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# Historical Review of Hydrogen Energy Storage Technology

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## Abstract

Hydrogen energy as a sustainable energy source has most recently become an increasingly important renewable energy resource due to its ability to power fuel cells in zero-emission vehicles and its help in lowering the levels of $CO_2$ emissions. Also, hydrogen has a high energy density and can be utilized in a wide range of applications. It is indeed the fuel of the future but, it is still not entirely apparent how to analyze the most successful ways for hydrogen storage based on technological configuration, nature, and efficiency mechanisms. The historical hydrogen storage technologies as they are presented by the current research have been evaluated, analyzed, and examined in this study. The two categories of hydrogen storage systems are physical-based and material-based. The first category involves storing hydrogen as liquid, cold/cryo-compressed, and compressed gas. Chemical sorption/chemisorption and physical sorption/physisorption are the two primary sub-groups of material-based storage, respectively. The quantitative and qualitative analyses of storage technologies for hydrogen are evaluated in this paper. Also, this report reviews the major safety and reliability issues currently facing hydrogen storage systems. Suggestions are made to assist lay the groundwork for future risk and reliability analysis to ensure safe, dependable operation.

## Keywords

Hydrogen Energy, Energy Storage, Sustainable Energy, Hydrogen Storage, Energy Source

## 1. Introduction

### 1.1. Background of Hydrogen Storage

Energy consumption is rising swiftly because of the expanding world population, industrialization, and urbanization [1]. Over 85% of the energy consumed worldwide today comes from coal, natural gas, and oil, which exacerbates already existing environmental (including global warming), economic, and political problems [2]. These resources are in short supply, and as their availability decreases internationally, their costs rise. A significant problem that must be resolved for a sustainable energy future is the depletion of finite fossil resources. Due to its sustainability advantages, renewable energy is becoming increasingly important on a global scale [2]. Due to the need to find an unlimited, infinite renewable source for the future energy supply while also reducing the consequences of climate change, the globe is starting to work towards energy transition. Numerous renewable energy sources, including wind, solar, and nuclear, have recently been researched to help solve these problems [1]. Wind, solar, and nuclear power cannot produce enough energy for a global green economy due to their intermittent nature caused by weather and geographic restrictions [1].

Hydrogen is considered the fuel of the future due to its outstanding energy efficiency and environmental friendliness [3]. The most prevalent element in the universe, hydrogen, can store and transfer electrical energy through chemical processes as opposed to combustion [3]. Additionally, it is easy to use in a range of applications, including heating homes and generating electricity for automobiles [4]. It only produces two by-products: water and heat. Hydrogen has a higher energy content per mass (120 MJ/kg) compared to gasoline [4] [5] as depicted in **Figure 1**. Before it can be stored and transported in a form that can be used for applications, hydrogen must first be created in an economically viable manner [5]. In nature, hydrogen is not a free gas. It requires a primary energy source, which could be either non-renewable (such as fossil fuel) or renewable (such as sun, wind, and biomass energy) [5]. Undoubtedly, one of the primary energy sources of the future will be hydrogen, and the generation of hydrogen from renewable energy sources will be essential to achieving the net-zero emissions objective by 2050 [5].

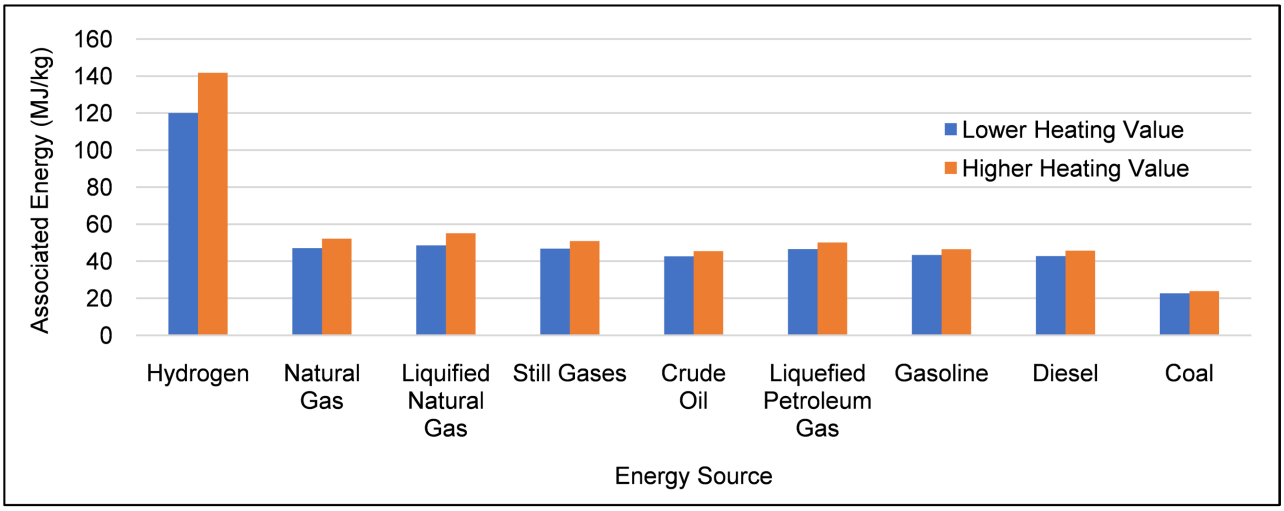

**Figure 1.** Comparison of energy associated with different sources [5].

Hydrogen storage is essential to advancing hydrogen applications in delivering fixed power, transit, and portable power systems. Therefore, very effective and reliable storage technologies would be required to build a clean hydrogen economy. Hydrogen storage is still challenging due to its extremely low density (0.082 kg/m$^3$ at STP) and significant flammability [5]. When storing hydrogen normally in its compressed form, there is always a chance of leakage and explosion [5]. As shown in **Figure 2**, the two major categories of hydrogen storage systems are physical-based and material-based. The first category involves storing hydrogen as liquid, cold/cryo-compressed, and compressed gas. Chemical sorption/chemisorption and physical sorption/physisorption are the two primary sub-groups of material-based storage,respectively [6]. Summarily, the categories of hydrogen storage technologies can be divided into two main groups: physical-based and material-based as given in **Figure 2**.

## 1.2. Problem Statement

There have been several works of literature on the storage of hydrogen, but most of them focus on either the physical storage of hydrogen (in compressed, liquid, or cryogenic tanks), the chemical storage of hydrogen (in sorbents, metals, or chemical hydrides), or various aspects of the hydrogen's storage efficiency.

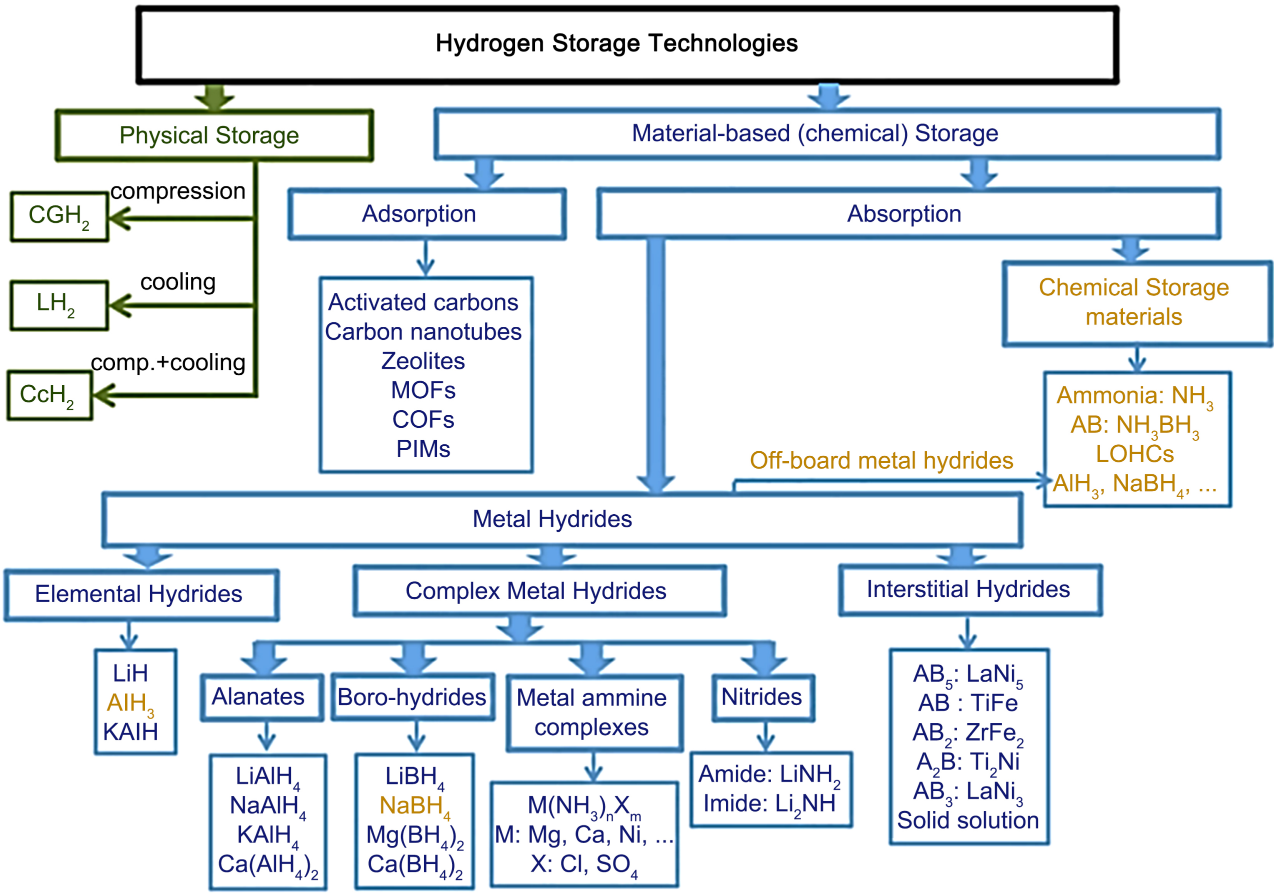

**Figure 2.** Hydrogen storage technologies [12].

However, there are very few research studies that compare the various hydrogen storage strategies to determine the most efficient technology. Industries have had a difficult time deciding which storage technology to use for their hydrogen because there hasn't been enough research in this sector.

### 1.3. Aim and Objectives of the Project

Regarding hydrogen storage, recent research has been observed to use several approaches, however the majority of them provide scant details on the reliability and safety of the mechanism used. We will examine the major hydrogen storage systems in this research, along with risk and reliability evaluations for each process. Additionally, a comparison and selection of the best technology for use in diverse applications will be made using quantitative and qualitative analysis.

## 2. Literature Review

### 2.1. History of Hydrogen Storage

In 2016, Zhang *et al.* stated that, hydrogen is an essential part of the energy supply chain and affects nearly every aspect of energy production, including the reliability of electricity grids, the utilization of renewable energy sources, the preservation of fossil fuels, and the environment. According to research, a combustion reaction that burns one cubic metre of hydrogen yields about 120 MJ/kg of energy (which is relatively high). Since methane has a 44 MJ/kg energy potential, the data show that hydrogen has a substantially lower energy potential [1].

In support of his research analysis, Zhang stressed that $H_2$ is not thought of as an energy source, but rather as an energy carrier because the energy produced from the generation of a unit of $H_2$ is smaller than that consumed. According to findings, hydrogen has been discovered to be an efficient energy carrier since it can be transported and stored as energy, which is backed by study [2]. Further highlighting the importance of hydrogen storage as a part of the hydrogen economy in 2016, Zhang *et al.* stated that developing secure, dependable, efficient, and effective storage techniques is one of the most essential and difficult applications [2]. According to specific study viewpoints by Zhang, there are three typical methods for storing hydrogen, physical storage as compressed gas, and physical storage as cryogenic liquid hydrogen, materials-based storage or solid-state storage.

Research has also shown that the first two methods of storing hydrogen as compressed gas and in liquid form—are the most established and often employed ones. Like this, the goal of a hydrogen storage system is to reach a density of 7.5 weight percent in gravimetric terms and 7.0 weight percent in volume terms, while also lowering the price of hydrogen to $266 per kilogram [2].

In 2022, Kumar *et al.* claimed that hydrogen will advance to become a better energy carrier and an eco-friendly working fluid. The results of his study also confirmed that hydrogen is the third most common element on earth which is a desirable feature that distinguishes it from other conventional fuels [3]. His studies also showed that hydrogen, a pollution-free fuel with a high calorific value, can be a great replacement. However, because hydrogen has a relatively low density (0.082 kg/$m^3$ at STP) and a high flammability, storing hydrogen is still difficult [3].

Mori *et al.* stated in 2009 that to reach a range that is equivalent to that of gasoline-powered vehicles, it is necessary to enhance both the amount of storable hydrogen and its efficiency. To conduct a thorough analysis of the best storage options, Wei *et al.* indicated in 2017 that hydrogen storage technologies should be considered from the energy production to the end-use [5]. In their research analysis from 2014, Anna *et al.* also noted that, in terms of hydrogen storage, hydrogen infrastructure will play a significant role in geologic storage to meet demand and lower costs associated with the storage of hydrogen within the same type of facilities, currently used for natural gas [6].

## 2.2. Development of Storage Materials for Hydrogen Storage

In 2022, Malleswararao *et al.*, stated that Metal hydrides (MH) have been widely investigated for their application in hydrogen storage because they are environmentally benign, can utilize low grade energy and offer high volumetric hydrogen storage densities at reasonable pressures [7]. Furthermore, research analysis by Hirscher *et al.* (2020) affirmed that hydrogen storage by physisorption in porous materials, using classical systems such as activated carbons and zeolites, has a long history. And maximum storage capacities are closely related to the surface area accessible to $H_2$ molecules [8]. However, Hirscher *et al.* (2020) further opined that an entirely new class of highly porous, crystalline materials called coordination polymers or Metal-Organic Frameworks (MOFs) was being developed and promising research results showed a sharp increase in the $H_2$ uptake at very low pressures and a maximum of 4.5 wt% below 1 bar at 77 k [8].

Marinelli *et al.* (2020), reported that Hydrogen can also be stored on surfaces of particular materials (adsorption) or within metallic matrices (absorption), their research analysis further showed that the reversible storage of hydrogen in metal hydrides have high potentials. Additionally, the reversible absorption-desorption pattern of hydrogen storage near ambient conditions was discovered for particular alloys which serve as storage materials, opening the way to a new research of hydrogen storage in intermetallic compounds, also known as hydrogen storage alloy, therefore hydrogen can be stored inside particular metallic matrices, known as hydrogen storage alloys [9]. Hydrogen fuel cell technologies also offer maximum energy storage densities ranging from 0.33 to 0.51 kWh/L depending on the $H_2$ storage method, while the highest value achieved for rechargeable Li-ion batteries does not exceed 0.14 kWh/L, and for pumped hydro the energy density is as low as 0.27 Wh/L [9]. Research results by Hermosilla-Lara (2007) indicated that the hydrogen storage in a solid-state matrix (e.g. metal hydride) has the advantage of being safe since chemisorption process makes in principle the pressure required to store a given amount of hydrogen much lower than what would be required if pure compression storage was used [10].

## 2.3. Methods and Technologies for Hydrogen Storage

Zhang *et al.* (2016), opined that Hydrogen can be compressed and stored as pressurised gas in the cylinders, containers, or even in the underground caverns, with pressure up to 700 bar in suitable cylinders, Zhang went further to state that there are three (3) major types of widely used high pressure hydrogen storage vessel, namely: stationary, vehicular, and bulk transportation [2]. Storing hydrogen in the liquid form can achieve higher density when compared with compressed hydrogen gas storage according to Zhang. Therefore, more energy can be stored per unit volume according to available research results by Zhang *et al.*, (2016). Similarly, in material-based hydrogen storage, hydrogen atoms or molecules are tightly bound with other elements. It is perhaps the most promising hydrogen storage method as it is possible to store large amount of hydrogen within a relatively small volume.

Research carried out by Zhang *et al.* (2016) concluded that produced hydrogen can be stored in compressed gas form or in liquid form, which are quite commonly used in industry. To increase the gravimetric density of hydrogen storage, so it can be more suitable for vehicular applications, material-based hydrogen storage has attracted a lot of research interest. Hydrogen atoms or molecules can be attached to the surface of the materials, or the atoms can be integrated in the lattice of the materials. However, the effectiveness of the storage is strongly dependent on the development of the novel materials so that large amount of hydrogen can be stored and released at a reasonable temperature and pressure.

## 2.4. Categories of Hydrogen Storage

According to Ramin *et al.* (2019) hydrogen storage is a key element in hydrogen energy systems, especially when it comes to large scale utilization of hydrogen. To address the current and potential future demands of hydrogen energy market, having a robust and reliable storage solution for each application is vital [11].

### 2.4.1. Storage of Hydrogen in Gaseous State

Hydrogen is the lightest gas that can occupy significant space under a standard condition of pressure, so to store and transport the hydrogen gas efficiently, the volume of the Hydrogen needs to be reduced significantly by compressing it and storing it in different types of storing media. Storing Hydrogen in its gaseous form is one of the most common, effective, and simplest technology that is being used. Nowadays, gaseous hydrogen gas is being stored by using different methods and technologies.

### 2.4.2. Compressed Hydrogen Storage

The common method of hydrogen storage is storing in thick-walled cylindrical or quasi-conformable tanks, which are made of high-strength material, ensuring the longevity and durability of the tanks. However, the design of the tanks is still not optimized. The tanks are oversized, and materials are used in-efficiently with poor pressure vessel lifetime [13]. According to the European Integrated Hydrogen Project EIHP, compressed hydrogen gas storage vessels are classified into four types, and the most advanced lightweight storage system for storage of compressed gas is an advanced composite tank using a non-load-bearing metallic (Type III) or plastic (Type IV) liner axial and hoop wrapped with resin impregnated continuous filaments [14]. According to Faye *et al.*, A high-pressure gas steel cylinder is the most known method to store hydrogen, with an operating pressure of 200 bar [15]. However, to maximize the storage capacity, new lightweight composite cylinders have been designed to withstand pressures up to 800 bar to enhance the volumetric density of hydrogen to 36 kg/m$^3$ which is approximately half of its liquid form at the standard boiling point of −252.87˚C [16].

### 2.4.3. Application of Compressed Hydrogen

There are many car manufacturing companies working on the development of compressed hydrogen gas storage tanks. Companies like Dynetek Industries working on Type II cylinders, EADS group, Faber, CEA, Ullit, and COMAT are developing Type IV Cylinders. Many car manufacturers in Japan are involved in the research and development of Fuel Cell Vehicles. Recently, Dynetek Industries have reported that they have tested the latest high-pressure cylinders at 825 bar, which will be used for storing Hydrogen for stationary applications [17]. They are made of aerospace-grade aluminium with good sealing performance. Another company, Quantum technologies, under its partnership with General Motors, has claimed to design of 700 bar composite tank which can increase the driving range up to 270 km of hydrogen-powered fuel cell vehicles, with the refueling process taking 5 min at most [17].

### 2.4.4. Benefits and Barriers

However, according to Faye *et al.*, when the transportation sector is considered, conventional hydrogen storage technology such as compressed gas cylinders and liquid tanks still need improvement to achieve a better driving range and performance compared to gasoline-operated vehicles. Talking about the benefits of storing Hydrogen in its gaseous phase, generally, the hydrogen storage is available to be stored up to the pressure of 200 bar, which makes this storage method low in cost; however, only a relatively small amount of Hydrogen can be stored at this pressure. High-pressure storage is still developing [15].

### 2.4.5. Challenges of Compressed Hydrogen

According to, the main areas that still require additional research and development include inexpensive materials for low-pressure storage to avoid hydrogen leakage, new manufacturing processes for mass production of high-pressure tanks at low cost, advancement of sensors to detect the possible leakage and there is a need of codes and standards for end-use requirements [15].

### 2.5. Liquid Storage Form of Hydrogen

The research results by Faye *et al.* stated that storing hydrogen in a liquid form is a practical way to enhance hydrogen energy density. The advantage of the liquefaction process is that high hydrogen storage densities can be attained at atmospheric pressure. For instance, the density of saturated liquid hydrogen at 1 bar is 70 kg/m$^3$, and more importantly, two main liquefaction processes used are the Linde cycle and the Joule-Thomson expansion cycle [16]. In line with this, liquid storage's challenges are significant, the boil-off (hydrogen evaporation because of heat transfer to the liquid) and hydrogen leakage. It also requires a refrigerator and an insulating container to minimize the energy lost during the liquefaction process.

### 2.6. Cryo-Compressed Hydrogen Storage

Faye *et al.* further proposed that cryo-compressed storage means storing hydrogen at cryogenic temperature in a vessel that can be pressurized in the pressure window (250 - 350 atm). In contrast, cryogenic vessels keep liquid hydrogen only around ambient pressures [16]. Furthermore, Cryo-compressed storage is safer because of the double-layer design with an outer vacuum enclosure protecting against chemical and mechanical intrusion, which allows keeping the vessel in a steady non-humid atmosphere [16].

### 2.7. Salt Cavern Hydrogen Storage

In the research carried out by Richard *et al.*, salt cavern hydrogen storage technology makes use of chambers formed through dissolution mining (leaching) of naturally occurring salt formations such as domes or layers (beds). These salt formations tend to be 2000 m above ground surface (bgs) as pressures and temperatures below this level make salt deformation more likely, posing stability issues even for well engineered cavern, it is worthy of note that unlike porous technologies, salt caverns do not require intense consideration of multiphase phenomena that could reduce injection rate as residual water gathers at the bottom of the cavity [18].

It is one of the underground hydrogen methods in which large quantities of hydrogen gas in pressure are stored in caverns and salt domes for many years. Salt caverns are artificial cavities created in geological salt deposits located at a depth of 500 to 1500 meters. Salt rock is suitable because of its good properties like low porosity and permeability, good plasticity, and damage self-healing characteristics. The first project for storing Hydrogen in salt caverns was started back in 1971 in Cologne, Germany, where industrial gas containing 62% of Hydrogen was stored in a salt cavern at a pressure of 8 - 10 MPa. It was 1972 when it began to store pure Hydrogen instead of mixtures in the underground salt caverns.

Many types of research have been carried out on the stability and tightness of salt cavern gas storage. Chen *et al.* [19], ascertained that the cause of tightness failure of the wellbore is due to the leakage in interlayer between salt cavern and casing shoe. In bedded salt rock strata, the surrounding rock often contains non-salt interlayers with various lithologies, but previous research considered that the lithologies were consistent. The porosity and permeability of salt caverns differ depending on the different lithologies of interlayers. Therefore, the effects of lithological diversity of the interlayers are necessary when evaluating the tightness of salt caverns [19]. According to research done by Zhang *et al.*, the lithologies of interlayers in bedded salt rocks of China are diverse, so their porosity and permeability are also different. With the increase in the buried depth of interlayers, these features decrease. In an earlier phase, hydrogen gas leakage increases with time because of the pore pressure but later gest stable. Therefore, the porosity and permeability play a key role in the tightness of the caverns, and $H_2$ leakage and reflux depend on this [20].

## 2.8. Aquifer Hydrogen Storage

Research analysis as conducted by Richard *et al.* (2021) stipulated that aquifer storage technology utilizes the inherent porous nature of subsurface rocks which occur in sedimentary basins across the world. The aim is to replace this water occupied porous spaces with hydrogen gas. This is accomplished at injection pressures greater than reservoir capillary pressure and less than that of the caprock capillary pressure. This is to allow evacuation of water within reservoir pores throats while preventing leakage from caprock [18].

### 2.8.1. Saline Aquifer

To supply energy in the GWh/TWh for a range of months, surface hydrogen storage facilities such as pipeline or compressed vessels is not applicable, so underground storage is needed. When it comes to underground hydrogen storage, there are two main options first one is the man-made salt caverns in thick evaporitic formations, and another is deep power formations such as saline aquifers or depleted oil fields. Deep Saline aquifer is distributed worldwide, considered to have the capability to provide larger storage capacity (hundreds of $Mm^3$), and has a potential to meet demand fluctuations for a longer period of time [21]. Though aquifers accumulate the majority of total natural gas storage in the subsurface, till today, no pure hydrogen storage exists, but if deep aquifers could be used, it could be used for decarbonization of the entire energy sector and regions and a most cost-effective option for the underground storage of Hydrogen [22].

Saline aquifers seem to be promising because of the above conditions for an alternative option for seasonal storage of Hydrogen, but there are still some critical questions on the stability and integrity of the overlying seal, allowing the aquifers to hold a limited range of pressure and low flow rates. As per another study conducted by Heinemann *et al.*, low density and viscosity of Hydrogen leads to large displacement and, eventually also leads to viscous fingering. As per another study conducted by Heinemann *et al.* [21], the injectivity, productivity and storage capacity of Hydrogen in a saline aquifer is related to the cushion gas (CG) and working gas (WG). Storage in deeper reservoirs with high permeabilities requires a lower CG/WG ratio. Tight anticlines make an injection of the gas more difficult and have little impact on the production.

### 2.8.2. Benefits and Barriers of Saline Aquifer

Underground hydrogen storage will enable the development of the renewable hydrogen sector. UHS allows large-volume storage and may be safely operated at pressures up to 200 bars due to their tightness depending on their depth and offers flexibility for injection and withdrawal cycles as per the need in the market. However, there are limited underground storage sites; therefore, to ensure the development of the concept and its viability for future projects, injection and withdrawal cycles should be frequent. Another potential problem is related to the reaction of hydrogen gas with the moisture and chemicals taking place, which might transform the composition of the gas. The geographical scarcity might also be a critical issue [23].

## 2.9. Hydrogen Storage in Depleted Oil/Gas Fields

Hydrogen Storage in depleted oil and gas fields is also another hydrogen storage technology that has been utilized over the years, Richard *et al.* (2021) that the main benefits to the utilization of depleted oil/gas fields for hydrogen storage are the availability of pre-existing infrastructure, their geographical availability and their reduced cushion gas capacity (CGC).

One critical point outlined by the author is that the use of existing infrastructure from the petrochemical industry contributes massively to it being estimated as the lowest costing technology analyzed Richard *et al.* (2021) [24].

### 2.9.1. Hydrogen Storage in Geological Structures

Underground energy storage for hydrogen is best for long-term and large-scale usage. Additionally, Thiyagarajan *et al.* (2022), the two types of geological locations used to store hydrogen in a gaseous form underground are the Porous media, in which the gas is stored in the pore space of sandstones or carbonate formations, and the cavern storage, in which the hydrogen gas is contained in excavated or solution-mined caverns in the thick rock [24]. Similarly, there are also additional technological, environmental, economic, and other considerations for hydrogen storage in geological structures. It is also crucial to select the right exploitation parameters, taking unique reservoir features into account, as well as injection and withdrawal pressures for hydrogen that do not exceed the fracture pressure of the subsurface formation [24].

### 2.9.2. Challenges Faced in Underground Hydrogen Storage

Underground hydrogen storage technology is faced with its unique challenges. These challenges as enlisted by Thiyagarajan *et al.* (2022) include the following: Site selection, Geochemical reactions and $H_2S$ formation, Microbial growth in reservoir, Well and Geological integrity of the caprock, Unstable displacement, and Biotic & Abiotic gas production.

**Figure 3** presents a comprehensive analysis of hydrogen storage behavior under specific temperature and pressure conditions. This figure provides valuable insights into the performance and characteristics of hydrogen as a storage medium in various applications. By investigating the relationship between temperature, pressure, and hydrogen storage, **Figure 3** shows an understanding of the thermodynamic properties and limitations associated with hydrogen storage systems. The findings presented in **Figure 3** offer a basis for optimizing storage strategies and enhancing the efficiency and safety of hydrogen storage technologies.

Additionally other challenges faced in underground hydrogen storage includes, $H_2$ contamination, seal quality reduction injectivity reduction, fault re-activation, chemical disequilibria, mineral dissolution, and precipitation, change in reservoir properties, mixing and diffusion and microbial clogging amongst others. In further research, these challenges have been critically analysed and emerging solutions have been developed to mitigate these current challenges.

In the same vein, Thiyagarajan *et al.* (2022) further opined that injecting hydrogen into a reservoir changes the chemical equilibrium between the formation pore water, dissolved gases, and the rock matrix, resulting in several geochemical reactions, including: high hydrogen loss, production of other gases that contaminate stored hydrogen, reduction or increase in injectivity due to mineral dissolution/precipitation, and Changes in mechanical properties. Based on this, for hydrogen storage, the contact angle, interfacial tension, and wettability of caprock must be studied for efficient hydrogen injection and withdrawal [24].

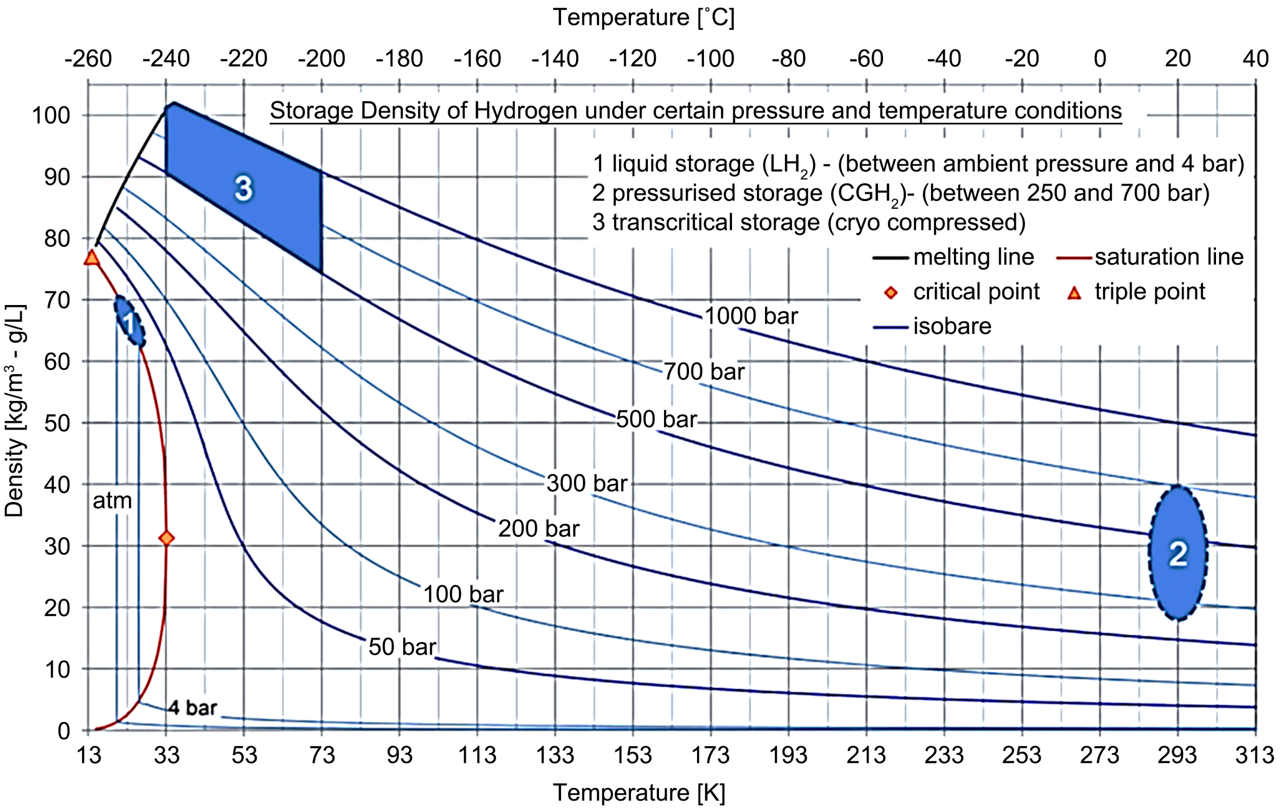

**Figure 3.** Hydrogen Storage under specific temperature and pressure [24].

### 2.9.3. Underground Hydrogen Storage Technology (UHST) Mechanisms and Effective Monitoring

This method is identified as an effective technology for storing a large volume of hydrogen gas in geological formations like empty salt caverns, deep aquifers, depleted hydrocarbon reservoirs, or underground coal seams for years, depending on the demands [25] [26]. According to earlier research, salt caverns were considered a feasible storage site for underground storage methods despite the fact that sedimentary reservoirs such as depleted hydrocarbon reservoirs, coal beds, or deep aquifers are abundant and widespread geographically [27]. For assessing the feasibility of such sedimentary reservoirs, the behavior of Hydrogen in the pore network of the storage rock in real storage conditions is assessed, and these pre-scale effects determine the multi-phase reservoir fluid dynamics associated. However, this pore-scale phenomenon still lacks good quality research. Another research conducted by Heinemann *et al.* concluded that carboniferous age sedimentary deposits of the D'Arcy-cousland. Anticline and the Balgonie Anticline are suitable storage sites and ideal for early research projects [28].

As a result of a review carried out by Pan *et al.*, the UHS is still in its infancy stage and differs significantly from CGS and natural gas reservoirs, so any extrapolation needs to be avoided. Much more fundamental review is required to evaluate the feasibility of UHS and get the data for $H_2$ systems, which will aid in the implementation of UHS for industrial-scale applications [29]. According to W. Lui *et al.*, Jintan salt mine in China satisfies the site requirement of UHS salt caverns to be constructed because of its trapping properties and scalability. The leakage through the rock salt can be neglected since the interlayer is the main channel for the seepage of the hydrogen gas [25]. The optimum efficiency of underground hydrogen storage technology is based on strong mechanisms and effectively monitoring them throughout the lifetime of hydrogen storage cycles. Depending on the energy demand required by the market and the type of field used for storage, an underground hydrogen storage system generally follows a cyclic operation with an alternate period of injection and withdrawal.

Also fingering in the subsurface where hydrogen has been stored is a potential activity that may contaminate the hydrogen and therefore, Thiyagarajan *et al.*, (2022) stated that it requires additional effort and equipment to mitigate this contamination and based on this, the flow rate must be optimized by ensuring that bottom-hole pressure, capillary entry pressure, and fracture pressure are all met to prevent fingering [24]. Among the obvious challenges involved in UHST, the mixing of cushion gases is one of the reasons for hydrogen loss in sub-surface storage, and since there is no comprehensive monitoring technique available for underground hydrogen storage, other geological storage techniques used in natural gas storage have influenced the monitoring systems employed for UHST. The monitoring techniques such as neutron log, cement bond log, sonic detection, spinner survey, camera inspection and radioactive tracer survey can also be considered for UHS monitoring, and for effective monitoring, chemical and electrochemical tools, gravity methods, electrical resistivity tomography (ERT), muon tomography, and seismic full waveform inversion (FWI) are being utilized for the monitoring of microbial, geochemical, and geo-hydraulic effects in subsurface storage [24].

## 3. Methodology

The methodology for this project focuses on conducting a comparative analysis of different hydrogen storage methods based on their energy efficiency, energy density, lifetime, and energy costs. Additionally, we will perform a quantitative risk reliability assessment of hydrogen storage systems, including transportation methodologies, for each storage method. Adequate quantitative analysis of these storage systems is essential in identifying the best and most appropriate method of hydrogen storage based on specific parameters. By examining the characteristics of different hydrogen storage systems, researchers can distinguish a storage method that is most suitable for utilization. In this study, we will analyze both liquefaction, underground storage, and other methodologies to identify the most feasible method of hydrogen storage, which we will propose for consideration.

### 3.1. Quantitative Analysis

A comparative analysis of the various hydrogen storage is a quality approach to utilize considering that compressed gas hydrogen storage, stores hydrogen gas by utilizing the high-pressure vessels for further commercial applications. Previous analysis of the several materials and vessel types for compressed hydrogen storage showed that the storage types for compressed hydrogen are classic metal pressure vessels that have low strength to weight ratios. Analytically, compressed hydrogen method is the most economical hydrogen storage method by comparison with others. Besides, it is manufactured with a filament-winding technique using high-strength UD carbon/epoxy composites on a metallic (type III) or polymer (type IV) liner. The main advantages of compressed hydrogen storage are; excellent impermeability, high anti-collision performance, and distinct plasticity that improve the entire performance of composite vessels by applying autofrettage pressure. Nevertheless, the low weight ratio (gravimetric energy density) operating performance is a critical drawback for type III composite overwrapped pressure vessel (COPV), leading to high manufacturing costs. To find an affordable vessel with a high weight ratio and satisfy large-scale industrial compressed hydrogen storage requirements, the internal pressure must be increased up to 70 MPa for maximum storage capacity. Type IV COPV which is composite made up of fibre and resin withstands 70 MPa with high hydrogen storage density. The unique design and material provide lightweight, high strength, and adequate resistance against corrosion and degradation. Moreover, in order to obtain the maximum internal pressure for maximum compressed hydrogen storage density, and based on the buckling and static analysis tests for the polymer liner's tension, results showed that filling and storing hydrogen gas ranges from 0.11 to 0.15 MPa, is superior as when compared with other hydrogen storage methods.

In the case of Metal Hydrides hydrogen storage process, the hydrogen gas to be stored with high pressure is pumped into a tank with metal to form metal hydrides, therefore enabling hydrogen to be stored as solid compounds. And during retrieval whenever hydrogen gas is needed, the counter reaction will be triggered. Metal hydrides at a high temperature will turn back to hydrogen and its original metal reactant, but the challenge of choosing the best metal reactant has consistently posed a challenge to this process.

Metal hydrides should have at least 10% weight as hydrogen capacity, which means 15 liters of hydrogen gas can be captured by 50 grams of metal. By looking at the lifetime comparatively, the lifetime cycle for Metal hydrides is short. And an experiment on magnesium hydride shows a cycle lifetime of 2000 hours with $LaNi_3$ as an amorphous modifier. Additionally, research data shows that the energy efficiency of 69% only considers the loss of hydrogen while necessary energy is used to push hydrogen gas in and to trigger the counter reaction. If the input and output electricity is taken into account, the electrical charging efficiency is 17%.

For liquid hydrogen storage, Liquid hydrogen storage (LHS) allows hydrogen to be liquefied in a very low temperature, usually −253.0˚C, and a high vacuum insulated container will be used to store the liquid. However, the actual processing of LHS meets several technical difficulties because $LH_2$ has a much lower boiling point than that of LNG under the same pressure, which will be about 90˚C lower in usual cases. Liquefaction can be adapted along with the production of $H_2$, and it has to occur before transportation in order to minimize the cost. Comparatively, a major obstacle in $H_2$ liquefaction is the consumption of energy to retain the low temperature. The electric bill can cost up to the price which one-third of the produced $H_2$ is worth. Another expensive factor happens to specific technologies when the $H_2$ liquefaction has a large scale. Similarly, the process of LHS is sensitive to several disruptions including heat leakage, ortho-para conversion, sloshing, and flashing. A boil-off of about 0.2% - 0.3% $LH_2$ may occur per day during storage and transportation. This leakage can be mitigated by upgrading the technology of thermal insulation, such as vacuum thermal insulation. As for the potential risk of sloshing during the transportation, it can be prevented by anti-slosh baffles.

On the other hand, while compressed, liquified, and metalized hydrogen can occur above ground, storage of hydrogen gas in geological formations or underground is the most dominant approach that is considered by direct field applications to be in connection with large-scale hydrogen use and to smooth out the fluctuations of energy demands. In terms of energy efficiency, LHS has an energy efficiency of about 40% - 50% in general: the efficiency during production is about 50% - 70%; during the transportation is about 95%; during utilization is about 50% - 70%. LHS also has a high energy density, over 800 times greater than the energy density of gaseous H2 with the same volume, *i.e.*, it saves a significant amount of space in storage and transportation. The lifetime of LHS is only about 300 - 500 days because boil-off often occurs during storage and transportation.

Underground Hydrogen Storage (UHS) in salt caverns, as one of the most promising technologies, allows a large quantity of hydrogen stored in constructed salt caverns. In addition, this technology has relatively high energy efficiency; and it's been estimated that the energy efficiency of underground compressed air storage is 70% to 89%, practical analysis also indicated that the energy efficiency of UHS in caverns can reach as high as 60%. On the other hand, the energy density of this technology can be up to 300 $kWh \cdot m^{-3}$, and this number is as high as the energy density of lithium batteries. In fact, a number of caverns have even higher energy capacity, and the entire storage capacity of 20 salt caverns equals 1700 GWh of electric power, which translates to a massive output of 14 GW. Therefore, UHS in salt caverns is a suggested technology that can be applied to large-scale energy storage of renewable sources in the future.

Underground hydrogen storage has a longer duration time period compared to other technologies analyzed above. The lifetime of compressed air stored underground can range from 20 to 40 years therefore enabling geological storage, namely salt caverns, has become the best option for long-term storage. Meanwhile, the underground storage of compressed air has a fast response time, which means the time that the underground hydrogen storage system requires to ramp up supply is fast. On this basis, UHS in salt caverns takes a low price: about 1.61 U.S. dollars for each kilogram of hydrogen. In comparison, the cost of storing hydrogen in aquifers and hydrocarbon deposits is 1.29 and 1.23 U.S. dollars per kilogram, respectively, storing hydrogen in salt caverns is more economic than other kinds of underground hydrogen storage. (**Table 1**)

**Table 1.** Comparison of different hydrogen storage methods.

| S/N | Hydrogen Storage Methods | Energy Efficiency | Energy Density | Lifetime | Response Time | Cost |
|---|---|---|---|---|---|---|
| 1 | Metal Hydrides | 69% | 244 $kWh \cdot m^{-3}$ | Less than 2000 hours | Fast | 9.01 - 19.67 USD/kg (Ni Metal Hydride) |
| 2 | Liquid Hydrogen Storage (LHS) | About 40% - 50% | 2359.30 $kWh \cdot m^{-3}$ | 300 - 500 days | Fast | 3.66 USD/kg in 2030 |
| 3 | CGH2 for Type IV Vessel | >85% | 3.3 - 1320 $kWh \cdot m^{-3}$ (depends on compression strength) | Over 20 years | Fast | Starts from 466 USD/kg |
| 4 | Underground Hydrogen Storage | More than 60% | Can reach about 300,244 $kWh \cdot m^{-3}$ | 20 - 40 years | Fast | 1.61 USD/kg |

### 3.2. Hydrogen Energy Storage Feasibility

Comparing the several mainstream hydrogen storage technologies that are discussed in the quantitative study section, different in phase and environment for storage. In brief, their characteristics, pros and cons, metal hydride hydrogen storage uses several metal materials as media to store the amount of hydrogen gas by condensing and metal absorption. Liquid hydrogen storage is stored in a high vacuum insulated container under very rigorous temperature, the type IV vessel CGH2 employs a novel design that combines metal and polymer (CFRP) parts to provide lightweight, high strength, and adequate corrosion and degradation resistance performance, but it is costly for the system material. Underground hydrogen storage (UHGs) is a technology that stores hydrogen gas in underground geological reservoirs (salt caverns) that offer large storage capacities and discharge rates while also minimizing environmental and need for surface space. On balance, as the underground energy storage technology is used on a large scale in the industry for long term storage capacity, it essentially considers several elements of control (e.g., cost-effectiveness), which corresponds to energy use and storage density, response time, and system lifetime. Under those statements and qualitative data as summarized in the table above, the most efficient and feasible technology for large scale industry storage systems would be underground hydrogen storage (UHGs) technology.

Although CGH2 for Type IV Vessel storage technology has the highest efficiency ratio for gas phase H2 storage and great storage capacity ranked only second to liquid hydrogen storage (LHS), the unique materials, the metallic parts fabricated by composite carbon fiber reinforced polymer (CFRP) liner are costly, which are not friendly for reconditioning.

Furthermore, due to the unique combination structure, the metallic parts are easily contacted with the dense hydrogen gas, where the surface interaction of the metal with hydrogen leads to hydrogen embrittlement and even mechanical property degradation. Similarly, the interaction leads to hydrogen gas always in a charged or discharged state, causing the polymer layer to penetrate and leading to the leakage of hydrogen gas from the vessel. Obviously, the liquid hydrogen storage technology is also not under consideration, because of its low efficiency and short lifetime, even though it has great storage capacity at a fairly lower cost. Furthermore, metal hydrides are still undesirable for selection not only because of their lifetime, and metal cost, but also attributed to the unmature technology, which bear more risks and uncertainty factors (e.g., imprecise storage capacity and overestimated metal wastage) and bring more challenges to the industry. Therefore, underground hydrogen storage meets the criteria of a technology that is relatively economically mature, long-term and applies to large-scale functions. In different types of underground hydrogen storage, namely aquifers, oil, and gas reservoirs, etc., salt caverns have some special advantages. First, caves are usually formed in impervious salt domes, which can minimize gas loss. In addition, rock salt has a dense matrix, low porosity, extremely low permeability, and self-healing ability. Therefore, salt caverns have been widely used for hydrogen storage. Finally, the technical characteristics of a salt cavern are more favorable than those of other reservoirs because a salt cavern typically has a volume of 10,100 × $10^4$ $m^3$ and a depth range of 600 - 2000 m, which is ideal for storing highly pressurized hydrogen. As a result, until recently, salt caverns have been the lone reservoir that has proved successful for subterranean hydrogen storage. (Table 2)

### 3.3. Hydrogen Energy Storage Analysis Based on Delivery Technologies

Another comparative analysis of Hydrogen Storage Methods is based on their delivery technologies for domestic and industrial activities. It is vital to note that hydrogen delivery is critical contributor to the cost, energy use and emissions associated with hydrogen pathways. In the case of centralized hydrogen storage and production, hydrogen delivery to end users includes two main phases: Transmission (delivery of hydrogen from production plants to the city gates), and Distribution (delivery from the city gates to the fuelling station from storage tanks or end users).

Based on the diverse methods of hydrogen storage as discussed earlier in the literature review, there are three main pathways for the delivery of hydrogen, and they include;

1) Gaseous hydrogen storage delivery system.
2) Liquid hydrogen storage delivery system.
3) Material based hydrogen carriers system.

**Table 2.** Comparative analysis of maximum hydrogen storage capacities (percentage of weight %wt) for chemical and physical hydrogen storage methods.

| S/N | Hydrogen Storage Methods | Material-based storage method | Maximum reported storage capacity (%wt) |
|---|---|---|---|
| 1 | Chemical | Ammonia Borane | 19.4 |
| | | Metal Hydrides | 12.6 |
| | | Alanates | 9.3 |
| | | Formic Acid | 4.4 |
| | | Carbohydrate | 14.8 |
| | | Liquid Organic Hydrogen Carriers | 7.2 |
| 2 | Physical | Carbon Materials | 8 |
| | | Zeolites | 9.2 |
| | | Glass Capillary Arrays | 10 |
| | | Glass Microspheres | 14 |

### 3.3.1. Gaseous Hydrogen Storage Delivery System

Gaseous hydrogen, due to its storage conditions is transported by either compressed H2 pressure vessels arranged in tube trailers, and or via gas pipelines. On the field, roughly 2600 km of hydrogen pipelines are available in the U.S mainly located close to refineries and ammonia plants, which are the mass hydrogen consumers. And a dedicated pipeline is necessary for hydrogen delivery over long kilometres, and based on this, the use of the current natural gas pipeline infrastructure to distribute hydrogen is gaining wide acceptability. Transportation of hydrogen gas via tube trailers has been developed and has an operating pressure of 250 bar considering the mass of hydrogen stored in large tanks, which is approximately 7% of the tank weight. The utilization of tube trailers is of interest since generally it is the easiest method in terms of infrastructure requirements. Tube trailers are advantageous because the hydrogen loss is minor and compression cost at fuelling stations is low and can be further reduced by 60% in comparison with liquid hydrogen. Hydrostatic burst test, penetration test, leak before burst and pressure cycle tests are some of the tests usually performed to ensure that the hydrogen tube trailer is safe for use.

### 3.3.2. Liquid Hydrogen Storage Delivery System

The liquid Hydrogen Delivery system is considered to be economical for high demands which are above 500 kg/day and for, mid range distances. Hydrogen fueling stations will be further supplied by liquid hydrogen due to much higher storage capacity. Cryogenic hydrogen delivery consists of three main stages and this includes; Liquefaction, Storage and transportation with cryogenic tanks the end users. The availability of liquefaction tanks with higher production rates, less specific energy consumption including lower capital cost, and higher efficiency would be vital for liquid hydrogen delivery.

### 3.3.3. Hydrogen Carriers Delivery System

The potential to offer higher safety levels due to low storage pressure, manageable properties at ambient conditions and good gravimetric density can be delivered by material-based hydrogen storage. But analytically, they cannot be effectively utilized for high demands of hydrogen by end users.

## 3.4. Comparatively Analysis of Hydrogen Energy Storage Technologies Based on Risk/Reliability Assessment

The reliability of hydrogen storage technologies is indeed important in accessing its efficiency and effectiveness. Therefore, there are a number of factors that could negatively influence the reliability of storage and delivery systems of hydrogen, and these reliability issues are outlined in **Table 3** below. However, in addition to the reliability issues, hydrogen storage handling issues that pose a risk to the reliability of hydrogen storage systems are also stipulated in **Table 3**.

**Table 3.** Risk and reliability analysis of factors that can negatively impact the reliability of hydrogen storage.

| Material Properties related issues | Hydrogen Storage handling related issues |
|---|---|
| • Hydrogen impact on materials.<br>• Liner blistering in pressure vessels.<br>• Damage mechanisms of carbon fibres.<br>• Resistance to fire and high temperature in storage vessels. | • Temperature variation<br>• Hydrogen leakage.<br>• Compression Process.<br>• Process fluctuations in pipelines.<br>• Contamination. |

## 4. Conclusions

This study provided an overview of the current hydrogen storage technology, and the following conclusions can be drawn from review research on hydrogen storage technologies. Firstly, it is vital to note that the most developed technologies are cryogenic and compression storage, but due to hydrogen's low density, compressed storage requires huge quantities and has poor energy efficiency. Material-based storage techniques are still in the early stages of development and will require more time to demonstrate their viability as long-term fixes.

In the case of Metal Hydrides hydrogen storage process, the hydrogen gas to be stored with high pressure is pumped into a tank with metal to form metal hydrides, therefore enabling hydrogen to be stored as solid compounds, and as the underground energy storage technology is used on a large scale in the industry for long term storage capacity, while compressed hydrogen method is the most economical hydrogen storage method by comparison with others. Due to intrinsic challenges around the various methods of hydrogen storage, underground hydrogen storage meets the criteria of a technology that is relatively economically mature, long-term and applies to large-scale functions.

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## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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