Wadi Alshatti University Journal of
Pure and Applied Sciences

مجلة جامعة وادي الشاطئ
للعلوم البحتة والتطبيقية

Volume 3, No. 2, July-December 2025 Online ISSN: 3006-0877 المجلد 3، الاصدار 2، يوليو- ديسمبر 2025

REVIEW ARTICLE **RENEWABLE ENERGY**

High-Pressure Compression, Liquefaction and Metal Hydrides for Hydrogen Storage

Abdulaziz H. Mohamed [1*]

[1]Department of Chemical Engineering, Engineering Faculty, Tobruk University, Tobruk, Libya.

ARTICLE HISTORY
Received 23 March 2025
Revised 20 May 2025
Accepted 25 May 2025
Online 28 May 2025

KEYWORDS
Hydrogen;
Compression;
Embrittlement;
Liquefaction;
Phase Inversion;
Metal Hydride.

ABSTRACT

Hydrogen storage involves the safe and efficient containment of hydrogen. Hydrogen storage is highly essential due to hydrogen's potential as a clean, green energy source that can enhance the utilization of energy derived from renewable resources. However, due to high flammability of hydrogen, its storage possess some challenges regardless of the storage method employed. To contribute to the existing literature and deepen the understanding of hydrogen storage methods, this review paper briefly examines the use of high-pressure hydrogen compression, hydrogen liquefaction and metal hydrides for hydrogen storage. The findings from the reviewed work suggest that each process comes with its own set of advantages and disadvantages regarding storage density, safety, energy requirements and more. Overall; however, the benefits outweigh the drawbacks. Effective hydrogen storage is crucial for the utilization of hydrogen across various applications, playing a crucial role in the decarbonization of associated sectors.

الضغط العالي والتسييل والهيدريدات المعدنية لتخزين الهيدروجين

عبدالعزيز الهمالي محمد [1*]

الملخص

يتضمن تخزين الهيدروجين احتواءه بشكل آمن وفعال. يعد تخزين الهيدروجين ضروريًا للغاية نظرًا لإمكانات الهيدروجين كمصدر طاقة نظيف وصديق للبيئة يمكنه تعزيز استخدام الطاقة المستمدة من الموارد المتجددة. ومع ذلك، نظرًا لقابلية الهيدروجين العالية للاشتعال، فإن تخزينه يواجه بعض التحديات بغض النظر عن طريقة التخزين المستخدمة.للمساهمة في الأدبيات الموجودة وتعميق فهم طرق تخزين الهيدروجين، تهدف هذه الورقة لدراسة استخدام ضغط الهيدروجين عالي الضغط وتسييل الهيدروجين وهيدريدات المعادن لتخزين الهيدروجين. تشير النتائج إلى أن كل عملية تأتي بمجموعة من المزايا والعيوب فيما يتعلق بكثافة التخزين والسلامة ومتطلبات الطاقة. بشكل عام، فإن الفوائد تفوق العيوب. يعد تخزين الهيدروجين الفعّال أمرًا بالغ الأهمية لاستخدام الهيدروجين في تطبيقات مختلفة، حيث يلعب دورًا حاسمًا في إزالة الكربون من القطاعات المرتبطة به.

الكلمات المفتاحية
الهيدروجين
الانضغاط
الهشاشة
التسييل
عكس الطور
هيدريد المعادن

Introduction

An ongoing and accelerated transition away from fossil fuels in energy systems is essential for attaining climate neutrality in the near future. In this process, hydrogen produced from renewable energy sources is expected to play a significant role as a secondary energy carrier. Hydrogen production technologies have been extensively reviewed in a previous study by Zainal, et al. [1]. Environmentally friendly hydrogen can be generated from water through an electrolysis process powered by solar panels or wind turbines. Zainal et al. have highlighted that the hydrogen produced can be converted into electricity using a fuel cell, resulting in no harmful emissions or waste, aside from water. Consequently, the economic potential of hydrogen has been acknowledged in several countries, including Australia, China, Germany, Japan, the UK and the USA.

In the last five years, over 30 countries have released hydrogen roadmaps; the industry has announced more than 200 hydrogen projects, and significant investment commitments have been made. Globally, governments have pledged over USD 70 billion in public support. If all proposed projects come to fruition, total investments in hydrogen could exceed USD 300 billion by 2030, which would represent 1.4% of global energy spending [2,3]. This gradual increase reflects a rising interest and investment in the hydrogen sector, likely fueled by a growing recognition of the advantages of hydrogen energy storage and its potential role in the shift toward more sustainable energy systems. Although hydrogen investments made up only 0.6% of total energy transition investments in 2023, this marks a six-fold increase compared to 2022 [4].

In fact, hydrogen is becoming increasingly important in the global energy transition and so its storage. By 2024, the hydrogen energy storage market is anticipated to reach $16.64 billion, with forecasts indicating it may surpass $20 billion by 2028. Therefore, it is important to address how hydrogen can be stored in a manner that is energy-efficient, safe and cost-effective for specific applications. Hydrogen storage has been achieved through various technologies, including but not limited to high-pressure hydrogen

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*Corresponding author

https://doi.org/10.63318/waujpasv3i2_08

compression, liquefaction and metal hydride. This review paper aims to examine these technologies in detail.

Storage of Hydrogen via High-Pressure

Hydrogen molecules can exist in various states depending on the pressure and temperature, as depicted in the phase diagram shown in Fig. (1). this diagram illustrates the different phases that hydrogen (H_2) can adopt based on these conditions. At low temperatures, hydrogen is solid, with a density of 70.6 kg/m³ at 11 K. As the temperature increases, it transitions to a gaseous state, exhibiting a much lower density of 0.089886 kg/m³ at 273 K and a pressure of 1 bar. Liquid hydrogen, which exists at 20 K, occupies a narrow region between the triple point and the critical point, with a density of 70.8 kg/m³ [6]. At the room temperature, hydrogen is in a gaseous state, and its behavior can be described by the Van der Waals equation as follows:

$$P(V) = \frac{nRT}{V - nb} - a\,\frac{n^2}{V^2} \tag{1}$$

In this equation, P represents the gas pressure, V denotes the volume, T is the absolute temperature, n indicates the number of moles, R is the gas constant, a represents the dipole interaction or repulsion constant and b is the volume filled by the hydrogen molecules [7]. The low critical temperature of hydrogen in its gaseous state (T_c= 33 K) leads to significant repulsive interactions between hydrogen molecules. Essentially, hydrogen storage involves minimizing the large volume that 1 kg of hydrogen gas occupies at the room temperature and atmospheric pressure, which is approximately 11 m³. To achieve this, work must be done to compress the hydrogen, the temperature must be lowered below the critical point, or hydrogen must interact with another material to mitigate repulsion; thereby, increasing its density within a storage system [6].

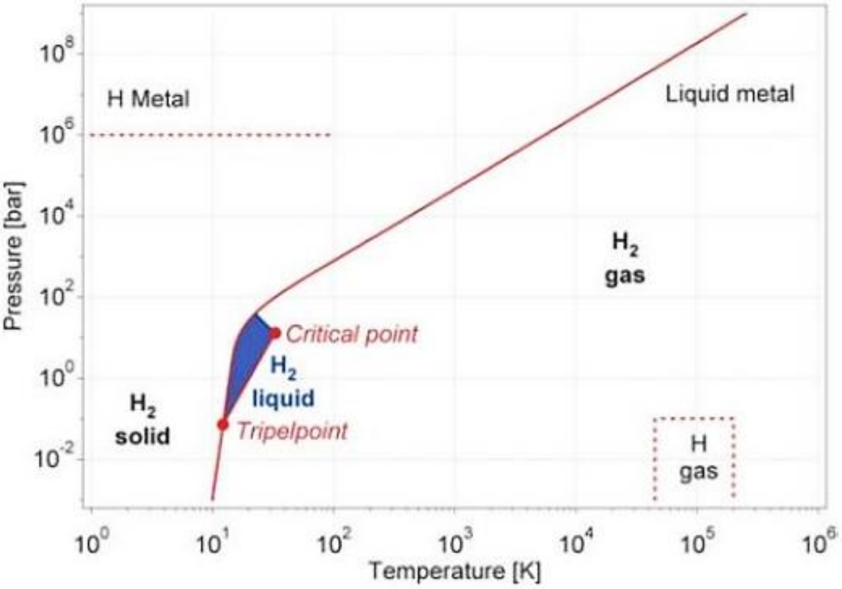

Fig. 1: Phase Diagram for Hydrogen [6].

The advancement of hydrogen infrastructure is significantly influenced by hydrogen storage methods. The way hydrogen is stored not only affects its transportation but also its applications. Therefore, improvements in hydrogen storage technology can further enhance and promote the various uses of hydrogen. The goal is to compress gaseous fuels, including natural gas and hydrogen, to achieve high pressure while reducing their volume, which results in increased storage capacity in the gas phase. For hydrogen storage, two primary compression methods are typically employed to enhance volume density: mechanical and non-mechanical compressors. Mechanical compressors, which account for over half of the capital costs of a hydrogen refueling station [8], include types such as reciprocating piston compressors, diaphragm compressors and screw compressors.

Conversely, non-mechanical compressors, which lack moving parts, include metal hydride compressors, electrochemical compressors, adsorption compressors and cryogenic compressors [9]. A discussion of these compressors typically necessitates a background in mechanical engineering, which is not applicable within the scope of this paper. On the other hand, those interested in a more in-depth analysis, a study by Orlova et al., which compares the performance of mechanical and non-mechanical compressors based on key factors such as efficiency, deliverable flow rates, pressure capability, cost and necessary maintenance [9], is recommended.

Once hydrogen is compressed, it can be stored in a specific type of storage vessel, as discussed next. The selection of the storage vessel involves a trade-off between performance and cost. Filling hydrogen storage vessels with highly compressed hydrogen gas at refueling stations necessitates strict safety protocols due to the inherent high-pressure conditions. To address this, numerous studies have been conducted to understand the charging process of high-pressure gaseous hydrogen into storage vessels. These investigations have led to the development of various computational fluid dynamic models that take into account the flow and temperature fields [10-16]. However, this review paper shall not delve further into this topic. At refueling stations, the high-pressure compressed hydrogen can; then. be dispensed as a fuel for fuel cell vehicles, similar to the operation of traditional gasoline or diesel pumps.

In the early days, hydrogen storage vessels were primarily made from steel or aluminum. For instance, steel vessels needed up to 500 kg to hold 25 Nm³ of hydrogen at a maximum pressure of 12 MPa [17]. Over time, high-pressure storage vessels have been developed using a range of materials, such as aluminum, steel, carbon fiber, polyethylene and epoxy resin. Expensive AISI type 316 austenitic stainless steel alloys with a minimum-required nickel content between 11.5 and 13 wt. % has been used for constructing hydrogen storage tanks. Also, a stainless steel known as Nitronic 40, which is composed of 21Cr-6Ni-9Mn was introduced for hydrogen storage applications. This material is particularly noteworthy as a low-nickel austenitic stainless steel that does not develop strain-induced at room temperature. The elastic-plastic fracture mechanics of this stainless steel show a notable decrease in both fracture initiation toughness and crack-growth resistance in thermally pre-charged specimens. A high concentration of hydrogen can alter the fracture mechanisms in 21-6-9 stainless steels. Additionally, considering the lack of strain-induced martensitic transformation, hydrogen-assisted fracture can be understood through deformation mechanisms that encourage localized deformation.

Another potential alloy for hydrogen applications is the 22Cr-13Ni-5Mn steel. This material offers several appealing properties, including excellent stability against strain-induced martensite formation, enhanced strength due to nitrogen addition and high fracture toughness when pre-charged with hydrogen. However, its relatively high nickel content may pose cost challenges for hydrogen applications. In pursuit of a stable austenitic matrix that combines high strength and ductility; Cr–Mn–N austenitic steels have also been assessed for their susceptibility to hydrogen embrittlement. Unfortunately, substituting nickel with manganese and adding nitrogen did not yield favorable results in slow strain rate tensile tests conducted in a 10 MPa hydrogen environment at −50 °C. The material's ductility response was significantly diminished, even though only a small portion of it transformed into strain-induced martensite. This brittle behavior was primarily linked to the influence of nitrogen, which encourages short-range ordering and increases the extent of planar slip during deformation [5].

However, steel vessels are considered too heavy for hydrogen storage in vehicles [18]. To store 5 kg of hydrogen enough for a vehicle to travel between 500 and 700 km, a high-pressure storage vessel with a volume of 0.18 m^3 is required [19]. In this scenario, optimizing both gravimetric and volumetric densities is essential to enhance hydrogen storage and increase the vehicle's range [20]. Additionally, utilizing steel vessels for the storage of high-pressure hydrogen, such as at pressures reaching 700 bar, is not advisable due to the risk of hydrogen embrittlement, which occurs when hydrogen diffuses into the steel. This risk is exacerbated by frequent charging and discharging of the vessels. To address the issue of embrittlement, steel storage vessels can be replaced with those made from composite materials that are chemically inert to hydrogen, such as polyethylene/carbon fiber or epoxy resin combined with a thin aluminum liner. However, it is important to note that the gravimetric storage density of these composite vessels is relatively low, at 0.01 kg H_2/kg. On the positive side, this technology offers an energy storage efficiency of 94% [18], which is significantly higher than the 75% efficiency associated with battery storage [21]. Nonetheless, while increasing pressure may enhance volumetric storage density, it could lead to a decrease in energy storage efficiency.

The first type of hydrogen storage vessels available today has the lowest pressure tolerance (15-30 MPa) compared to other types. They are also the most economical, costing $240 per kilogram of stored hydrogen, and have the lowest gravimetric storage density, which measures the ratio of the mass of the stored gas to that of the vessel itself [8,20,22-25]. Due to its low gravimetric density, this type is primarily used for stationary applications to store hydrogen as an industrial gas [17,23]. To enhance its ability to withstand higher pressures, it is advisable to hoop-wrap the central section of the vessel with resin-impregnated fiber [17]. The second type consists of a metal combined with a glass and fiber composite [8,17,24]. This type demonstrates the greatest tolerance to pressure, accommodating a broad range from 10 to 100 MPa. This capability may explain its elevated cost of $360 per kilogram of stored hydrogen [22]. These vessels are utilized in stationary applications, such as refueling stations [26]. The third and fourth types of hydrogen storage vessels are fully composite wrapped with a metal liner and fully composite with a high-density polyethylene inner layer reinforced with glass or carbon fiber. Both types are relatively lightweight and can handle pressures ranging from 30 to 70 MPa [8,17,24]. However, they are more costly than the first two types and are only suitable for use in hydrogen tube trailers and on-board hydrogen storage systems [17,27]. Among these storage vessels, the gravimetric storage density improves as we progress from the second type to the fourth.

Hydrogen embrittlement is not the only concern when it comes to the damage of steel hydrogen storage vessels, whether of the first or second generation. Other issues, such as hydrogen-induced cracking and high-temperature hydrogen attack, also pose significant risks, with hydrogen embrittlement being the most critical due to its potential for severe damage [28-29]. Given their importance, these phenomena have been the focus of numerous research studies [28-37]. It has been observed that all these issues stem from a common factor: the tendency of hydrogen molecules to react with steel.

In general, high-strength materials are particularly vulnerable to hydrogen embrittlement when exposed to hydrogen environments in high pressure hydrogen storage tanks. Materials that are prone to this phenomenon include high-strength steels of higher than 1000 MPa [38], high manganese steels, aluminum alloys, titanium, magnesium and magnesium alloys among others [39-44]. Hydrogen embrittlement is a well-recognized phenomenon in high-strength materials. It causes the material to fail catastrophically at stress levels lower than its intended strength. Hydrogen embrittlement results in the material become brittle and cracked due to the development of hydrogen atoms during the corrosion reaction and dislocation emission or microvoid coalescence, respectively. It was revealed that the combination of dislocation emission and microvoid coalescence can contribute to crack growth, ultimately leading to material fracture and failure below its allowable limits [45].

The occurrence of hydrogen embrittlement in steel storage vessels is a complex process involving the interaction of hydrogen with the metal of the storage vessel or its additives, creating stable solid solutions, metal hydrides, molecular hydrogen and gaseous byproducts such as methane. This interaction may weaken the bonding strength at metal grain boundaries, resulting in decreased vessel plasticity and potentially causing brittle fractures, microscopic cracks, or pitting, as illustrated in Fig. (2). A study conducted by Melaina and colleagues indicates that the occurrence of hydrogen embrittlement is influenced by several factors, including the pressure, purity and moisture content of the stored hydrogen, as well as the strength and deformation rate of the storage vessel. Additionally, the surrounding temperature contributes to the risk of hydrogen embrittlement. Generally, increasing the pressure within a storage vessel raises the likelihood of hydrogen embrittlement [46]. However, it has been noted that this relationship is evaluated on a case-by-case basis, and there is no consensus among experts regarding all the mechanisms of hydrogen embrittlement and their interactions [29,33]. Nevertheless, advancements in molecular simulations and cutting-edge technologies, such as transmission electron microscopy and atom probe technology, have gradually improved understanding the mechanisms behind hydrogen embrittlement [29].

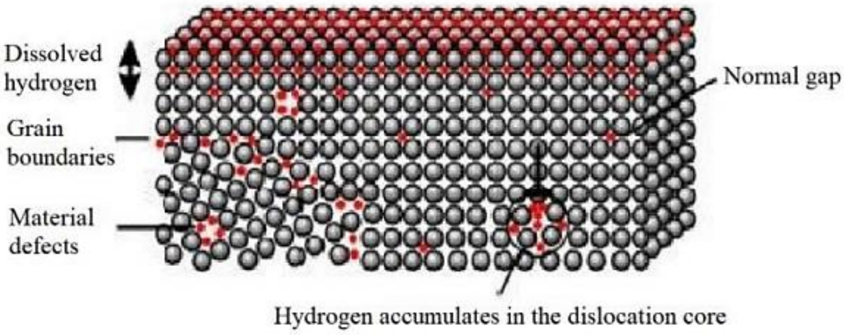

Fig.2: Location of Material Defects and Hydrogen Accumulation [29].

Elsewhere, factors contribute to hydrogen embrittlement were further extended to include factors that relate to the susceptibility of the material of storage vessel to embrittlement, surrounding environment and stress, as depicted in Fig. (3). Such factors include storage material's microstructure, metallic coatings and specific precipitates, heat treatment method(s) applied to the storage material. levels of temperature and exposure time, the presence of hydrogen traps, reactive solutions that interact with metals (such as acidic solutions) and residual stress of the storage vessel along with rate of applied strain and the material's surface condition [38]. That being said; nevertheless, it was highlighted that to prevent hydrogen embrittlement, three key conditions must be met. First, the ultimate tensile strength of the material in contact with hydrogen should not exceed 1000 MPa. Additionally, hydrogen should neither be present in the alloy nor introduced from the surrounding environment. Lastly, there should be no tensile stresses during service conditions, and the surface hardness must be sufficiently high [45].

Fig.3: Causes of Hydrogen Embrittlement [45].

Additionally, hydrogen-induced cracking in steel storage vessels is often linked to stress corrosion cracking, which can lead to vessel failure characterized by cracks in the steel. This issue arises from the combined effects of hydrogen and the stress experienced by the steel vessel, resulting in significant hydrogen embrittlement [28]. Another critical phenomenon to consider is high-temperature hydrogen attack. This occurs when hydrogen comes into contact with steel at elevated temperatures and pressures. Under these conditions, hydrogen molecules or atoms penetrate the metal, becoming absorbed within its internal structure due to the synergistic effects of high temperatures (exceeding 473 K) and pressure. The absorbed hydrogen; then, interacts with the alloying elements and impurities present in the metal's microstructure, leading to the formation of insoluble gaseous products, which is the essence of high-temperature hydrogen attack. The dissolved hydrogen within the metal microstructure is highly mobile and tends to accumulate at grain boundariesinclusions and other microstructural defects. In these areas, the dissolved hydrogen atoms can recombine to form hydrogen molecules. These molecules; then, react with carbon impurities at the grain boundaries, resulting in the formation of methane, alongside the processes of decarbonization and the decomposition of cementite in steel at elevated temperatures (above 473 K). Consequently, the methane gas generates internal pressure, leading to the formation of bulges and cracks in the steel storage vessel, manifesting as elongated holes. This phenomenon can significantly exacerbate hydrogen embrittlement, potentially causing catastrophic damage to hydrogen storage vessels [47].

Nevertheless, in high-pressure gas compression, the energy demands for compressing hydrogen are high due to its lower specific gravity compared to other gaseous fuels [48]. Regarding environmental and safety concerns, both the materials used for constructing high-pressure storage vessels and the stored hydrogen itself are not harmful to the environment or dangerous. In the event of a hydrogen leak, it rises and disperses quickly into the atmosphere because it is lighter than air. Consequently, hydrogen is likely to accumulate near the source of the leak. However, it is important to consider hydrogen's flammability; its flammability range is between 4.1% and 74.8% by volume at 1 bar in dry air. To ensure safe operation, adequate ventilation is essential to dilute hydrogen and prevent ignition [49].

Storage of Hydrogen via Liquefaction

Compared to storing hydrogen in its gaseous state through high-pressure compression, liquefying hydrogen allows for safer storage in a liquid state, which offers higher volumetric and gravimetric storage densities [14]. Hydrogen liquefaction is a crucial step in the complete liquid hydrogen supply chain [48]. Liquefied hydrogen can serve as fuel for internal combustion engines, fuel cells and other applications. A study by Takeichi *et al.* indicates that the volumetric and gravimetric storage densities of liquefied hydrogen range from 20 to 50 kg H2/m^3 and 8 to 25 kg H2/kg, respectively, with a calorific value of 120 MJ/kg [15]. It is; however, important to emphasize that the liquefaction of hydrogen is a complex and energy-demanding process when compared to the liquefaction of other gases or the previously mentioned hydrogen compression process [14].

The challenges associated with hydrogen liquefaction are partly due to its low evaporation temperature at atmospheric pressure, with a critical point of 20.28 K and 33 K, respectively. Additionally, the small size of hydrogen molecules, which is comparable to that of ideal gas molecules at higher temperatures, contributes to these difficulties. The Joule-Thomson coefficient for hydrogen is only positive at temperatures below 202 K; above this threshold, the temperature increases, which contradicts the principles of liquefaction. Ultimately, the liquefaction of hydrogen is further complicated by the lower enthalpy of vaporization of para-hydrogen, which is 447 KJ/kg at hydrogen's critical point (20 K). This is in contrast to the exothermic process of converting normal hydrogen to equilibrium hydrogen, which has an enthalpy of 532 KJ/kg at the same temperature due to the conversion from ortho- to para-hydrogen [49].

Numerous methods for hydrogen liquefaction have been proposed and implemented, which can be generally classified into two main categories based on their thermodynamic cycles: conventional liquefaction cycles and magnetic refrigeration cycles. To date, several conventional hydrogen liquefaction cycles have been commercialized for practical applications. Conventional liquefaction cycles include: Linde-Hampson liquefaction cycle including: pre-cooled Linde-Hampson cycle, dual-pressure Linde-Hampson cycle and pre-cooled dual-pressure Linde-Hampson cycle. Refrigerant liquefaction cycle with no isentropic expansion. Claude liquefaction cycle including: pre-cooled Claude cycle and pre-cooled dual-pressure Claude cycle. Expander liquefaction cycle including: Brayton cycle**,** Collins Cycle (multi-stage Brayton cycle) and mixed refrigerant cycle with isentropic expansion [48]. On the other hand, magnetic refrigeration cycles rely on the magneto-thermal effect of magnetic materials. Magnetic refrigeration cycles offer several potential benefits, including low cost, a simple and compact design, lightweight construction, quiet operation, easy maintenance and environmentally friendly performance [50]. This paper; however, does not aim to delve into the specifics of these cycles. The main features and important parameters of these hydrogen liquefaction cycles, such as liquid hydrogen capacity, liquid yield, specific energy consumption and energy efficiency, have been the aim of several studies [51-62].

The most basic liquefaction cycle is the Joule-Thomson cycle. Subsequent advancements have led to the development of newer cycles and systems for hydrogen compression, including the Linde-Hampson cycle and the Claude system. To liquefy hydrogen, it is first compressed and; then, cooled using heat exchangers until it reaches a temperature below the phase inversion point 202 K to around 80 K. The hydrogen must; then, be cooled to a temperature below 30 K using a closed-loop refrigeration cycle. During this process, an adsorption method is employed to eliminate impurities, and a catalytic conversion process is conducted to facilitate the ortho-to-para conversion. To produce storable cryogenic liquid hydrogen with a boiling point of 20-30 K and a pressure of 0.1–0.2 MPa, the liquefied hydrogen is directed to an adiabatic expansion unit (a throttle valve) where Joule-Thomson expansion takes place. For simplicity, further details of hydrogen liquefaction cycle are not included in this review but can be found in other sources [6,48,63-71]. A flow sheet of a standard Joule-Thomson cycle of hydrogen liquefaction is illustrated in Fig. (4).

To prevent phase inversion of stored liquefied hydrogen, such as evaporation (boil-off loss) caused by heat absorption from the environment, thermally insulated vessels are typically employed. This involves using support system materials that possess low thermal conductivity and high mechanical strength to minimize heat transfer through conduction. Additionally, to decrease heat transfer via conduction in the interconnecting piping system, it is recommended to reduce the pipe cross-section while increasing their length [14].

To mitigate heat transfer through natural convection, it is important to minimize heat flows, and evacuated double walls are utilized for this purpose. Reflective metallic foils are also employed to block radiation [6].

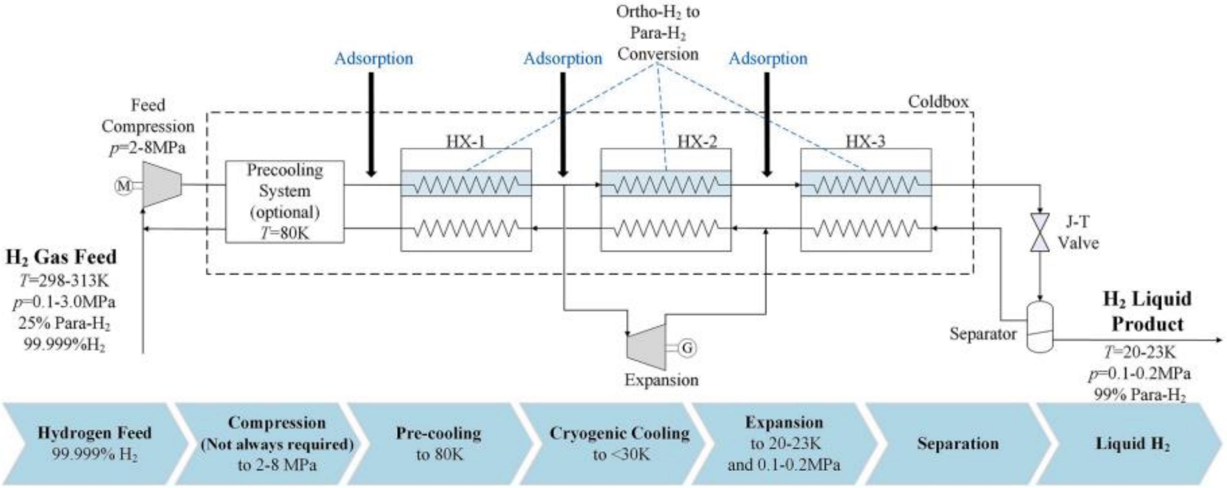

Fig.4: A Standard Joule-Thomson cycle of hydrogen liquefaction [71]

Whereas to reduce radiation intensity, it is beneficial to shield the storage vessel with baffles, enhance its design and prevent direct exposure of the vessel's warm sections. Additionally, employing multilayer insulation technology combined with a high vacuum can significantly decrease hydrogen boil-off to just 0.01% per day for large storage vessels and 0.04% per day for smaller ones [14]. By integrating a refrigeration system with multilayer insulation technology, it is possible to achieve zero hydrogen boil-off loss [67]. Safety is a key concern, as the metallic inner vessel is not intended to withstand higher pressures [14]. Therefore, it is essential to ensure proper ventilation away from any potential ignition sources to manage hydrogen vapor safely [72]. The recent adoption of hydrogen vapor for cooling hot compressed hydrogen gas has become increasingly popular, resulting in a decrease in boil-off loss to less than 1% per day [68-69]. Typically, the generation of hydrogen vapor (boil-off) occurs around three days after a vessel is filled with liquid hydrogen, accounting for roughly 0.1% to 3% of the total liquid hydrogen stored [73].

The boil-off of hydrogen is influenced by the shape, size and thermal insulation of the liquid hydrogen storage vessel. Small spherical storage vessels are the most effective in minimizing hydrogen boil-off due to their optimal surface-to-volume ratio and their ability to evenly distribute stress and strain. In contrast, large spherical storage vessels are less commonly used because of their higher costs. Since boil-off is directly related to the surface-to-volume ratio, it significantly decreases as the size of the storage tank increases. For instance, a double-walled vacuum-insulated spherical Dewar vessels with capacities of 50, 100 and 20,000 m³ experiences daily boil-off losses of 0.4%, 0.2%, and 0.06%, respectively [6]. When considering energy storage density for hydrogen, it is estimated to be around 5 MJ per liter [74].

However, this method demands more energy compared to compressing hydrogen gas at high pressure [6,73]. Hydrogen liquefaction is an energy-intensive process, although ongoing advancements aim to further lower energy consumption and improve efficiency. These enhancements are essential for reducing the costs associated with the liquid hydrogen supply chain, making it more competitive within the hydrogen economy [48]. Additionally, the continuous boil-off of hydrogen restricts the practicality of liquid hydrogen storage systems [6]. Furthermore, unless high gravimetric and volumetric storage densities are crucial, such as in vehicles and space applications, liquefying hydrogen is generally not a favorable option due to its low energy efficiency, high costs and significant energy requirements [6,75].

In terms of the environmental and safety concerns, hydrogen storage via liquefaction is less environmentally friendly compared to high-pressure hydrogen gas compression. This is primarily due to its lower energy efficiency, which correlates with higher pollutant emissions [75]. Additionally, multiple studies have indicated that liquefaction for hydrogen storage poses greater hazards than high-pressure hydrogen gas compression storage [76-78]. This can be explained by several factors: First, when liquid hydrogen leaks, it is denser than air, causing it to flow downward and accumulate before it vaporizes. Additionally, proper ventilation is necessary to manage any hydrogen vapor [72]. Furthermore, the cooling of moist air can lead to ice formation, which may clog safety valves and vents of the storage vessel. This can result in a pressure buildup that could rupture the storage vessel. During this process, there is a significant risk of ignition, fire, or explosion, especially if the stored liquid hydrogen becomes contaminated with air during the charging and discharging phases. However, maintaining the storage vessel at a pressure higher than atmospheric pressure may help mitigate this risk [75].

Use of Metal Hydrides for Hydrogen Storage

Hydrogen is the lightest of all chemical elements and can form hydrides with nearly all elements, except for the inert gases. The formation of a metal hydride occurs when hydrogen is exothermically absorbed by a metal or alloy (the host metal) at a low ratio of hydrogen atoms to metal atoms (less than 0.1) [6,79]. Metal hydrides play a crucial role in various applications, including chemical processing (as reducing agents, strong bases, strong reductants and catalysts), physical separation processes (such as desiccants, isotope separation, gas separation and hydrogen purification), nuclear engineering (serving as neutron moderators, reflectors, and shields), thermal applications (heat pumps), energy storage (in hydrogen fuel tanks and secondary batteries) and energy conversion through alkaline fuel cells, among others [80]. Given that renewable energy is fundamental to many of these applications, metal hydrides and their applications have become a prominent focus of research [14,79-82].

Metal hydrides can be produced through reactions occurring in the gas phase, in solution, or within solids derived from other hydrides. Many metal hydrides are formed as a result of hydrogenation reactions, which involve the direct interaction of hydrogen with elemental metals, intermetallic compounds or alloys, refer to Eq. 1 [82]. A typical hydrogenation process begins with an initial heating phase, reaching elevated temperatures of up to 1100 K, where the rate of hydrogen absorption is relatively low yet rapid. To achieve the optimal temperature that balances the kinetics and capacity of the hydrogenation reaction, and at which the hydride is formed, a gradual cooling step in a hydrogen-rich environment is necessary. Due to its exothermic nature, a hydrogenation reaction can proceed to completion even with heat loss during the cooling phase. In some cases, a spark may be necessary to 'ignite' the reaction and overcome the activation barrier.

$$H_2 + Metal \leftrightarrow Metal\ hydride + \Delta H_R \qquad (1)$$

Where ΔH_R Is the enthalpy of hydrogeneation reaction in kJ/kg. The pressure dependence of hydrogenation reactions varies; certain metal hydrides require high hydrogen equilibrium pressures ranging from 103 to 109 Pa for synthesis, while others can form at lower pressures, depending on the specific metal or intermetallic compounds involved. Additionally, utilizing high pressures in hydrogenation reactions can stabilize new phases of metal hydrides that exhibit high coordination numbers or elevated metallic oxidation states. In many cases, lower temperatures can hinder the kinetics of hydrogenation reactions, although they can be beneficial for hydrogen storage [79]. A study conducted by Young, K. has revealed that metal systems with larger unit cell volumes generally form more stable hydrides. Conversely, metal systems characterized by high levels of disorder can display a range of local cell volumes, which enhances their storage capacity and improves adsorption/desorption kinetics [80].

The use of metals in this application dates back to 1866, when T. Graham discovered that palladium (Pd) could absorb significant amounts of hydrogen. Subsequently, metal hydrides gained attention in this field due to their ability to reversibly absorb and release hydrogen in intermetallic compounds [81]. Among these metal hydrides, those based on magnesium (MgH2) and its related alloys are considered the most promising materials for solid-state hydrogen storage applications. Hydrogen storage benefits from a high hydrogen absorption capacity, relative cost-effectiveness and favorable reversibility [83]. However, to maintain the cycle stability of the formed MgH2, it is essential to adequately protect magnesium particles from gaseous impurities like O, N_2, CO and CO_2 to prevent such impurities hindering or slowing down the hydrogen absorption process [84].

The use of metal hydrides for hydrogen storage is relatively recent when compared to traditional methods such as high-pressure gas compression and liquefaction. Storing hydrogen in metal hydrides involves keeping it in a solid state, which is considered the safest option and offers the highest volumetric hydrogen storage density [14,79]. Metal hydrides have a unique capability to absorb and release hydrogen as needed, a process that is influenced by the conditions under which they are hydride. The most important feature of metal hydrides is their ability to reversibly store hydrogen, making them effective solid hydrogen storage materials [79]. Metal hydrides are formed through the chemical bonding of hydrogen molecules with metals, alloys or certain intermetallic compounds. However, intermetallic compounds like $LaNi_5$ and Ti-based body-centered cubic alloys, such as the FeTi alloy, exhibit low gravimetric hydrogen storage densities of only 1.28 wt.% and 1.9 wt.%, respectively, which limits their use in hydrogen storage applications. Consequently, the focus has shifted to lighter metals and metal alloys that offer more favorable hydrogen storage densities, including Al, B, Be, Li, Mg, Na and compounds like Pd, Mg_2Ni, MgN_2, NaAl, Ti and Ti_2Ni [14,81,85]. Additionally, various other combinations are also utilized, such as $LaNi_5$, TiFe, $LiBH_4$, $NaAlH_4$, MgH_2, 6 Mg $(NH_2)_2$-9LiH-$LiBH_4$, among others [82,86]. The corresponding features of such metal hydrides are displayed in Table (1).

Hydrogen can be exothermically absorbed by metals or alloys when the ratio of hydrogen atoms to metal atoms is low (< 0.1). In the metal lattice, hydrogen atoms occupy interstitial sites, forming interstitial hydrides that allow for the absorption of significant amounts of hydrogen at a constant pressure. This process generates heat, ranging from 9300 to 23250 KJ per kilogram of hydrogen absorbed [6]. This heat can be recovered simultaneously to mitigate the reduction in storage capacity caused by temperature increases. Conversely, a consistent supply of heat is necessary for the desorption of hydrogen, which takes place at temperatures exceeding 773 K during the hydrogen discharge process [73]. This required heat can be obtained by utilizing the heat recovered from the adsorption phase or by harnessing the waste heat from fuel cells. While the operating pressure may exceed 100 bar, the charging and discharging pressures should be limited to 30 and 2 bar, respectively, for practical and economic reasons. In all metal-hydrogen systems, the principles that govern the interaction between hydrogen and metal are consistently similar. When atomic hydrogen is introduced into a matrix of host metal atoms, the resulting compounds are referred to as interstitial hydrides, which are suitable for hydrogen storage. However, it is important to note that certain sites can be occupied by hydrogen simultaneously. For hydrogen, which has an atomic radius of 0.53 Å, a site must meet specific criteria: its radius must exceed 0.40 Å, and the distance between two adjacent occupied sites must be greater than 2.10 Å [80]. For further details on the chemistry of hydrogen storage in metal hydrides, an additional resource is available [6].

In this hydrogen storage method, it is essential to consider not only the gravimetric hydrogen storage density, as previously mentioned, but also several other factors. These include safe operation with rapid kinetics, as well as the chemical and thermal stability of the resulting metal hydride(s) during multiple charge/discharge cycles [75]. Extreme caution is necessary when handling metal hydrides that contain heavy alkaline metals due to their high reactivity. Additionally, given the sensitivity of various metal hydrides, they should exhibit strong stability against oxygen, air and moisture; if not, they must be stored in an inert atmosphere [79]. Several key factors are crucial including a low dissociation temperature combined with moderate pressure, minimal heat dissipation during the exothermic formation of a metal hydride and a low heat of formation to decrease the energy needed for hydrogen release. Additionally, the infrastructure for recycling and charging should be cost-effective. Furthermore, it is essential to minimize energy loss during the hydrogen charge and discharge processes to help prevent spontaneous hydrogen release.

Storing hydrogen using metal or alloy hydrides is an effective option for stationary power plants, where the weight of the storage medium is not a significant concern. This method boasts a high volumetric storage density, exceeding 100 kg H_2/m^3 [75]. However, its gravimetric storage density is relatively low, ranging from 0.015 kg H_2/kg to 0.07 kg H_2/kg [73,75]. This low density is primarily due to the substantial internal structure of the metals or alloys used. Additionally, the considerable weight of the storage material poses challenges for mobile applications, such as in vehicles. In contrast, utilizing this technology for hydrogen storage is associated with an energy storage efficiency of 88%. However, there are environmental and safety concerns related to the storage of hydrogen using metal hydrides. These include the disposal of spent metal/alloy materials, as well as the components of compressors and storage vessels used for high-pressure hydrogen storage, along with the energy demands involved in the process [75].

Table.1: Parameters of some Metal Hydrides [82,86].

Metal Hydride	Range of Operating Temperature, K	Volumetric Energy Density, KgH_2/m^3	Storage Capacity, wt.%	Charging Rate, KgH_2/min.	Reference(s)
$LaNi_5$	293-353	106	1.3	<1.0	[81,85-86]
TiFe	303-343	98	1.5	<1.0	[81,87]
$LiBH_4$	733-1273	91	13.4	<1.0	[81,88]
$NaAlH_4$	373-573	47	3.7	0.5	[81,89]
TiH_2	923-1023	39	1.0	<1.0	[81,90]
MgH_2	523-773	80	5.5	<1.0	[81,91]
6Mg $(NH_2)_2$-9LiH-$LiBH_4$	363-453	74	4.2	<1.0	[81,92]

Conclusions

In this paper, the reviewed hydrogen storage methods include high-pressure compression, hydrogen liquefaction and the use of metal hydrides. Each of these storage techniques can be utilized for various hydrogen applications. Highly compressed hydrogen gas is suitable for both stationary industrial applications and mobile onboard uses. Hydrogen liquefaction allows for storage in liquid form, making it ideal for internal combustion engines and fuel cells. Metal hydride storage is particularly advantageous for stationary power plants, where the weight of the storage material is less of a concern. Among these methods, the use of chemically stable metal hydrides for hydrogen storage appears to be the most efficient and safest option. Additionally, it offers a high volumetric storage density, although its gravimetric storage density is lower due to the heavy internal structure of the metal or alloy used. That being said; however, each of the storage processes examined has its own set of advantages and disadvantages regarding storage density, safety, energy requirements, with the benefits generally surpassing the drawbacks. Storage vessels used for high-pressure hydrogen gas; in particular, are particularly vulnerable to issues such as embrittlement, hydrogen-induced cracking and high-temperature hydrogen attack, all of which can result in catastrophic failures of the storage vessels.

From an environmental perspective, the high-pressure compression method for storing hydrogen in a storage vessel is considered to be environmentally friendly. In contrast, the liquefaction process for hydrogen storage is less eco-friendly and poses greater hazards due to its lower energy efficiency, which correlates with higher pollutant emissions. In line with this, the use of metal hydrides for high-pressure hydrogen storage raises concerns regarding the disposal of spent metal/alloy materials, as well as the energy demands associated with compressors and storage vessels

Author Contributions: “It's a single-author article."

Funding: "This research received no external funding."

Data Availability Statement: “The data are available at request.”

Conflicts of Interest: “The authors declare no conflict of interest.”

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