thermal expansion, small thermal changes in the cavern wall can induce relatively large stress changes, effecting the integrity of the cavern (Leister et al., 2018). The internal cavern pressure should be limited to ensure that the stresses of the cavern remain below the dilatancy boundary. This will mitigate the potential of micro-cracking and damage propagation occurring which may result in fatigue failure (Khaledi et al., 2016). Thermal and geomechanical stresses may also affect the wellbore/cement/salt formation interface as creep deformation or interbed slip could lead to casing collapse or loss of well sealing so this must be monitored. Cavern geometry is an important consideration in reducing roof collapse, where a slim cavern, with a large depth:radius ratio and a capsule (ovoidal) shape preferable. With increasing burial depths the pressure difference within a cavern can be increased which allows for a large storage capacity and increased hydrogen energy density (with increasing temperatures and pressures) (InSPEE, 2016).

For hydrogen storage in porous media the geomechanical aspects are similar in many respects to those of natural gas storage, and include (annual/seasonal) cyclical pressure changes, short- and long-term chemical interaction of hydrogen with intact rock and faults, and stress-strain-sorption on mechanical and transport behaviour (Heinemann et al., 2021a). For storage site integrity it is crucial to keep pressure increases below the capillary entry pressure and fracture initiation pressures of cap and fault rocks (Miocic et al., 2019; Vavra et al., 1992). Hydrogen-rock-brine wettability and the role of capillary sealing in geological hydrogen storage has been studied extensively in the past years (Ali et al., 2022; Hashemi et al., 2022, 2021). Results of contact angle measurements indicate that conditions remain water-wet at storage conditions, but organic content on mineral surfaces may result in close to hydrogen wet condition (Al-Mukainah et al., 2022; Iglauer et al., 2021), highlighting that the composition of potential caprocks needs to be well known to ensure safe storage of hydrogen. In cases where hydrogen is stored in depleted gas reservoirs, reservoir pressures should not be increased above initial pre-production pressures as this may induce geomechanical failure of the reservoir (Tenthorey et al., 2013). Note that due to the lower density of H_2 compared to CH_4, the volume of H_2 that will be able to be safely stored in depleted gas reservoirs are lower than the original gas in place. Pressure changes are not limited to the immediate storage formation but can also impact wider regions (Birkholzer et al., 2009), highlighting that monitoring of pressure may be needed. A storage site connected to an open large-scale aquifer will allow for more pressure dissipation, but local pressure increase during injection via a single or multiple injection wells has to be managed effectively in order to inject the desired volume of hydrogen within the given time (usually a few months). The hydrogen production cycle is generally effective due to the low density and the low viscosity of the fluid. However, computer modelling indicates that a massive production of hydrogen from a saline aquifer could lead to a low-pressure zone around the production well due to the inertia of the brine to dissipate the pressure drop during production (Heinemann et al., 2021b). This pressure drop could then lead to the curtailment of targeted production rates. If the site is compartmentalized, and no or limited pressure communication exists between the actual site and the surrounding formation, the storage capacity is very low if no further measures are applied. However, if these compartmentalized structures are depleted gas fields, such as in the Southern North Sea, their post-production pressure and water saturation can be relatively low. Injected gas would fill up a low-pressure zone and capacity is mainly dependent on gas compression. Optimization strategies to ensure large-scale hydrogen storage operations in porous media are still rare but recent research shows that a carefully designed cushion gas to working gas ratio as well as a detailed site selection procedure can increase the efficiency of the operation (Heinemann et al., 2021b).

The cyclicity of hydrogen storage will lead to stress fluctuations within the reservoir and nearby faults which may cause reservoir compaction, subsidence, or fault reactivation (Hettema et al., 2002; Nagel, 2001). Currently, knowledge about the response of porous reservoirs and faults to cyclic stresses relevant to hydrogen storage conditions is limited, however lessons may be learned from other subsurface utilization operations including natural gas storage, nuclear waste storage, unconventional hydrocarbon and geothermal production. Hydrogen storage in porous media may lead to dissolution-precipitation reactions which may alter load-bearing grains and cements (Heinemann et al., 2021a), which subsequently may lead to increased elastic and inelastic deformation of the reservoir (Peng et al., 2020; Pijnenburg et al., 2019). Additionally, swelling or drying-out of clays within the reservoir and caprock may occur during hydrogen storage operations, which may lead to leakage pathways (Wentinck and Busch, 2017).

To reduce geomechanical related leakage there are a number of mitigation steps that can be implemented at the early site assessment stages, particularly for storage in depleted gas fields, including (a) accurate determination of upper injection pressure limits, (b) optimise well locations to manage pressure, (c) undertake a detailed assessment of the historical data on reservoir pressure, stimulation procedures and energy-related production management history, (d) ensure new well drilling designs mitigate development of new fractures and importantly (e) undertake wellbore integrity testing of all existing wells.

5 Major obstacles and future opportunities

While the available experience with hydrogen storage is sparse, this technology is comparable with natural gas storage in terms of operation and the experience gained in underground natural gas storage will be an asset for the development of research, pilot and industrial scale hydrogen storage projects. Decade-long experience in the storage of hydrogen-rich gas mixtures enables the reduction of the risk of biological and geochemical reactivity in the subsurface due to hydrogen injection through site selection and mitigation strategies. Laboratory based research efforts made in the last two decades on geological CO_2 storage can inspire the much-needed fundamental research on hydrogen, with experimental equipment and workflows adjusted for hydrogen flow and reactivity. Finally, the basic concepts and technical challenges of seasonal hydrogen injection, storage and production are well known from decades of successful natural gas storage operations at all scales, and as such, important information from operational procedures, site management and safety protocols can be used and reworked for hydrogen according to the latest advantages in hydrogen research and development.

For large-scale seasonal hydrogen storage (i.e. in the TWh range), for example to supply energy to domestic homes during the winter season, subsurface storage in saline aquifers and depleted gas fields represents an opportunity which has not been fully explored yet (Heinemann et al., 2021a). Hydrogen storage plays, featuring a suitable reservoir formation, a caprock that will retain the stored hydrogen and a trap structure which allows efficient reproduction, could provide a geographically more flexible solution for large-scale energy storage than salt caverns (Zivar et al., 2021). Due to the low density of hydrogen, large volumes of injected hydrogen will displace the in-situ pore fluids, usually brine and/or residual hydrocarbons, and lead to an increase in formation pressure, which, if not managed properly, can compromise the integrity of the storage site. This highlights the need for pressure monitoring within the storage reservoir but also in surrounding aquifers/units to identify pressure issues early and allow for mitigations such as the production of the stored hydrogen.

Recent research on alternative cushion gas, such as nitrogen, methane or CO_2, to either decrease costs or store additional greenhouse gas, are another promising prospect (e.g. Dussaud, 1989; Oldenburg and Pan, 2013; Pfeiffer and Bauer, 2015). The use of these alternative cushion gases can greatly reduce the capital expenditure in saline aquifer storage, where about 60% of the gas in place might not be recoverable (Misra et al., 1988). Additionally, all considered cushion gases alternatives can reduce the density contrast between the low-density hydrogen and the dense formation water, potentially reducing unstable displacement. However, the risk of gas mixing is a concern, especially when the degree of mixing between the working gas and cushion gas reaches levels which lead to production quality reductions, so that topside component separation would be required to yield the required hydrogen purity (Pfeiffer and Bauer, 2015). However, depending on the uses of produced hydrogen topside processing may be needed regardless of the level of mixing within the reservoir.

There are important differences between hydrogen and other subsurface fluids, such as natural gas or CO_2 (Hassanpouryouzband et al., 2020). In porous media storage, hydrogen could potentially undergo geochemical reactions with the formation rocks and fluids, which could improve or decrease reservoir quality but recent research has not confirmed this risk (Hassanpouryouzband et al., 2021). Additionally, for hydrogen storage, the impact of these reactions on the storage operation requires validation as mineral reactions often occur over long periods and may not be relevant for hydrogen storage operations, where storage sites are anticipated to be in operation for less than 50 years. Added to this, hydrogen has low solubility in water (Tab. 1) and as such will not drive significant mineral dissolution, the risk of which is reduced further by progressive dehydration of water saturation over subsequent injection and production cycles.

Hydrogen is an electron donor for many subsurface microbial processes, so the elevation of hydrogen concentrations in a storage site may stimulate the growth of hydrogen-oxidising microorganisms which are expected to have some impact on porous media storage. Their growth could lead to the consumption of hydrogen, production of methane, biofilm growth plugging fluid flow pathways, mineral precipitation and hydrogen sulphide production which could lead to corrosion of metal infrastructure. Further research has to clarify to what degree their presence impacts storage, how it can be mitigated and if their occurrence and activity can be mitigated by a critical site selection process, as proposed by (Thaysen et al., 2020).

For the actual storage operation, hydrogen's fluid properties have to be taken into account. Buoyancy pressures will be higher compared to CO_2 storage and natural gas storage, and hydrogen's low density and viscosity lead to an unfavourable phase mobility ratio compared to brine which results in a higher tendency for unstable, inefficient displacement, including gravity overriding and viscous fingering (Feldmann et al., 2016; Paterson, 1983). Relative permeability and capillary pressure measurements for the hydrogen / brine system are still rare. Yekta et al. (2018) suggest that interfacial tension and contact angle are almost constant in the hydrogen-water system under storage conditions. However, the authors' conclusion is based on only two experiments and there is a requirement for more measurements to obtain reliable relative permeability and capillary pressure curves for hydrogen storage. Linked to this, it has been demonstrated that CO_2 can change wettability towards intermediate at pressures over 10 MPa particularly in micas (Espinoza and Santamarina, 2010) and that pressure and temperature have different effects on wettability for CO_2 and CH_4 (Pan et al., 2019), leading to depth constraints regarding storage capacity (Miocic et al., 2019). Research is needed to identify if hydrogen influences wettability and what could be the potential impact of cyclic injection and extraction on wettability. The diffusion mobility of hydrogen is calculated to be higher than other gases due to its smaller molecular size, however Amid et al. (2016) estimated hydrogen losses from a storage site through diffusion at less than 0.1%.

Ultimately, a successful hydrogen storage operation has to be conducted in a secure and transparent manner. Uncertainties related to potential leakage as well as other risks have to be investigated and quantified, and monitoring programs, designed for storage operations of other fluids, require investigation and calibration.

Salt caverns, depleted gas fields and saline aquifers provide a wide range of hydrogen storage scales and deliverability, capable of meeting both our seasonal and daily energy demands. Salt caverns provide shorter term delivery flexibility along with an established cyclic operational system with reduced risk of producing contaminated hydrogen. They are however geographically constrained and cannot provide the scale of hydrogen storage capacity required for the necessary deep decarbonisation of the global energy system. Depleted gas fields and saline aquifers have the potential to provide many 1000's of TWh of hydrogen storage capacity that are capable of providing seasonal energy storage.

Recent work by Mouli-Castillo et al. (2021) and Scafidi et al. (2021) has demonstrated that only one offshore depleted gas field has enough static capacity to store enough hydrogen to meet the UK domestic heating demand, highlighting that there will be limited competition for subsurface space by alternative low-carbon applications, such as CAES (compressed air energy storage) or CO_2 storage. This is particularly relevant if hydrogen produced from steam reformation of methane along with carbon capture and storage (blue hydrogen) in the transitional phase is used while the required technology advances and cost reductions for the production of hydrogen from renewable energy and electrolysis (green hydrogen) are achieved. However, these calculations are based on static volumetric estimations and firstly do not accurately account for cushion gas requirement, and secondly do not provide information about achievable injection and production rates. Gas fields have been emptied over decades whereas only a short production window of a few months is available for hydrogen production. Research on how to use these depleted fields most effectively, or alternatively saline aquifer trap structures, is still work in progress.

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CRediT statement+

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Figure 1. Hydrogen properties versus reservoir depth. (a) Hydrogen density, with scaled circles representing hydrogen energy density for one cubic meter reservoir rock with a porosity of 20% (b) viscosity and (c) solubility in pure water with depth. Based on a geothermal gradient of 0.033°C per m and a hydrostatic gradient of 10 kPa/m. Note that salinity of the reservoir brine influences solubility.

Figure 2. Geological storage options of hydrogen with their corresponding storage power and discharge time. Ranges for each option reflect variations in storage site size and operational management (e.g., number of production wells).

Figure 3 Underground hydrogen storage options include storage in depleted hydrocarbon fields, saline aquifers, and salt caverns. Geological storage of by-product CO2 will also be required depending on the source of the hydrogen. Adapted from Griffioen et al. (2014).

Figure 4: Life cycle emissions in kg CO2 equivalents per kg of H2 produced by the various hydrogen production processes. Bars indicate ranges given in the literature, white lines indicate the median value, which is also given as a number. Data from Parkinson et al. (2019), which is based on an extensive review of case studies and models reported in the literature. CG = coal gasification, CCS = carbon capture and storage, NG = natural gas, SMR/ATR = steam methane reformation / autothermal reformation, BM = biomass.

Table Caption

Table 1. Physical properties of hydrogen (NIST, 2022). [1]NTP (Normal temperature and pressure): 293 K, 101325 Pa.

Table 2. Table showing different hydrogen production processes and colours used to describe them. The feedstock is the substance from which hydrogen is extracted, the energy source is how the energy is produced to extract hydrogen from the feedstock and the production method is the process used to extract hydrogen from the feedstock, powered by the energy source. The primary colour is the most commonly referred to colour when describing hydrogen extracted in that process. Alternative colours refers to less common colours used when describing hydrogen extracted in that process. See text for further details. *Orange refers specifically to hydrogen produced from renewable energy in the North Netherlands.

Table 3. Historical record of underground hydrogen storage projects.

Parameter		**Hydrogen**
Molecular weight (u) Molar mass(g/mol)		1.09784 2.01594
Triple point	Temperature (K) Pressure (kPa)	13.8 7.041
Critical point:	Temperature (K) Pressure (MPa)	32.938 1.2858
Density @ NTP[1] (kg/m^3)		0.08990
Viscosity @ NTP[1] (μPoise)		89.48
Solubility in water @ NTP[1] (g gas per kg water)		0.0016
Diffusion coefficient @ NTP[1] (m^2/s)		0.000061
Diffusion velocity @ NTP[1] (m/s)		<0.02
Buoyant velocity (m/s)		1.2 - 9
Specific heat constant of gas @NTP[1] (kJ/ (kg K)		14.85
Thermal Conductivity of gas @ NTP[1] (W/m K)		0.187
Flammability limits in air (vol%)		4 – 75
Explosion limits in air (vol%)		15-59
Auto ignition temperature in air (K)		793 – 1023 (858)

Table 1

Bituminous coal	Coal	Gasification	9	Black	Grey/Black/Brown	14.72-30.9
Brown coal			9	Brown		N/A
Natural Gas	Natural Gas	SMR/ATR	9	Grey		10.09-17.21
		SMR/ATR + CCS	7-8	Blue	Green	2.97-9.16
		Pyrolysis	3-5	Turquoise	-	4.2-9.14
Any fossil fuel	Any fossil fuel	Gasification, SMR/ATR	9	Grey	Black/Brown	10.09-30.9
		Gasification + CCS, SMR/ATR	6-7	Blue	-	1.09-10.35
	Hydrogen generated electricity	In-situ (underground) gasification + selective filtering	3-5	Clear		N/A
Biomass	Biomass	Gasification	5-6	None	Green	0.31-8.63
Biomass	Biomass	Gasification + CCS	3-5	None	Green	-17.5 - -11.66
Water	Solar generated electricity	Electrolysis	9	Green	Yellow	1.32-2.21
	Wind generated electricity		9	Green	Orange*	0.52-1.14
	Renewable generated electricity		9	Green	-	N/A
	Mixed grid generated electricity		9	White	Yellow	N/A
	Nuclear generated electricity		9	Pink	Red/Purple	0.47-0.96
Naturally occurring hydrogen	Deep Earth processes	Drilling	3-5	Gold	White	N/A

* based on (Parkinson et al., 2019; The Royal Society, 2018)

Table 2

Location	***Storage type***	***Gas composition***	***Storage Volume (m^3)***	***Mean depth (m)***	***Status***	***Year***
Teesside, UK	*Salt cavern (bedded salt)*	*95% H2, 3-4% CO2*	*3x 70.000*	*350*	*Active*	*1972*
Clemens, USA	*Salt cavern (domal salt)*	*95% H2*	*580.000*	*1000*	*Active*	*1983*
Moss Bluff, USA	*Salt cavern (domal salt)*	*H2*	*566.000*	*1200*	*Active*	*2007*
Spindletop, USA	*Salt cavern (domal salt)*	*95% H2*	*906.000*	*1340*	*Active*	*2017*
STOPIL-H2, Etzel, France	*Salt cavern*		*570.000*		*Under development*	
Kiel, Germany	*Salt caverns*	*Town gas*	*7.8 *10^7*	*1330*	*Repurposed as natural gas storage*	
Bad Lauchstädt, Germany	*Salt cavern & porous reservoir (depleted gas field)*	*Town gas*	*6.7 * 10^8*	*800*	*Repurposed as natural gas storage*	
Underground Sun Storage, Austria	*Porous reservoir (depleted field)*	*10% H2*	*115.000*	*1200*	*Under development*	*2017*
Yakshunovskoe Field, Russia	*Porous reservoir (depleted field)*				*Active*	*2010*
Hychico, Argentina	*Porous reservoir (depleted field)*	*10% H2*	*750.000*	*815*	*Under development*	*2010-2018*
Kirchheiligen, Germany	*Porous reservoir (depleted field)*	*Town gas*	*2.4 *10^8*	*900*	*Repurposed as natural gas storage*	
Hähnlein, Germany	*Porous reservoir (aquifer)*	*Town gas*	*1.6* 10^8*	*500*	*Repurposed as natural gas storage*	
Eschenfelden, Germany	*Porous reservoir (aquifer)*	*Town gas*	*1.68*10^8*	*600*	*Repurposed as natural gas storage*	
Engelborstel, Germany	*Porous reservoir (aquifer)*	*Town gas*	*?*	*?*	*Decommissioned*	*1955-1998*

Ketzin, Germany	*Porous reservoir (aquifer)*	*Town gas*	*1.30 x 10^8*	*250-400*	*Decommission ed*	*1964-2000*
Lobodice, -Czech Republic	*Porous reservoir (aquifer)*	*Town gas*	*1 * 10^8*	*400-500*	*Repurposed as natural gas storage*	*1965-1995*
Beynes, France	*Porous reservoir (aquifer)*	*Town gas*	*3.3 x 10^8*	*430*	*Repurposed as natural gas storage*	*1956-1972*
HyBRIT, Sweden	*Rock cavern*	*100% hydrogen*	*100*	*30*	*Under development*	*2016*

Table 3

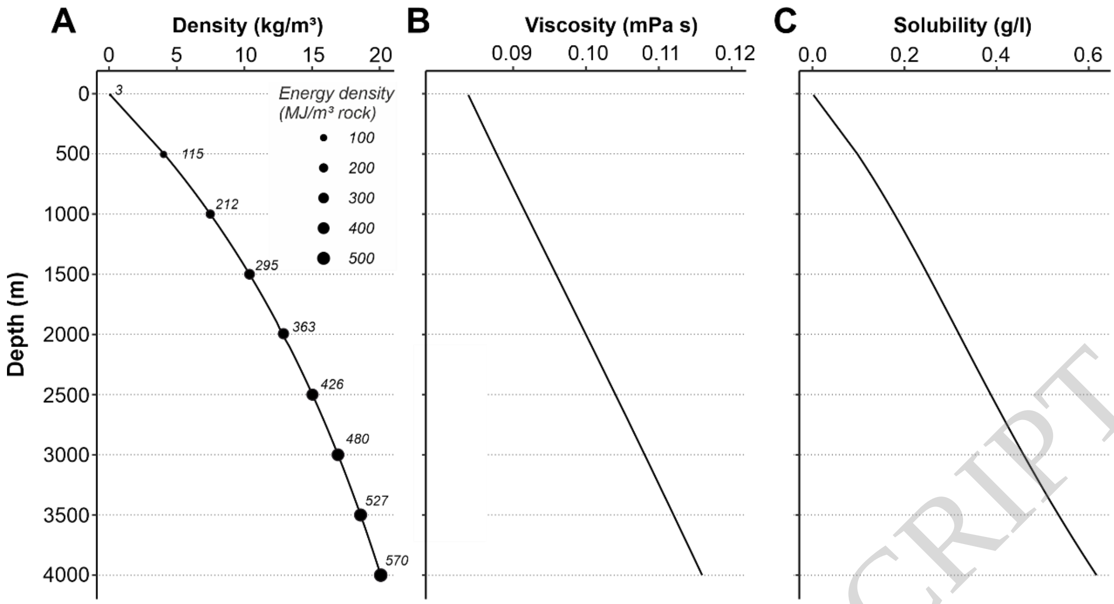

A
Density (kg/m³)
0 5 10 15 20
Depth (m)
0 500 1000 1500 2000 2500 3000 3500 4000
Energy density (MJ/m³ rock)
100
200
300
400
500
3
115
212
295
363
426
480
527
570
B
Viscosity (mPa s)
0.09 0.10 0.11 0.12
C
Solubility (g/l)
0.0 0.2 0.4 0.6

Figure 1

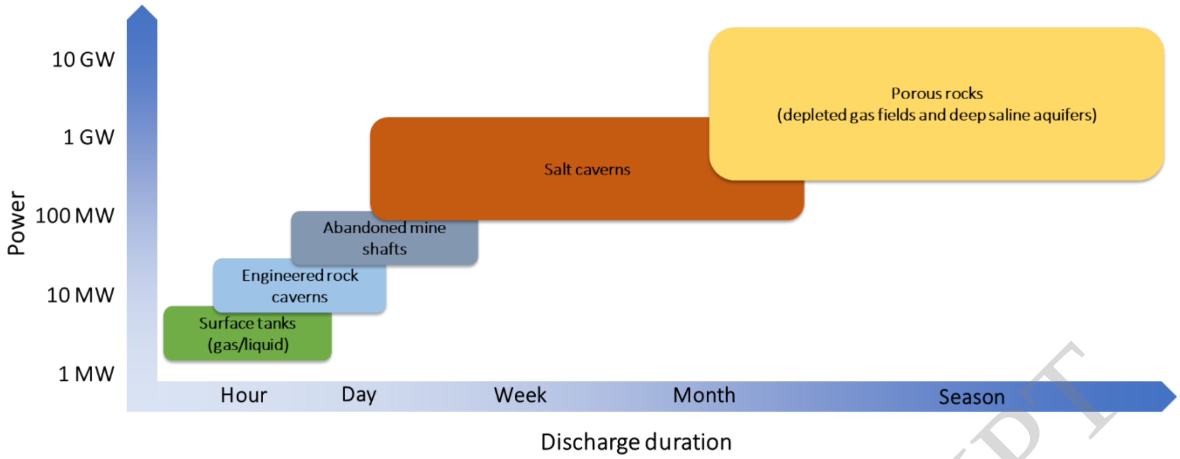

10 GW
1 GW
100 MW
10 MW
1 MW
Power
Porous rocks
(depleted gas fields and deep saline aquifers)
Salt caverns
Abandoned mine
shafts
Engineered rock
caverns
Surface tanks
(gas/liquid)
Hour
Day
Week
Month
Season
Discharge duration

Figure 2

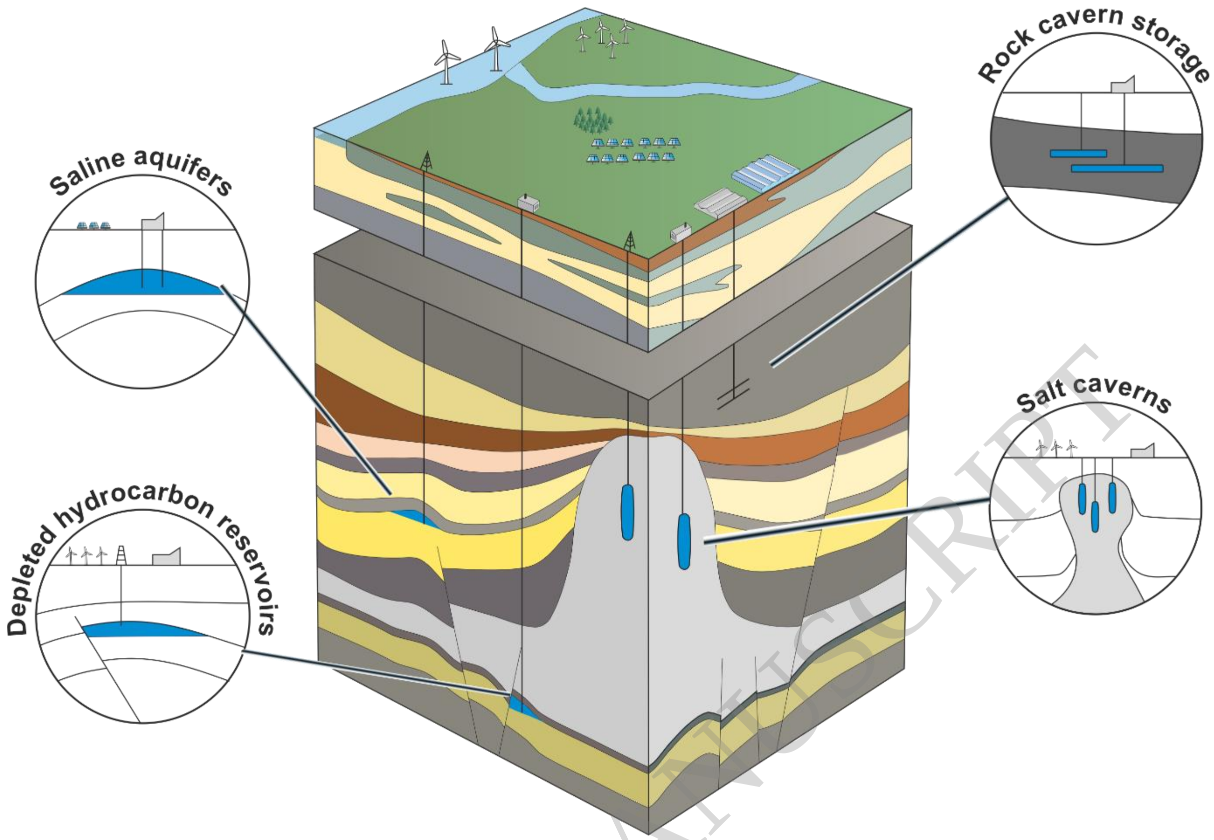
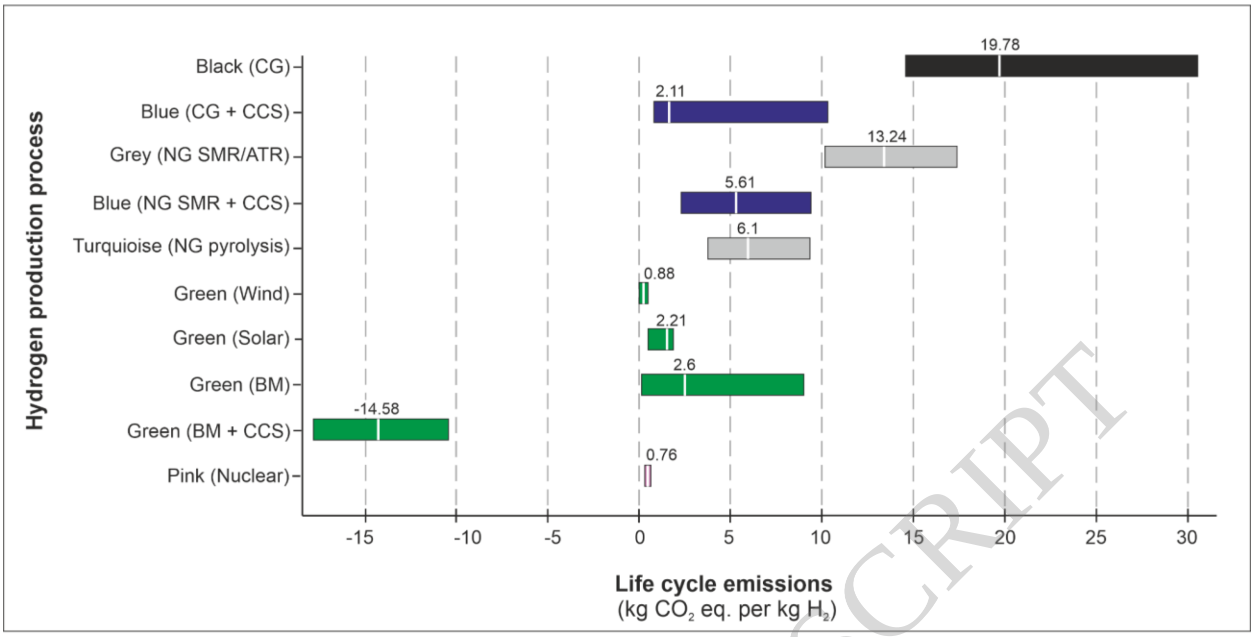

Saline aquifers
Rock cavern storage
Salt caverns
Depleted hydrocarbon reservoirs

Figure 3



Hydrogen production process
Black (CG)
Blue (CG + CCS)
Grey (NG SMR/ATR)
Blue (NG SMR + CCS)
Turquioise (NG pyrolysis)
Green (Wind)
Green (Solar)
Green (BM)
Green (BM + CCS)
Pink (Nuclear)
19.78
2.11
13.24
5.61
6.1
0.88
2.21
2.6
-14.58
0.76
-15
-10
-5
0
5
10
15
20
25
30
Life cycle emissions
(kg CO_2 eq. per kg H_2)

Figure 4