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Underground hydrogen storage: A UK perspective

Amir Jahanbakhsh ^{a,b,*}, Alexander Louis Potapov-Crighton ^a, Abdolali Mosallanezhad ^{c,d}, Nina Tohidi Kaloorazi ^a, M. Mercedes Maroto-Valer ^{a,b}

- ^a The Research Centre for Carbon Solutions (RCCS), School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh, UK
- ^b Industrial Decarbonisation Research and Innovation Centre (IDRIC), Heriot-Watt University, Edinburgh, UK
- ^c Department of Petroleum Engineering, School of Chemical and Petroleum Engineering, Shiraz University, Shiraz, Iran
- ^d Formation Damage and Well Treatment Research Group, IOR/EOR Research Institute, Shiraz University, Shiraz, Iran

ARTICLE INFO

Keywords: Energy storage Underground hydrogen storage Hydrogen strategy Salt caverns Saline aquifers Depleted oil and gas reservoirs

ABSTRACT

Hydrogen is anticipated to play a key role in global decarbonization and within the UK's pathway to achieving net zero targets. However, as the production of hydrogen expands in line with government strategies a key concern is where this hydrogen will be stored for later use. This study assesses the different large-scale storage options in geological structures available to the UK and addresses the surrounding uncertainties moving towards establishing a hydrogen economy. Currently, salt caverns look to be the most favourable option, considering their proven experience in the storage of hydrogen, especially high purity hydrogen, natural sealing properties, low cushion gas requirement and high charge and discharge rates. However, their geographical availability within the UK can act as a major constraint. Additionally, a substantial increase in the number of new caverns will be necessary to meet the UK's storage demand. Salt caverns have greater applicability as a good short-term storage solution, however, storage in porous media, such as depleted hydrocarbon reservoirs and saline aquifers, can be seen as a long-term and strategic solution to meet energy demand and achieve energy security. Porous media storage solutions are estimated to have capacities which far exceed projected storage demand. Depleted fields have generally been well explored prior to hydrocarbon extraction. Although many saline aquifers are available offshore UK, geological characterizations are still required to identify the right candidates for hydrogen storage. Currently, the advantages of depleted gas reservoirs over saline aquifers make them the favoured option after salt caverns.

1. Introduction

Energy has been fundamental in helping shape the modern world. Almost all daily activities require energy in some form, but since the industrial revolution in the mid-1870s global temperatures have risen by almost 1°C, with forecasts predicting this trend to continue due to increased emissions of heat-trapping greenhouse gases [1]. The Keeling Curve shown in Fig. 1 depicts the build-up of carbon dioxide (CO₂) concentration in the atmosphere as one of the main drivers of global warming [2]. Fig. 2 highlights the global share of energy consumption by fuel source along with the associated carbon emissions for oil, coal and gas. It can be seen that heavy reliance is still placed on fossil fuels, with oil, coal and gas contributing to over 70% of the world's energy consumption. These three sources of energy are also some of the most common carbon pollutants, accounting for nearly 95% of energy-related carbon emissions and collectively expelling almost 35 billion tonnes of

carbon dioxide in 2021 [3].

To help shift this current trajectory away from a 1.5°C increase by 2050 aligned with goals set out in the Paris Agreement, the UK and other countries have been transitioning away from carbon-intensive and polluting energy generation, instead pursuing low-carbon or renewable alternatives. Figs. 3 and 4 show the UK's fuel mix for electricity generation and the territorial carbon dioxide emissions from power stations from 1990 to 2021. Reliance on gas has remained fairly constant since 1997, however, the last decade shows that significant measures have been made to phase out energy generation from coal and place a greater uptake of energy generated from renewable sources [6]. Total UK's GHG emissions had a 44% reduction, from 1990 to 2019, and the energy sector has contributed to about half of the total reduction. The energy sector was the largest GHG producer in the UK until 2016 which came second after the transportation sector. Energy sector emissions accounted for about 21% of total GHG emissions in the UK in 2019, compared to 34% in 1990 [7]. To achieve a reduction in GHG emissions,

^{*} Corresponding author.The Research Centre for Carbon Solutions (RCCS), School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh, UK. *E-mail address:* a.jahanbakhsh@hw.ac.uk (A. Jahanbakhsh).

| Abbreviations | WGC-90 a rate-limited capacity for 90 days of withdrawal |
|--|---|
| CAES Compressed Air Energy Storage CCUS Carbon Capture, Utilisation and Storage Hystories HYdrogen STORage In European Subsurface HyStorPor Hydrogen Storage in Porous Media HyUSPRe Hydrogen Underground Storage in Porous Reservoirs Hydrogen TCP Hydrogen Technology Collaboration Programme LEL Lower Explosion Limit MICP Minimum Internal Cavern Pressure NZHF Net Zero Hydrogen Fund Pce Capillary entry pressure PEM polymer-electrolyte membrane SRL Storage Readiness Level UEL Upper Explosion Limit UHS Underground Hydrogen Storage UGS Underground Gas Storage | CO ₂ Carbon Dioxide H ₂ Hydrogen CH ₄ Methane Units bcm billion cubic metres g/L grams per litre GW giga Watt kWh kilo Watt hour TWh tera Watt hour kg/h kilogram per hour kg/m ³ kilogram per cubic metre mD millidarcy MJ/kg mega Joule per kilogram kWhel/m ³ kilo Watt hour (electrical energy) per cubic metre Nm ³ /h normal cubic metre per hour |

the UK increased the capacity of renewable electricity connected to the grid by about 500 % from 2009 to 2020 which led to a reduction in greenhouse gas emissions of electricity generation, by 72% between 1990 and 2019 [8].

In the recent UK Powering Up Britain plans published in March 2023, the government has set out plans to move towards energy independence by aiming for a doubling of Britain's electricity generation capacity by the late 2030s, in line with the aim to fully decarbonise the power sector by 2035. However, the role that the UK's oil and gas sector will play in that transition will be recognized. The government plans cover expanding renewable energy generation, launching Great British Nuclear, development of Carbon Capture, Utilisation and Storage (CCUS) projects and growth of low carbon hydrogen economy [9].

A key issue which remains with most renewable technologies is their intrinsic intermittency, and moreover, how inconsistent supply of energy generated from renewable sources can be matched to variable demand. A common phenomenon in renewable energy technologies, particularly evident in wind power, is curtailment, which is defined as the reduction of energy generation, and arises when surplus energy is

frequently available but remains unnecessary [10]. The curtailment of wind power not only leads to the dismissal of clean energy but also imposes significant economic repercussions on the government [11]. In 2020, 3.70 TWh of wind electricity generation was curtailed, costing the UK government £274 million. Predictions indicate that by 2030, wind energy rejection could reach up to 7.72 TWh, incurring a cost of £573 million for the government. This projection is attributed to the planned installation of 40 GW of offshore wind capacity in the UK by 2030 [12]. Consequently, the expansion of wind capacity will inevitably lead to increased curtailment of this valuable clean energy source.

One method of matching the energy generated from renewable sources to variable demand and avoiding the curtailment is through energy storage, such as batteries, compressed air, pumped storage hydropower, flywheels and thermal energy storage systems. These storage systems present some technical challenges, such as varied roundtrip efficiencies, greater associated risks and higher upfront costs on a grid-connection scale when compared with conventional methods such as gas-fired power plants [13]. Hydrogen storage offers a promising solution by converting surplus electricity into hydrogen or producing it from

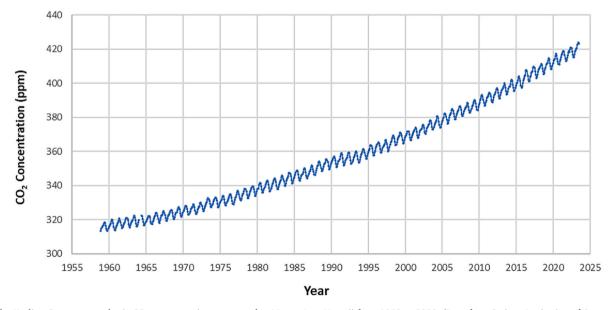


Fig. 1. The Keeling Curve: atmospheric CO₂ concentrations measured at Mauna Loa, Hawaii from 1958 to 2023. (Data from Scripps Institution of Oceanography at UC San Diego) [2].

low-carbon processes to then be released again by using the gas as a fuel in combustion engines or fuel cells [14]. Moreover, hydrogen has the highest energy per mass of any fuel and can play a significant role in helping to decarbonise the global energy mix.

The EU already considers hydrogen to be essential in reaching carbon neutrality by 2050, as well as meeting the global commitments made as part of the Paris Agreement. The UK has made its own commitments, pledging to reduce carbon emissions to a net zero level by 2050 (2045 for Scotland) and an interim reduction target of 78% by 2035 [15]. To facilitate achieving these targets the UK Government produced a 10-point plan listing out key areas for growth in an accelerated pathway towards net zero. Low carbon hydrogen together with carbon capture, utilisation and storage both featured heavily, with greater technological advances and an increase in scale necessary to accommodate for a low hydrogen economy.

To expedite the deep decarbonization of energy systems, hydrogen is anticipated to serve various energy-intensive sectors such as industry, electricity, transportation, and heating, tailored to each country's requirements. As previously mentioned, hydrogen storage is a pivotal component in hydrogen energy systems, making it essential to have robust and reliable storage solutions tailored to each application. The hydrogen storage application can be classified as stationary or mobile. Stationary storage includes on-site storage at the point of production or use and stationary power generation, while mobile storage encompasses vehicle fuel and hydrogen transportation. For large-scale stationary applications, underground hydrogen storage technologies are the most feasible. In contrast, mobile applications demand alternative technologies [16–18].

Although the mass-energy density of hydrogen is nearly three times that of liquid hydrocarbons, its volumetric energy density is notably lower. Liquid hydrogen possesses a density of 8 MJ/L, whereas gasoline is at 32 MJ/L. Consequently, for transportation applications, it is imperative to significantly increase hydrogen's density to minimize the requisite storage space. Hydrogen storage systems are categorized into physical-based and material-based storage. With physical-based storage, hydrogen can be stored as compressed gaseous hydrogen by increasing the pressure, as liquid hydrogen by cooling it below its boiling point, or as cryo-compressed hydrogen by adjusting both pressure and

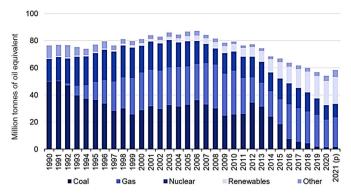


Fig. 3. Fuel mix for UK electricity generation (Million tonnes of oil equivalent), 1990–2021 [6] (Reprinted with permission).

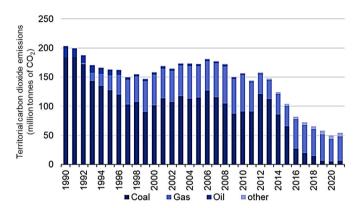


Fig. 4. Territorial carbon dioxide emissions (Million tonnes of CO₂) from power stations, UK, 1990–2021 [6] (Reprinted with permission).

temperature. In fuel-cell-powered vehicles, hydrogen is compressed and stored in large, high-pressure containers. However, this occupies substantial space, reducing the area available for passengers and cargo. An

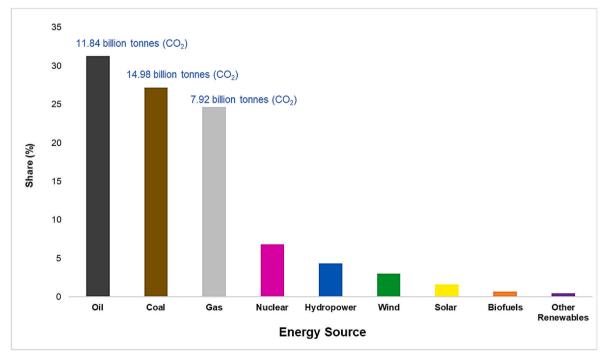


Fig. 2. World share of energy consumption by Source in global CO₂ Emissions of oil, coal and gas, 2021 [4-6].

alternative is storing hydrogen in liquid form, but this approach is technically complicated and expensive. It must be cooled to cryogenic temperatures to liquefy hydrogen, given that its boiling point at atmospheric pressure is -252.8° C. Thus, it's stored in insulated tanks to maintain this extreme cold and reduce evaporation. While hydrogen typically does not corrode storage containers, it can induce cracks in certain metals, affecting storage safety [16–19].

In comparison to physical-based storage systems, material-based storage addresses potential safety concerns through its lower storage pressure and controllable properties at ambient conditions. For this storage method, additional materials, called carriers are used. These carriers can form bonds with hydrogen molecules or atoms, either physically or chemically, improving storage density and safety. Metal hydrides, ammonia, and liquid-organic hydrogen carriers are examples of chemical sorption materials. Porous physical sorption materials offer new avenues for high-capacity and reliable storage. Among these, Metal-Organic Frameworks and porous carbon materials stand out as the most promising [18,20,21]. However, it's worth noting that many material-based storage technologies are still in the research and demonstration phases [18,20,21]. For more information on emerging storage technologies refer to Yang et al. (2023), Moradi and Growth (2019) and Usman (2022) comprehensive reviews [16–18].

For stationary storage applications, hydrogen can be stored above ground using three main methods: gas holders at regular air pressure, round pressure containers with pressures of up to 20 bar, and pipe storage with pressures of up to 100 bar [22]. Around the world, there are numerous aboveground hydrogen storage projects of different sizes. These projects are mainly designed to help manage the electricity produced by renewable energy sources, like solar and wind, which means they can store a moderate amount of energy and release it over a few hours [23]. For larger-scale storage needs, using aboveground vessels is much more expensive compared to storing underground. Above-ground storage also requires a lot of land, which might have other uses or may not be available in some cases. Moreover, because of pressure restrictions and the materials needed for high-pressure conditions, above-ground storage is not suitable for extensive storage purposes [24, 25]. For long-term storage on the scale of days and months, as well as for large-scale energy production on the order of GWh or TWh, underground energy storage stands out as one of the most promising solutions [26,27]. Subsurface geological storage can provide the required capacity for long discharge time for hydrogen storage compared with other technologies [28], as shown in Fig. 5.

Several studies have examined energy storage requirements, with hydrogen playing a significant role [26,27,29]. Olabi et al. conducted a comprehensive assessment of hydrogen production and storage technologies, discussing the associated challenges [30]. Zivar et al. (2021) reviewed the feasibility and technical challenges of underground hydrogen storage (UHS) in porous media [31]. Heinemann et al. (2021)

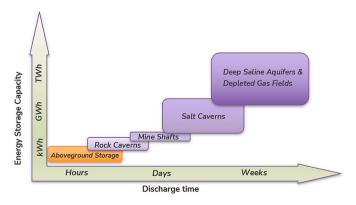


Fig. 5. Hydrogen storage potential options based on their energy storage capacity and discharge time (adapted from Ref. [28]).

identified key processes impacting underground hydrogen storage in porous media (UHS), concluding that safely and efficiently storing hydrogen in subsurface areas is more complex than storing CH₄ or CO₂ in the same formations [32]. Tarkowski et al. (2019) analysed how physio-chemical properties of common gases for underground storage -H₂, CO₂, and CH₄ - along with storage site formation and conditions can influence the storage process [33]. However, there is still limited experience with hydrogen storage in underground porous media, especially for pure hydrogen storage [22]. In contrast, decades of experience exist in utilizing subsurface porous media formations for storing other gases, such as carbon dioxide (CO2) and natural gas (mainly CH4). There are mainly three potential types of underground hydrogen storage in scale above GWh. The first option is salt caverns which is a proven technology for storing hydrocarbons [34], including natural gas, liquefied hydrocarbons such as LPG, crude oil or refined products. The worldwide count of salt caverns surpasses 1900.

Even though there is a significant need for energy storage, the capacity of underground hydrogen storage has yet to widely be explored [35]. However, in recent years several collaborative projects such as Hystories (HYdrogen STORage In European Subsurface), HyUnder (Hyunder.eu), HyStorPor (Hydrogen Storage in Porous Media), HyUSPRe (Hydrogen Underground Storage in Porous Reservoirs) and Hydrogen TCP (Hydrogen Technology Collaboration Programme) have performed thorough investigations to explore the potentials of different underground storage options mainly across European countries.

For instance, the primary goals of the "Hystories" project were threefold: 1) to advance technical methodologies that can be applied across a broad spectrum of future aquifer or depleted hydrocarbon fields. 2) to undertake techno-economic feasibility studies. And 3) to offer valuable insights into underground hydrogen storage for policymakers in both government and industry sectors in 17 European countries. The HyUnder project investigated and compared all known underground storage technologies for hydrogen including salt caverns, depleted hydrocarbon reservoirs, saline aquifers, conventional mined rock caverns, abandoned conventional mines, and pipe storage across 6 different European countries including Germany, Spain, France, Romania, the Netherlands and the UK. The HyUSPRe project investigates the viability of storing renewable hydrogen in Europe's porous reservoirs (depleted hydrocarbon fields and aquifers) on a large scale. The study identifies ideal geological reservoirs and assesses their technological and economic feasibility for use by 2050. Addressing technical challenges, the project offers both a techno-economic evaluation and considers environmental, social, and regulatory implications. The project's first objective is to analyze the feasibility and risks of underground hydrogen storage in Europe's porous reservoirs, including cost estimation, potential business cases, and storage site identification (Technical Assessment). Secondly, developing a roadmap through creating a strategy for geological hydrogen storage integration up to 2050, considering the proximity to renewable energy infrastructures and energy storage capacities to meet varying demands, setting the stage for future demonstrations [36].

Established in 1977 under the International Energy Agency, the Hydrogen Technology Collaboration Programme (Hydrogen TCP) has pioneered hydrogen research, development, and demonstration through 40 tasks among its 26 Contracting Parties. With a history spanning over 40 years and contributions from 7 Sponsor Members, the Hydrogen TCP stands as a leading global resource in hydrogen technical expertise. The program envisions a future where hydrogen is central to a sustainable global energy supply across all sectors. Currently, the TCP activities encompass 47 tasks, with Task 42 focusing on underground hydrogen storage (UHS). This Task aims to accelerate the safe deployment of UHS by fostering coordinated collaborations and spreading knowledge. Its goal is to advance research in the field, monitor ongoing and emerging technologies, and aid in shaping a regulatory framework for UHS in the participating countries [37].

The Hydrogen Knowledge Centre, established in the UK by the

Institution of Gas Engineers and Managers, is a digital library featuring resources from academic and research institutions, public agencies, supply chain organisations, as well as energy and engineering experts. These contributors provide technical research and leading-edge reports. The Centre's objective is to disseminate pivotal research, foster global hydrogen learning, and support the transition to a net-zero carbon emissions future. Serving industry professionals, policy influencers, students, and academics, this repository plays a key role in empowering those active in the future energy domain. Thus, the Hydrogen Knowledge Centre promotes global knowledge exchange [38].

International collaboration is pivotal in accelerating the deployment of hydrogen projects because it combines expertise, resources, and research from multiple nations, fostering innovation and overcoming technical challenges more efficiently. Collaborative efforts lead to shared standards and best practices, ensuring safety and efficacy across borders. Moreover, by uniting on a global scale, countries can drive down costs, enhance supply chains, and foster a more sustainable and universally adopted hydrogen-based energy system. Although several international initiatives and research projects have started in recent years, limited experience exists globally with underground hydrogen storage in practice.

While many research projects and programmes are exploring the capacity of underground hydrogen storage, there are still limited operational projects. Table 1 presents a selected list of existing underground hydrogen storage projects worldwide. While salt cavern storage is the predominant form of underground hydrogen storage, there have been only a handful of projects reported in aquifers and depleted gas reservoirs. A shared characteristic of all experiences with hydrogen storage in porous media is that hydrogen is typically stored in a blend with other gases, primarily methane and $\rm CO_2$. While hydrogen purity in salt caverns can reach very high purity (up to 95 %), it is less than 60 % in aquifers and below 10 % in depleted gas reservoirs [39–44]. Fig. 6 shows the current and future hydrogen storage projects in salt caverns and geological formations on different continents.

To have a better understanding of storage requirements and objectives of to the existing hydrogen storage projects in aquifers and depleted gas reservoirs listed in Table 1, a brief introduction to each project is presented here.

Beynes (France) - An aquifer in a depth of 366 m (1200 ft) near the city of Beynes in France was used to store hydrogen from 1956 to 1972 [45]. The aquifer is an unconsolidated sandstone with a thickness of 10 m and permeability of 3–5 Darcy. The total capacity of the aquifer is about 500 million m³ of which 360 million m³ can be used for storage purposes. The hydrogen-rich low-Btu stored gas in this reservoir was a manufactured gas named town gas or city gas which had 50–60% hydrogen. The extracted gas after one year of storage consisted of a trace amount of nickel and iron carbonyls. Before scrubbing out the carbonyls it was required to desulfurize, dry and oxygenate the gas. In 1973, the by-product gas was not available any longer therefore the reservoir was converted to a natural gas storage site since then [45].

Ketzin (Germany) - In this project, manufactured hydrogen-rich gas

has been stored in the aquifer in a depth of 200-250~m of sandstone located 40~km west of Berlin [41]. The monitoring system in this project, between 1964 and 1985, indicated that gas compositions changed over the storage time. An increase in the composition of hydrogen, methane and carbon dioxide and a decrease in the composition of carbon monoxide was reported. Moreover, a $30-40^{\circ}\text{C}$ temperature increase was observed [46].

Lobodice (Czech Republic) - A major function of the Lobodice UGS was to store excess coke gas produced in Ostrava that was surplus to demand. Conversion of the facility to the storage of natural gas was completed in 1991 [47]. In the Lobodice project, town gas was a blend of 50% hydrogen and 25% methane which was stored in an aquifer. The reservoir is at a depth of 430 m and the operating temperature and pressure are 43°C and 90 bar respectively. This project has reported gas losses caused by a diversity of mechanisms, including dispersion, structural trapping, partial hydrogen leakage, and main changes in the composition of the gas [48].

Underground Sun Storage (Austria) - Underground Sun Storage is a pilot project for storing hydrogen in depleted gas reservoirs. The stored gas is a mixture of natural gas (90%) and green hydrogen (10%). This project was the first real-world test which addressed the possibility of hydrogen storage in a subsurface sandstone reservoir at a depth of 1027 m with an average temperature of 40°C [49]. The project demonstrated that it is possible to store renewable energy in the form of hydrogen in underground porous media. Hydrogen did not have any negative influence on the integrity of the reservoir and cap rock. The project studied the complete life cycle of hydrogen injection, storage and recovery. In the last stage, the volume balance showed that about 82% of the injected hydrogen could be recovered. The rest of the hydrogen was diffused, dissolved, or converted chemically via microbial activities. This field test showed that a limited percentage of hydrogen (up to 10%) blended in natural gas, will not damage the existing infrastructures of a gas field [49]. Following up on the Sun Storage project, RAG Austria executed another project called "Underground Sun Conversion" in which hydrogen conversion to methane in a sandstone reservoir due to microbial activities was investigated. A series of laboratory experiments, numerical simulations and small field tests were performed to understand how microbes react to the injection of hydrogen and metabolize the hydrogen to generate methane. They found that the conversion happens in the reservoir but not as fast as in the laboratory [50].

Hychico (Argentina) - The Hychico project in Argentina is a combination of the wind farm, hydrogen production and underground storage. The $6.3~\mathrm{MW}$ wind park and hydrogen production facilities with a capacity of $120~\mathrm{Nm}^3/\mathrm{h}$ are located near a depleted gas reservoir in Patagonia. A $2.3~\mathrm{km}$ pipeline transfers green hydrogen to an injection well. To confirm the sealings of the reservoir, natural gas was first injected. In the subsequent step, hydrogen is injected into the reservoir to study the behaviour of the blending of natural gas and hydrogen in an underground reservoir. The share of hydrogen in this stage is 10~%. In the final step to study the tightness of the reservoir and cap rock in the presence of hydrogen, natural gas is injected to raise the pressure [51].

Table 1List of existing underground hydrogen storage including salt caverns (selected projects), aquifers and depleted gas reservoirs [39–41].

| Project | Type of Reservoir | H ₂ Percentage | Depth (m) | Capacity ($10^3 \times \text{sm}^3$) | Electric Energy (GWh) | Status |
|------------------------------------|------------------------|--|-----------|--|-----------------------|-------------------|
| Kiel (Germany) | Salt Cavern | 62 % H ₂ | 1,335 | 32 | NA | Operating with NG |
| Teesside (UK) | Salt Cavern | 95 % H ₂ 3–5% CO ₂ | 370–400 | 3 × 70 | 30 | Operating |
| Spindletop (USA) | Salt Cavern | 95 % H ₂ | 850-1400 | 600 | NA | Operating |
| Clemens Dome (USA) | Salt Cavern | 95 % H ₂ | 850 | 580 | 92 | Operating |
| Moss Bluff (USA) | Salt Cavern | _ | 850-1,400 | 566 | 80 | Operating |
| Beynes (France) | Aquifer | 50-60 % H ₂ | 430 | 1,185,000 | NA | Operating with NG |
| Ketzin (Germany) | Aquifer | 62 % H ₂ | 200-250 | _ | NA | Closed |
| Lobodice (Czech Republic) | Aquifer | 40-50 % H ₂ 25 % CH ₄ | 400–500 | 400,000 | NA | Operating |
| Underground Sun Storage, (Austria) | Depleted Gas Reservoir | 10 % H ₂ | 1,000 | NA | NA | Operating |
| Hychico (Argentina) | Depleted Gas Reservoir | 10 % H ₂ | 600-800 | NA | 24.6 | NA |

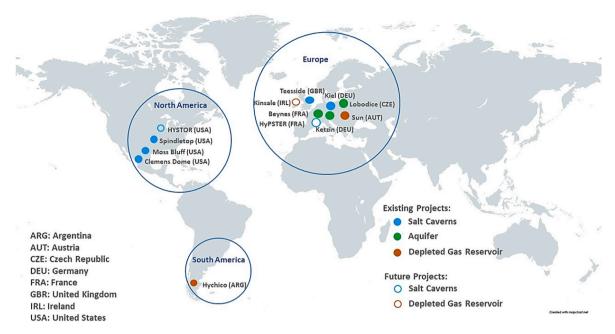


Fig. 6. Current and future hydrogen storage projects in salt caverns and geological formations across the globe. (Authors created with MapChart).

The selected reservoir is a glauconitic type with a depth of 815 m and a temperature of 55°C. The reservoir porosity is about 25% and the permeability is 300–500 mD. The final goal of the project is to produce methane in the underground reservoir from methanological reactions by storing hydrogen and carbon dioxide. In this process, the reservoir will be a natural reactor to generate green methane which can be directly used as a fuel [51].

This study aims to investigate the potentials and hurdles of UHS options, focusing on salt caverns and porous media such as saline aquifers and depleted hydrocarbon fields. In this research methodology, a multifaceted approach, including a comprehensive investigation into several key areas, is adopted to achieve the research objectives. Salt caverns, depleted hydrocarbon reservoirs and saline aquifers, their advantages, disadvantages and economics are reviewed. A detailed analysis of the primary key characteristics of each storage option is undertaken. Lessons learned from other underground storage processes such as CO2 storage are leveraged for building insight into the storage site selection process. Government strategies related to decarbonization are critically reviewed, with a focus on their potential impact on the research. Future scenarios regarding hydrogen production are projected and analysed, considering various influencing factors and trends. The novelty of this work lies in its approach to integrating technical and operational aspects of UHS with economic and policy considerations. However, as the energy needs and resources, government priorities, infrastructure development, technological advancement and market conditions of countries are different, the focus of this work is the United Kingdom. In the UK, commitment to clean energy and decarbonization has developed a significant emphasis on harnessing the potential of hydrogen as a versatile and sustainable energy carrier. In this research, recent storage capacity estimates have been identified and discussed with respect to how they align with the UK's hydrogen strategy. Gaining a comprehensive understanding of the necessary and accessible storage types and their characteristics, capacities, pros, and cons holds paramount importance in aligning with the country's strategic expansion of the hydrogen economy. By delineating the storage requirements and capacities, this study intends to help decision-makers and stakeholders ascertain the infrastructural investments, technological advancements. and regulatory frameworks needed to facilitate the envisioned hydrogen economy's development. This review offers valuable insights into the international effort to align energy storage strategies with hydrogenbased economies. The evolution of the global hydrogen economy and the formulation of corresponding strategies and policies are dynamic and constantly evolving processes. Although there has been notable progress in hydrogen storage in salt caverns, the widespread adoption of large-scale hydrogen storage in depleted reservoirs and saline aquifers remains relatively limited. This scarcity of practical experience has led to a deficiency in measurements and empirical data, forcing many assessments to rely on laboratory experiments or numerical simulations, which, in turn, introduce certain limitations. To ensure precision and efficacy within the ever-changing hydrogen sector, it is imperative to reassess assumptions, review new policies and regulations and perform fresh analyses once real-project data becomes available.

2. Hydrogen production

2.1. Current state of hydrogen production and applications in the UK

Since its discovery in 1766 by British scientist and philosopher Henry Cavendish, hydrogen has been an integral component within a vast array of applications which help to benefit civilisation. From being used in the production of ammonia to fertilise crops to petroleum refinement, the production of methanol and in applications related to wielding, metalworking, glass, electronics, food, medicine and aeronautics, the usefulness of hydrogen is insurmountable [52]. Recently, however, with global shifts towards decarbonization and pathways towards net zero objectives, the role of hydrogen as an energy carrier is becoming increasingly vital. According to the UK Climate Change Committee (CCC) analysis, in the balanced net zero pathway, there will be about 225 TWh of low-carbon hydrogen demand in the UK in 2050. This demand will be in different sectors including power, aviation, shipping, surface transport, buildings, manufacturing, and construction. National Grid has developed Future Energy Scenarios (FES) in the UK and predicted that 21-59% of energy demand in 2050 will be supplied by hydrogen [53].

Currently, the known natural sources of pure hydrogen are of little abundance, meaning hydrogen needs to be manufactured for commercial use [54]. The most common way to produce so-called 'grey' hydrogen uses natural gas or methane, which reacts with steam in an exothermic reaction to produce hydrogen, carbon monoxide and carbon dioxide [55]. The process known as steam methane reformation (SMR)

accounts for 95% of the world's current hydrogen production. If the emission of the harmful greenhouse gas by-products from SMR were to be avoided through carbon capture, utilisation and storage technologies, the production process would be sustainable, resulting in what is known as blue hydrogen. Another important method of extracting hydrogen is through electrolysis, to split water into its components of hydrogen and oxygen, with no carbon by-products. By using electricity generated through renewable energy sources the product of this much-cleaner process is aptly referred to as green hydrogen [54]. Other colours of hydrogen with different carbon impacts exist. Black and brown hydrogen use black or brown (lignite) coal as feedstock in a process known as gasification and they are the most environmentally damaging hydrogen as by-product carbon dioxide and carbon monoxide are released into the atmosphere. Pink hydrogen uses nuclear power for electrolysis which can be one of the most efficient production processes. Turquoise hydrogen is created through methane pyrolysis when methane is decomposed at very high temperatures to generate hydrogen and solid carbon. This production process is classified as low carbon as no carbon dioxide or carbon monoxide is produced. Yellow hydrogen which solely uses solar power in its electrolysis has a moderate carbon impact as the electricity comes from renewable sources. Finally, white hydrogen is naturally occurring hydrogen found in underground geological formations [56].

Steam methane reforming is the predominant, well-established, fully commercialized method for hydrogen production and is also the least expensive. As of the end of 2021, nearly 47% of the world's hydrogen was produced using this technology. Notably, the cost of hydrogen derived from SMR is heavily influenced by the price of its natural gas feedstock [57]. As per the International Renewable Energy Agency, by the close of 2021, a mere 4% of global hydrogen production was attributed to water electrolysis. Electrolysis becomes cost-effective when there's a need for small amounts of exceptionally pure hydrogen. The pivotal operational cost component in electrolysis is electricity, however by 2021, the cost of producing hydrogen via electrolysis was becoming more competitive, especially in regions with abundant and cheap renewable electricity [57].

As the UK hydrogen strategy emphasizes both blue and green hydrogen production, it is worth comparing the respective production processes known as SMR and electrolysis. SMR is primarily employed for industrial applications such as ammonia production and oil refining, where hydrogen serves as a crucial input. Electrolysis is versatile and suited for a wide array of applications, including fuel cells for transportation, energy storage, and industrial feedstock. While SMR can utilize existing natural gas infrastructure, potential modifications are required to accommodate carbon capture technologies. In contrast, the development of new infrastructure is necessary for green hydrogen production through electrolysis. Although blue hydrogen production might be constrained by CCUS requirements, SMR stands as an established process with high efficiency. Conversely, electrolysis efficiency largely hinges on the source of electricity used [58,59].

In summary, SMR and electrolysis represent two primary methods of hydrogen production, each with distinct advantages and challenges. While SMR boasts established efficiency, it can lead to carbon emissions unless integrated with carbon capture. However, electrolysis offers cleaner hydrogen production potential when powered by renewable energy. However, concerns persist regarding infrastructure development and efficiency enhancement. The choice between these methods hinges on factors such as carbon emissions goals, energy availability, and intended hydrogen applications [60,61].

Presently, practically all the hydrogen produced in the UK is used as industrial feedstock in oil refineries and chemical plants, roughly the equivalent of 27 TWh of energy [62]. For these applications, hydrogen will commonly be produced and used on-site, and therefore, it can be directly integrated and coupled with the industrial process it is aiding to Ref. [54]. As a fuel source, hydrogen is a lot less utilised, with the main applications presently being related to transport. Fleets of hydrogen

buses, trucks, cars and marine vessels are already under operation, supported by an infrastructure of refuelling stations with additional developments and investments towards a multi-modal hydrogen transport hub located in northern England's Tees Valley [63]. A further application of hydrogen is to blend it with natural gas to reduce its carbon intensity. HyDeploy Winlaton is a project currently underway investigating the blending of to 20% blending of natural gas with hydrogen being supplied to 668 homes, a primary school and other small businesses on a trial basis, leading up to the first target driven timeline set ahead for 2023 [24,64]. Similarly, the H100 project in Fife (Scotland) is aiming to be the world's first pilot project to use green hydrogen as the heat supply in 300 local homes by 2023 using its own dedicated electrolysis plant powered by an offshore wind turbine nearby [65].

2.2. Timeline of the UK's hydrogen strategy for large-scale production

Moving forward, the UK government have put a significant reliance on the role that hydrogen technologies will play in the UK's transition towards net zero targets. The publication of 'The Ten Point Plan for a Green Industrial Revolution' directly states out plans to grow the low carbon hydrogen industry in the UK, as well as push towards greener ships and planes and increased investments in CCUS technologies up to £1 billion, among others [66]. Investing in CCUS technologies is a key component of producing blue hydrogen. The Department for Business, Energy and Industrial Strategy (replaced by Department for Energy Security and Net Zero, Department for Science, Innovation and Technology, and Department for Business and Trade in 2023) published their 'UK Hydrogen Strategy' in 2021, outlining a roadmap to establish and facilitate a thriving hydrogen economy moving ahead in the 2020s. In line with The Sixth Carbon Budget (2020) and the pathway towards meeting net zero commitments, the report stated a hydrogen production target of 5 GW set for 2030. The strategy emphasizes both blue hydrogen (produced from natural gas with carbon capture) and green hydrogen (produced through renewable energy-powered electrolysis). This target was recently doubled to a 10 GW capacity of low carbon hydrogen production with at least half expected to come from electrolytic hydrogen, requiring a growth four order of magnitudes greater (10,000 times) [67].

A closer look into the hydrogen economy roadmap shows key actions and milestones broken down into smaller timeframe targets. After the successful announcement of the industrial cluster schemes in 2021 and the recent launch of the £240 million Net Zero Hydrogen Fund (NZHF) the successive key stage will be announcing the winning projects to receive the funding grants. There are currently twenty shortlisted projects from the 2022 NZHF allocation round, with an aim of finalising contracts by the end of 2023 [9,68]. Pushing ahead to 2025, it is aimed that there will be 1 GW of low hydrogen production capacity already installed and the deployment of two industrial CCUS clusters announced as HyNet North-West and the East Coast Cluster, located around the industrial powerhouses of Liverpool, Manchester, Humber and Teesside [69]. By 2030, this is intended to have developed into 10 GW of production and an additional two clusters.

With low-carbon hydrogen still in the early development stages, there are a number of challenges that need to be overcome. Hydrogen is still much more expensive than existing fossil fuels due to the high upfront costs for the electrolyser and balance of plant; however, it is predicted that this may reduce as much as 30% by 2030 as a result of declining costs of renewable energy and greater scaling of hydrogen production [70]. Another barrier to overcome is the lack of technical knowledge, skills and financial uncertainty going ahead into future years. Although there are cases where these technologies already exist, it is still relatively unclear how they will operate on a much larger scale and require a fully trained workforce. Furthermore, to properly accommodate for this increased deployment the surrounding infrastructure will also need to be scaled up and upgraded. CCUS technologies will need to advance and integrate with the production of hydrogen,

as well as further installations and enhancements to gas and electricity networks with capable transport and storage systems [54]. Additional uncertainty remains with regard to the regulation and policy of these technologies. There must also be assurances on the safety and quality, incentives and financial support, direction on supply chains and a range of other strategic decisions to be considered. Without these, and due to the nascent nature of low carbon hydrogen, there is a fear that 'first-of-a-kind' deployment poses a greater risk to its investors and as such there will be a hesitancy to move early, especially without secured offtake.

3. Underground hydrogen storage

The aim of this section is to succinctly discuss the primary underground storage options: salt caverns and geological porous media, specifically depleted hydrocarbon reservoirs and saline aquifers. Numerous comprehensive reviews already exist on underground hydrogen storage, delving into the intricacies of salt caverns, depleted oil and gas reservoirs, and saline aquifers. As such, the focus of this section is kept on the UK's perspective, steering clear of redundant information, and offering pertinent references for in-depth understanding [31,32,71–82].

3.1. Salt caverns

Salt caverns are artificial cavities created in underground salt formations through the dissolution of rock salt by water injection during the solution mining process [83]. This process involves drilling wells into the targeted salt formations, followed by the injection of fresh water to facilitate salt leaching and subsequent brine production through the wellbore. Residual traces of brine are subsequently evacuated using gas injection [73,84]. The practice of using these underground cavities for the storage of natural gas has been around for decades, and the knowledge gained from their widespread deployment is now being transferred to hydrogen storage due to the similarities in cavity design, construction and operation [85]. Furthermore, salt caverns are a highly regarded storage option due to the cost-effective construction, efficient injection and withdrawal rates, high sealing capacity of rock salt, low cushion gas requirement (see section 4.5 for more information), high purity hydrogen, and inert nature to liquid and gaseous hydrocarbons as well as hydrogen, helping protect against contamination [85].

Salt caverns can achieve depths of up to 2000 m, typically spanning heights between 300 m and 500 m, with diameters ranging from 50 to 100 m. A single salt cavern can be designed with a maximum storage capacity of around 1,000,000 m³ [30]. Given hydrogen's inherently low density (0.08988 g/L), its effective storage poses significant challenges [86]. To optimize hydrogen's storage density, it's imperative to leverage energy, e.g., via compression. Operating at pressures between 60 and 180 bar, salt caverns can efficiently store hydrogen, achieving energy densities of up to 300 kWhel/m³. Although salt caverns have low risk associated with microbial activities, considering the potential impact of microorganisms on gas purity, comprehensive studies on microbial characteristics and their implications are essential. Such studies should be undertaken for each selected site, complemented by robust monitoring systems [86]. The high salinity and brine content within these caverns alleviate osmotic stress on microbial cells, potentially reducing bacterial diversity [87]. During the withdrawal stage, salt caverns' stability can be compromised due to the unloading of the adjacent rock, potentially leading to induced micro-cracking [88]. The structural integrity of the cavern must be maintained. While the risk is low, a breach or collapse could lead to significant losses. One of the environmental challenges associated with the process of leaching to create the cavern is the production of large amounts of brine. Its primary influence on hydrogen storage lies in hydrogen diffusion through the salt walls. Additionally, the presence of brine and sumps can contribute to an increase in hydrogen humidity [89]. Proper disposal or utilisation of this brine can represent an added operational challenge and cost.

Establishing a salt cavern storage facility requires navigating various regulatory, environmental, and safety standards, which can influence the timeline and cost. For a comprehensive exploration of the challenges associated with underground hydrogen storage, including in salt caverns, refer to Navaid et al. (2022), Muhammed et al. (2022) [73,77,90].

In the UK, the industrial hub of Teesside has been storing hydrogen in three elliptically shaped salt caverns since 1972, giving a total storage volume of 210,000 m 3 [91]. Three larger hydrogen storage facilities exist in Texas, USA, with each of the sites having a storage capacity of 566,000 m 3 , 580,000 m 3 and >580,000 m 3 , the largest of which with an estimated energy capacity of more than 120 GWh. According to Caglayan et al. [85], these projects have long since demonstrated that the underground storage of hydrogen is technically possible.

Fig. 7 shows a map of the UK's rock salt deposits (halite) distribution. The map illustrates the thick-bedded halite formations from both the Permian and Triassic ages found in several onshore and offshore sedimentary basins. However, it does not show the thin, aerially restricted onshore lateral equivalents of offshore Triassic halite formations [92]. In the UK, there exist four predominant sedimentary basins known for their rich halite mineral deposits, making them prime candidates for cavern construction. These basins are strategically located in Wessex, Eastern England (mainly in East Yorkshire), Cheshire, and the East Irish Sea [90, 93]. Whilst the distribution of thick and continuous halite formations is spatially restricted, they may provide a storage option for several large industrial centres, including Teesside, Humber, Northwest, and Solent [92]. The Fordon Evaporite Formation from the Permian era, which spans a significant portion of England's east coast and continues beneath the southern part of the North Sea, has already been tapped for natural gas storage. Several vast gas storage caverns in Hornsea and Aldbrough are hosted in this formation, where it reaches depths of more than 1600 m and has a thickness of nearly 300 m. Typically, the cavern's diameter is comparable to its height, around the order of 100 m. These caverns operate at depths ranging from 1700 to 1800 m [92].

The Northwich Halite Member is one of two significant bedded halite formations in the fault-bounded Cheshire Basin, which is Triassic in age. Compared to older Permian halite formations, Triassic halite formations are typically found at shallower depths. In the UK, the Cheshire Basin stands as the preeminent region for natural gas storage caverns, currently boasting a minimum of 73 caverns, either operational or slated for construction [86,90,92]. In the Wessex Basin located in southern England, the Dorset Halite Member is identified as a saliferous unit within the Triassic Mercia Mudstone Group [94,95]. In this basin, while the presence of salt has been confirmed through multiple boreholes, the nature and lateral continuity of many of these accumulations remain inadequately defined. The regional structure is intricate, characterized by a series of sub-basins that interrupt the continuity of the Dorset Halite member. Owing to the prevailing geological uncertainties and the lack of operational precedents for underground gas storage in the area, Williams et al. (2022) concluded that detailed site investigations are imperative before developing Triassic halites. Halite beds in the area have shown variable thickness and might also contain considerable impurities and mudstone interbeds, especially closer to basin margins. Such geological features could limit potential cavern dimensions and their storage capacities [92].

The Boulby Halite, located within the Teesside salt field, is characterized by depths ranging from 274 m to 366 m and exhibits a maximum thickness of approximately 45 m. Within this geological region, multiple former brine caverns, such as Saltholme and Wilton, have been designated for the storage of nitrogen, natural gas, and an array of liquid hydrocarbons. Historically, solution-mined caverns in Teesside are well known for storing town gas - with up to 60% hydrogen content - since as early as 1959 [76]. However, three former brine caverns have been used for the strategic storage of hydrogen for industrial applications since the early 1970s. The constrained halite thickness in the Teesside salt field has resulted in caverns of an elliptical morphology, typically spanning heights between 15 m and 40 m and exhibiting diameters proximate to

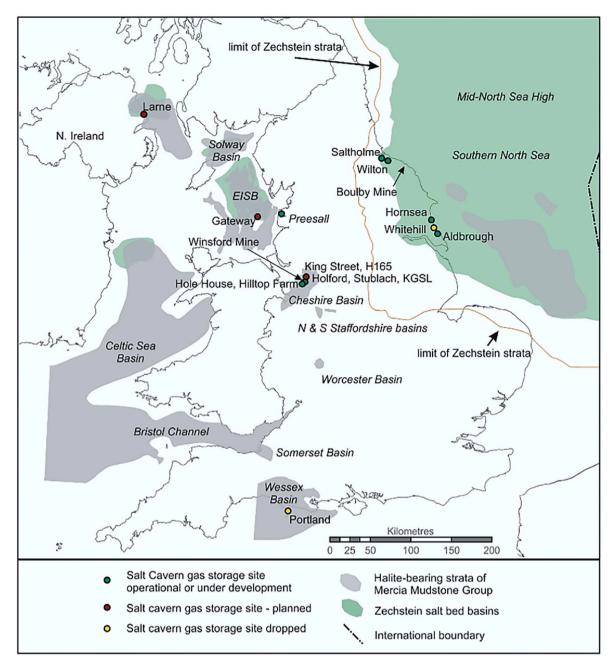


Fig. 7. UK rock salt deposits (halite) distribution, adopted and updated by Williams et al. [92] from Evans et al. [90]. Red ellipses determine the three regions in the Williams et al. study. (Reprinted with permission).

70 m. Despite this limitation, either constructing new caverns or repurposing the existing ones bring potential opportunities for hydrogen storage in the Teesside area [92].

In the East Irish Sea Basin and the Larne area of Northern Ireland, halite deposits of Permian age, though relatively thin, have been preserved at depth [86,96,97]. Evaluations have been conducted on the Permian halite beds in proximity to Larne, exploring the feasibility of cavern construction for gas storage and compressed air energy storage [86,98]. Triassic halite formations are also present in both the southern and northern regions of Northern Ireland. Further investigation is needed to characterize these halite formations in terms of their thickness and continuity to determine their suitability and potential storage volume for hydrogen. While onshore salt cavern development is technically simpler and typically less expensive than offshore developments, and while co-location with existing gas distribution networks is advantageous, offshore development could become more prominent in the

future. This shift could be driven by the potential to repurpose existing oil and gas infrastructure, the co-location opportunities with future offshore wind farms, and the integration of floating electrolyser facilities for hydrogen production [30]. It is worth mentioning that there is significant storage potential in offshore salt caverns. Extensive halite beds and halokinetic structures are evident in offshore areas, including the East Irish Sea Basin, the North Sea, and the Southern North Sea [92].

Williams et al. (2022) applied a modelling approach to assess the theoretical hydrogen storage capacity in potential new salt caverns in the Fordon Evaporite Formation of East Yorkshire, the Northwich Halite Member of the Cheshire Basin, and the Dorset Halite Member of the Wessex Basin in the UK. The theoretical storage calculations for these three onshore UK areas relied on the distribution of bedded halite formations appropriate for creating new gas storage caverns ranging in size from 10 to 441 GWh. Their findings suggest an upper bound potential for hydrogen storage of over 64 million tonnes, equating to a storage

capacity of 2150 TWh, spread across three distinct salt basins in more than 13,000 potential cavern locations. Nonetheless, the storage estimates reduced considering the associated geological uncertainty of approximately $\pm 36\%$ and wider cavern spacings [92]. Their calculations indicate that even when accounting for uncertainties in key geological variables, hydrogen storage caverns could provide a lower-bound estimate of at least 612 GW. This is significantly higher than the UK's peak heat demand [99]. The estimated inter-seasonal storage capacity in these caverns could be seamlessly incorporated into a hydrogen transmission system. Moreover, their analysis underscores that the potential storage capacity in salt caverns presents more opportunities than limitations for fostering a low-carbon hydrogen heat network in the UK. However, A substantial increase in the number of caverns (up to around 1000) relative to the current inventory of natural gas caverns will be necessary [92].

The uneven distribution of suitable salt formations across the UK poses a significant challenge for future salt cavern developments. Usually, not all potential hydrogen users will have the advantage of being situated near storage sites. Regions such as industrial clusters in Scotland and South Wales, lack proximate access to onshore storage caverns. This geographical disparity underscores the critical importance of addressing the spatial distribution of salt formations in planning for future developments [92]. To conclude, salt caverns can provide an economically attractive solution for large-scale hydrogen storage. However, the economic feasibility is contingent on various factors, including location, cavern size, and the specifics of the project. As the demand for hydrogen grows, especially green hydrogen, the need for large-scale storage solutions like salt caverns will likely increase, potentially driving further research, development, and cost optimizations in this area.

3.2. Depleted oil and gas reservoirs, and saline aquifers

Depleted oil and gas fields were at one time filled with hydrocarbons and a certain amount of these hydrocarbons have since been withdrawn to be used in a variety of different applications. These hydrocarbons would have accumulated over time by essentially being trapped due to the natural geological formations that occurred, often consisting of a reservoir, seal and aquifer [100]. Depleted oil and gas reservoirs stand out as ideal candidates for underground gas storage due to their inherent geological characteristics [33,101]. A hydrocarbon reservoir is encircled by an impermeable caprock layer, often with an aquifer providing structural support either from beneath or along its edges [45]. Historically, these reservoirs have been primary choices for natural gas storage because of the distinctive combination of their well-defined geological structures, proven integrity, tightness of their caprocks, and existing infrastructure both above and below ground [45,102]. The advantage is twofold: pre-existing surface and subsurface installations cut down on development time and costs, and the geological knowledge accumulated over years of operation provides assurance of their tightness and integrity [31,45].

Moreover, the remaining gas that remains post-extraction in gas reservoirs can function as cushion gas, facilitating the stable storage of additional gases. However, this residual gas can sometimes act as a double-edged sword; while it aids in maintaining pressure, it also has the potential to compromise the purity of stored hydrogen. For effective underground gas storage (UGS) in such depleted gas deposits, halting gas extraction at the right moment is pivotal, enabling faster and more cost-effective conversion to storage facilities [45,103]. In the context of UHS, depleted gas reservoirs offer several advantages over aquifer storage. The residual gas present in these reservoirs can function as cushion gas, thus reducing the required volume typically needed in aquifers [12,103]. Depending on the reservoir structure and operational criteria, anywhere between 50 and 60% cushion gas is essential to ensure pressure stability and avoid hydrogen trapping by aquifer water encroachment [103]. Their operational pressures vary, lying in the

range of 1.5–30 MPa, and depths span between 300-2700 m [104]. Their transformation from gas fields to storage entities can span from 3 to 10 years, and the rates of injection and withdrawal are primarily determined by rock permeability [103,104].

Interestingly, some UGS units often hit their intended operational parameters within an estimated five years, mainly due to the production of formation waters that invade post gas extraction [33,45]. This process occasionally results in the underground storage site experiencing pressures that exceed the reservoir's original readings, paving the way for a more substantial gas storage than what was previously possible [33]. However, challenges arise when considering the transition of depleted oil reservoirs for hydrogen storage. Residual oil can trigger chemical reactions, mostly converting hydrogen to methane, thereby diminishing the purity of stored hydrogen. This phenomenon stems from the intricate interplay occurring at the hydrogen-oil interface [45,104].

When contrasting hydrogen storage in aquifers with that in depleted reservoirs, the former necessitates a greater injection pressure to counterbalance natural reservoir pressures, pushing water away from the injection site. However, the flow characteristics of hydrogen, due to its reduced viscosity and density, make it prone to phenomena like fingering and gas overriding [105]. Over time, aquifer conditions during hydrogen injection undergo variations influenced by several factors, including porosity, permeability, and reservoir geometry, to name a few [105,106]. An understanding of flow behaviour, which is crucial for optimizing storage efficiency, is contingent on a myriad of variables, ranging from viscosity and density to gravitational forces and flow direction [106–109]. It is also noteworthy that specific geological features, such as steeply inclining structures and thick formations, can serve as bulwarks against undesirable events like fingering [110].

In terms of practical application, no reservoir has yet stored pure hydrogen. However, blends, such as the mix of 10% hydrogen and 90% methane tested by RAG Austria, indicate the feasibility of such an endeavour [111]. Encouragingly, the horizon for pure hydrogen storage in depleted gas fields is near, with expectations set for its operational debut in 2030, spearheaded by RAG Austria [112]. In summary, while the expansive volumes, seasoned infrastructure, and rich geology of depleted gas fields make them potential powerhouses for hydrogen storage, challenges, particularly concerning purity and geological interactions, remain. Yet, with continued research, pilot projects, and rigorous studies, these reservoirs may soon solidify their role in the future of hydrogen storage.

Unlike hydrocarbon reservoirs, aquifers, are vast underground layers of porous and permeable rock predominantly filled with fresh or saline water [33]. These basins have no presence of hydrocarbons and as such are referred to as geological traps [28,29]. They are widely present in sedimentary basins around the world, making them viable for underground storage applications [33]. Aquifers' suitability for gas storage resembles that of depleted hydrocarbon reservoirs, emphasizing porous media such as sandstone that lie thousands of feet underground. These natural formations are developed for storage by displacing water, achieved by injecting cushion gas followed by hydrogen through strategically placed wells [33]. The replacement of water by hydrogen causes a density disparity, leading to water displacement and increasing the pressure in the porous media, thereby reshaping the liquid-gas interface [33,113]. The injection of hydrogen can lead to water production alongside gas upon withdrawal, a challenge attributed to the movement of the gas-liquid interface [31,113].

A prime concern is ensuring gas confinement, requiring impermeable layers or caprock to prevent gas migration [33]. While depleted reservoirs have historically proven their containment capabilities, guaranteeing the reliability of aquifers mandates rigorous geological studies to confirm caprock integrity [31,114]. Such studies often lead to increased costs, particularly in scenarios lacking requisite infrastructure [103]. Operational pressures in aquifers typically range from 3 to 30 MPa, with depths spanning 400 to 2300 m [115]. An essential component for aquifers is cushion gas, which aids in maintaining pressure. Unlike

depleted reservoirs, which may contain natural gas to serve as cushion gas, aquifers often necessitate an additional cushion gas injection, sometimes up to 80% of the storage volume [103,104].

Hydrogen storage in aquifers faces challenges related to potential gas leakage along undetected faults, geochemical and microbial reactions, and the possible interaction of hydrogen with reservoir rock minerals [33]. For example, sulfate-reducing bacteria have been known to contaminate stored gas in deep aquifers [116]. Gas drying infrastructure, vital due to water being a frequent impurity in gas stored within aquifers, adds another layer of complexity to the process [45,104]. It's pertinent to mention that, as of the recent literature, no pure hydrogen storage in aquifers has been successfully reported. However, several European projects, including those at Engelbostel, Bad Lauchstädt, Lobodice, Beynes, and Ketzin, have showcased storage potential by storing town gas or coal gas with significant hydrogen content [117]. In conclusion, while aquifers present a widespread and accessible option for hydrogen storage, their development demands a deeper understanding of geological intricacies, meticulous planning, and robust infrastructure to optimize their potential.

3.2.1. Porous media storage site selection

While there is some experience in storing hydrogen mixtures in porous media, there is currently no established standard practice for screening and ranking depleted oil/gas fields and aquifers for pure hydrogen storage. This is because there's no prior experience in this specific area. However, the principle of hydrogen storage in porous media is akin to that of natural gas storage. At the end of 2019, there were 661 underground gas storage (UGS) facilities in porous media in operation in the world (76 in aguifers and 488 in depleted hydrocarbon fields), representing a global Working Gas (WG) capacity of 386 bcm (47 bcm in aquifers and 339 bcm in depleted hydrocarbon fields) [118]. Therefore, it might be beneficial to adapt site selection procedures from underground natural gas storage (UGS) and draw from experiences in converting natural gas storage fields into storage for hydrogen. However, it is critical to add new criteria to address fundamental processes unique to subsurface hydrogen storage, such as diffusion, geochemistry, and microbial activity.

The Hystrories project investigated the applicability of selection criteria used in natural gas storage to hydrogen storage. Although the development of storage is mainly site-specific, a reasonable set of environmental, geological and reservoir selection criteria was recommended to be adopted for hydrogen storage site selection [119]. To summarize the recommendations from the Hystroeis project: a net thickness of the reservoir ranging between 3 and 100 m, a total area of the site between 0.3 and 60 km², and a maximum top depth of 2500 m have been advised. These parameters ensure reasonable capacity, structural integrity, safety and pressure range sufficient for supplying at grid pressure. Beyond these dimensions, the reservoir should have good petrophysical characteristics, mostly in terms of porosity and permeability. The geological structure should be delineated, featuring a significant closure height. The sealing overburden formation must be effective, although assessing and verifying this for an aquifer can pose challenges. Lastly, the formation fluids at the site should not compromise the storage gas quality. This implies a low likelihood of corrosion issues, especially when encountering sweet gas, low salinity formation water, and other compatible materials [119].

While the objectives of underground hydrogen storage differ from those of CO₂ storage, the frameworks developed for assessing potential geological formations for CO₂ storage can be adapted for hydrogen. For instance, in the ALIGN-CCUS project, a framework featuring nine Storage Readiness Levels (SRL) was established to communicate a site's progress toward operational storage for both depleted hydrocarbon fields and saline aquifer sites. These SRLs were standardized by leveraging three decades of national experience in planning, appraisal, permitting, and project development from the UK, Norway, and the Netherlands. Adapting or creating a similar framework for underground

hydrogen storage would significantly aid in determining the readiness level of potential storage sites, ultimately resulting in a diverse storage portfolio with sites spanning various readiness levels. The nine SRLs proposed by ALIGN-CCUS for CO_2 storage are as follows [120–122]:

 $SRL\ 1$ - First pass assessment of storage capacity at country-wide or basin scales.

SRL 2 - Site identified as theoretical capacity.

SRL 3 - Screening study to identify an individual storage site and an initial storage project concept.

SRL 4 - Storage site validated by desktop studies and storage project concept updated.

SRL 5 - Storage site validated firstly by detailed analyses, and then in a real world setting.

SRL 6 - Storage site integrated into a feasible CCS project concept or portfolio of sites.

SRL 7 - Storage site is permit ready or permitted.

 $\ensuremath{\mathsf{SRL}}$ 8 - Commissioning of the storage site and test injection at the site.

SRL 9 - Storage site on injection.

Clearly, owing to the intrinsic differences between hydrogen and CO_2 storage, other factors come into play for the former. For instance, the proximity to wind farms can influence the feasibility and efficiency of hydrogen storage due to potential synergies in renewable energy. Additionally, the development or existence of production and surface facilities becomes crucial. These components must be meticulously considered and incorporated to ensure optimal hydrogen storage and withdrawal.

Recently the Hystories project released a geological database viewer which showcases geological data on depleted fields and aquifers relevant for assessing underground hydrogen storage suitability in porous media, at a European scale [119]. This database can be used as a starting point for the site screening and selection for hydrogen storage purposes [119,123]. Fig. 8 shows the locations of saline aquifers and hydrocarbon fields in the UK, with further geological studies required to truly quantify how many are suitable for the storage of hydrogen. It is evident that a large number of saline aquifers are available offshore UK compared to depleted fields. Both these geological storage options have been used in storing natural gas for decades, arising from the need to supply gas to consumers during peak demand and enabling much greater volumes and pressures than can be reached using surface gas tanks [124]. For the conversion from natural gas to hydrogen to take place, the reservoirs must first meet certain prerequisites to ensure they have the necessary subsurface properties to accommodate the storage requirements. Because of this, depleted oil and gas are generally favoured as they will have already undergone a series of characterisations on their subsurface conditions [100].

3.3. Prospects and hurdles in underground hydrogen storage (UHS)

While the intricate challenges of UHS involve diverse domains such as geology, engineering, economy, and societal considerations, certain variables are more flexible (like financial concerns, policy influences, engineering methods, and socio-legal dynamics) than the more static factors (like geological constraints) [126]. For UHS to be successfully implemented, all these aspects must be cohesively assessed. Geological constraints are paramount and should be prioritized when evaluating potential UHS sites. While caverns stand out due to their accessibility, minimal microbial interference, and ability to endure intense reservoir conditions, their scarcity limits their application. Following this are the depleted hydrocarbon reservoirs, whereas aquifers come last owing to unidentified complexities unique to them. Typically, determining the right proportion of cushion gas, essential for any UHS operation post working gas injection, varies across storage types. For instance, while depleted hydrocarbon reservoirs require around 33% of H2 and 50% of CH₄, aquifers might demand between 33% and 66% of H₂ and up to 80% of CH₄ [35].

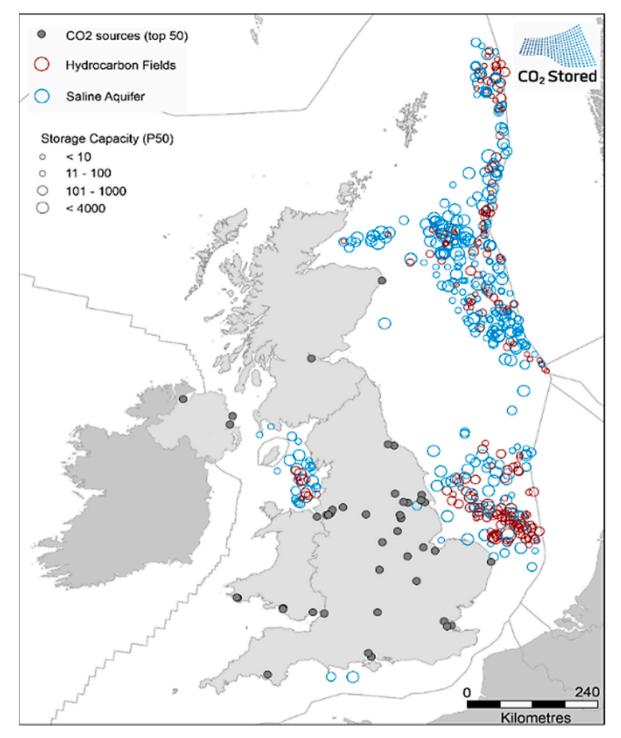


Fig. 8. UK Map of hydrocarbon reservoirs and saline aquifers from CO₂ Stored Database, CO₂ emissions and storage capacity are in megatons [125] (Reprinted with permission).

Knowledge about a site's depth and potential storage is indispensable. Although depth might be less critical than volume, it still plays a role in planning and execution. Conversely, understanding storage capacity is pivotal. While caverns typically have lower storage, aquifers and depleted hydrocarbon reservoirs offer higher capacities, ensuring efficient hydrogen turnover. The prospect of seismic activities poses safety issues for UHS. It is vital for prospective UHS sites to access seismic hazard mappings, particularly at larger operational scales, to mitigate the potential repercussions of overlooked seismic activities in the vicinity [102]. Pressure fluctuation in cyclic operations of the

storage sites is the main cause of seismic risks. Depleted gas reservoirs have been encountered with pressure fluctuations and there is sufficient knowledge about the geological and mechanical properties of these reservoirs. However, limited knowledge and experience are available for aquifers [127]. To reduce seismic risks, it is important to maintain a minimum pressure in the reservoir. Cushion gas will provide this stability in the reservoir and remain in the reservoir permanently. Depleted gas reservoirs and aquifers because of their scale and geometry need quite a high volume of cushion gas. In depleted gas reservoirs, the remaining methane can be a potential cushion gas. If pure hydrogen

storage is required, H_2 itself can be used as a cushion gas however this will not be cost-effective. Nitrogen as a neutral gas is a potential candidate for cushion gas.

Recognizing fluid-fluid and rock-fluid Interactions is critical. In this spectrum, salt caverns fare better than porous mediums, mainly because they're relatively non-reactive. Nevertheless, certain microbial species present might influence the environment during hydrogen storage. In porous media, factors like rock nature, bacteria, ions, and pre-existing fluids matter, particularly because they can trigger various reactions. Here, aquifers have an edge since their primary fluid is water or brine, whereas depleted hydrocarbon reservoirs pose additional complications due to residual hydrocarbons [128–131].

The UHS site's inherent conditions, like the absence of oxygen in aquifers, can be advantageous in preventing ignition and potential flammable situations compared to sites like depleted hydrocarbon reservoirs. Enhanced monitoring mechanisms must be in place to ensure the secure storage of hydrogen [132,133]. It is evident that many specialists are converging on the idea that hydrogen, perhaps in conjunction with electricity, will dominate the future energy landscape. Transitioning to this hydrogen-centric energy paradigm will undoubtedly be accompanied by numerous scientific, technical, and economic challenges. Stepping towards this future will necessitate a combination of heightened awareness, intensive research, and strategic planning to navigate the multifaceted opportunities and challenges presented.

3.4. Economics of underground hydrogen storage

Underground geologic storage of hydrogen not only offers substantial cost reductions and buffer capacity to address disruptions in supply or seasonal demands but also presents a sizable financial asset, ensures continuity of delivery, and helps control congestion in the pipeline system [103]. Therefore, it is essential to understand the economic aspects of underground hydrogen storage however it represents an extensive topic which is out of the scope of this work. More detailed information and analysis can be found in Refs. [103,134-137]. The economic viability of hydrogen storage in salt caverns, depleted hydrocarbon reservoirs, and saline aquifers was tested by Lord et al. using the Hydrogen Geological Storage Model (H2GSM) developed by Sandia National Laboratories. Their cost model addresses both capital expenditures (CAPEX) and operating expenses (OPEX) for the operations and maintenance of the site, compressors, cushion gas, and wells and pipelines. However, the costs of hydrogen delivery to the storage site, withdrawal, and monitoring after injection are not included. The CAPEX and its various sources for all three storage options in Lord et al.'s study are presented in Fig. 9. The volume of cushion gas (hydrogen) required for depleted hydrocarbon reservoirs and saline aquifers was assumed to

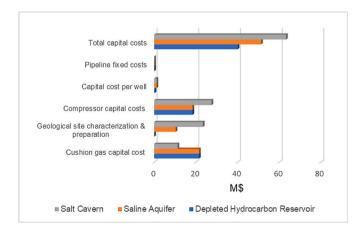


Fig. 9. CAPEX and its various sources for, depleted hydrocarbon reservoir, saline aquifer and salt cavern. (Data from Lord et al., 2014).

be 50% of the total volume, while for salt caverns, it was assumed to be 30%. However, in reality, the cushion gas volume for saline aquifers could be closer to 80%, which would result in a higher capital cost for the cushion gas. According to their study, depleted oil and gas reservoirs and aquifers are economically attractive options, ranging from \$0.04 to \$0.06/kg. The salt cavern storage options have a higher levelized cost of storage, with costs starting at \$1.61. One of the main findings of the Lord et al. study was that geological limitations, rather than city demand, cause a more significant disparity in the costs of salt cavern development from one city to another. Hydrogen storage within salt caverns in cities located near thinly bedded salt formations may cost multiple times more than in cities close to thick salt formations, where the development of larger and fewer caverns is required [103].

The HyUnder project studied the economics of underground hydrogen storage in six European countries including France, Germany, the Netherlands, Romania, Spain, and the United Kingdom. Key findings suggest that, aside from future electricity costs, investment in electrolysers is a major cost component. While the initial costs of creating salt caverns, which involve drilling, leaching, and infrastructure setup, are high, their operational costs are low. Yet, periodic monitoring is essential for safety. The feasibility of salt caverns also depends on the availability of suitable salt deposits in a region. Larger caverns can be more cost-effective due to economies of scale. HyUnder estimated a €28 M investment for a 500,000 m³ hydrogen storage cavern at a depth of 1000 m³ [134]. This project indicated that developing hydrogen storage in aquifers has greater uncertainties and is generally more costly than using salt caverns or depleted hydrocarbon fields. The site selection process for aquifers is expensive and intensive, requiring numerous seismic surveys and exploration wells due to the lack of prior exploration data and production history. Once potential storage sites are identified, further costs arise from storage characterization, involving appraisal wells and lab tests. During the construction phase, the need to drill new wells and install surface infrastructure like compressors adds to the expenses, especially since repurposing existing infrastructure, as done in depleted fields, is not possible for aquifers [134]. Operationally, aquifers may necessitate higher injection pressures, leading to elevated compressor costs. They also demand more cushion gas than other methods and require a significant amount of gas to establish the initial gas bubble. A large portion of this gas becomes unrecoverable after decommissioning, influencing overall costs. Contamination issues, which need expensive gas treatment facilities, further increase both investment and operational expenses [134].

Le Duigou et al. (2017) carried out a study evaluating the technoeconomic feasibility of large-scale underground hydrogen storage in salt caverns in France. Their findings suggest that while geologically storing hydrogen in these salt caverns is technically viable in France, capital expenditures make up over 40% of the total costs. This is threefold the annualized operation and maintenance (O&M) costs. To attain profitability, they identified a necessary incentive range of €1 to €2.5/kg. This falls between 22% and 65% of the overall target hydrogen cost of €4/kg [135]. Tarkowski (2019) suggested that the economics of underground hydrogen storage in the future will largely hinge on demand from three sectors: the power industry, transport (hydrogen fuel cells), and hydrogen-consuming industries. Decarbonization of the power industry, the primary stakeholder, necessitates the expansion of hydrogen energetics. This is connected with utilizing surplus energy from the intermittent outputs of renewable energy sources. Countries that have a significant proportion of wind and solar energy in their mix could benefit first from this shift, rendering projects economically viable [33]. Currently, the primary economic barrier to the broader adoption of this technology is the cost of electricity for electrolysis. Consequently, enhancing electrolyser efficiency and durability has become the main focus of research in this field. Meanwhile, the costs associated with transport and storage are comparatively lower and are expected to decline as the technology becomes more widespread [33].

4. Key characteristics of underground hydrogen storage

In this section the main properties, effects and risks of hydrogen (states of hydrogen, energy density, diffusivity, solubility, flashpoint and autoignition, embrittlement, detection, and contamination) along with the key storage medium properties and challenges will be reviewed briefly. It is beyond the scope of this work to delve into all the characteristics, instead the objective is to provide sufficient information on the crucial parameters in UHS for the ensuing discussion. Several comprehensive studies are available in the literature which are recommended here for more details [31,32,71–82].

4.1. Summary of hydrogen properties

Hydrogen, like all chemical elements, can occur as a gas, solid or liquid depending on the temperature and pressure it is subjected to. At a standard temperature of 25°C and a pressure of 1 bar it is a diatomic gas with a density of 0.089 kg/m³. At extremely low temperatures below -262° C, hydrogen exists in its solid state, with a density of 70.6 kg/m³. Hydrogen's liquid state exists in a small zone between its triple point and critical, yielding a density of 70.8 kg/m³ at a temperature of around -253°C [33,77]. It is special in that it has the highest gravimetric density of any substance, estimated at around 120 MJ/kg. This means that on the basis of mass, hydrogen has an energy content close to nearly three times that of gasoline which is around 44 MJ/kg. In electrical terms, hydrogen therefore contains 33.6 kWh of useable energy per kg, compared to only 12.2 per kg for gasoline and up to 14 kWh per kg for diesel [138]. This is reversed when considering hydrogen in terms of its volumetric density, where it is comparatively lower than other fuel sources [139]. This highlights the need to be able to compress or liquefy hydrogen using high pressures and cryogenic temperatures, as this process raises the volumetric energy density of hydrogen and makes it easier to store, transport or use in applications such as vehicles.

All fuels share a common trait in that they can only burn in a gaseous or vaporous state. Hydrogen and methane already occur as gas under atmospheric conditions, however, fuels such as petrol and diesel are commonly seen to be liquids which first must convert to a vapour before they can burn [140]. The temperature at which fuels produce enough vapour to form an ignitable mixture is known as the flashpoint, an important characteristic in determining how susceptible a fuel source is with regard to its flammability. Essentially, the lower the flashpoint temperature, the greater the risk of the substance's flammability. Hydrogen has an extremely low flashpoint at −253°C, compared to methane (-188°C), propane (-104°C), Gasoline (-43°C) and methanol (11°C). This low flashpoint means that hydrogen is flammable between 4 and 75% concentrations in air and explosive between 15 and 59%, a much greater range in comparison to other fuels. Therefore, even small leaks of hydrogen can quickly reach flammable and explosive levels within an enclosed environment, leading to potentially devastating consequences if not safely contained [132,133,140]. Conversely, hydrogen does have a relatively high autoignition temperature at around 585°C, meaning that it is unlikely that it will ignite solely due to heat alone without the presence of an ignition source.

One noteworthy aspect is that hydrogen only necessitates a 5% oxygen concentration to sustain combustion, while hydrocarbon-based fuels require a higher 12% oxygen concentration [132]. This underscores that hydrogen is not inherently explosive unless an ignition source is present. The lower flashpoint of hydrogen in comparison to methane ($\rm CH_4$), indicates its broader flammability range. Consequently, a lower flashpoint value for a gas corresponds to a wider flammability range. The extensive flammability range, spanning from the lower explosion limit (LEL) to the upper explosion limit (UEL), offers numerous possibilities for utilizing hydrogen as a fuel source for combustion engines or turbines [141,142]. LEL and UEL values denote the concentrations of fuel in the air required to render a mixture flammable. Therefore, mixtures with fuel contents below the LEL or above the UEL

are incapable of ignition due to either insufficient fuel or an inadequate amount of oxygen in the mixture, respectively [143].

A key characteristic of hydrogen in relation to underground storage is its high penetrability (diffusivity), resulting from it being the smallest chemical particle currently known to exist. This means that it diffuses through solids faster than other gasses, such as methane and carbon dioxide. In addition, the solubility of hydrogen is a further important factor to consider. Solubility refers to the degree in which a substance dissolves in a solvent to form a solution, often in relation to water. For hydrogen, its solubility is highly dependent on the temperatures and pressures [107]. In its liquid and gas states (close to the critical point), hydrogen has a very low solubility in water, calculated at around a couple of hundred parts per million (molar) in high pressures between 100 and 1000 bar [77]. When hydrogen is stored in salt caverns, its high diffusivity can pose challenges for long-term storage, especially when compared to deep aquifers and depleted hydrocarbon reserves. This is primarily because salt caverns are typically quite dry. As a result, the tightness of storage is not enhanced by the presence of water in pore spaces, given hydrogen's low solubility in water [33]. During the lifetime of a storage site, estimated diffusion-driven hydrogen losses are in the range of 0.1–1% [32]. Although salt caverns may contain brine and sump, the diffusion would still be largely influenced by the salt walls which make up the bulk of the surface area within the cavern. For storage purposes, the solubility of hydrogen is an essential characteristic to consider. In porous media, like deep aquifers and depleted hydrocarbon reservoirs, the pore spaces often contain water. The presence of water enhances porous media's capacity to contain hydrogen due to its low solubility.

Hydrogen's interaction with metals can lead to significant alterations in their physical properties, a phenomenon known as embrittlement. Embrittlement refers to the effects that hydrogen can have on the mechanical properties of metals it comes in contact with. Metals can become brittle or undergo structural faults such as fractures as a result of hydrogen atoms diffusing into the metal under storage or transportation conditions. These faults occur as the hydrogen reacts with impurities or isotopes present within the metal, creating imbalances within the structures due to the formation of gases or hydrides after absorption into the metal lattices [89]. More commonly, embrittlement is caused by hydrogen diffusing into the metal grain boundaries, forming bubbles which exert pressure on the metal grains [107]. Over time, this build-up in pressure induces stress, potentially reducing the metal's strength and ductility and leading to structural defects.

Hydrogen is also colourless, odourless and has no taste, making it highly difficult to detect. Furthermore, odorants such as butanethiol (commonly mercaptan) which are used as a safety measure in natural gas are incompatible with hydrogen gas as the hydrogen can be contaminated by the present sulfur. Instead, hydrogen requires more complex odorants which are also light enough to match its high dispersion rate [38]. Hydrogen flames are also difficult to detect during daylight as they burn with a pale blue flame and with the absence of soot. Heat ripples which emanate from the flame help to increase detection, along with thermal radiation in the ultraviolet (UV) and infrared (IR) spectral ranges [144]. Additionally, the flames may become more visible as a result of reacting with impurities in the air such as sulfur, or by spreading to surrounding materials which produce smoke and soot when combusted due to the presence of carbon particles [140].

4.2. Porosity and permeability (porous media)

Porosity is a fundamental property of porous media, such as underground reservoirs, that quantifies the proportion of void space relative to the total volume of the medium. It is defined as the ratio of the volume of the voids or pores to the total volume, usually expressed as a fraction between 0 and 1 or as a percentage. This parameter is essential in reservoir engineering and hydrogeology as it dictates the storage

capacity of the reservoir for fluids like water, oil, or gas. A high porosity indicates a large amount of pore space, which can potentially store significant amounts of fluid. However, it is also crucial to consider the permeability of the reservoir, as this parameter determines the ease with which fluids can flow through the porous media. The interconnectedness, size, shape, and distribution of these pores can greatly influence both the porosity and permeability. For instance, a rock might have high porosity due to many small, disconnected pores but low permeability because these pores do not provide effective pathways for fluid flow. As a result, in the study of underground reservoirs, understanding porosity alongside other petrophysical properties is vital for predicting reservoir performance and devising effective extraction strategies. In porous media, the storage capacity and the efficacy of injection or production operations at a prospective storage site are primarily dictated by reservoir rock attributes, especially porosity and permeability. Within these formations, pore spaces are typically occupied by resident fluids. During hydrogen injection, these fluids are displaced, yet not entirely evacuated, leading to an outward movement and consequent pressure increase due to the introduced hydrogen [100]. An illustration of this mechanism can be seen in Fig. 10. Several factors can modulate this pressure surge, including the porous media's dimensions, the nature of the reservoir system (connectivity), and the compressibility characteristics of both the entrapped fluids and the surrounding rock matrix. Moreover, permeability, being a measure of the fluid's ease of movement through the porous substrate, impacts the permissible injection velocities. This ensures that injected hydrogen can efficiently disperse throughout the

The storage capacity of aquifers for UGS ranges from hundreds to thousands of million cubic meters, and this can be equivalently applied to hydrogen storage. Depleted gas reservoirs used for UGS typically exhibit porosities between 15% and 30%. A permeability exceeding 300 mD is generally deemed suitable for UGS [27,30,127]. In UK-based projects, permeabilities have been reported as 2-184 mD for the Rough gas storage, 38.4 and 284 mD for two reservoirs in Hatfield Moors, and 20 mD for the Humbly Grove gas storage [115]. For natural gas, the average charge/discharge rate stands at approximately 85,000 kg/h, whereas for hydrogen in a formation with comparable permeability, a flow rate between 14,000 and 20,000 kg/h can be anticipated. Depleted gas reservoirs, like aquifers, have significant volumetric capacity, making them less sensitive to pressure fluctuations. Consequently, they exhibit lower charge/discharge rates compared to salt caverns. For instance, while a salt cavern can release up to 450 Mm³/day of stored gas, the peak discharge rate for a porous reservoir, be it an aquifer or a depleted gas reservoir, is estimated at around 1 Mm³/day. The injection/recovery rate for natural gas in these reservoirs ranges between 80,000 and 800,000 kg/h. Given the distinct viscosity and density of hydrogen, a plausible rate for hydrogen storage is projected to fall between 8000 and 80,000 kg/h [127].

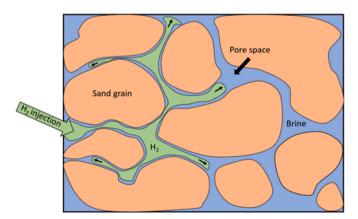


Fig. 10. Gas migration in brine filled pore space. (authors' own drawing).

4.3. Caprock integrity (porous media)

Another critical element in being able to safely store hydrogen is the sealing capabilities of the overlying caprock. Pre-existing faults, shrinkage fractures and prior boreholes all provide pathways from which the stored hydrogen may escape, facilitating the need for extensive geo-mechanical studies [145]. Furthermore, uncontrolled leakage and the long-term upward migration of hydrogen through the caprock may induce seismic activity, leading to groundwater contamination or affecting marine ecology depending on the storage location [146,147]. Espinoza and Santamarina (2017) also stated that the sealing capacity of the caprock may degrade over time due to hydraulic fractures and fault reactivation caused by overpressure of the storage reservoir [148]. Additionally, in the case of carbon dioxide being the gas stored, this could potentially lead to aqueous diffusion into the caprock water causing water acidification and mineral dissolution, although this may be less of a concern in the case of hydrogen storage due to its low solubility.

Underground hydrogen storage relies heavily on the integrity of the caprock which acts as a barrier to prevent hydrogen escape. Ensuring the integrity of caprock is paramount and is influenced by a myriad of factors such as physical discontinuities, hydrogen-driven redox reactions, wettability, temperature and pore size distribution. Natural occurrences like existing faults, shrinkage fractures, and past human activities like boreholes can undermine the caprock's sealing capabilities. Comprehensive geo-mechanical studies become indispensable in such scenarios [145,149]. Hydrogen can initiate redox reactions with iron-bearing minerals, particularly hematite, goethite, or Fe³⁺ bearing clays and micas. If these reactions lead to the removal of hematite-containing cements or clays at grain-grain contacts in sandstone reservoirs, it can modify the mechanical strength of the rock matrix. This alteration might form new leakage pathways, although the magnitude of such reactions has been reported as limited [150]. Studies on reservoir sandstones under subsurface conditions (40-100°C, 10-20 MPa) have shown the dissolution of carbonate and sulfate cements during hydrogen exposure, leading to increased porosity [151]. Examinations of natural gas storage sites show a decline in permeability due to alterations in clay minerals [152]. Notably, minerals such as quartz and feldspar remained unaffected by hydrogen. Some potential storage reservoirs are situated in carbonate formations, making the dissolution of carbonate and sulfate minerals crucial. This can potentially weaken the reservoir rock or carbonate/sulfate-cemented faults in the caprock, hinging on the distribution of these cements and the fluid to rock ratio [32,153]. To predict the chemical reactions over the lifespan of a hydrogen storage site, advanced geochemical modelling is required. A geochemical database, akin to those made for CO2 storage [32], should be developed to quantify the extent of these reactions in the reservoir and caprock. It should encompass hydrogen's reactions with dissolved ions and mineral surfaces, their kinetics, and any potential catalysis. Moreover, to fine-tune these predictions, flow-through experiments at genuine in-situ conditions, using rock samples from potential storage sites and insights from natural hydrogen fields, should be placed side by side against reactive transport models [154].

A paramount factor in the efficiency of caprock sealing is the capillary entry pressure (Pce). Invasion of the storage gas into the caprock is another key consideration and it is important that the capillary entry pressure can sufficiently resist the force created by upward buoyancy pressure built up under the caprock [155]. Recent studies have underscored the importance of Pce for the structural trapping of hydrogen underneath caprocks [148,156,157]. Findings reveal that the water wettability of caprocks is influenced by pressure, organic acid concentration, total organic carbon, and temperature. Usually, wettability decreases with the first three but increases with temperature. Moreover, the capillary sealing efficiency of the caprocks, especially in oil shales, is temperature-sensitive. Variations in the interfacial tension between gas and water and the contact angle with temperature contribute to changes

in Pce. Additionally, Smaller pore sizes and evaporites exhibit superior sealing and storage capacities. Crucially, to analyze the capillary sealing efficiency more effectively, it is imperative to accurately determine parameters like the interfacial tension between gas and water, the receding contact angle, which determines the wettability of the rock/gas/water system, and the pore throat radius of the caprock and mineralogy. Such accurate assessments are instrumental for the success of hydrogen geo-storage projects [129–131,149]. By understanding these elements, the challenges of ensuring caprock integrity become clearer. Continued research, regular monitoring, and integration of newer insights will be pivotal in preventing potential hydrogen leakage and ensuring secure storage.

4.4. Cushion gas requirements (salt cavern & porous media)

Cushion gas (also referred to as buffer gas or base gas) refers to the volume of gas required as a permanent inventory within a storage (salt cavern or reservoir) to maintain adequate pressure whilst undergoing operational cycles of compression and decompression. The volume of gas which is continually extracted and refilled into the reservoir is therefore referred to as the working gas. The ratio of cushion gas to working gas is dependent on geological parameters, such as the shape, depth and permeability of the porous media [158]. As such, the cushion gas requirements and associated concerns differ greatly between salt caverns, saline aquifers and depleted hydrocarbon fields.

The cushion gas serves multifaceted roles, from ensuring consistent pressure maintenance and acting as a barrier against water ingress, to providing a buffer during cyclic operations [159]. The depth and geological parameters of the storage site, such as reservoir shape, trap, porosity and permeability, play a pivotal role in determining the cushion gas to working gas ratio. For instance, deeper reservoirs, often require a lower cushion gas to working gas ratio [158]. Salt caverns can reach depths of approximately 2 km, and depth directly influences the amount of cushion gas needed due to the increased pressure at such depths [103]. Safety and stability are paramount, influencing factors such as the size, shape, and targeted pressure level of the working and cushion gas. The transient behaviour of rock salt creep, for instance, must be analysed before operational commencement.

Mechanical interactions in UHS systems, especially those resulting from the adsorption and desorption of hydrogen to swelling clays, can introduce stresses between individual grains. Over prolonged periods, these stresses might lead to mechanical fatigue of the reservoir, affecting its structural integrity [160]. The balance between active forces in the reservoir, combined with injection rates, can also influence hydrogen losses, such as those due to residual gas saturation and solution into connate water [106,107]. Geochemical interactions between hydrogen and minerals in the reservoir, caprock, and even pre-existing faults can significantly influence the mechanical response of the system. Reactions leading to the dissolution and precipitation of minerals can weaken the reservoir's load-bearing framework, potentially resulting in increased deformation [161]. Moreover, the interactions of certain cushion gases such as CO2 with rock and in-situ fluids can promote the dissolution of caprock minerals, posing a risk of leakage and compromising the storage's efficiency [162,163].

Gases such as low-value gas, CO_2 , CH_4 , and N_2 are used as cushion gas to pressurise or maintain reservoir pressure, increasing the efficiency of the withdrawal cycle. However, the mixing of hydrogen with cushion gases causes hydrogen with lower purity. Typically, the amount of hydrogen mixed into cushion gas is around 1–3% [164]. A similar issue may arise in the case of a depleted oil reservoir where a portion of the hydrogen gets dissolved in the oil phase. Cushion gas is considered not to participate in production and is assumed to be a permanent inventory in the storage media. It undergoes alternate compression and expansion during the injection and withdrawal cycles, respectively, to maintain the required pressure and deliverability rate [165]. The production of cushion gas together with hydrogen can cause an issue. Observations

from natural gas storage, where low-value town gas has been used as cushion gas, reveal that only 1% of cushion gas can be produced after several injection-withdrawal cycles, which hardly influences the safety and economics of storage [45]. The injection of hydrogen or an alternative cushion gas into storage reservoirs will result in the mixing of the injected hydrogen, leading to contamination of the stored hydrogen. The degree of mixing of the gases depends on various factors, including the cycling rate, injection and reproduction rates, reservoir properties, and the type of cushion gas used. Limited experimental data is available for multicomponent hydrogen-rich fluids, making it challenging to validate and tune existing thermodynamic models.

Despite the lower investment costs for nitrogen as cushion gas, it has a higher viscosity and density than hydrogen and even methane. Hence, the displacement of water is more efficient. The disadvantage is the intensive mixing of hydrogen and nitrogen when cyclic operation starts. Carbon dioxide can also be used as cushion gas because of its density compared to other gases. The presence of cushion gas minimizes hydrogen losses, but it can be produced during the withdrawal period in the form of a mixture of hydrogen and cushion gas in a single stream. This requires suitable separation techniques to separate the mixed gas stream into its original constituents. Drawing from experiences in underground natural gas storage, the selection of cushion gas often hinges on its interaction with the storage medium, economic considerations, and the density difference between the working and cushion gas [113, 166].

Salt caverns are highly favourable, as they typically require the least cushion gas, estimated at around 30% [167]. Moreover, a key advantage over porous rocks is that hydrogen does not react with salt, reducing the risk of contamination and therefore the need for gas cleaning. However, the bottom of the caverns will likely contain residual brine, known as cavern sump, which will evaporate over time increasing the present moisture and depending on the intended use of the hydrogen may require drying prior to its application [100,167]. For depleted oil and gas fields, the cushion gas requirements are estimated at around 50-60%, necessary to prevent degradation and breakdown of the reservoir rock. Furthermore, contamination of the hydrogen is much more likely due to the previous presence of hydrocarbon, potentially requiring gas upgrade units for purification [85]. The advantages of depleted fields are that they have generally been well explored prior to hydrocarbon extraction and for gas fields there will often be leftover gas which can reduce the cushion gas requirements but increase the likelihood of contamination with pressurised hydrogen [100]. For aquifers, deep reservoirs with a high degree of permeability are generally favoured [158]. Even so, due to their porous nature, they require greater cushion gas requirements than salt caverns and depleted hydrocarbon fields, ranging from 45% to as high as 80%, depending on the individual geological parameters [12]. During the withdrawal phase, there will be a certain amount of gas that will be trapped within the pore spaces and is therefore classed as physically unrecoverable. Using cushion gas reduces direct contact of hydrogen with brine and eventually may result in less hydrogen trapping; however, there is still a risk of mixing with the cushion gas.

To limit the incurring capital loss that using hydrogen as the cushion gas will have it may be possible to instead use other, less expensive, gasses such as carbon dioxide, nitrogen or methane. In a study by Kanaani et al. [168], it was found that methane had higher hydrogen recovery rates than both carbon dioxide and nitrogen for the same heterogeneous depleted oil reservoir. This is thought to be a result of its low molecular weight. As hydrogen itself is extremely light, it tends to rise to the upper layers of a reservoir but is influenced by the density difference of the phases in contact. As methane has a lower molecular weight than carbon dioxide and nitrogen, the intensity of upward migration is higher when it comes in contact with the hydrogen, leading to increased recovery rates.

4.5. Microbial activity (salt cavern & porous media)

Subsurface microorganisms which dwell in underground storage resources can cause undesirable side effects when considering their suitability for large-scale hydrogen storage. These living organisms use hydrogen in their metabolism, potentially leading to hydrogen losses as well as the formation of methane, hydrogen sulfide and acid, which can cause clogging and corrosion [87]. Microbial activities depend on specific conditions, including mineral content and the presence of certain microorganisms. The temperature range within which these activities occur is from $-15^{\circ}\mathrm{C}$ to $121^{\circ}\mathrm{C}$ [169,170]. The most common hydrogen-consuming reactions in underground reservoirs and the prerequisite conditions are presented in Table 2.

In a recent study by Schwab et al. [171], microbiomes found in the brine solution of five underground salt caverns (previously used for natural gas) were investigated to gain a better understanding of microbial activity in underground caverns. Factors found to determine microbial growth were the cavern's pH levels, temperature and osmolarity. The five caverns were found to have a neutral average pH level of 6.2 and a moderate average temperature of 26.4°C, making them more suited to the growth of mesophilic microorganisms which prefer moderate temperatures of between 20 and 45°C. High salinity was also observed in the caverns, which is thought to limit microbial activity through high osmotic pressures in the solution, potentially being a leading factor in the composition of the microbial community found in these caverns. Furthermore, Muhammed et al. [172] suggest that the presence of brine and sump might modify the cavern's humidity. This could increase moisture levels within the cavern, possibly promoting microbial growth. It is important to mention that, unlike carbon dioxide storage, hydrogen might not be stored for extended periods. Operational conditions are likely to involve the regular usage and replenishment of hydrogen in the storage facility.

Thaysen et al. [173] looked into microbial growth and hydrogen consumption for hydrogen storage in porous media, finding that reservoirs of low salinity and low temperature indicated increased microbial growth rates and hydrogen consumption, with losses of 3% by methanogens and 2–4% by sulfate reducers. Additionally, these factors are thought to further increase with repeated storage cycles which may replenish nutrients through mineral weathering, inflowing water and decaying microbial cells. However, their work also showed that in storage conditions with high temperatures, such as deep reservoirs, microbial life can be excluded but increasing depths will likely lead to greater operational difficulties and further costs [173]. Microbial activities have been observed in five projects listed in Table 1, namely Beynes, Lobodice, Ketzin, HyChico, and SunStorage [87]. While in some underground storage scenarios, it might be beneficial to prevent

Table 2Major hydrogen-consuming reactions [87].

| Process | Reaction | Effective parameters |
|---|--|--|
| Methanogenesis | $^{1/4}HCO_{3}^{-} + H_{2} + ^{1/4}H^{+} \rightarrow ^{1/4}CH_{4} + ^{3/4}H_{2}O$ | High temperature CO ₂ /Carbonate content |
| Acetogenesis | $^{1/2}HCO_{3}^{-} + H_{2} + ^{1/4}H^{+} \rightarrow$ $^{1/4}CH_{3}COO^{-} + 2H_{2}O$ | Less active at pH>7 CO ₂ /Carbonate content Higher active at pH<7 |
| Sulfate reduction | $^{1}/_{2}SO_{4}^{2-} + H_{2} + ^{1}/_{2}H^{+} \rightarrow ^{1}/_{2}HS^{-} + H_{2}O$ | Sulfate/sulfidic mineral content |
| Iron reduction | $2FeOOH + H_2 + 4H^+ \rightarrow 2Fe^{2+} + 4H_2O$ | Iron mineral content |
| Denitrification | $\frac{2}{5}NO_3^- + H_2 + \frac{2}{5}H^+ \rightarrow \frac{1}{5}N_2 + \frac{1}{5}H_2O$ | Nitrate content in water |
| Sulfur reduction Aerobic H ₂ oxidation | $H_2 + S \rightarrow H_2 S$ $H_2 + \frac{1}{2} O_2 \rightarrow H_2 O$ | Sulfur content Oxygen ingress in the system |

microbial activities, in others, such as integrated UHS and CCUS sites, methanogenesis processes can be harnessed to produce green methane. Specifically, the primary objective of hydrogen storage in the HyChico and SunStorage projects (the instances of hydrogen storage in depleted gas reservoirs) was to generate CH_4 by leveraging in-situ microbial activities to consume CO_2 and green hydrogen. As such, the ultimate goal of storage can play a pivotal role in determining site selection criteria [49,174–176].

4.6. Viscoelasticity of salt formations (salt cavern)

A key characteristic of salt caverns for the purpose of large-scale gas storage is the self-sealing nature of salt. In underground conditions rock salt may be considered to exhibit behaviours similar to a viscoelastic fluid, thought to originate from deformation mechanisms observed at microscopic levels such as pressure solution creep and dislocation creep [172,177]. Salt exhibits intrinsic characteristics that make it ideal for storage. It is ductile and has a viscoplastic behaviour under stress, allowing it to recover from induced cracks and faults [178]. This unique behaviour is attributed to salt's near-isotropic stress state that resists hydrofracturing. The complex stress state is influenced by a myriad of factors including depth, geological stress, internal gas pressure, and operational rates [179].

The deformation physics of salt is predominantly nonlinear, stemming from creep processes. A material undergoing creep witnesses timedependent deformation under constant mechanical stress. Salt experiences three distinct phases of creep: primary (transient), secondary (steady), and tertiary. The tertiary phase is particularly notable as it is linked to microcrack formation leading to potential rupture [180]. Salts can undergo creep at temperatures between 20 and 200°C, emphasizing the thermal sensitivity of the storage caverns [181]. Pressure solution creep is mainly dependent on the applied stress or overburden as well as time, but is also dependent on additional factors such as temperature, grain size, porosity, diffusive transfer and pore fluid composition [182]. Increased pressure solution creep can occur when brine present within the cavity permeates through the excavation damaged zone [183]. Dislocation creep occurs in crystalline materials such as rock salt and involves the movement of dislocations through the crystal lattices, making it more dominant at high rates of deformation but is also dependent on stress and temperature [177].

Deformation in salt encompasses two main mechanisms. The first, dislocation creep, relies on dislocation motion within the salt's crystalline grains [178]. This behaviour exhibits stress exponents between 3.5 and 5.5 and activation energies around 60 kJ/mol [184]. In scenarios with minute brine quantities, new crystal formations emerge, subtly augmenting the creep rate. The second mechanism, solution-precipitation or pressure solution creep is more prominent in fine-grained materials under low stresses and temperatures [178,185]. For maintaining cavern integrity, attention must be paid to the site's geological heterogeneities, especially non-salt interbeds, as they can alter permeability and affect steady-state creep [33,186]. The gas temperature within the cavern is subjected to thermodynamic fluctuations which can impact the salt's stress state. Furthermore, careful management of internal cavern pressure is essential to prevent potential microcracking and consequential fatigue failure [187]. Ideal cavern shape can avert potential pitfalls like roof collapses. Deep caverns with a high depth-to-radius ratio and an ovoidal form are preferable. Deeper burial allows for a more significant pressure difference, thus enhancing storage capacity and the density of the stored hydrogen. To ensure the long-term safety and viability of salt cavern storage facilities, predictive models are vital. These models encompass a variety of formulations, from the power-law to the Hampel/Schulze model [178,188,189]. Predictive modelling aids in understanding the long-term implications of stress and deformation, potentially spanning centuries.

Salt caverns, primarily designed for short-term underground hydrogen storage, present unique challenges when subjected to cyclic

loading. The internal pressure of these caverns undergoes significant fluctuations, leading to intricate underground processes that could result in issues like cavern convergence, hydrogen leakage, and salt dilatancy. A crucial factor influencing these challenges is the minimum internal cavern pressure (MICP). Balancing MICP is imperative; while decreasing MICP can lead to structural issues, endlessly increasing it diminishes the cavern's storage capacity [190]. Another aspect of cavern management is the rock salt's low tensile strength, especially problematic during hydrogen storage. Cyclic operations can cause notable temperature swings, resulting in stresses that might exceed the rock salt's tensile strength, leading to microcrack formations [191]. These cracks can potentially enable hydrogen to infiltrate the surrounding rock mass, a problem which can exacerbate with continuous thermal expansion and contraction. Thus, moderating temperature fluctuations and curtailing storage cycle frequency emerge as essential strategies for optimal cavern operation [192,193].

In summary, when aiming for hydrogen storage in salt caverns, salt domes are the preferred choice over bedded salt formations. This is because bedded salt formations contain interlayers which can serve as channels for hydrogen migration during cyclic usage. It's also essential during the construction phase to closely monitor and regulate the temperature of the brine. This oversight prevents unintentional shape modifications of the cavern due to thermal-induced stresses. It's crucial for project managers to define clear objectives before embarking on the construction since the specific shape of the cavern will influence its overall deliverability [73]. While the cavern is in use, it's imperative to keep a close eye on changes in pressure and temperature. This vigilant monitoring ensures that no undue thermal or tensile stresses arise on the cavern walls, which might alter its shape or even cause fractures over time with cyclic operations. To maintain a safety net, the inside operating pressure of the cavern should ideally fall within the range of 24-80% of the overburden pressure of the surrounding rock. Additionally, it's advisable to steer clear of very high operating temperatures and exceedingly low minimum operating pressures [73].

5. UK's hydrogen production targets and storage capabilities

5.1. Hydrogen production targets

The publication of the UK Hydrogen Investor Roadmap in April 2022 showed ambitious targets being made with regard to the production of hydrogen alongside investment commitments necessary to meet these targets. After originally aiming for a 5 GW low carbon hydrogen capacity by 2030, this target was revised up to 10 GW with current estimates predicting that hydrogen demand in the UK could be between 20 and 35% of final energy consumption by 2050 [54]. Fig. 11 breaks this down into the expected ranges of hydrogen demand for key sectors in both 2030 and 2035, highlighting the significant increase of scale expected

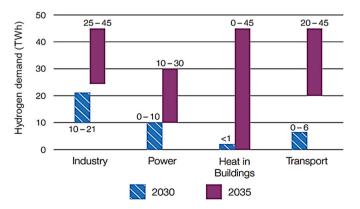


Fig. 11. Forecasted future hydrogen demand (TWh) [54] (Reprinted with permission).

within this period given that the UK currently only produces 10–27 TWh of hydrogen, almost all of which is used outside of the energy system and derived from fossil fuels without the use of carbon capture [194].

In their Future Energy Scenarios document published in July 2021, the National Grid detailed four distinct scenarios pushing ahead to 2050 [195]. They are named as Consumer Transformation, System Transformation, Leading the Way and Falling Short. In the Consumer Transformation scenario, the net zero target for 2050 emphasizes high consumer engagement, with homeowners making significant changes for enhanced energy efficiency. Most electricity demands will be intelligently managed for system flexibility. Typical homes will feature electric heat pumps and low-temperature heating systems, and residents will use electric vehicles (EVs). Peak electricity demands will be tackled using energy storage and smart energy management. Compared to the Consumer Transformation scenario, in the System Transformation scenario, the net zero target is realized by 2050 with most significant changes occurring on the energy supply side. A typical consumer will have a hydrogen boiler and either an electric vehicle or a fuel cell vehicle. Homes will have fewer energy efficiency updates, and there will be less flexibility provided by consumers to the energy system. The primary source of hydrogen will be natural gas, processed with Carbon Capture, Usage, and Storage. In the Leading the Way scenario, the net zero target is expected to be met in 2046, earlier than the first two scenarios. The UK undergoes swift decarbonization through substantial investments in cutting-edge decarbonization technologies. Assumptions regarding various decarbonization areas are accelerated to the earliest feasible timelines. Consumers actively participate in decreasing and managing their energy consumption. The scenario emphasizes extensive energy efficiency upgrades, including homes being equipped with features like triple glazing and external wall insulation, coupled with a surge in smart energy services. Hydrogen, primarily produced from electrolysis driven by renewable electricity, is employed to decarbonise particularly challenging sectors like certain industrial processes. In the last scenario, Falling Short, the net zero target will not be achieved by 2050. Though there's some progress in decarbonization, it lags behind other scenarios. Homes see better insulation, but natural gas remains predominant, especially for heating. Electric vehicle adoption is gradual, primarily replacing petrol and diesel for personal use, while Heavy Goods Vehicles still largely depend on diesel. By 2050, substantial annual carbon emissions persist, failing to meet the net zero objectives [195-198].

The three scenarios which met with the net zero targets but with largely different hydrogen production supplies. The 'Consumer Transformation' scenario had the smallest hydrogen supply at close to 150 TWh, relying on the cost of electrolysis dropping below methane reformation by 2023 and estimating green hydrogen to make up 70% of the total supply by 2050, followed by 23% for blue hydrogen and 7% for nuclear electrolysis. The 'Leading the Way' scenario had nearly double the hydrogen supply at around 280 TWh. This scenario's supply comes almost entirely from green hydrogen and relies on electrolysers being built near where renewable electricity is produced in addition to expected government support to meet their production targets. 'System Transformation' had by far the largest hydrogen supply at 470 TWh but places a heavy significance on blue hydrogen which contributes to 70% of the 2050 supply, with combined biomass gasification and CCUS making up a further 10%. This scenario was also assumed to be the most commercially attractive, as the biomass gasification supply could offset the methane emissions with the incentive of payments for negative emissions [195].

As of April 2023, the total amount of planned hydrogen production in the UK pipeline is estimated at up to 20 GW, the equivalent of 175.2 TWh [66]. Fig. 12 shows the approximate locations of low carbon hydrogen projects across the UK supported through the Net Zero Hydrogen Fund (NZHF), the first Hydrogen Allocation Round and CCUS-enabled projects. Additionally, indicative figures published as part of the investor roadmap indicate the predicted end-use

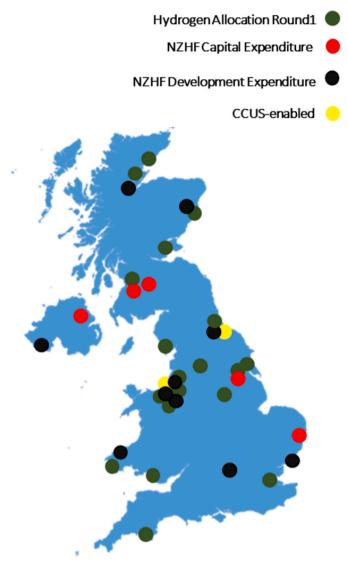


Fig. 12. Map of sample of potential hydrogen projects in the UK based on different funding schemes [66] (adapted from Hydrogen Net Zero Investment Roadmap).

consumption for electrolytic hydrogen based on sector, with 42% going to the mobility sector (road, aviation, maritime), 31% to industry, 15% to heat and the remaining 12% to the power industry [66].

5.2. Storage capabilities

As the production of hydrogen advances in line with the UK's targets, it is vital that there are the necessary storage capabilities in place to support them. Currently, the UK has a natural gas storage capacity of around 15 TWh, considerably less than the capacities of closely neighbouring European countries such as Germany and France, at one seventeenth and one ninth respectively. Contributing factors to this include the UK's prior reliance on North Sea production reserves, the decommissioning of Rough back in 2017 (the UK's largest gas storage facility) and presently choosing to import an approximate 500 TWh of natural gas annually through just-in-time deliveries [36]. This provision is made up of six onshore salt caverns totalling 11 TWh in storage capabilities, and two onshore depleted gas fields which make up the additional 4 TWh [199]. Fig. 13 displays these natural gas facilities in terms of their shared capacities, with green shades representing salt caverns and blue shades indicating depleted gas reservoirs [12].

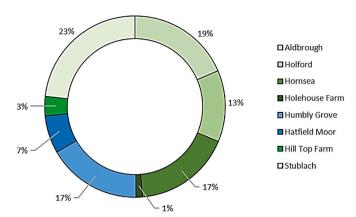


Fig. 13. UK's natural gas storage facilities, green shades represent salt caverns and blue shades indicate depleted gas reservoirs [12]. (Reprinted with permission).

Currently, 40% of the UK's electricity is reliant on natural gas and 85% of homes use gas for heating [200]. However, as the UK transitions away from relying on fossil fuels and pushes forward with hydrogen production, it may be possible to repurpose natural gas storage facilities to instead store hydrogen. According to Wallace et al. [12] this could provide 4.85 TWh of storage capacity, or, roughly a third of the required capacities to reach net zero under the 'Consumer Transformation' and 'Leading the Way' scenarios. Further to this, Centrica (operator of Rough) have put forward plans to repurpose the facility to store hydrogen, claiming that converting the UK's salt cavern natural gas storage facilities alone is not enough to stay on track with the National Grid's projected scenarios. Rough is a large offshore depleted gas field located in the southern North Sea that operated for over three decades. Natural gas production from the Rough field began in October 1975, from a sandstone reservoir approximately 9000 feet deep [72,201]. However, by 1985, after the field's depletion, it transitioned to function as a seasonal gas storage facility. This facility provided for 10% of Britain's peak natural gas demand in 2007 [72,201-203]. For a significant portion of the last decade, Rough accounted for 30 TWh or 70% of UK storage capacity [202].

Amid et al. [202] used numerical simulations to assess the feasibility of hydrogen storage in Rough, wherein green hydrogen gas was compressed and injected at pressures ranging from 5 to 10 MPa. Their findings suggest that Rough can effectively store and supply hydrogen as efficiently as natural gas. Potential losses due to the dissolution and diffusion of hydrogen into underlying aquifers or the pores of overlying cap rocks are projected to be minimal, less than 0.1%. However, the biological reduction of sulfur minerals to hydrogen sulfide was identified as a potential issue [202]. By keeping Rough open in the interim as a blend of natural gas and hydrogen storage, Centrica believes this to be a more cost-effective solution than investing in new storage facilities [204]. Additionally, this would strengthen the UK's security of natural gas supply whilst Rough transitions to 100% hydrogen, all the more relevant given the political pressures and resource constraints as a result of the Russia-Ukraine conflict. Centrica anticipated that 10 to 40 TWh of the total 150 and 600 TWh UK hydrogen demand require storage by 2050 [204,205]. With Rough potential hydrogen storage capacity of 10-15 TWh (a rate-limited capacity for 90 days of withdrawal -WGC-90) Centrical has proposed a £1.6bn repurposing plan in the 2030s. Besides Rough, there are two neighbouring depleted gas fields, Baird and Deborah, which together could contribute an additional 27 TWh of hydrogen storage (WGC-90) [36].

Research carried out by the HyStorPor team and the University of Edinburgh estimates that the UK's hydrogen storage requirement will be much greater than the figures posed in the National Grid's scenario analysis in order to decarbonise the heating sector. Mouli-Castillo et al.

[206] estimate the true storage requirement to be \sim 77.9 TWh, which is roughly 25% of the energy required for domestic heating through natural gas and that fluctuations in seasonal demand are a key contributor within their estimation to maintain balance in the supply of heat. However, even considering this revised storage figure the study presents a new method for comparing the geological storage capacity to estimated need, claiming that the total working gas capacity of UK gas fields could be as great as 2661.9 TWh, 34 times that of which is required (77.9 TWh). Fig. 14 shows a map of the potential storage sites as well as the centroids of the local gas distribution zones. Importantly, the study also notes that at least twelve of the analysed fields can meet the estimated storage need alone. As such, this would allow for other low-carbon solutions to utilize the available storage resource, such as compressed air energy storage (CAES) or carbon storage [206].

The UK would account for 65 TWh of an estimated 500 TWh European hydrogen storage capacity based on the population share (67 of 514 million citizens). According to the results of the HyUSPRe project, the UK has a total hydrogen capacity of 44 TWh (WGC-90) in porous media which would increase capacity to 63 TWh (WGC-90) by including the planned salt caverns development [36].

Another study estimated the UK's continental shelf may contain a working gas capacity of 9100 TWh at a P50 level of confidence, split a between 6900 TWh capacity for gas fields and 2200 TWh for saline

aguifers assuming a working gas requirement of 50% [115]. These figures were derived by applying high-level assumptions and calculations to geological data available through CO2 Stored, an online database developed by the UK Storage Appraisal Project (UKSAP) and commissioned by the Energy Technologies Institute on behalf of the British Geological Survey and The Crown Estate between 2013 and 2018 [125]. The UKSAP project's primary focus was to ascertain the geological storage potential of the UK waters with regards to carbon storage, identifying reservoirs and categorising them into types as well as documenting key characteristics such as stratigraphic age, depth, theoretical storage capacity and a range of additional geological parameters. By having these data readily available, Scafidi et al. [115] were able to filter down from the original 574 entries to instead identify reservoirs deemed capable of storing hydrogen. Of the remaining reservoirs, 95 are gas fields, 70 are saline aguifers with identified structures and 12 are saline aquifers with no identified structure. As a result of having certain aquifers without properly identified structures, in addition to uncertainties surrounding the size and location of useable pore spaces, the study notes that the working gas estimation for saline aquifers is of low confidence and that there are considerable barriers needed to be overcome to achieve an accurate result hydrogen storage estimation using the CO2 stored database. Additionally, in contrast to the 77.9 TWh seasonal storage estimate by Mouli-Castillo et al. [206], this assessment

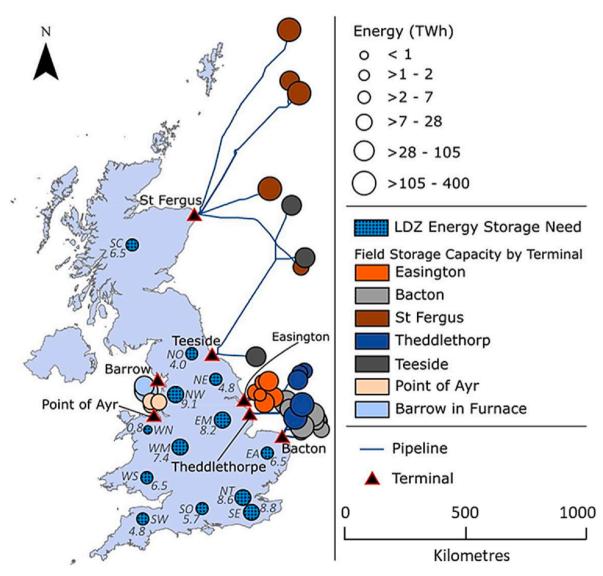


Fig. 14. Map of potential UK gas field storage sites [206] (Reprinted with permission).

predicts the necessary hydrogen storage capacity may be close to double that at 150 TWh, although this figure was considered to be the highest of the forecasted assumptions and assumes a linear relationship between storage capacity and gas demand which may not truly representative given the number of uncertainties moving into the future.

6. Discussion

From the material researched as part of this study, it is clear that the UK considers hydrogen to be a key component in the transition towards achieving net zero and the decarbonization of its energy infrastructure. Despite this, the maturity of hydrogen technologies is still relatively novel both globally and in the UK. As such, the UK is committed to significantly scaling up its hydrogen economy and seems to be wellpoised in helping to lead the way in hydrogen development and establishing itself as a modern-day progenitor in the utilisation of hydrogen as an energy resource. However, significant barriers remain to be overcome to accomplish this and stay on course with the ambitious targets set ahead for 2030 and 2050, the issue of storage being especially pertinent [9,54,66,194]. As previously discussed, a wide range of emerging technological solutions is required to address the storage challenges of hydrogen in both stationary and mobile applications. From compressed gas, liquefaction, and material-based storage for mobile applications to above-ground and underground storage solutions for large-scale stationary applications, all are essential for achieving deep decarbonization of energy systems [16-18].

Salt caverns have the obvious advantage of being a proven resource in the storage of hydrogen to be used as a feedstock in industrial processes, with one of the four existing facilities being operational in the UK's industrial hub of Teesside since 1972 [91]. In terms of the desired geological properties, salt caverns also appear to have dominance over the porous media alternatives. Their rheology, inertness, low gas permeability and small cushion gas requirement are all well-suited to being able to store hydrogen especially pure hydrogen, in addition to their self-sealing nature and mechanical stability which allows for the potential of medium to short-term storage as opposed to just seasonal, making them more adoptable in a wider range of applications [12,77, 114]. The purity of hydrogen is a crucial factor as it directly pertains to the specific application for which it is intended. For instance, cells, proton-exchange membrane fuel also known polymer-electrolyte membrane or PEM fuel cells, are currently being promoted as alternatives to traditional internal combustion engines for a pollution-free solution. However, they demand an exceptionally high purity of hydrogen, specified at 99.97% as per ISO 14687:2019. Moreover, impurities can degrade the performance of PEM fuel cells, potentially causing irreversible damage to the membrane electrode assemblies over time [89]. Conversely, when hydrogen is intended for blending with natural gas, the purity requirements are more lenient, making contamination with other elements a less pressing issue. Williams et al. (2022) showed that hydrogen storage in new caverns in three major basins (Fordon Evaporite Formation of East Yorkshire, the Northwich Halite Member of the Cheshire Basin, and the Dorset Halite Member of the Wessex Basi) could theoretically provide at least 612 GW which is significantly higher than the UK's peak heat demand [99]. However, A substantial increase in the number of caverns (up to around 1000) relative to the current inventory of natural gas caverns will be necessary [92]. Unfortunately, key concerns remain as to caverns local capacity when considering projected hydrogen storage scenarios as well as their geographic availability. In the UK, salt caverns reside exclusively within England, acting as a major constraint for implementation amongst its other countries. According to Wallace et al. [12], this could be particularly problematic for Scotland, which may depend more heavily on hydrogen for heating and where there is a disproportionate number of onshore wind farms, representing a 59% share of the UK's total onshore wind capacity [207]. [92]. With these considerations in mind, additional subsurface storage options will likely be essential in accommodating the hydrogen requirements as part of the net zero pathway. The overall risk of using salt caverns for hydrogen storage is relatively low, given their existing use and minimal research and development needs. However, specific projects face geological uncertainties, including the presence of non-halite interbeds and proximity to salt structure edges, which can compromise cavern integrity. Technical challenges, like well cementation failures, can result in leaks. Furthermore, managing brine disposal, especially far from the sea, can elevate costs and necessitate environmental impact assessments, potentially delaying the permitting process [134].

Both depleted hydrocarbon reservoirs and saline aquifers have more widespread geographical availability across the UK and significantly greater storage capacity potentials when compared to salt caverns. Despite this, the majority of these fields are found in the Southern North Sea basin and are exclusively offshore, although this creates a large potential for the generation of green hydrogen as many of these site locations coincide with areas currently operating or developing wind farms with substantial installed capacities [206]. Given the current levels of wind energy curtailment required to balance energy supply, it is estimated that the UK loses approximately 3.70 TWh of electricity generation annually which could potentially increase to 7.72 TWh if the 40 GW offshore wind by 2030 target is met. Utilizing this to produce hydrogen would help to reduce the associated costs to consumers which were reported at £507 m for 2021 alone and converting this excess of electricity generation could produce up to 4.43 TWh of hydrogen in 2030 [12,208].

Of the two porous media storage options discussed, depleted oil and gas fields appear to have the edge over saline aquifers. By having long relied on these hydrocarbon reserves for industrial expansion, economic growth and energy security, there is already a greater understanding into their storage mechanisms with more studies already carried out on their geological structures. Furthermore, depleted gas fields typically require a smaller cushion gas requirement compared to saline aquifers, although both have sufficient permeability to manage operational flows and a trapping system which helps to prevent fluid migration through leakage [77]. The remnants of resident gas in depleted reservoirs can further help to satisfy the cushion gas requirements, but this can also be viewed as a hindrance in maintaining the purity of the stored hydrogen due to the risk of contamination [100,103]. Additional minor losses (other than the necessary cushion gas requirement) may result from gas escaping through the caprock, efficiency losses through pumping, diffusion into the surrounding groundwater and dissolution into connate water which is thought to be dependent on the extent of fingering [103,

Aquifers require the greatest cushion gas requirements, acting as an instant loss of capital as this gas is then deemed as unrecoverable. Attempted extraction of the cushion gas may cause damage to the reservoir structure, but a more likely problem is that the gas will be trapped in pore spaces and unable to escape the capillary pressure if reservoir heterogeneity is high [103]. Instead, it may be possible to use cheaper gasses as cushion gas, such as carbon dioxide, nitrogen or methane [168]. Further losses are similar to those shared by depleted oil and gas fields, although, in contrast to these hydrocarbon deposits the tightness of an aquifer is less known and therefore requires extensive studies and tests to determine their suitability. As such, this can make the creation of an aquifer storage facility more costly than of depleted fields [12,33]. The HyUnder project presented multiple risks for storing hydrogen in saline aquifers, chiefly concerning the storage container's integrity to prevent leakage and economic loss. In aquifers, risks include potential leakage through the top seal, sideways migration, and along fractures. Additionally, the aquifer's size can only be ascertained post-drilling, posing a project risk if the formation is too small. Biodegradation of hydrogen is a significant technical and economic risk, especially since fuel cells require minimal methane contamination. While some operational risks, like well integrity, are manageable, their mitigation can be costly [134].

Depleted fields, like aquifers, offer limited flexibility, typically allowing only one turnover annually. The rates are influenced by the storage formation's permeability and complexity, as well as the number and performance of production wells for a specific site. Natural gas rates for large storages, like Bierwang operated by E.ON Gas Storage, suggest possible hydrogen flow rates of 8000 to 80,000 kg/h. However, withdrawn gas from these storages can have high water and impurity contents. As such, specific gas treatment processes are necessary to ensure purity [134].

Saline aquifers and depleted hydrocarbon reservoirs have yet to be practically utilised in the storage of pure hydrogen, although both have experience in the successful storage of natural gas [33]. As such, many uncertainties remain as to the long-term effects that the highly pressured storage of hydrogen can have on the environmental geology of a potential site. In a study looking into earthquake triggering as a result of the large-scale geological storage of carbon dioxide, Zoback and Gorelick [209] argue that pre-existing faults are evident in brittle rocks all throughout the Earth's crust and that even slight increases in pore pressure can be enough to cause failure. The triggering of an earthquake could seriously alter the storage integrity of a reservoir, a major concern for hydrogen storage due to its high flammability and difficulty in detecting. This issue may not be much of a concern for the UK which sees little seismic activity but reinstates the significance of site selection and being able to understand the relationship that the large-scale storage of hydrogen and carbon dioxide (needed for blue hydrogen) can have on the surrounding bedrock strata.

Due to differences in the estimation techniques applied, there is a wide disparity between the two studies aimed at evaluating the hydrogen storage potential of the UK. Mouli-Castillo et al. [206] puts the storage potential of UK gas fields at around 2661.9 TWh, whereas Scafidi et al. [115] estimate a much greater potential of 6900 TWh with an additional 2200 TWh capacity from just saline aquifers. Importantly, both studies agreed that individual sites have the capacity to meet the seasonal storage demand, despite also having opposing requirement figures of 77.9 TWh and 150 TWh respectively. This means that only a few sites may be needed to accommodate demand and therefore hydrogen storage would not have to compete for geological space with other gas storage applications, however, this presents an imbalance in site selection as a greater number of potential sites are situated in the Southern North Sea [115,206].

7. Conclusions

The principal objective of this review has been to critically assess the storage capabilities of salt caverns, depleted hydrocarbon fields and saline aquifers in line with the UK's prospective hydrogen targets. While the primary focus of this work is the UK, it's important to note that all the technical, operational, and economic aspects of UHS investigated herein hold global significance. This research extends its relevance both nationally and internationally by highlighting lessons learned from other contexts and identifying applications of the key findings from worldwide projects. This study offers valuable insights applicable not only to the UK but also to the broader global landscape. To achieve this, research has been carried out to investigate the available resources and the key advantages and disadvantages of each storage option as well as the review of government strategies related to decarbonization, forecasted scenarios on future hydrogen production, research works relative to underground hydrogen storage and recent studies focussed on evaluating the hydrogen storage potential. The development of the hydrogen economy, along with the formulation of strategies, policies, and regulations, is a rapidly evolving global process. Large-scale hydrogen projects in salt caverns have seen significant progress, while large-scale hydrogen storage in depleted reservoirs and saline aquifers remains relatively limited. Many analyses in this field draw from comparisons with underground gas storage and experiences from geological CO2 storage projects, as there are few real-world large-scale underground hydrogen storage projects to reference. This shortage of practical experience results in limited measurements, data, and information. Many of the observations and assumptions in this context are based on either lab experiments or numerical simulations, which introduces certain limitations. One significant limitation is the lack of data from the actual UHS project. Additionally, the use of analogies can lead to potential errors in the analysis. Given these challenges, it is crucial to prioritize available storage options based on reliable available data, especially in cases where multiple options are feasible. This approach acknowledges the complexity of hydrogen storage and underscores the importance of making well-informed decisions in the absence of comprehensive empirical data. Revisiting assumptions and conducting fresh analyses upon the acquisition of substantial real-project data is pivotal. This iterative approach guarantees the precision and effectiveness of strategies and policies in the dynamic hydrogen sector. Formalizing the integration of this iterative methodology is proposed. This enables adaptation, the extraction of insights from actual field experiences, and a perpetual enhancement of understanding in underground hydrogen storage. This enduring commitment to improvement is indispensable in managing the intricacies and challenges posed by the hydrogen economy.

The key findings of this research are as follows:

- Three out of four future energy scenarios of the UK which meet the net zero target rely on hydrogen production and supply. As the UK pursues its ambitious hydrogen production goals, it is expected that the UK will need a robust storage strategy and portfolio of different storage options for both stationary and mobile applications. Material-based storage including both liquid and solid carriers are promising solutions for mobile storage applications. However, for large-scale stationary applications, underground storage options including salt caverns and porous media such as depleted hydrocarbon reservoirs and saline aquifers satisfy storage requirements. Individual sites of both saline aquifers and depleted fields are estimated to be able to satisfy forecasted hydrogen storage requirements, reducing competition with other gas storage applications and highlighting the significance of appropriate site selection. Curtailment of wind energy acts as a major loss in potential energy and incurs high losses in associated revenue. This excess power could be utilised in the production of green hydrogen. Colocating storage sites and wind farms would be a key advantage to the improvement of the energy systems and the development of the UK's hydrogen economy.
- The UK's industrial hub of Teesside has successfully stored hydrogen in salt caverns since 1972. Salt caverns have proven experience in the storage of hydrogen especially high purity hydrogen. Their natural sealing properties, low cushion gas requirement and high charge and discharge rates make them favourable short-term storage over depleted fields and saline aquifers; however, their geographical availability within the UK and low total capacity act as major constraints. Additionally, a substantial increase in the number of new caverns (up to around 1000) relative to the current inventory of natural gas caverns will be necessary to address future storage demand. Having said that, storage in porous media can be seen as a long-term and strategic solution to meet energy demand and achieve energy security.
- Depleted hydrocarbon reservoirs and saline aquifers offer broader geographical availability and greater storage capacity potential compared to salt caverns. The Southern North Sea basin, where many of these fields are located, aligns with the region's substantial wind energy generation. Recent revised geological storage capacity claims that the total working gas capacity of UK gas fields could be as great as 2661.9 TWh, 34 times that of which is required, 77.9 TWh.
- There is a broader range of geological data collected about depleted fields over saline aquifers due to prior developments for hydrocarbon recovery. Additionally, remaining gas left over in depleted reservoirs

may be used as cushion gas for a cost-efficient solution, although concerns remain over the risk of hydrogen contamination which may ultimately be dependent on the end-use application of the stored hydrogen. Repurposing of the Rough gas field, which historically contributed to 70% of the UK's storage capacity, could significantly contribute to the UK's future hydrogen storage needs. Research studies indicated Rough's viability in storing hydrogen, with a potential capacity of 10–15 TWh. Coupled with the neighbouring gas fields, this contribution can further burgeon by an additional 27 TWh.

- Recent economic analysis suggests that demand from the power industry, transport (hydrogen fuel cells), and hydrogen-consuming industries will drive the economics of underground hydrogen storage in the future. The economic analysis of different hydrogen storage can be significantly site (location) dependant. One study in the US showed that depleted hydrocarbon reservoirs and aquifers are more economically attractive options than salt caverns, ranging from \$0.04 to \$0.06/kg compared to \$1.61/kg. While the HyUnder project indicated that developing hydrogen storage in aquifers has greater uncertainties and is generally more costly than using salt caverns or depleted hydrocarbon fields. Currently there is no established site selection standard for pure hydrogen storage in depleted oil/gas fields and aquifers. As a result, adapting procedures from natural gas storage and adding criteria specific to hydrogen's subsurface processes can be a great starting point.
- Despite the limited number of operational underground hydrogen projects worldwide, numerous international research initiatives are active in this domain. International collaboration is essential for accelerating the deployment of these projects. By pooling expertise, resources, and research from various nations, such collaboration fosters innovation and more efficiently addresses technical challenges. These joint efforts help establish shared standards and best practices, promoting safety and efficacy. Unified global efforts also reduce costs, optimize supply chains, and encourage the adoption of a sustainable hydrogen-based energy system. However, practical experience with underground hydrogen storage remains sparse on the global stage.

One central message conveyed by this study revolves around the importance of resource availability and the influence of multiple technical, operational, environmental, financial, and political parameters. These factors distinctly shape the priorities and choices in underground hydrogen storage. This study shows that salt caverns should continue to be favoured hydrogen storage solutions but recognises that underground porous media will almost certainly be required to accommodate for future storage requirements necessary to meet the UK's targets for decarbonization and hydrogen production. Of the secondary options available, depleted gas reservoirs hold several key advantages over saline aquifers and it is predicted that they will be the favoured alternative until a greater geological understanding is gained through characterization, modelling and testing.

Authors contributions

Amir Jahanbakhsh: Conceptualization, Methodology, Investigation, Visualization, Supervision, Project administration, Writing - Original Draft, Writing- Reviewing and Editing, Alexander Louis Potapov-Crighton: Methodology, Investigation, Visualization, Writing - Original Draft, Abdolali Mosallanezhad: Investigation, Writing - Original Draft, Writing- Reviewing and Editing, Nina Tohidi Kaloorazi: Investigation, Visualization, Writing - Original Draft, M. Mercedes Maroto-Valer: Supervision, Writing- Reviewing and Editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

all used data in this article was obtained from published literature which have been cited and are available to readers.

Acknowledgements

The authors would like to acknowledge that this work was supported by the UKRI ISCF Industrial Challenge within the UK Industrial Decarbonization Research and Innovation Centre (IDRIC) award number: EP/V027050/1.

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