Exploring the prospects and challenges of underground hydrogen storage for a sustainable energy future

Azlinda Azizi[1*]**,** Nurul Haziqah Abdul Aziz[2], Atikah Kadri[3], Nurul Fadhilah Kamalul Aripin[4], Fazlena Hamzah[5], Nur Sabrina Ahmad Neezam[6], Prashant Jadhawar[7]

[1,2,3,4,5,6]*School of Chemical Engineering, College of Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia*
[7]*University of Aberdeen, King's College, Aberdeen, United Kingdom*

ARTICLE INFO

ABSTRACT

Compared to fossil fuel-based energy sources, renewable energy sources are gaining momentum worldwide due to climate control agreements. The Hydrogen Energy Roadmap proposes to generate hydrogen using renewable energy sources such as hydro, biomass, and solar. However, renewable energy source like hydrogen often has an unstable flow of energy supply, which can lead to temporary underproduction of the required supply. Underground storage options like depleted gas or oil reservoirs, aquifers, and salt caverns are used to address this issue. These underground gas storage alternatives have been used for various applications, including hydrogen storage. Underground hydrogen storage is possible in two geological sites: porous media and cave storage. Salt caverns are suitable for seasonal hydrogen storage at high pressures, while aquifers have the potential for hydrogen storage due to their widespread distribution. However, it is crucial to note that adequate reservoir properties and an impermeable layer are necessary for hydrogen storage in underground structures to prevent gas migration. Microbial and geochemical activities, often overlooked but crucial in hydrogen storage, can pose challenges due to their existence.

1. INTRODUCTION

Worldwide, the primary energy sector has acknowledged a 2.2% increase in demand for the year 2017. Coal, oil, natural gas, and other fossil fuels dominate the global energy supply, accounting for 85% of the total energy production. The increased of global energy demands and strain on global energy supply are resulting from economic expansion and the expansion of global markets (Sáez-Martínez et al., 2016). However, the world's heavy reliance on fossil fuels has generated concerns about energy security, as global

[1*] Corresponding author. *E-mail address*: azlinda68@uitm.edu.my

oil and natural gas reserves are expected to run out in 60 years if they are mined at current rates (Rahman and Wahid, 2021).

Fossil fuel combustion produces greenhouse gas emissions (GHG), contributing to the global warming. GHG emissions at current levels would result in a 4 °C increase in global temperature, resulting in changes such as rising sea levels and shifting climate zones that would affect social, biological, and economic systems (Solomon et al., 2007). Moreover, 195 member countries that took part in the Paris Agreement agreed to aim for global average temperature rises of no more than two degrees Celsius and to strive to limit warming to 1.5 °C above pre-industrial levels. The importance of a low-carbon economy for the country's (Malaysia) long-term future is underlined by its commitment to reduce GDP-related greenhouse gas emissions by 45% in 2030, which is much lower than the expected level of emissions in 2005 (Lian, 2018). It is evident the crucial role of sustainable energy in facilitating this transition.

Renewable sources of energy are gaining attention globally, as a means to fulfill the goals of global climate agreements. Given their minimal emissions of greenhouse gases (GHGs), renewable sources play a crucial role in mitigating the adverse impacts of climate change attributable to fossil fuel consumptions (European Commission, 2020; United Nation, 2015). Solar and wind energy are various existing renewable energy sources. These energy sources rely heavily on factors such as sunlight availability, wind speed, and directions. On the other hand, hydrogen has been viewed as an appealing renewable energy source with significant potential for reducing dependency on imported non-renewable power sources, particularly fossil-based energy sources like oil. Alternative to fossil fuels, hydrogen does not produce unfavourable emissions when burned, making it an attractive option for reducing greenhouse gas emissions and fighting climate change (Mazloomi and Gomes, 2012). Hydrogen is commonly used in various applications, including powering fuel, serving as fuel cells, as a refinery, in agriculture, and more. It has also been proven to be a viable secondary energy source (Seo et al., 2020). The distinctive property of hydrogen is its high energy density, which means that it possesses a lot of energy per unit of mass (Posso et al., 2022). This makes it a possible fascinating fuel source for transportation, where weight is a critical factor. Hydrogen is a flexible energy carrier that can be manufactured from different sources, including natural gas, coal, and renewable sources such as wind and solar (Hassan et al., 2023). Global production and consumption of hydrogen have been growing in recent years as countries seek to transition to cleaner and more sustainable energy systems. Fossil fuel-based hydrogen production continues to dominate, but there has been increasing interest in renewable hydrogen production to minimise greenhouse gas emissions.

However, a potential problem with renewable energy, mainly hydrogen, is that the production of renewable energy can be inconsistent, resulting in a shortfall of supply. In the Hydrogen Energy Roadmap (2005-2015), sustainable hydrogen production sources such as hydro, biomass, and solar are anticipated for the deployment of a hydrogen infrastructure for hydrogen fuel cell vehicles. These sources have rekindled interest in hydrogen generation and storage. As a result, a proposed concept to balance the energy shortfall is to gather excess energy in hydrogen and install an underground hydrogen pipeline. Moreover, the potential storing of hydrogen energy is economically viable since it may be kept as a high-pressure gas, a liquid, or through improved chemical and physical storage techniques (Akademi Sains Malaysia, 2017). This potential storing of hydrogen energy allows Renewable Energy (RE) to be stored in hydrogen form and converted to electricity when required. One of the critical strategies in harnessing hydrogen's potential is underground storage in sedimentary formations — in depleted hydrocarbon reservoirs, aquifers, and even decommissioned wellbores (Kalam et al., 2023).

2. HYDROGEN STORAGE METHODS

Generally, hydrogen can be stored in various ways such as physical storage as compressed gas, physical storage as cryogenic liquid hydrogen and materials-based storage or solid-state storage (Zhang et al., 2016). Fig. 1 below summarises various strategies for storing hydrogen.
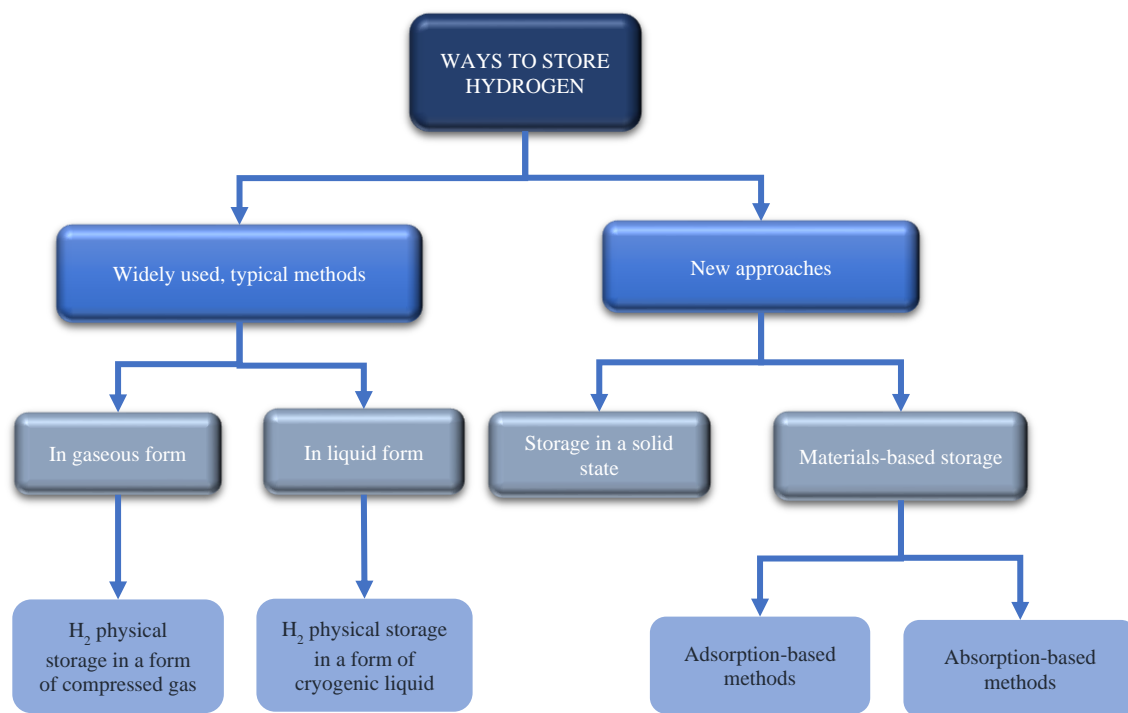

Fig. 1. A variety of strategies for storing hydrogen

Source: Author's illustration

2.1 Compressed gas

Along with hydrogen's low density (0.089 kg/m^3), storing and transporting are challenging tasks to accomplish in relation to its volume. The most proven hydrogen storage method is physically storing compressed hydrogen gas storage (Prachi et al., 2016; Salameh, 2014; Sheriff et al., 2014; Ozturk & Demirbas, 2007). At the moment, current fuel cell programs demand hydrogen to be pushed to a pressure of between 35 and 70 MP. According to anecdotal evidence, pressurising affects between 11% to 13% of hydrogen energy composition negatively (Pesonen & Alakunnas, 2017). Because of its extraordinary lightness, hydrogen can escape from containment vessels under high pressure. Aluminium and steel are the usual materials for commercialised hydrogen storage containers.

Furthermore, carbon fibre-enhanced plastic composite vessels with suitable strength and shock resistance for incident safety are a lighter option. Still, they are extremely expensive, creating another issue for future cost reductions when compared to aluminium or steel vessels (Salameh, 2014), (Sheriff et al., 2014; Mazzolai, 2012; Pesonen & Alakunnas, 2017; Janot et al., 2005; Jorgensen, 2011; Hirscher et al.,

2010). Additionally, engineers and scientists are investigating more cost-effective and viable alternatives. If the amount of hydrogen to be stored is substantial or the storage period is lengthy, pressurised hydrogen can be stored in huge underground storages such as caves or salt domes.

For an extended period, there have been no sufficient infrastructures for burning pure hydrogen successfully, but these infrastructures are now being built (Pesonen & Alakunnas, 2017; Iordache et al., 2014c; Deveci, 2018; Schitea et al., 2019; Wolf, 2015; Ozarslan, 2012; Iordache et al., 2014b; Iordache et al., 2019). One recent example is the hydrogen subsurface storage project in Romania, a component of the European assessment program entitled HyUnder and funded by the FCHJU (Fuel Cell and Hydrogen Joint Undertaking). This initiative aims to facilitate the subterranean storage of sufficient hydrogen in salt caverns for future use, particularly in the transportation sector, the chemical industry and the salt industry (Schitea et al., 2019; Iordache et al., 2014b; Iordache et al., 2014a; Iordache et al., 2019). Considering potential choices, the low hydrogen density, incredibly high gas pressures, expense, and system safety concerns continue to be substantial impediments to this technically straightforward and well-established technology (Prachi et al., 2016; Hwang & Varma, 2014; Züttel, 2003; Niaz et al., 2015; Jia et al., 2015).

2.2   Cryogenic liquid

Another approach to storing hydrogen physically is using cryogenic liquids. Liquid storage has a higher density. At its usual boiling point of 20 K, liquid hydrogen does have a density of roughly 71 g/L, which would be around 1.8 times the density of hydrogen pressured to 70 MPa at 288 K. Due to liquid hydrogen's weak boiling point, its cooling technology needs extremely low temperatures that use around 30% of its overall energy content (Webb, 2015; Midilli et al., 2005). As a result, unique double-walled vessels equipped with effective insulating systems are required to minimise heat loss.

With that being the case, cryogenic pressure vessels are more condensed and lighter than pressurised hydrogen pressure vessels, providing more excellent safety features. Conversely, the prolonged boil-off of hydrogen and the high energy necessary for liquefaction limit the possible utilisation of liquid hydrogen storage systems with implementations requiring high energy density, including those in which the expenditure of hydrogen is irrelevant. Consumption occurs quickly, such as space, air and automotive applications (Schitea et al., 2019; Midilli et al., 2005; De Jongh & Adelhelm, 2010; Mazloomi & Gomes, 2012; David, 2005).

It is challenging to store hydrogen in either solid or liquid form. Because of hydrogen's relatively low density in the gaseous phase (0.09 kg/NA m^3) and extraordinarily high density (70.9 kg/NA m^3) when liquid, traditional storage techniques for stationary and mobile uses have considerable limits. Other issues limit the approaches that are now in use. Condensation loses 40% of its energy, and compressing gas can lose up to 20% of its hydrogen content because of the significant energy loss. A further crucial issue is a requirement to restrict the use of cryogenic and high-pressure storage, primarily due to the unfavourable social factors associated with the usage of gas under pressure and the difficulties in controlling the escape of liquid H$_2$ (Edwards et al., 2008). Fig. 2 depicts the most used methods of storing hydrogen storage.

However, more extensive research is required to improve storage materials' hydrogen absorption and desorption characteristics. Physical or chemical adsorption at volume densities greater than liquid hydrogen is the most promising method for storing hydrogen. (Sharma & Ghoshal, 2015). From Fig. 2, hydrogen storage technologies can be summarised into three main categories: (1) hydrogen may be stored as a gas or a liquid in pure, molecular form, excluding any significant physical or chemical bonding to other materials; (2) molecular hydrogen may be adsorbed onto or into a material, held by relatively weak physical van der Waals bonds; (3) atomic hydrogen may be chemically bonded (absorbed). Further, storage technologies based on chemical bonding can be categorised into two subcategories: metal hydrides and chemical hydrides. This division makes sense due to these materials' fundamentally different hydrogen storage properties.
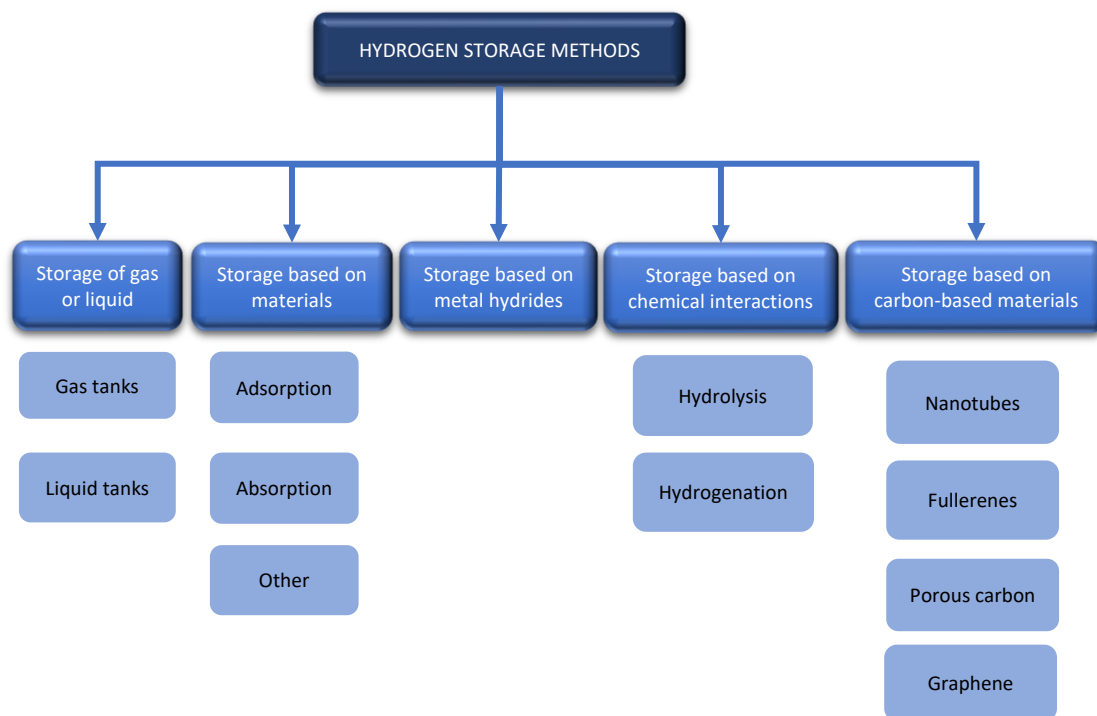
Fig. 2. Widely used methods of hydrogen storage

Source: Author's illustration

2.3   Solid-state Storage System

Storage systems must be highly secure, efficient, affordable, light, and small to support a hydrogen economy (Hwang & Varma, 2014; Ozturk & Demirbas, 2007; Prachi et al., 2016; Wolf, 2015; Zhang et al., 2015; Edalati et al., 2018; Zhang et al., 2017; Sleiman & Huot, 2017). On the other side of the coin, as previously stated, conventional pressurised hydrogen gas and cryogenic liquid hydrogen demand a large amount of space in a bulky storage system, which poses safety concerns, and are therefore extremely expensive; thus, they fall short of meeting future goals for a hydrogen economy (Zhang et al., 2015; Eftekhari & Fang, 2017; Sadhasivam et al., 2017; Shan et al., 2009). Hydrogen storage, compared to pressurised hydrogen gas and cryogenic liquid hydrogen, clearly requires a more significant technological leap. Mainly, for a fuel cell car, sufficient hydrogen storage onboard is a considerable challenge to overcome. A hydrogen storage system is anticipated to have a volumetric and large gravimetric capacity in order to actualise commercially viable hydrogen-powered vehicles.

As shown in Fig. 3, a great deal of attention is currently being dedicated to storing hydrogen in solid-state materials to accomplish this objective (Kaya et al., 2021). Solid-state materials can collect and emit hydrogen reversibly, making them a more advantageous option because they are not consumed immediately (Prachi et al., 2016; Salameh, 2014; Mazzolai, 2012; Webb, 2015; Zhang et al., 2015; Eftekhari & Fang, 2017; Sahaym & Norton, 2008; Zhang & Wu, 2017; Lim et al., 2010). The development of innovative solid-state devices for hydrogen storage can escort a dramatic shift in the present flawed framework of hydrogen storage and have a significant influence on the road to a functional hydrogen economy (Zhang et al., 2015; Zhang & Wu, 2017; Lim et al., 2010; Crabtree et al., 2004).
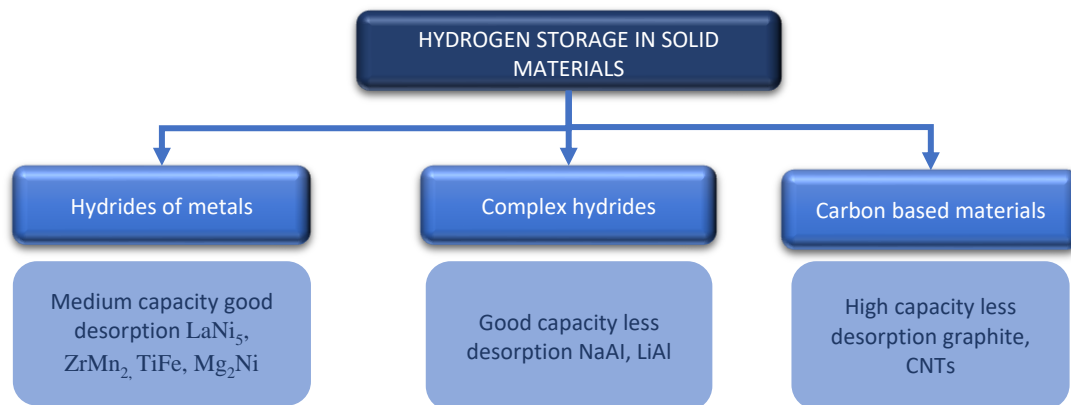
Fig. 3. Approaches for hydrogen storage in solid materials

Source: Author's illustration

Hydrogen is stored in solid-state storage systems via physisorption or chemisorption (Prachi et al., 2016; Salameh, 2014; Mazzolai, 2012; Jia et al., 2015; Eftekhari & Fang, 2017; Sadhasivam et al., 2017). Physisorption and chemisorption of hydrogen are regarded as the safest methods of storing hydrogen in solid form. There are two types of solid-state hydrogen storage: complex hydrides and metal hydrides. Palladium was the first metal identified to absorb hydrogen, allowing it to store a substantial amount of hydrogen at ambient temperatures. Hydrogen storage applications involving intermetallic compounds were later investigated. In comparison to cryogenic liquid hydrogen and compressed gas hydrogen, the volumetric hydrogen density of metal hydride types AB_5 (e.g., $LaNi_5$), AB_2 (e.g., $ZrMn_2$), A_2B (e.g., Mg_2Ni), AB (TiFe) is higher. In this case, the reversibility of hydrogen sorption is adequate to allow for adequate hydrogen recovery. Because heat is used to desorb hydrogen from metal hydride, it limits the risk of leakage. Hydrogen can also be stored in complex metal hydrides like lithium, sodium, and potassium borates and alanates. Relatively speaking, the hydrogen storage capacity of complex hydrides is much higher than that of metal hydrides.

In physisorption, molecular hydrogen is adsorbed on the surface of solids via van der Waals interactions, as in carbon-based materials such as fibres, carbon nanotubes, fullerenes, zeolites, activated carbon, covalent organic frameworks (COFs), metal-organic frameworks (MOFs) and lately, intrinsic microporosity polymers (PIMs). Thermal stimulation or other acceptable approaches can then free the hydrogen whenever required. While the reversibility and rapid kinetics of all these materials make them attractive candidates, their low hydrogen storage volume at ambient temperatures. Meanwhile, the requirement for low temperatures to achieve high hydrogen storage capacity imposes severe limitations on their use in practical applications (Niaz et al., 2015; Jia et al., 2015; Webb, 2015**;**, Zhang et al., 2015; Lim et al., 2010**;** Chamoun et al., 2015; Wagemans et al., 2005, Züttel, 2003; Lai et al., 2015).

As for Chemisorption occurs when atomic hydrogen reacts chemically with solids to generate hydrides (complex metal and chemical hydrides). While some prototype vessels (with $NaAlH_4$ and $Mg\ (NH_2)_2$-LiH) for complex hydrides have been established and examined with recorded high energy densities, its complexity of dehydrogenation and hydrogenation reactions of complex hydrides, as well as the lack of reversibility, currently prohibit potential applications (Hwang & Varma, 2014; Jia et al., 2015; Webb, 2015; Lai et al., 2015; Lim et al., 2010; Babu et al., 2014). Hydrogen storage capacity in solid-state alternatives is shown in Fig. 3 at near-ambient conditions. A complex hydride has a high heat of reaction but a low thermal conductivity. Physisorption can also be used to store hydrogen as a solid. Because of their porous microstructure, large specific surface area (SSA), and low mass density, these carbon materials have a high

hydrogen storage capacity (Sharma & Kumar, 2018; Sharma & Kumar, 2017). Although the metal hydrides may yield a substantial amount of hydrogen, the chemisorption mechanism that occurs when hydrogen molecules and metal hydrides interact usually results in high enthalpies (Sharma & Kumar, 2017). As a result, reversibility is hindered because of the high temperature of materials arising from reaction enthalpy (Umegaki et al., 2009, MacDonald and Rowe, 2006, Sakintuna et al., 2007). When hydrogen molecules engage with a storage material by physisorption, the amount of energy generated is significantly smaller than when hydrogen molecules connect with the storage material via chemisorption (Schimmel et al., 2003; Panella et al., 2005). This shows that the future of hydrogen's economy relies heavily on finding an efficient and affordable storage technology for various uses.

Studies in the literature dictate the hydrogen-storage capacity of a wide variety of different types of carbon nanostructure, such as a variety of activated carbon, carbon nanotubes, carbon nanofiber, and carbon nano horn. Physisorption and/or chemisorption are two methods by which these materials can store hydrogen. Micropores (pores smaller than 2 nm), mesopores (pores between 2 and 50 nm), and macropores (pores more than or equal to 50 nm) all have a role in hydrogen adsorption on carbon materials. The total capacity of physisorption-based hydrogen storage can be found by adding the amount of hydrogen that is adsorbing on a solid surface and the amount of hydrogen that is compressing the slit pores together. When hydrogen adsorbed phase density is more remarkable than hydrogen unadsorbed phase density above the critical point, hydrogen adsorption occurs. The hydrogen storage capabilities of adsorbent material could be decided using micropore SSA.

Schimmel et al. (2003) found that for any carbon material, the bond between hydrogen and material isn't very strong, and low adsorption energy meant that hydrogen could not be bound to nanotubes in the limited interstitial channels between them because it would have too much energy to do so. This means that the more surface area there is, the more storage space there will be, like with ACs (activated carbon). Panella et al. (2005) discovered that hydrogen adsorption depends on the SSA, regardless of the operating temperature or type of carbon material used. Agarwal et al. (1987) also said that the total capacity increases with SSA if the surface is changed a little. Noh et al. (1987) found that oxygen treatment makes the surface acidic, increasing hydrogen storage capacity but not changing SSA. By analysing the hydrogen storage principle and performance of various types of hydrogen storage technologies, current status of hydrogen storage systems volumetric and gravimetric hydrogen density of existing developed hydrogen storage systems with respect to US DOE targets is shown in Fig. 4 below.
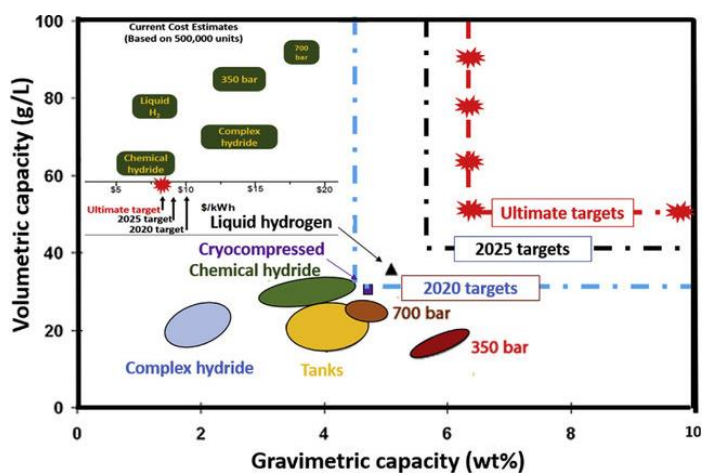

Fig. 4 Current status of hydrogen storage systems volumetric and gravimetric hydrogen density of existing developed hydrogen storage systems with respect to US DOE targets

Source: US Department of Energy, 2017; Read et al., 2007

3.   UNDERGROUND HYDROGEN STORAGE

Currently, underground gas storage alternatives include depleted gas/oil reservoirs, aquifers, and salt caverns, the three most common forms of underground gas storage choices. Underground gas storage alternatives such as those stated above are employed for various gas storage applications, including CO_2 storage, methane storage, town gas storage and, more recently, hydrogen storage. CO_2 storage is the most common sort of underground storage, and it has been implemented with CO_2 being deposited in all the types of underground storage sites indicated above.

Various other storage solutions, such as lined hard rock caverns, abandoned coal mines, and refrigerated mining caverns, are projected to gain popularity throughout the coming years as the need for hydrogen gas storage increases, particularly in places without aquifers, depleted reservoirs, or salt deposits. There are two categories of suitable geological sites in underground hydrogen storage:

i)   porous media, whereby the gas is confined inside the pores of sandstones or carbonate formations and

ii) cavern storage, whereby the gas is trapped within excavated or solution-mined cavities in a dense rock.

3.1   Depleted oil/gas reservoirs

Since the beginning of the Industrial Revolution, oil reservoirs and depleted gas have been the most obvious and widely employed reservoirs for natural gas storage. Depleted reservoirs are oil fields and old gas thousands of miles underground, and most of their recoverable resources have been withdrawn. From a geological standpoint, the reservoirs have demonstrated their ability to keep gas because they previously held hydrocarbons that migrated upwards from the underlying source rock. Nevertheless, some cautionary factors should be considered. After a few occurrences, reservoirs that originally contained gas were found to have been constantly depleted of gas across geologic time until the time of production. During this case, when operational pressure is increased, stored natural gas is lost (Foh et al., 1979). To have an effective gas storage container, reservoirs must have an efficient trap that keeps the gas in the reservoir. For porosity, an appropriate porosity (in relation to reservoir volume) will meet storage requirements. As for the flow, an adequate flow rate in a reservoir will meet operational demands. It is often that effective gas traps are either structural, such as stratigraphical, or an anticline in nature, which includes an impermeable layer, including a caprock (Foh et al., 1979).

Furthermore, to ensure that reservoir pressure and discharge rates are maintained, cushion gas in a reservoir must at least contain 50% of the reservoir volume. Although the producing reservoir has been discontinued, remnants of gas, oil, and water will remain there. Formerly abandoned natural gas could be utilised to satisfy the cushion gas requirements (Foh et al., 1979). It is a financial loss to inject cushion gas when the available natural gas supply is deficient to satisfy cushion gas criteria. Thus, more gas must be supplied to maintain the appropriate pressure. Aside from the loss of gas due to irretrievable cushion gas, the most logical path for gas to escape will be through leaky wells. Even so, the majority of potential minor gas losses can occur via the caprock, gas fingering with reservoir water, breakdown into connate water, contaminating with pre-existing hydrocarbons, and dispersion into the surrounding groundwater (Wagemans et al., 2005; Chamoun et al., 2015).

3.2   Salt caverns

A salt cavern is an underground storage facility suitable for storing various compounds in specialised gases at high pressures. They are typically cylindrical in shape, artificial pits formed in thick subterranean salt deposits by injecting a measured amount of water into a hole drilled into the salt rock. The term used for this operation is Solution mining (Züttel, 2003;Lai et al., 2015). A Salt cavern can be constructed up to

1,000,000 m^3 in volume, 2000 m deep, 50-100 m in diameter, and 300-500 m in height, depending on the conditions and technical feasibility (Michalski et al., 2017). This allows them to store massive volumes of gas. Traditionally, the operating pressure for Underground Hydrogen Storage (UHS) in salt caverns is between 30% and 80% of the lithostatic pressure (Crotogino et al., 2010). Due to its unique geological characteristics, such as its tightness, the advantageous mechanical qualities of salt, and its resistance to chemical reactions, salt caverns are an attractive alternative for hydrogen storage (Tarkowski, 2019). Furthermore, the viscoplastic characteristics of evaporitic rocks contribute to their improved sealing function, and the fact that salt caverns are mechanically robust makes the procedure (injection-withdrawal) versatile and suited for short- and medium-term storage. Similarly, the excessive saline environment inhibits microorganisms' ability to consume hydrogen (Sainz-Garcia et al., 2017). The depth of salt caverns impacts storage capacity. A cave's deeper depth increases pressure and, thus, more compressed gas. On the other hand, less cushion gas is required at a shallower depth, which contributes to the operation's cost savings.

Hydrogen storage within salt caverns has promising potential for seasonal hydrogen storage at high pressures because the hydrogen can be extracted within a reasonable time frame. Salt caves are particularly resistant to hydrogen, even at high pressures, efficiently preventing leaking. Whereby during calm or cloudy weather, when solar and wind energy are inactive, hydrogen might be retrieved from caves and burnt in a combined-cycle power station to produce electricity. A critical challenge is supplying an adequate flow of water in order to leach the cavern and adequately dispose of the brine created in the process. Additionally, technical, and economic variables such as the availability of technological water and distance from pipelines must be taken into account. These caverns (salt) are less expensive to build than other subsurface projects since all activities are carried out from the surface via a single well with sufficient casing and construction to transfer gas dependent on the cavern's area. The methods mentioned above are effective during both the injection and withdrawal phases.

Salt cavern storage facilities are simple to manage, and gas can be injected and retrieved numerous times each year. Among the best options for storing gas reserves during peak periods (Tarkowski, 2019). Considering the capacity constraint, the success of underground cavern storage has already been extensively explored in a variety of published research (Tarkowski & Czapowski, 2018; Liebscher et al., 2016; Stone et al., 2009; Tarkowski, 2019; Michalski et al., 2017).

3.3 Aquifers

Aquifers are permeable and porous mediums through which fresh or saline water can pass and fill the pore spaces. Due to their widespread distribution, aquifers are an excellent alternative for UHS. Numerous gas storage activities in aquifers have been documented in the literature, demonstrating the aquifer's potential for hydrogen storage (Tarkowski, 2019). Hydrogen storage in underground structures such as deep aquifers requires two criteria: 1) adequate reservoir properties of the host rock, and, 2) the availability of an impermeable layer to hinder gas migration (Tarkowski, 2019). As hydrogen is pumped into a water-filled aquifer, the fluid will shift downhill or away to the sides due to its differential in density between liquid and gas. Hence, the pressure of porous media is augmented in this situation. This is due to the addition of hydrogen with the same volume and no liquid drawdown, and as a result, the liquid-gas contact is also altered during the injected operation. In aquifers, when hydrogen is being extracted, liquid can be created synchronously with the gas because of the movement of the gas-liquid interface (Tarkowski, 2019; Sainz-Garcia et al., 2017; Pfeiffer & Bauer, 2015). This is the downside of using aquifers to store hydrogen.

Numerous events can affect the performance of a hydrogen storage facility, including leakage along unsuspected faults, biological reactions, and hydrogen interactions with minerals in the reservoir rock. Furthermore, in comparison to depleted gas and oil reservoirs, critical characteristics such as the structure and geological characteristics of aquifers, such as underground formations, are rarely accessible to identify.

As a result, additional wells and laboratory investigations are required to eliminate any potential risk that might increase the operation's cost.

There is currently no report in the literature on pure hydrogen storage in aquifers. Nonetheless, European cities such as Bad Lauchstädt and Engelbostel in Germany, the Republic, Beynes in France, and Lobodice in the Czech have documented municipal gas storage projects with a mixture of approximately 50% hydrogen and 50% methane (Panfilov et al., 2006; Kruck et al., 2013).

3.4 Microbial and geochemical activities

A detailed investigation into the reactions that occur among injected gas and pre-existing minerals, gases, ions, microorganisms, and other factors is required for a safe (to minimise the likelihood of hydrogen leakage) and effective (to minimise hydrogen conversion into other gas or a decrease in its quality) storage procedure. Hydrogen is an electron donor, making it an energy source for bacteria (Gregory et al., 2019; Reitenbach et al., 2015; Liebscher et al., 2016). Additionally, the hydrogen concentration would be used to influence the reaction rate. Since a large concentration of hydrogen is present in the hydrogen storing procedure, the reaction activity is speeded up.

Two types of processes primarily drive hydrogen generation and consumption in the subsurface: 1) abiotic, and 2) biotic. Abiotic elements are non-living components in porous media, including rock, minerals, water, and gases. In subsurface porous medium, the term "biotic" refers to a living component, which includes bacteria. The temperature required for an abiotic operation might reach 600 °C throughout its operations. Nevertheless, it is smaller for a biotic process at temperatures similar to those in storage sites.

The hydrogen generation during the operation of a hydrogen storage facility has not been described in the literature, and it is unclear whether this could be beneficial. Based on the prospective process for hydrogen generation, nitrogenases are considered to be the most likely process able to produce hydrogen gas as a by-product of nitrogen fixation (Shafaat et al., 2013). This process generates one mole of hydrogen and two moles of ammonia, both produced by consuming one mole of nitrogen (Gregory et al., 2019). Nitrogenases are critical in applications that employ nitrogen gas as a cushion gas. While the abiotic activity happens at high temperatures, which makes it unsuitable for storage locations, methanation has been observed in Turkey's Chimaera seep, as previously stated. In this instance, the transition of hydrogen into methane at 50 °C has been documented (Etiope et al., 2011). Recent research in the literature has revealed that clay minerals can act as hydrogen traps, which is crucial for hydrogen storage operations in porous media such as soils. According to the literature, the existence of clay minerals in the rock mineralogy, including illite, chlorite, and kaolinite, might create entrapment and complicate the storage process (Truche et al., 2018). Due to the numerous microbiological processes associated with molecular H_2, subsurface hydrogen storage can have various adverse effects. Microbial activity is already common in certain industries, such as oil and gas and geothermal, and it may continue to be prevalent during the H_2 UGS. Each microbiological concern is discussed in detail, along with a few critical performance indicators for the various processes. In many circumstances, microbial activity will have multiple effects on the system due to its existence.

4. CHALLENGES IN UNDERGROUND HYDROGEN STORAGE

Hydrogen storage methods are chosen based on the stored volume, the duration of storage, the required speed of discharge, and the geographic availability and cost of the various alternatives (Hematpur et al., 2023). Currently, at the small scale, hydrogen is stored as a gas or liquid in tanks for stationary or mobile applications. Table 1 summarises a comparison of different storage types including the future research and development (R&D) needed (Lord et al., 2014; Cihlar et al., 2021).

Table 1: Comparison for Underground Hydrogen Storage

Storage Type	Depleted Gas Field	Aquifer	Salt Cavern	Lined Rock Cavern
General suitability for hydrogen	Site-Specific	Site-Specific	High	High
Typical type of operation	Seasonal	Seasonal	Peaking	Peaking
Typical number of cycles per year	1-2	1-2	10	10
Working/total gas capacity	50-60%	20-50%	70%	70+%
Depth	300-2700 M	400-2300 M	300-1800 M	1000 M
Operating pressure	1.5-30mpa	3-30mpa	3.5-20 Mpa	2-20mpa
Cost of development (relative)	Low	Low	Low	High
Cost of operation (relative)	Low	Low	Medium	Medium
Largest expenses (new development)	Well Infrastructure, Cushion Gas, Compression	Exploration And Determination Of Geology, Well Infrastructure, Cushion Gas, Compression	Formation Of The Cavern, Disposal Of Brine, Cushion Gas, Compression	Blasting Of The Cavern, Steel Lining, Cushion Gas, Compression
Geographic availability	Most Countries	Most Countries	Limited	Anywhere With Igneous And Metamorphic Rocks
Suitability for hydrogen	Hydrogen-Methane Blending Proven, Pure Hydrogen Storage Under Study	Under Study, But Experience From Depleted Fields Can Be Used	Proven	First Hydrogen Storage In Development (2022)
Suitability factors	Operational Conditions, Fluid And Rock Composition, Bacteriological Activity	Operational Conditions, Fluid And Rock Composition, Bacteriological Activity, Tightness (For New Storage Development Only)	Salt Domes Are Superior To Bedded Salt Structures	Metamorphic Or Igneous Rock; Low Steel Price
R&d needed	Effect Of Residual Natural Gas, Bacterial Reactions	Bacterial Reactions, Tightness Of Rocks	Accuracy Of The Timing Of Injection And Withdrawal	Compatibility Of Lining Materials With Hydrogen

Source: Author's illustration

The knowledge gained by natural gas (methane) storage for decades can be easily transferred to hydrogen (Ozarslan, 2012). The main differences between hydrogen and natural gas storage are the materials required in access wells, the well head and also transmission infrastructure (Ozarslan, 2012). However for hydrogen, embrittlement due to long-term diffusion can cause fracturing, followed by leakage, especially in the steel components, which reduces the strength and stresses that can be safely applied to metal components (Caglayan et al., 2020).

4.1 Microbial-induced plugging

The exopolymers, microbial biomass, or microbially influenced mineral deposits can all promote plugging or clogging of the pore space within rock/pipelines. Proliferation and cell growth in a porous region, such as a pipeline or its connected surfaces, as well as the formation of biofilm structures, results in the clogging of the pipeline or its related surfaces. Given the case of a porous system such as rock or sediment, the reduction in pore space will result in a reduction in permeability and, as a result, a drop in injectivity (Kryachko, 2018). This is frequently most noticeable in the area closest to the injection site, where the concentration of nutrients and cells is often the highest. Because bacteria accelerate redox processes, various mineral deposits can be induced, which might result in clogging, deteriorating injectivity, and other consequences.

Lowered sulphate produces H_2S, which reacts with dissolved ferrous iron and precipitates primarily as FeS crystals (see souring and H_2S formation). Iron-oxidizing microorganisms will precipitate ferric iron minerals such as goethite, ferrihydrite, magnetite, etc., in dissolved iron and either low oxygen content or nitrate. Iron-oxidizers, particularly microaerophilic iron-oxidizers, produce considerable volumes of minerals and biofilms and are frequently responsible for system clogging due to oxygen ingress. Microbial-induced carbonate precipitation (MICP) is another possibility for plugging (Martin et al., 2013).

Carbonate minerals are formed due to chemical changes driven by a wide range of various metabolic processes. For example, nitrate- and sulphate reduction can result in MICP when an extra carbon source is degraded, resulting in the creation of CO_2 in the immediate vicinity of the degraded carbon source and an elevation in pH. This mechanism is frequently connected with biofilms because the restricted diffusion within the matrix promotes the formation of isolated conditions for precipitation. It is possible to detect all plugging occurrences because of reduced injectivity and/or increased injection pressure (Dopffel et al., 2018). Both biomass plugging and mineral precipitation have the potential to be severe and long-lasting. In contrast, mineral deposition is regarded as more stable when compared to mineral deposition and biomass plugging.

4.2 Souring & H_2S formation

Microbial sulphate reduction in the presence of oxygen can generate poisonous and corrosive H_2S gas (Kleinitz & Boehling, 2005). For the microbial activity to take place, sulphate must be present either as dissolved sulphate in the water or as sulphidic minerals (e.g. gypsum, anhydride) (Wolicka &Borkowski, 2007). The presence of H_2 and commonly available carbon sources such as ethanol or organic acids considerably accelerates the process. Apart from the generation of H_2S, a reduction in the sulphate level of the re-produced fluid would be detectable. Usually, community analysis indicates growth in microbes or Proteobacteria with the suffix "Desulfo," such as *Desulfobacterium, Desulfovibrio or Desulfotomaculum*, and sulphate-reducing Archaea. Sulphate removal is a widespread metabolic process in various microbial species (Oren, 2008; Pallud & Cappellen, 2006). Sulphate reduction is a highly efficient process; even small amounts of sulphate can produce a substantial amount of hydrogen sulphide. H_2S will form precipitation with accessible Fe^{2+} in the presence of dissolved ferrous iron or iron minerals. This process is characterised by a decline in dissolved iron and sulphide amounts in the re-produced liquids.

4.3   Gas mixture changes

As stated previously, microorganisms can consume H_2 as part of their metabolism, which can result in a reduction of H_2 content, a rise in other gases, for example, H_2S or methane, and an elevation or reduction in CO_2 concentrations. This could directly affect the utility of the H_2 that has been re-produced. Already low levels of H_2S can have a detrimental effect on several elements of gas quality, along with safety, material integrity and health, necessitating gas treatment. According to the field site activities, the cell counts, and the electron acceptors introduced or available during operations can drop the total H_2 concentration inside the total amount of gas and/or injected gas mixture.

This H_2 leakage will presumably remain constant or growing over the course of the H_2 production or injection cycles, compared to the original H_2 lost due to dispersion, which will be most significant during the first cycle and gradually diminish over time. The simultaneous decrease in CO_2 in the mixture will indicate microbial activity, most likely acetogenesis or methanogenesis. The latter can be characterised by a rise in the content of methane within a generated mix and a change in the microbial habitat toward Archaea, particularly members of the *Methanobacteriales, Methanococcales, Methanosarcinales, Methanopyrales,* and *Methanomicrobiales* families (Basso et al., 2009; Enzmann et al., 2018). Lately, field pilots have already been done to harness this metabolism by feeding commercial H_2 and CO_2, where methanogens may convert to CH_4 (Sato et al., 2013, Strobel et al., 2020).

Additionally, acetogenesis can result in H_2 depletion and simultaneously growing acetate contents in the generated fluids when Bacteria, most frequently *Acetoanaerobium, Acetobacterium, Clostridium,* or *Acetogenium,* are present, as well as a pH decrease (Borja & Rincón, 2017; Zabranska & Pokorna, 2018). The field's high buffer capability and acetate-consuming bacteria can conceal this pH drop in a gas storage tank.

4.4   Steel corrosion by microbes and H_2S

The corrosion of steel infrastructure caused by microbial influences (MIC) is a prevalent concern in various contexts, including water and oil pipelines and underground installations. The general process and its consequences are discussed in greater detail elsewhere (Enning & Garrelfs, 2014; Skovhus et al., 2017). The intricate interaction of abiotic and biotic corrosion activities can result in primarily localised corrosion of steel infrastructure and eventual equipment failure. This mechanism is intimately connected to developing microbial biofilms on metal surfaces. Biofilms are a highly prevalent mode of microorganism emergence in the environment (Donlan, 2002). A biofilm is made up of cells that are encased in a kind of matrix of exopolymeric components such as proteins, sugars, nucleic acids, and, frequently, minerals. As chemical and physical stressors are decreased, the biofilm environment creates a safe environment for the cells. Corrosion frequently occurs beneath a biofilm due to the fact that diffusion within the biofilm can drastically alter redox and chemical variables. Sulphate-reducing methanogens, microorganisms, and acid-producing bacteria are mainly connected with the formation of biofilms and developing corrosion problems. Detecting MIC remains challenging because it is difficult to differentiate from chemical corrosion that exhibits a wide variety of corrosion occurrences and rates (Javaherdashti & Alasvand, 2019).

For the most part, a mixture of corrosion products, corrosion properties, and microbial activity could be utilised to establish whether MIC is present and, if so, in what form. In liquid-phase and/or solid-phase samples, high and growing cell counts of sulphate reducers, specifically sulphate reducers, can be indicative of continuing corrosion (Eckert & Skovhus, 2018 l; El-Shamy et al., 2009). Hydrogen can operate as an electron donor in this process, promoting the formation of microbial biofilms, and it can also be a result of corrosion activities (Atta et al., 2011). Additionally, H_2S produced by bacteria can accelerate corrosion and result in H_2S-induced stress-cracking (Bai et al., 2015).

4.5 Possible effects of H_2 leakage

Numerous reports have reported natural gas leaks from subterranean gas storage facilities (Evans & Chadwick, 2009). Operations failures, well integrity loss, diffusion through the cap rock, and geological cracking can all result in this type of leakage. H_2 has a more significant diffusion coefficient (m^2/s) than other gases, such as CH_4 (Reitenbach et al., 2015), and throughout long-term storage, saturated liquids or H_2 gas mixtures may migrate to higher geological strata, such as groundwater or Earth's surface. The emission of H_2 will have consequences for groundwater and soil microbial communities and the accompanying nutrient cycles, in addition to the visible risk of explosion connected with the emission of H_2. The results of one study (Berta et al., 2018) focused on the issue of response in groundwater throughout the case of an H_2 spillage at alkaline pH8.5–10, whereby accelerated H_2 consumption in association with acetate formation, sulphate reduction, and pH increase observed were shown to be true. If this is true in practice, it would indicate that H_2 plumes would dissipate quickly and that the products created would serve as a helpful tracer for leakage detection.

Additional laboratory studies revealed that acetogenesis no longer occurred at a far more balanced groundwater pH of roughly 7, while formate and CH_4 generation increased. The researchers also discovered that the microbial operations were suppressed over a period or after pH shifts. According to some theories, this could be due to a decrease in the availability of trace elements in the groundwater over time, which could result in a persistent H_2 plume in the groundwater (Lagmöller et al., 2019). Long-term adjustments in the microbiology of topsoil and groundwater caused by H_2 may also result in long-term changes in the chemistry of topsoil and groundwater that should be monitored. The first tests on tracer and particular piezometer measurements were published as part of the ongoing ROSTOCK-H project to identify probable H_2 plumes (Lafortune et al., 2020), which is still ongoing. A greater number of in-depth research on this vital topic is required to gain a better understanding of the potential environmental consequences.

4.6 Dissolution of minerals & change in reservoir properties

Due to the apparent activity of acid-producing microorganisms, also known as acetogens or heterotrophic bacteria, the pH of a reservoir fluid can drop, and the acids created by these microbes can cause the dissolution of carbonate and other weakly dissolvable minerals. This can potentially result in a substantial release of CO_2/HCO_3. The procedure will be supported by an increase in the amount of dissolved CO_2 in the replicated fluids. In addition, the dissolved carbonates can be used as a carbon source by bacteria, resulting in increased development and activity and an increase in the number of cells in the environment. In the case of a carbonate-containing reservoir, the dissolving of the minerals will change permeability and porosity, resulting in a change in fluid flow and flow behaviour in the reservoir. Exothermic enzymatic reactions can occur due to high levels of microbial activity (Bayne-Jones & Rhees, 1929), resulting in the emission of excess heat. It has been hypothesised that a minor temperature increase within a reservoir could indicate high microbial activity. However, this has not yet been seen within an H_2 storage site.

5.   CONCLUSIONS

To conclude, underground gas storage options are crucial for various gas storage applications, including CO_2 storage, methane storage, town gas storage, and hydrogen storage. While depleted oil/gas reservoirs, salt caverns, and aquifers are commonly used for gas storage, new solutions like lined hard rock caverns and refrigerated mining caverns are projected to gain popularity as hydrogen gas storage needs increase. Despite the challenges that hydrogen storage in underground structures faces, including microbial activity, microbial-induced plugging, and steel corrosion caused by microbial influences, these can be overcome through increased R&D funding, collaboration with relevant sectors, altering energy subsidies, and raising public awareness. If the right solutions are found, hydrogen can be an efficient energy storage medium for renewables, contributing to a sustainable future.

CONFLICT OF INTEREST STATEMENT

The authors agree that this research was conducted in the absence of any self-benefits, commercial or financial conflicts and declare the absence of conflicting interests with the funders.

AUTHORS' CONTRIBUTIONS

Azlinda Azizi: Conceptualisation and design, methodology, draft manuscript preparation, supervision, review and editing the manuscript, validation; **Nurul Haziqah Abdul Azizi**: Wrote the paper, related papers collection, formal analysis and summarised the related papers ; **Atikah Kadri**: Project administration and conceptualisation; **Nurul Fadhilah Kamalul Aripin**: Project administration and conceptualisation, review and editing the manuscript, validation; **Fazlena Hamzah**: Project administration and conceptualisation; **Nur Sabrina Ahmad Neezam**: Review and editing the manuscript; **Prashant Jadhawar**: Project administration and conceptualisation.

REFERENCES

Akademi Sains Malaysia (2017). *The blueprint for Fuel Cell Industries in Malaysia*. https://www.akademisains.gov.my/asm-publication/fuel_cell___hydrogen-2017/

Agarwal, R. K., Noh, J. S., Schwarz, J. A., & Davini, P. (1987). Effect of surface acidity of activated carbon on hydrogen storage. *Carbon*, *25*(2), 219–226. https://doi.org/10.1016/0008-6223(87)90119-9

Atta, N. F., Fekry, A. M., & Hassaneen, H. M. (2011). Corrosion inhibition, hydrogen evolution and antibacterial properties of newly synthesized organic inhibitors on 316L stainless steel alloy in acid medium. *International Journal of Hydrogen Energy*, *36*(11), 6462–6471. https://doi.org/10.1016/j.ijhydene.2011.02.134

Babu, A. R. V., Devunuri, N., Manisha, D. R., Prashanthi, Y., Merugu, R. & Teja, A. J. R.R. (2014). Magnesium hydrides for hydrogen storage: A mini review. *International Journal of Chemtech Research, 6*(7)*,* 974–4290.

Bai, P., Zhao, H., Zheng, S. & Chen, C. (2015). Initiation and developmental stages of steel corrosion in wet H_2S environments. *Corrosion Science, 93,* 109–119. https://doi.org/10.1016/j.corsci.2015.01.024

Basso, O., Lascourreges, J-F., Le Borgne, F., Le Goff, C., & Magot, M. (2009). Characterization by culture and molecular analysis of the microbial diversity of a deep subsurface gas storage aquifer. *Research in Microbiology*, *160*(2), 107–116. https://doi.org/10.1016/j.resmic.2008.10.010

Bayne-Jones, S., & Rhees, H. S. (1929). Bacterial Calorimetry II. relationship of heat production to phases of growth of bacteria. *Journal of Bacteriology*, *17*(2), 123–140. https://doi.org/10.1128/jb.17.2.123-140.1929

Berta, M., Dethlefsen, F., Ebert, M., Schäfer, D., & Dahmke, A. (2018). Geochemical effects of millimolar hydrogen concentrations in groundwater: An experimental study in the context of subsurface hydrogen storage. *Environmental Science & Technology*, *52*(8), 4937–4949. https://doi.org/10.1021/acs.est.7b05467

Borja, R., & Rincón, B. (2017). Biogas production. In *Reference module in life sciences* (pp.1–24). http://doi.org/10.1016/B978-0-12-809633-8.09105-6

European Commission (2020). *A strategy for competitive, sustainable and secure energy*, Communication From The Commission To The European Parliament, The Council, The European Economic and Social Committee and The Committee of The Regions.

Caglayan, D. G., Weber, N., Heinrichs, H. U., Linßen, J., Robinius, M., Kukla, P. A., & Stolten, D. (2020). Technical potential of salt caverns for hydrogen storage in Europe. *International Journal of Hydrogen Energy*, *45*(11), 6793–6805. https://doi.org/10.1016/j.ijhydene.2019.12.161

Chamoun, R., Demirci, U. B., & Miele, P. (2015). Cyclic dehydrogenation–(re)hydrogenation with hydrogen-storage materials: An overview. *Energy Technology*, *3*(2), 100–117. **https://doi.org/10.1002/ente.201402136**

Cihlar, J., Mavins, D., & van der Leun, K. (2021). *Picturing the value of underground gas storage to the European hydrogen System.* Gas Infrastructure Europe . https://www.gie.eu/wp-content/uploads/filr/3517/Picturing the value of gas storage to the European hydrogen system_FINAL_140621.pdf

Crabtree, G. W., Dresselhaus, M. S., & Buchanan, M. V. (2004). The hydrogen economy. *Physics Today*, *57*(12), 39–44. https://doi.org/10.1063/1.1878333

Crotogino, Fritz & Donadei, Sabine & Bünger, Ulrich & Landinger, H. (2010). Large-scale hydrogen underground storage for securing future energy supplies. *Proceedings of the 18th World Hydrogen Energy Conferenece,* 37–45.

David, E. (2005). An overview of advanced materials for hydrogen Storage. *Journal of Materials Processing Technology*, *162–163*, 169–177. https://doi.org/10.1016/j.jmatprotec.2005.02.027

De Jongh, P. E., & Adelhelm, P. (2010). Nanosizing and nanoconfinement: New strategies towards meeting hydrogen storage goals. *Chemsuschem*, *3*(12), 1332–1348. https://doi.org/10.1002/cssc.201000248

Deveci, M. (2018). Site selection for hydrogen underground storage using interval type-2 hesitant fuzzy sets. *International Journal of Hydrogen Energy*, *43*(19), 9353–9368. https://doi.org/10.1016/j.ijhydene.2018.03.127

Donlan, R. M. (2002). Biofilms: Microbial life on surfaces. *Emerging Infectious Diseases*, *8*(9), 881–890. https://doi.org/10.3201/eid0809.020063

Dopffel, N., Kögler, F., Hartmann, H., Costea, P. I., Mahler, E., Herold, A., & Alkan, H. (2018). Microbial induced mineral precipitations caused by nitrate treatment for souring control during microbial enhanced oil recovery (MEOR). *International Biodeterioration &Biodegradation*, *135*, 71–79. https://doi.org/10.1016/j.ibiod.2018.09.004

Eckert, R. B., & Skovhus, T. L. (2018). Advances in the application of molecular microbiological methods in the oil and gas industry and links to microbiologically influenced corrosion. *International Biodeterioration & Biodegradation*, *126*, 169–176. https://doi.org/10.1016/j.ibiod.2016.11.019

Edalati, K., Uehiro, R., Ikeda, Y., Li, H.-W., Emami, H., Filinchuk, Y., Arita, M., Sauvage, X., Tanaka, I., Akiba, E., & Horita, Z. (2018). Design and synthesis of a magnesium alloy for room temperature hydrogen storage. *Acta Materialia*, *149*, 88–96. https://doi.org/10.1016/j.actamat.2018.02.033

Edwards, P. P., Kuznetsov, V. L., David, W. I. F., & Brandon, N. P. (2008). Hydrogen and fuel cells: Towards a sustainable energy future. *Energy Policy*, *36*(12), &4356–4362. https://doi.org/10.1016/j.enpol.2008.09.036

Eftekhari, A., & Fang, B. (2017). Electrochemical hydrogen storage: Opportunities for fuel storage, batteries, fuel cells, and supercapacitors. *International Journal of Hydrogen Energy*, *42*(40), 25143–25165. https://doi.org/10.1016/j.ijhydene.2017.08.103

El-Shamy, A. M., Soror, T. Y., El-Dahan, H. A., Ghazy, E. A., & Eweas, A. F. (2009). Microbial corrosion inhibition of mild steel in salty water environment. *Materials Chemistry and Physics*, *114*(1), 156–159. https://doi.org/10.1016/j.matchemphys.2008.09.003

Enning, D., & Garrelfs, J. (2014). Corrosion of iron by sulfate-reducing bacteria: New views of an old problem. *Applied and Environmental Microbiology*, *80*(4), 1226–1236. https://doi.org/10.1128/aem.02848-13

Enzmann, F., Mayer, F., Rother, M., & Holtmann, D. (2018). Methanogens: Biochemical background and biotechnological applications. *AMB Express*, *8*(1). https://doi.org/10.1186/s13568-017-0531-x

Etiope, G., Schoell, M., & Hosgörmez, H. (2011). Abiotic methane flux from the Chimaera seep and Tekirova Ophiolites (Turkey): Understanding gas exhalation from low temperature serpentinization and implications for Mars. *Earth and Planetary Science Letters*, *310*(1–2), 96–104. https://doi.org/10.1016/j.epsl.2011.08.001

Evans, D. J., & Chadwick, R. A. (2009). Underground Gas Storage: An introduction and UK perspective. *Geological Society, London, Special Publications*, *313*(1), 1–11. https://doi.org/10.1144/SP313.1

Foh, S., Novil, M., Rockar, E., & Randolph, P. (1979). *Underground Hydrogen Storage. Final Report. [Salt Caverns, Excavated Caverns, Aquifers and Depleted Fields]*(BNL-51275). Brookhaven National Lab. https://doi.org/10.2172/6536941

Gregory, S., Barnett, M., Field, L., & Milodowski, A. (2019). Subsurface microbial hydrogen cycling: Natural occurrence and implications for industry. *Microorganisms*, *7*(2), 53. https://doi.org/10.3390/microorganisms7020053

Hassan, Q., Sameen, A. Z., Salman, H. M., Jaszczur, M., & Al-Jiboory, A. K. (2023). Hydrogen Energy Future: Advancements in storage technologies and implications for sustainability. *Journal of Energy Storage*, *72*, 108404. https://doi.org/10.1016/j.est.2023.108404

Hematpur, H., Abdollahi, R., Rostami, S., Haghighi, M., & Blunt, M. J. (2023). Review of underground hydrogen storage: Concepts and challenges. *Advances in Geo-Energy Research*, *7*(2), 111–131. https://doi.org/10.46690/ager.2023.02.05

Hirscher, M., Züttel, A., & Schlapbach, L. (2010). *Handbook of Hydrogen Storage*:New Materials for Future Energy Storage*, 202*. https://doi.org/10.1002/9783527629800

Hwang, H. T., & Varma, A. (2014). Hydrogen storage for fuel cell vehicles. *Current Opinion in Chemical Engineering*, *5*, 42–48. https://doi.org/10.1016/j.coche.2014.04.004

Iordache, I., Schitea, D., Gheorghe, A. V., & Iordache, M. (2014a). Hydrogen underground storage in Romania, potential directions of development, stakeholders and general aspects. *International Journal of Hydrogen Energy*, *39*(21), 11071–11081. https://doi.org/10.1016/j.ijhydene.2014.05.067

Iordache, I., Schitea, D., Gheorghe, A. V., & Iordache, M. (2014b). Hydrogen underground storage in Romania, potential directions of development, stakeholders and general aspects. *International Journal of Hydrogen Energy*, *39*(21), 11071–11081. https://doi.org/10.1016/j.ijhydene.2014.05.067

Iordache, M.-D., Bioucas-Dias, J. M., & Plaza, A. (2014c). Collaborative sparse regression for hyperspectral Unmixing. *IEEE Transactions on Geoscience and Remote Sensing*, *52*(1), 341–354. https://doi.org/10.1109/tgrs.2013.2240001

Iordache, M., Schitea, D., Deveci, M., Akyurt, İ. Z., & Iordache, I. (2019). An integrated aras and interval type-2 hesitant fuzzy sets method for underground site selection: Seasonal hydrogen storage in salt caverns. *Journal of Petroleum Science and Engineering*, *175*, 1088–1098. https://doi.org/10.1016/j.petrol.2019.01.051

Janot, R., Latroche, M., & Percheron-Guégan, A. (2005). Development of a hydrogen absorbing layer in the outer shell of high pressure hydrogen tanks. *Materials Science and Engineering: B*, *123*(3), 187–193. https://doi.org/10.1016/j.mseb.2005.07.016

Javaherdashti, R., & Alasvand, K. (2019). *Biological treatment of microbial corrosion: Opportunities and challenges*. Elsevier. https://doi.org/10.1016/C2017-0-04219-2

Jia, Y., Sun, C., Shen, S., Zou, J., Mao, S. S., & Yao, X. (2015). Combination of nanosizing and interfacial effect: Future perspective for designing MG-based nanomaterials for hydrogen storage. *Renewable and Sustainable Energy Reviews*, *44*, 289–303. https://doi.org/10.1016/j.rser.2014.12.032

Jorgensen, S. W. (2011). Hydrogen storage tanks for vehicles: Recent progress and current status. *Current Opinion in Solid State and Materials Science*, *15*(2), 39–43. https://doi.org/10.1016/j.cossms.2010.09.004

Kalam, S., Abu-Khamsin, S. A., Kamal, M. S., Abbasi, G. R., Lashari, N., Patil, S., & Abdurrahman, M. (2023). A Mini-Review on underground hydrogen storage: Production to field studies. *Energy & Fuels*, *37*(12), 8128–8141. https://doi.org/10.1021/acs.energyfuels.3c00841

Kaya, E., Dasdemir, G., Mustafov, S. D., Goksu, H., Gerengi, H., & Sen, F. (2021). Nanocatalysts for hydrogen evolution reactions from hydrazine borane. *Nanomaterials for Hydrogen Storage Applications*, 197–218. https://doi.org/10.1016/b978-0-12-819476-8.00015-3

Kleinitz, W. & Böhling, E. (2005). Underground gas storage in porous media - operating experience with bacteria on gas quality. *All Days*. https://doi.org/10.2118/94248-ms

Kruck, O., Crotogino, F., Prelicz, R. & Rudolph, T. (2013). Assessment of the potential, the actors and relevant business cases for large scale and seasonal storage of renewable electricity by hydrogen underground storage in Europe. *Environmental Science*.

Kryachko, Y. (2018). Novel approaches to microbial enhancement of oil recovery. *Journal of Biotechnology*, *266*, 118–123. https://doi.org/10.1016/j.jbiotec.2017.12.019

Lafortune, S., Gombert, P., Pokryszka, Z., Lacroix, E., Donato, P. De, & Jozja, N. (2020). Monitoring scheme for the detection of hydrogen leakage from a deep underground storage. Part 1: On-site validation of an experimental protocol via the combined injection of helium and tracers into an aquifer. *Applied Sciences*, *10*(17), 6058. https://doi.org/10.3390/app10176058

Lagmöller, L., Dahmke, A., Ebert, M., Metzgen, A., Schäfer, D. & Dethlefsen, F. (2019). *Geochemical effects of hydrogen intrusions into shallow groundwater—An incidence scenario from underground gas storage*[PowerPoint slides]. https://www.uee.uliege.be/cms/c_3483013/en/urban-environmental-engineering

Lai, Q., Paskevicius, M., Sheppard, D. A., Buckley, C. E., Thornton, A. W., Hill, M. R., Gu, Q., Mao, J., Huang, Z., Liu, H. K., Guo, Z., Banerjee, A., Chakraborty, S., Ahuja, R., & Aguey-Zinsou, K. (2015). Hydrogen storage materials for mobile and stationary applications: Current state of the art. *Chemsuschem*, *8*(17), 2789–2825. https://doi.org/10.1002/cssc.201500231

Lian, K. F. (2018). The implications of the Paris Climate Agreement for Malaysia. *International Journal of Science Arts and Commerce,* 3, 27–239.

Liebscher, A., Wackerl, J., & Streibel, M. (2016). Chapter 26–Geologic storage of hydrogen–Fundamentals, processing, and projects. In D. Stolten & B. Emonts (Eds.), *Hydrogen science and engineering: materials, processes, systems and technology* (pp. 629–658). Wiley. https://doi.org/10.1002/9783527674268.ch26

Lim, K. L., Kazemian, H., Yaakob, Z., & Daud, W. R. (2010). Solid-state materials and methods for hydrogen storage: A critical review. *Chemical Engineering & Technology*, *33*(2), 213–226. https://doi.org/10.1002/ceat.200900376

Lord, A. S., Kobos, P. H., & Borns, D. J. (2014). Geologic storage of hydrogen: Scaling up to meet city transportation demands. *International Journal of Hydrogen Energy*, *39*(28), 15570–15582. https://doi.org/10.1016/j.ijhydene.2014.07.121

MacDonald, B. D., & Rowe, A. M. (2006). A thermally coupled metal hydride hydrogen storage and fuel cell system. *Journal of Power Sources*, *161*(1), 346–355. https://doi.org/10.1016/j.jpowsour.2006.04.018

Martin, D., Dodds, K., Butler, I. B., & Ngwenya, B. T. (2013). Carbonate precipitation under pressure for bioengineering in the anaerobic subsurface via denitrification. *Environmental Science & Technology*, 130709154540005. https://doi.org/10.1021/es401270q

Mazloomi, K., & Gomes, C. (2012). Hydrogen as an energy carrier: Prospects and challenges. *Renewable and Sustainable Energy Reviews*, *16*(5), 3024–3033. https://doi.org/10.1016/j.rser.2012.02.028

Mazzolai, G. (2012). Perspectives and challenges for solid state hydrogen storage in automotive applications. *Recent Patents on Materials Science*, *5*(2), 137–148. https://doi.org/10.2174/1874465611205020137

Michalski, J., Bünger, U., Crotogino, F., Donadei, S., Schneider, G.-S., Pregger, T., Cao, K.-K., & Heide, D. (2017). Hydrogen generation by electrolysis and storage in salt caverns: Potentials, economics and systems aspects with regard to the German energy transition. *International Journal of Hydrogen Energy*, *42*(19), 13427–13443. https://doi.org/10.1016/j.ijhydene.2017.02.102

Midilli, A., Ay, M., Dincer, I., & Rosen, M. A. (2005). On hydrogen and hydrogen energy strategies. *Renewable and Sustainable Energy Reviews*, *9*(3), 255–271. https://doi.org/10.1016/j.rser.2004.05.003

Niaz, S., Manzoor, T., & Pandith, A. H. (2015). Hydrogen storage: Materials, methods and perspectives. *Renewable and Sustainable Energy Reviews*, *50*, 457–469. https://doi.org/10.1016/j.rser.2015.05.011

Noh, J. S., Agarwal, R. K., & Schwarz, J. A. (1987). Hydrogen storage systems using activated carbon. *International Journal of Hydrogen Energy*, *12*(10), 693–700. https://doi.org/10.1016/0360-3199(87)90132-7

Oren, A. (2008). Microbial life at high salt concentrations: Phylogenetic and metabolic diversity. *Saline Systems*, *4*(1), 2. https://doi.org/10.1186/1746-1448-4-2

Ozarslan, A. (2012). Large-scale hydrogen energy storage in Salt Caverns. *International Journal of Hydrogen Energy*, *37*(19), 14265–14277. https://doi.org/10.1016/j.ijhydene.2012.07.111

Ozturk, T., & Demirbas, A. (2007). Boron compounds as hydrogen storage materials. *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects*, *29*(15), 1415–1423. https://doi.org/10.1080/00908310500434572

Pallud, C., & Van Cappellen, P. (2006). Kinetics of microbial sulfate reduction in estuarine sediments. *Geochimica et Cosmochimica Acta*, *70*(5), 1148–1162. https://doi.org/10.1016/j.gca.2005.11.002

Panella, B., Hirscher, M., & Roth, S. (2005). Hydrogen adsorption in different carbon nanostructures. *Carbon*, *43*(10), 2209–2214. https://doi.org/10.1016/j.carbon.2005.03.037

Panfilov, M., Gravier, G., & Fillacier, S. (2006). Underground storage of H_2 and H_2-CO_2-CH_4 mixtures. *ECMOR X-10th European Conference on the Mathematics of Oil Recovery,* cp-23-00003. https://doi.org/10.3997/2214-4609.201402474

Pesonen, O. & Alakunnas, T. (2017). *Energy storage : A missing piece of the puzzle for the self-sufficient living*. *Publication series B: Research reports and Compilations Publication Series 12/2017*. UAS Publication Series B. https://urn.fi/URN:ISBN:978-952-316-186-3

Pfeiffer, W. T., & Bauer, S. (2015). Subsurface porous media hydrogen storage – scenario development and simulation. *Energy Procedia*, *76*, 565–572. https://doi.org/10.1016/j.egypro.2015.07.872

Posso, F., Galeano, M., Baranda, C., Franco, D., Rincón, A., Zambrano, J., Cavaliero, C., & Lópes, D. (2022). Towards the hydrogen economy in Paraguay: Green hydrogen production potential and end-uses. *International Journal of Hydrogen Energy*, *47*(70), 30027–30049. https://doi.org/10.1016/j.ijhydene.2022.05.217

Prachi, P. R., Wagh Mahesh M., Gangal Aneesh C. (2016). A review on solid state hydrogen storage material. *Advances in Energy and Power, 4*4(2), 11–22. https://doi.org/10.13189/aep.2016.040202

Rahman, M. N., & Wahid, M. A. (2021). Renewable-based zero-carbon fuels for the use of power generation: A case study in Malaysia supported by updated developments worldwide. *Energy Reports*, *7*, 1986–2020. https://doi.org/10.1016/j.egyr.2021.04.005

Read, C., Thomas, G., Ordaz, G., & Satyapal, S. (2007). U.S. Department of Energy's system targets for on-board vehicular hydrogen storage, *Materials Matters, 2,* Article 2. https://www.sigmaaldrich.com/MY/en/technical-documents/technical-article/materials-science-and-engineering/batteries-supercapacitors-and-fuel-cells/on-board-vehicular-hydrogen-storage

Reitenbach, V., Ganzer, L., Albrecht, D., & Hagemann, B. (2015). Influence of added hydrogen on underground gas storage: A review of key issues. *Environmental Earth Sciences*, *73*(11), 6927–6937. https://doi.org/10.1007/s12665-015-4176-2

Sadhasivam, T., Kim, H.-T., Jung, S., Roh, S.-H., Park, J.-H., & Jung, H.-Y. (2017). Dimensional effects of nanostructured Mg/MgH$_2$ for hydrogen storage applications: A Review. *Renewable and Sustainable Energy Reviews*, *72*, 523–534. https://doi.org/10.1016/j.rser.2017.01.107

Sáez-Martínez, F. J., Lefebvre, G., Hernández, J. J., & Clark, J. H. (2016). Drivers of sustainable cleaner production and sustainable energy options. *Journal of Cleaner Production*, *138*, 1–7. https://doi.org/10.1016/j.jclepro.2016.08.094

Sahaym, U., & Norton, M. G. (2008). Advances in the application of nanotechnology in enabling a hydrogen economy. *Journal of Materials Science*, *43*(16), 5395–5429. https://doi.org/10.1007/s10853-008-2749-0

Sainz-Garcia, A., Abarca, E., Rubi, V., & Grandia, F. (2017). Assessment of feasible strategies for seasonal underground hydrogen storage in a saline aquifer. *International Journal of Hydrogen Energy*, *42*(26), 16657–16666. https://doi.org/10.1016/j.ijhydene.2017.05.076

Sakintuna, B., Lamaridarkrim, F., & Hirscher, M. (2007). Metal hydride materials for solid hydrogen storage: A Review. *International Journal of Hydrogen Energy*, *32*(9), 1121–1140. https://doi.org/10.1016/j.ijhydene.2006.11.022

Salameh, C. M. (2014). *Synthesis of boron or aluminum based functional nitrides for energy applications (Hydrogen production and storage)*(NNT: 2014MON20157. tel-01743835v2) [Doctoral Thesis, Université Montpellier II-Sciences et Techniques du Languedoc]HAL Open Science. https://theses.hal.science/tel-01743835v2

Sato, K., Kawaguchi, H., & Kobayashi, H. (2013). Bio-electrochemical conversion of carbon dioxide to methane in geological storage reservoirs. *Energy Conversion and Management*, *66*, 343–350. https://doi.org/10.1016/j.enconman.2012.12.008

Schitea, D., Deveci, M., Iordache, M., Bilgili, K., Akyurt, İ. Z., & Iordache, I. (2019). Hydrogen Mobility Roll-up site selection using intuitionistic fuzzy sets based WASPAS, COPRAS and EDAS. *International Journal of Hydrogen Energy*, *44*(16), 8585–8600. https://doi.org/10.1016/j.ijhydene.2019.02.011

Seo, S.-K., Yun, D.-Y., & Lee, C.-J. (2020). Design and optimization of a hydrogen supply chain using a centralized storage model. *Applied Energy*, *262*, 114452. https://doi.org/10.1016/j.apenergy.2019.114452

Shafaat, H. S., Rüdiger, O., Ogata, H., & Lubitz, W. (2013). [NiFe] hydrogenases: A common active site for hydrogen metabolism under diverse conditions. *Biochimica et Biophysica Acta (BBA) - Bioenergetics*, *1827*(8–9), 986–1002. https://doi.org/10.1016/j.bbabio.2013.01.015

Shan, Xi & Payer, Joe & Jennings, Wayne. (2009). Mechanism of increased performance and durability of Pd-treated metal hydriding alloys. *International Journal of Hydrogen Energy, 34*(1). 363–369. https://doi.org/10.1016/j.ijhydene.2008.09.040

Sharma, S., & Ghoshal, S. K. (2015). Hydrogen the future transportation fuel: From production to applications. *Renewable and Sustainable Energy Reviews*, *43*, 1151–1158. https://doi.org/10.1016/j.rser.2014.11.093

Sharma, V. K., & Anil Kumar, E. (2017). Metal hydrides for Energy Applications - classification, PCI characterisation and simulation. *International Journal of Energy Research*, *41*(7), 901–923. https://doi.org/10.1002/er.3668

Sharma, V. K., & Kumar, E. A. (2018). Metal hydrides. *Kirk-Othmer Encyclopedia of Chemical Technology*, 1–21. https://doi.org/10.1002/0471238961.koe00039

Sheriff, S. A., Yogi, S. D., Stefanakos, E. & Steinfield, A. (Eds.). (2014). *Handbook of Hydrogen Energy* (1st Edition). CRC Press, Taylor & Francis Group. https://doi.org/10.1201/b17226

Skovhus, T. L., Enning, D., & Lee, J. S. (Eds.). (2017). *Microbiologically influenced corrosion in the upstream oil and gas industry* (1st Edition). CRC Press, Taylor & Francis Group. https://doi.org/10.1201/9781315157818

Sleiman, S., & Huot, J. (2017). Microstructure and hydrogen storage properties of $Ti_1V_{0.9}Cr_{1.1}$ alloy with addition of x wt% Zr (x = 0, 2, 4, 8, and 12). *Inorganics*, *5*(4), 86. https://doi.org/10.3390/inorganics5040086

Solomon, S. , Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M. & Miller, H.L (Eds.). (2007). *AR4 Climate Change 2007: The Physical Science Basis*. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change IPCC. https://www.ipcc.ch/report/ar4/wg1/

Stone, H. B., Veldhuis, I., & Richardson, R. N. (2009). Underground hydrogen storage in the UK. *Geological Society, London, Special Publications*, *313*(1), 217–226. https://doi.org/10.1144/sp313.13

Strobel, G., Hagemann, B., Huppertz, T. M., & Ganzer, L. (2020). Underground bio-methanation: Concept and potential. *Renewable and Sustainable Energy Reviews*, *123*, 109747. https://doi.org/10.1016/j.rser.2020.109747

Tarkowski, R. (2019). Underground hydrogen storage: Characteristics and prospects. *Renewable and Sustainable Energy Reviews*, *105*, 86–94. https://doi.org/10.1016/j.rser.2019.01.051

Tarkowski, R. & Czapowski, G. (2018). Salt domes in Poland – potential sites for hydrogen storage in caverns. *International Journal of Hydrogen Energy*, *43*(46), 21414–21427. https://doi.org/10.1016/j.ijhydene.2018.09.212

Truche, L., Joubert, G., Dargent, M., Martz, P., Cathelineau, M., Rigaudier, T., & Quirt, D. (2018). Clay minerals trap hydrogen in the Earth's crust: Evidence from the Cigar Lake Uranium Deposit, Athabasca. *Earth and Planetary Science Letters*, *493*, 186–197. https://doi.org/10.1016/j.epsl.2018.04.038

Umegaki, T., Yan, J.-M., Zhang, X.-B., Shioyama, H., Kuriyama, N., & Xu, Q. (2009). Boron- and nitrogen-based chemical hydrogen storage materials. *International Journal of Hydrogen Energy*, *34*(5), 2303–2311. https://doi.org/10.1016/j.ijhydene.2009.01.002

United-Nations (2015). United Nations, "Paris Agreement, United Nations Treaty Collection".

US Department of Energy. (2017). *DOE Technical Targets For Onboard Hydrogen Storage For Light-Duty Vehicles*. Office of Energy Efficiency & Renewable Energy, US Department of Energy. https://www.energy.gov/eere/fuelcells/doe-technical-targets-onboard-hydrogen-storage-light-duty-vehicles

Wagemans, R. W., van Lenthe, J. H., de Jongh, P. E., van Dillen, A. J., & de Jong, K. P. (2005). Hydrogen storage in magnesium clusters: Quantum Chemical Study. *Journal of the American Chemical Society*, *127*(47), 16675–16680. https://doi.org/10.1021/ja054569h

Webb, C. J. (2015). A review of catalyst-enhanced magnesium hydride as a hydrogen storage material. *Journal of Physics and Chemistry of Solids*, *84*, 96–106. https://doi.org/10.1016/j.jpcs.2014.06.014

Wolf, E. (2015). Large-scale hydrogen energy storage. *Electrochemical Energy Storage for Renewable Sources and Grid Balancing*, 129–142. https://doi.org/10.1016/b978-0-444-62616-5.00009-7

Wolicka, D., & Borkowski, A. (2007). The geomicrobiological role of sulphate-reducing bacteria in environments contaminated by petroleum products. *Geomicrobiology Journal*, *24*(7–8), 599–607. https://doi.org/10.1080/01490450701672117

Zabranska, J., & Pokorna, D. (2018). Bioconversion of carbon dioxide to methane using hydrogen and hydrogenotrophic methanogens. *Biotechnology Advances*, *36*(3), 707–720. https://doi.org/10.1016/j.biotechadv.2017.12.003

Zhang, B., & Wu, Y. (2017). Recent advances in improving performances of the lightweight complex hydrides Li-mg-n-H System. *Progress in Natural Science: Materials International*, *27*(1), 21–33. https://doi.org/10.1016/j.pnsc.2017.01.005

Zhang, F., Zhao, P., Niu, M., & Maddy, J. (2016). The survey of key technologies in Hydrogen Energy Storage. *International Journal of Hydrogen Energy*, *41*(33), 14535–14552. https://doi.org/10.1016/j.ijhydene.2016.05.293

Zhang, Y., Li, J., Zhang, T., Wu, T., Kou, H., & Xue, X. (2017). Hydrogenation thermokinetics and activation behavior of non-stoichiometric zr-based laves alloys with enhanced hydrogen storage capacity. *Journal of Alloys and Compounds*, *694*, 300–308. https://doi.org/10.1016/j.jallcom.2016.10.021

Zhang, Yang-huan, Jia, Z., Yuan, Z., Yang, T., Qi, Y., & Zhao, D. (2015). Development and application of hydrogen storage. *Journal of Iron and Steel Research International*, *22*(9), 757–770. https://doi.org/10.1016/s1006-706x(15)30069-8

Züttel, A. (2003). Materials for hydrogen storage. *Materials Today*, *6*(9), 24–33. https://doi.org/10.1016/s1369-7021(03)00922-2