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RESEARCH ARTICLE

From Grey to Green: Hydrogen Storage for a Net-Zero Earth

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ABSTRACT

Hydrogen storage is essential for enabling hydrogen as a clean energy source, supporting the transition to a sustainable and reliable energy system. This review paper uncovers a comprehensive analysis of the current technologies and challenges associated with hydrogen storage, including solid-state, compressed gas, and liquefied hydrogen storage methods such as chemical hydrides, metal hydrides, and carbon-based materials. Emphasis is placed on the energy density, safety, efficiency, and cost-effectiveness of each storage method, as well as the advancements in materials science that are driving innovations in storage solutions. Additionally, the paper explores the potential applications of hydrogen storage in renewable energy integration, transportation, and grid stabilization, highlighting its role in enhancing energy security and reducing carbon emissions. The review concludes with an outlook on future research directions and the potential for hydrogen storage technologies to contribute to a low-carbon economy.

Keywords: Chemical Hydrides, Cryo-compressed, Hydrogen Storage, Physical Sorption.

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1. INTRODUCTION

For centuries, the prime energy source on the globe has been fossil fuels, mainly coal, gas, and oil. International Energy Agency (IEA) estimated that conventional fossil fuels contributed 84% of the primary energy consumed worldwide in 2020 [1]. Unfortunately, this extensive use of fossil fuels results in serious environmental issues around the world, which emit approximately 78% of greenhouse gases [2]. World leaders have come to the realization that hydrogen must be allocated a fair and substantial portion of the total energy required within the first half of the 21st century to limit the rise of global temperature to at least 1.5°C [3], [4]. As the shift to renewable energy sources continues, the intermittent nature of these sources is becoming increasingly evident and new energy storage technologies are needed to balance demand and supply [5].

Because of its long-term energy storage capacity, hydrogen has become one of the major reliable renewable energy carriers that helps the intermittent problems related to renewable energy sources [6]–[8]. However, the chemical properties of hydrogen raise principal concerns about storage. One kg of hydrogen gas occupies more than 11 m^3 under normal temperature and atmospheric pressure, making hydrogen gas the lightest molecule in terms of density [9]. Therefore, hydrogen storage needs to be made more economically viable by increasing its storage density. There are numerous ways to store hydrogen at higher densities. All these techniques evolve around hydrogen-binding compounds or need energy input as heat and work.

Despite being a colorless gas, hydrogen has been assigned a color depending on methods of production and the effects these methods have on the environment (Fig. 1). Approximately 8 kg–9 kg of CO_2 is released to produce each kg of H_2 through methane steam reformation (ammonia generation). "GREY hydrogen" is the term for the hydrogen created when CO_2 is released into the atmosphere. H_2 generated with power extracted from renewable energies is classified as "GREEN" [10]. There are some other colors of hydrogen as well.

Statistics for hydrogen production in 2022 show that it was over 95 Mt (Metric ton), increasing by almost 3% per year. The government has prioritized promoting the production of low-emission hydrogen, paying less attention to the demand side. Less than half of the government's aims for boosting demand, just 14 Mt are centered on increasing the use of hydrogen, compared to the 27–35 Mt total of all targets for low-emission hydrogen production [11]. While storage options for hydrogen are still far behind those for other fuels, storage is also receiving attention in addition to production. The physical hydrogen storage market is

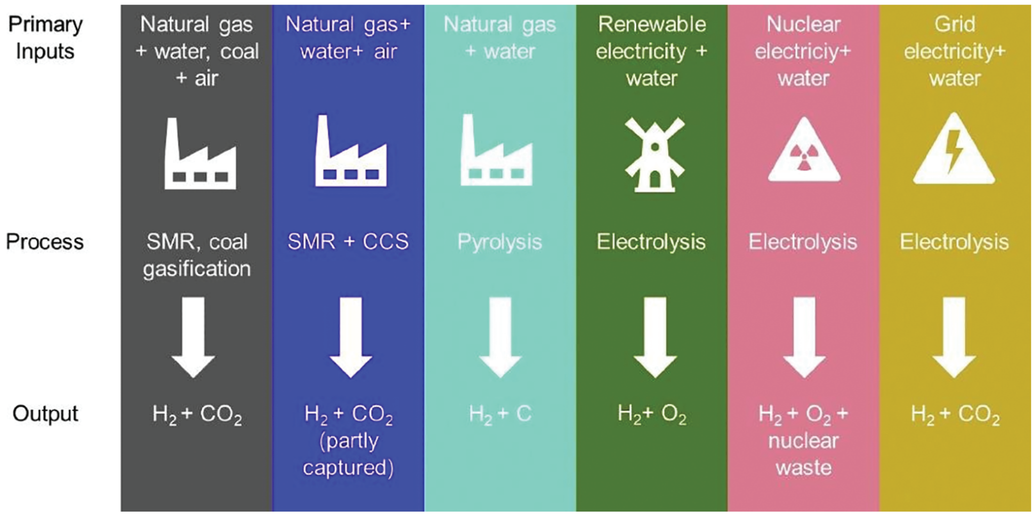


Fig. 1. Colors of hydrogen [10].

growing at a rate of 21.5% annually over the course of the forecast period, to USD 6.3 billion by 2030 from USD 1.5 billion in 2023 [11].

It's possible that earlier studies didn't fully assess how scalable the green hydrogen storage options they suggested were. This may raise questions regarding how well the suggested techniques can achieve large-scale storage applications. As a result, a more thorough examination of the various large-scale hydrogen storage solutions is necessary. Furthermore, it's possible that earlier studies did not fully consider the potential effects of outside variables on the viability and execution of their suggested remedies. Future studies addressing these constraints will help build a deep understanding of the opportunities and problems closely related to large-scale green hydrogen storage, which will result more efficient and knowledgeable decision-making in this crucial field.

This work reviews and analyzes the options for large-scale hydrogen storage. By reviewing global progress and challenges of hydrogen storage technologies and infrastructures, like liquid organic hydrogen carriers, solid-state hydrogen storage, compressed, and cryogenic, it also aims to close this gap in the literature. It is not, however, tried here to unveil the exhaustive list of all hydrogen storage materials. This paper satisfies the ultimate goal of identifying and evaluating advanced hydrogen storage methods in the major categories above the surface.

2. Green H_2 Production Methods

Photocatalytic water splitting process can be utilized to produce green hydrogen from excess renewable energy [12]. This hydrogen can then be stored and eventually oxidized to generate electricity when required. This enables the utilization of renewable energies in certain industries where fossil fuels now hold a dominant position, as well as the replacement of blue or grey hydrogen by exchanging green hydrogen. Renewable sources are unstable because of the weather and climatic variations, and they are dissociated from the energy demand unless backup systems are in place to stabilize swings. This makes the energy supply chain sustainable, which is essential for growing the renewable energy-based hydrogen economy.

Every link in the energy supply chain, from generation and distribution points of view, which consists of homes, businesses, and other sectors, needs some level of buffering. For hydrogen to be incorporated into the energy supply chain at every stage, storage methods for the gas must be developed successfully. There are two types of hydrogen applications in energy economy: mobile and stationary. In manufacturing and static power generation, terminals, or end-use points, are the stationary uses for on-site energy storage. Static applications, in contrast to mobile ones, can function at higher pressures and temperatures and be comparatively big [13]. Stationary applications include things like home power generators, backup power supplies, and power delivery to off-grid areas [14]. The main benefit of hydrogen is its current 94 million tons of annual global demand, which is mostly fulfilled by stationary storage systems (SSS) [15]. Many industries are currently using hydrogen with SSS, including food processing, heat generation, electronics, cosmetics, fertilizers, methanol, ammonia, glass, and steel, and refining processes like desulfurization and hydrocracking [16], [17]. With the significant inclusion of the mobile industry, it is anticipated to surpass 2.3 gigatons [17]. In contrast to fixed uses, mobile hydrogen storage is anticipated to become the norm in the future. It can be used for both carrying and burning as a fuel for vehicles [18]. Quick energy intake and release kinetics, high gravimetric and volumetric energy densities, suitability for normal operating temperature and pressure settings, built-in safety, and economic viability are all desirable characteristics of an ideal hydrogen storage system. In contrast to other fuels, its volumetric energy content is relatively low at 9.9 MJ/m^3, although the energy density is greater by weight [19]. For integrating hydrogen across several sectors, there isn't a single storage option that is universal. To meet the unique operating conditions, capacity, and demand, several storage systems are needed depending on the end-user application and the supply chain component. Large-scale storage facilities are needed to implement hydrogen on a national basis to use it as a major export commodity or to power electrical grids

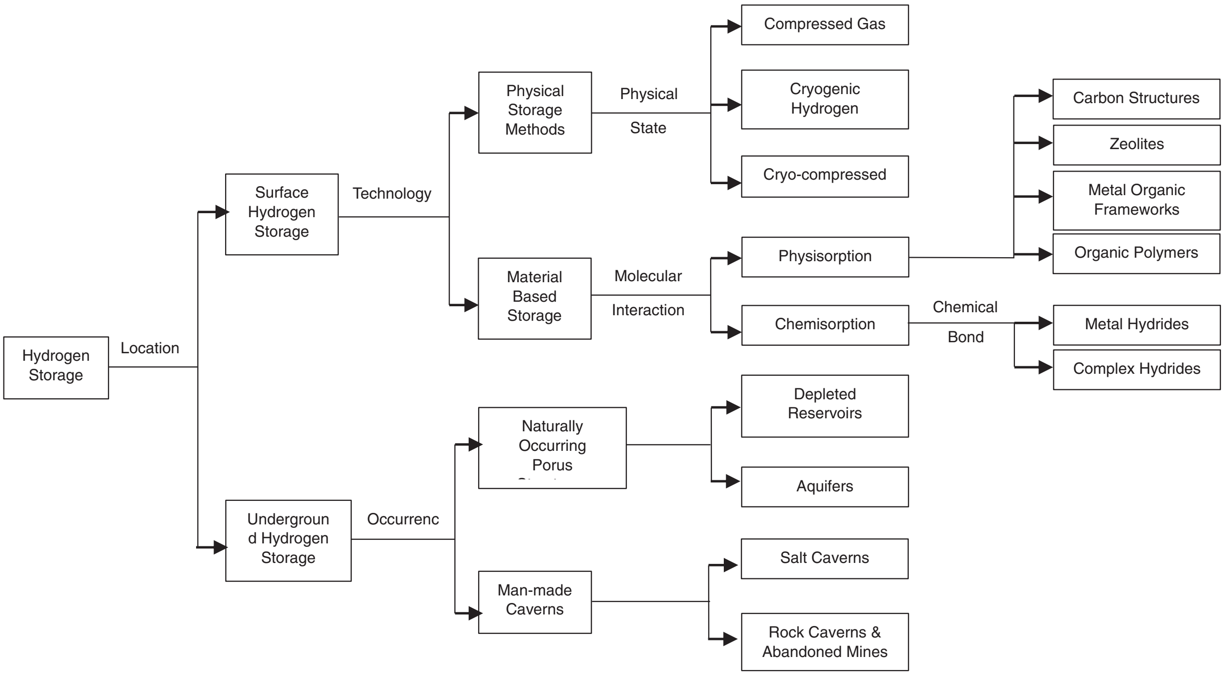


Fig. 2. Different types of hydrogen storage solutions [20].

and establish it in place of natural gas. When renewable energy is abundant, based on climate conditions, it may be used to make green hydrogen and can be stored in these mediums and produced again when the demand is at its highest. The daily energy consumption needs of end-user applications must be satisfied by the lower-capacity storage systems. The literature has examined a wide range of storage options for various application kinds. As seen in Fig. 2, they can be categorized into two classes according to where they are located: surface and subsurface facilities. Depending on how they occur, the subsurface storage media can be further divided into different categories. The methods for storing hydrogen that the surface storage alternatives use can be categorized. The ensuing sections and subsections go into greater detail on the various kinds of hydrogen storage solutions. Only the surface facilities are discussed here.

3. COMPRESSED GAS STORAGE OF HYDROGEN

A commonly used physical storage method for hydrogen is high-pressure storage in vessels [18], [21]–[24]; over 80% of processes use this method [25]. The petroleum industry already uses compressed hydrogen to hydroprocess and hydrocrack crude oil. Additionally, natural gas can be replaced by gaseous hydrogen usable in the transportation sector, heat generation, and combustion, which account for 33% of all emissions and 50% of the world's energy consumption [26], [27].

Compressing hydrogen is energy-intensive, with a very low density of 0.083 kg/m^3 at NTP. It typically kept at 200 bars of pressure in steel cylinders. These are the most typical hydrogen tanks seen in regular industrial settings. A very low gravimetric density of about 1% can be achieved with these cylinders [9]. To avoid risking the volumetric and gravimetric capabilities, the storage pressure must be raised. When considering hydrogen storage for mobile applications, these restrictions increase even further [28]. US Department of Energy (DOE) has set up the necessary volumetric and gravimetric targets for light cars, which are 0.030 kgH_2/L and 4.5 wt%, respectively [29]. However, only Type IV vessels are close to achieve these predetermined goals, with 4.2 wt% and 0.024 kgH_2/L [30]. The totally composite Type V vessels without liners have the capacity to exceed the 6.5 wt% goal. The set target is only approached by Type IV vessels, whereas Type V vessels under development are anticipated to meet the objective [28].

As illustrated in Fig. 3, pressure vessels that are often used in industry today can be divided into five categories [31]. The key points of every type of pressure vessel are presented in Table I. Most vessels in use today are Type I, which are comparatively easiest to design and produce. Type II, III, and IV vessels typically use a plastic or metal liner for gas containment and carbon fiber to handle all or partial structural load, referred to as composite overwrapped pressure vessels (COPVs). Type IV vessels have the highest storage densities and the lightest weight, which makes them ideal for many ground transport applications. The Honda Clarity and Toyota Mirai, two popular hydrogen fuel cars, use Type IV vessels that maintain 700 bar operating pressure [49], [50].

Fig. 3 depicts the construction diagram of cylindrical Type IV vessels. To stop gas leaks, a plastic liner, usually made of HDPE or PA [32], is put in use. Polar apertures with metal end bosses are located at the ends of the vessel. The boss is used to fill and empty the vessel, and it frequently has sensor access [33]. In Type I–III vessels, the boss is usually incorporated along the liner; Type IV–V vessels, on the other hand, have separate metal or composite components.

Compared to other types, as Type IV vessels are still relevant to many businesses today, there is a lot of research

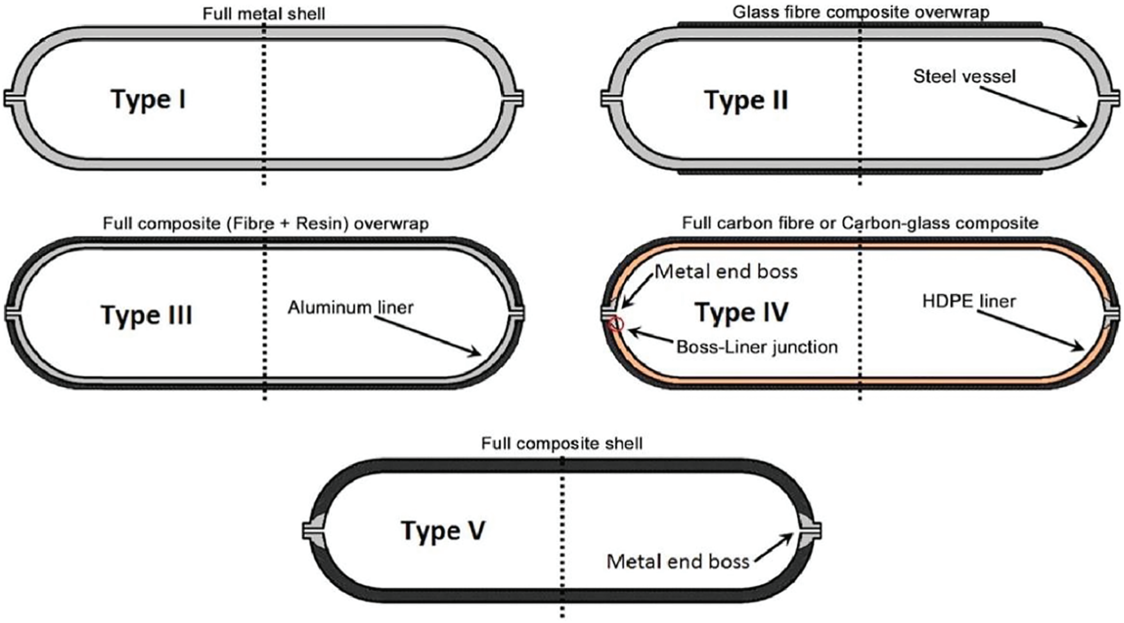


Fig. 3. Types of hydrogen gas storage in pressurized vessel [28].

TABLE I: Classification and Key Points of Every Type of Pressure Vessel

Attributes	Type–1	Type–2	Type–3	Type–4	Type–5
Construction	Metal tank (Steel or Aluminum)	Steel or aluminum liner composite hoop wrap	Steel or aluminum liner composite full wrap	Plastic liner composite full wrap	Composite cylinder without liner
Load sharing	Withstands the whole load	Equally shares the load	Most load is carried by the composite structure	Composite material carries the load	Composite material carries the load
Operating pressure (MPa)	25–70	70–87.5	87.5–110	70–87.5	87.5–110
Density (gm/liter)	Around 15	Around 20	Around 25	Around 40	Around 20
Weight	Relatively high	Lighter than Type I	Half of Type II	Lighter than Type III	Lightest among others
Cost (USD/kg of H_2)	7–10	5–8	4–6	7–10	5–7

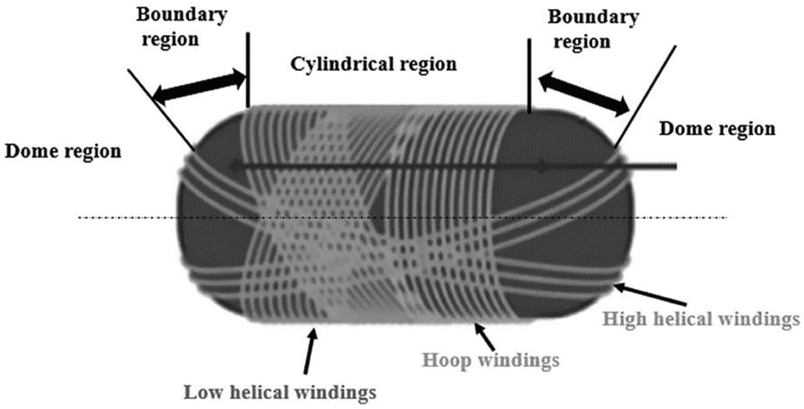
being done on them. However, Type V represents composite pressure vessel technology. Since there is no internal liner in Type V or linerless vessel, the composite serves as both the load-bearing structure and the gas barrier. They can be called composite pressure vessels, or CPVs for short, because the composites are not overwrapped on the liner. When using a no-liner strain, compatibility between composite and liner is no longer required, which improves fatigue performance and may result in weight savings of 10% to 20% [34], [35]. There is still more research to be done on Type V tank operating pressures [30].

The compressed hydrogen storage tanks experience temperature fluctuations and cyclic loads at high pressures. Temperature- and cycle-related stresses must be taken into consideration throughout the design phase. Burst pressure is typically the cause of pressure vessel collapse. It is the pressure at which internal fluid leaks from the vessel due to a crack [35].

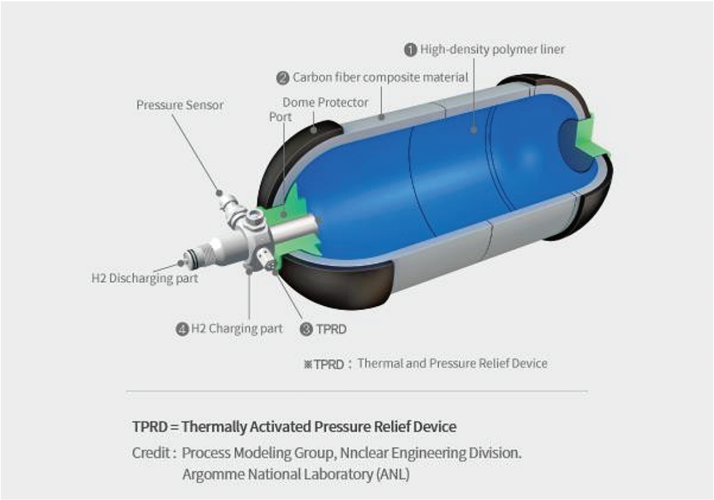
Nevertheless, high-pressure tank failure is a complicated process that can be brought on by thermal (thermal fatigue) or mechanical (burst pressure) factors. Coupling these two events during charging and discharging may result in more difficulties. For an extended tank life, the liner strain should need to coincide with the composite strain. If there is not enough resin binding, local strain in fibers can cause holes that allow penetration [36]. The property of hydrogen is that it is a small, odd molecule and has the propensity to leak, which makes this worse.

Most of the metallic components found in high-pressure tanks are made with steel and aluminum 6061 or 7061. Fiber, like carbon fiber reinforced polymer (CFRP), is utilized to reinforce the polymer sections. By offering great strength at a much-reduced weight, this technique improves storage performance [37], [38]. They are made via continuous filament winding, which involves winding fiber filaments infused with resin around a mandrel that rotates. Following that, the fiber solidifies with the resin–i.e., epoxy, vinyl ester, etc. Finishing the winding process, the whole assembly is heated at a very high temperature for curing. Finally, a hollow structure is obtained by eradicating the mandrel [35]. The strength of the fiber-reinforced component can be optimized by aligning the principal stress direction and fiber orientation together. The strength of the final composite is, therefore, also influenced by the form of fiber tape winding, whether it be unidirectional, angle-ply, or cross-ply [39]. Helically stacked fiber is the norm. Nonetheless, additional layers are added circumferentially; they can be referred to as hoop layers since hoop tension in a pressure vessel is double the axial stress.

Without any support, it is impossible to layer the fiber circumferentially on the dome. Because of this, the dome is strengthened by introducing in-plane perpendicular doilies [40], [41]. The type-IV tank's layering design is seen in



(a)



(b)

Fig. 4. Type-IV tank made by Doosan Mobility (a) Winding pattern and (b) Sectional view.

Fig. 4a. Optimization studies are conducted for lowering carbon fiber without sacrificing its strength since carbon fiber causes most of the tanks' costs. Numerous research works have examined the impact of winding design and thickness on tank strength [42]–[44]. A typical type-IV tank made by the South Korean UAV manufacturer Doosan Mobility is depicted in Fig. 4b. The tank includes carbon fiber-reinforced composite body and high-density polyethylene liner. In discharge mode, there is a fast-coupling mechanism that enables a plug-and-play connection.

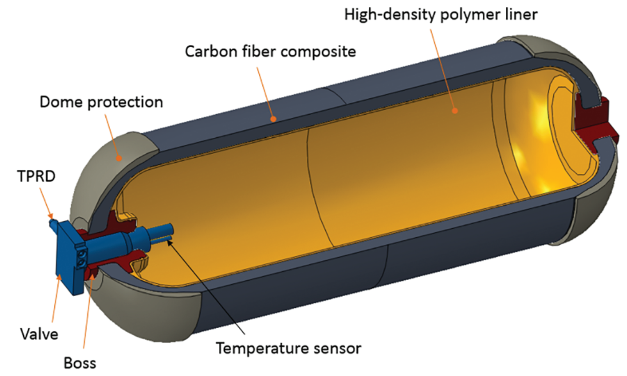
4. Liquid Hydrogen

For large-scale storage, it is thought to be the most promising technology due to a number of benefits, including larger gravimetric energy density and lower volumetric densities, which are 2.7 times and 3.7 times, respectively, compared to petrol. Japan commissioned an Air Liquide hydrogen plant that boasts the world's largest liquid hydrogen (LH2) storage capacity of 1250 tons [45].

As illustrated in Fig. 5, hydrogen needs to be chilled to cryogenic temperatures, which are normally lower than −253°C, for liquefication. After that, delivery vehicles are filled with LH2, and they are driven to distribution centers, where they vaporize it to create a high-pressure gaseous product that can be distributed. Several industries have used LH2 for commercial applications. To make the liquefaction process energy-efficient, several challenges, such as green LH2 boil-off and security for large-scale storage, restrict its widespread use despite its numerous advantages.

4.1. *Materials Selection for Liquid Hydrogen Storage*

The materials used as LH2 storage must fulfill several unique specifications, such as being fire and heat-resistant, mechanically strong, and resistant to hydrogen embrittlement and penetration. The term "hydrogen embrittlement" describes how exposure to hydrogen can cause the metal to shatter under room temperature and cryogenic temperatures. Temperature, pressure, and alloy strength all have an impact on the properties of hydrogen embrittlement [46]. Hydrogen embrittlement has macroscopic impacts, such as weakened steel alloy material strength, although hydrogen is primarily entrained into the metal lattice at the atomic level [47]. Compared to gaseous hydrogen, liquid one causes much less hydrogen embrittlement because of



TPRD = Thermally Activated Pressure Relief Device
Credit: Process Modeling Group, Nuclear Engineering Division. Argonne National Laboratory (ANL)

Fig. 5. Liquid hydrogen storage technology [20].

the reduced hydrogen material penetration at low temperatures [48]. Stainless steels are the predominant materials popularly utilized for storing liquid hydrogen, offering varying qualities and uses depending on their alloy components and grades. For instance, 316L stainless steel that has had molybdenum (Mo) added to it resists chloride ion corrosion better, making it perfect to be used in maritime environments. The titanium (Ti)-infused 321 stainless steel performs comparatively well in terms of high strength at elevated temperatures and resistance to intergranular corrosion. Studies on laser shot peening, ion implantation, cathodic protection, and surface coating have also been conducted to strengthen stainless steel's resistance to hydrogen embrittlement corrosion [49]. One of the biggest obstacles to the use of lightweight composite materials is hydrogen penetration.

Since the freezing point of hydrogen is −423.17°F and cooled to low temperatures in order for liquefaction, the process takes a lot of energy [50]–[52]. Therefore, for LH2 to be successful in considering future energy demands, it is necessary to boost energy efficiency and decrease specific energy consumption (SEC). SEC is typically referred to as the amount of energy needed for creating, transporting, and storing one unit of LH2. Several ideas with energy efficiencies between 40% and 60% and SEC values between 5 kWh/kg-LH2 and 8 kWh/kg-LH2 have been presented. From cutting-edge plants to recently created large-scale liquefaction operations, Cardella lays forth a roadmap to enhance hydrogen liquefaction technology. For each kg, 100 tpd (tonnes/day) LH2 plant's SEC was found to between 5.9 kWh to 6.6 kWh while upscaling from 5 tpd to 50 tpd resulted in a 60% reduction in specific liquefaction costs in five years [53]. Yunus created a unique method of liquefying hydrogen that is based on cycles of liquefaction cooled by helium. They mentioned that the liquefaction process's efficiencies are recorded as 70.12% in energy and 57.13% in exergy, respectively [54]. Shaimaa suggested a unique strategy that combines isobutene and geothermal power plants with a large-scale hydrogen liquefaction technology. It is discovered that SEC drops to 4.7 kWh/kg-LH2 when hydrogen pressure is raised to 20 bar and mass flow rate is decreased by 9 kg/s. In particular, the energy, energy efficiency, and SEC are respectively 19.8%, 63.7%, and 6.47 kWh/kg-LH2 [55]. Several more contemporary and efficient configurations (such as the Brayton cycle liquefaction and Linde-Hampson system) have been proposed for efficiency enhancement, but most hydrogen liquefaction plants are normally operating under the pre-cooled Claude system, which remained un-upgraded in the last 50 years.

The boil-off phenomenon is a significant obstacle in the storage and transportation of LH2. The LH2 starts to evaporate and turn into a gas when it hits its boiling point, which can cause the system's pressure and temperature to drop as well as cause fuel to be lost. In order to lessen or completely eradicate this boil-off, zero-boil-off (ZBO) technology has been developed that combines insulation and heat management strategies. Gyu-Mok Jeon studied the effects of filling ratio (FR) on boil-off gas and thermodynamic properties for cryogenic liquid fuel tanks. They discovered that it is of high priority to increase the heat and flow transfer properties at the bottom of the tank [56].

However, the development and deployment of ZBO technologies are accompanied by a number of difficulties. One is its performance and dependability over the long run. For large-scale storage, the technology might be more difficult to scale up, and its long-term performance and reliability might not be well understood. Furthermore, ZBO technology can be costly because it requires sophisticated refrigeration systems and insulation materials. More research may be necessary to assess whether ZBO technology can be widely implemented across a range of businesses.

5. Cryo-Compressed Hydrogen Storage

It combines the characteristics of both liquid and gaseous hydrogen, referred to as hybrid technology. In place of liquified hydrogen storage systems, it is compressed roughly at 233°C [14]. Compared to other hybrid methods discussed above, the storage density of supercritical cryogenic hydrogen is 80 g/L, making it safer for storage. It is possible that cryo-compressed hydrogen will reach the capacity targets established by DOE [57]. The automobile driving range allowed by cryo-compressed vessels is comparable to that of a car driven by petrol. The integration of heavy-duty vehicles is favored by the pumps and boats that offer cost-effectiveness and speedy refueling. The cost of the big storage containers for buses with 350 bars of design pressure is anticipated to be $10/kWh [58].

The cryogenic fluid's internal pressure must be able to be maintained within the storage vessel. Since the tanks are made to function at high pressure, boiling off has no effect on the cryo-compressed hydrogen [13]. At higher temperatures, an additional factor in prolonged dormancy is the release of hydrogen from high-pressure tanks [57]. Aceves proposes a Type III cryo-compressed tank that is utilized for cars, has an alloy liner wrapped in carbon fiber, and is separated from the elements by a thin layer of stainless steel or aluminum, as depicted in Fig. 6. At low temperatures, the carbon fiber-covered alloy liner becomes stiffer and stronger. To provide insulation, a vacuum void filled with vacuum superinsulation separates the inner and outer shells. BMW pursued the concept, which came from Lawrence Livermore National Laboratory [59]. The hybrid

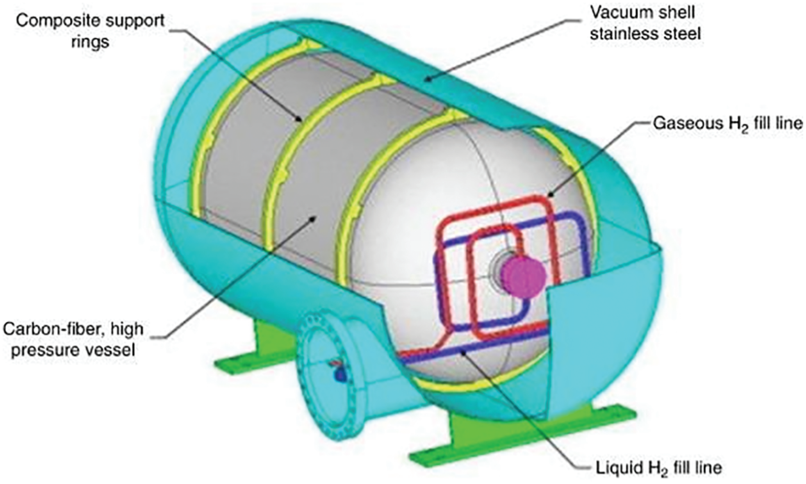


Fig. 6. Type III cryo-compressed tank [60].

hydrogen model Toyota Prius used for this storage system was able to store 10.7 kg of hydrogen to roughly transport 1050 km, and it successfully holds hydrogen for two weeks without evaporation [60]. This hydrogen storage technique gives the highest storage efficiency for both volumetric and gravimetric values.

The technology with the lowest capital cost is cryo-compressed storage. But because liquefaction is so expensive, the total cost of cryo-compressed hydrogen is higher, roughly $4.8/kg, and its energy efficiency is reduced [61]. More so than the other two forms of physical storage, it is also impacted by power usage and heat loss [30], [62].

6. CHEMICAL HYDRIDES HYDROGEN STORAGE

Liquid organic hydrogen carriers (LOHC), currently considered the most popular, are discussed in this section, along with the most recent industrial demonstrations that have been put into service globally. Articles [63]–[66] contain a number of reviews based on storage methods. To power the fuel cell vehicles, Mori and Hirose [63] explained that high-pressure and cryogenic storage were not practical. As a result, they combined high-pressure storage and metal hydride, developing a hybrid system. Satyapal *et al*. [64] conducted an assessment for numerous hydrogen storage techniques to fulfill the 2010 target set by the US DOE. They came to the conclusion that individual material's gravimetric and volumetric storage capacities must be prioritized over system-level limits. Ren *et al*. [65] did some critical studies on absorption and adsorption-based hydrogen storage materials finds no significant benefit to adsorption-based storage when it comes to scaling up from lab prototypes to commercial applications. Lai *et al*. [66] also reported similar findings. Furthermore, it can be inferred from recent studies that very few studies have been conducted for large-scale hydrogen storage [67], [68]. Conventional storage methods, like liquid hydrogen and compressed gas form, have drawbacks that include high cost, safety risk, and transportation-related problems [69], [70]. These significant problems are resolved by the liquid organic hydrogen carrier, which also offers very safe, inexpensive hydrogen storage with extended storage times and long-distance hydrogen transportation [28], [71].

There are several chemical hydrides that demonstrate the capacity to store hydrogen in literature, including liquid hydrogen organic carriers, formic acid, methanol, and ammonia [72]. The chemical hydrides are appropriate for handling and transporting hydrogen since they are known to remain liquid across a large temperature range. Attracting these kinds of hydrides has the major benefit of not requiring extra infrastructure for their manufacture. The industrial facilities for storage and transportation that are currently in place are quite beneficial.

Operating conditions with pressures ranging from 10 to 80 bar and temperatures ranging from 220°C to 280°C are required for methanol to be used as a storage medium (hydrogenation) of hydrogen. Excess amount of heat is released throughout the process and can be recovered in many ways [73], [74]. Once more, heat energy is needed for separating hydrogen by the dehydrogenation process.

The primary benefit for using ammonia as hydrogen storage is its huge 17.6 weight percent (10 bar) storage capacity. The main drawback, however, is that larger storage plants must operate at high temperatures during the release (dehydrogenation) process [74]–[76]. An alternative to the dehydrogenation issues encountered in hydrogen storage based on methanol and ammonia is formic acid. On the other hand, formic acid only has a 4 wt.% storage capacity.

Hydrogen is stored using LOHC technology in two stages: first, it is loaded, called hydrogenation, and then emptied, known as de-hydrogenation. A catalytic reaction results in storage in the LOHC method, and the same process occurs during dehydrogenation [28], [77], [78]. Due to low energy density, compressed and liquid hydrogen storage systems are inefficient, as noted by Preuster *et al*. [78] and Aakko-Saksa *et al*. [77]. They also provided a critical analysis of circular energy carriers, including liquid hydrogen organic carriers, toluene, and methanol. Additionally, they concluded that as these energy carriers offer safe, dependable, and adaptable options in terms of storage capacity and sustainable renewable energy supply chain, they have been easily integrated with existing technologies in nations like Japan and Germany. The most well-known LOHCs for the hydrogenation process are N-ethyl carbazole [79], [80], dibenzyl toluene [81], and toluene [82]. The working temperature and pressure for this process are 200°C and 10 bar, respectively. However, the industries already have a system in place for the dehydrogenation of LOHC. Methylcyclohexane and toluene are two examples of the various compounds that make up the LOHC, which is considered the best hydrogen storage method [67], [82], [83]. Typically, the storage densities of LOHCs are ranges between 5 wt% to 6 wt%. By adjusting the working conditions appropriately, the experiment done by Jorschick *et al*. [84] revealed that a single catalyst is quite feasible and usable for hydrogenation and dehydrogenation processes. The carrier molecules must be transported to the location of re-hydrogenation by the LOHC following dehydrogenation.

7. SOLID-STATE HYDROGEN STORAGE (SSHS)

Physisorption [85], [86] or chemisorption [22], [87] principles are utilized for hydrogen storage in solid state. In comparison to gaseous or liquefied forms of storage, solid-state technologies need less energy and may even be safer [88]. The main solid-state hydrogen storage techniques are physisorption-based and chemisorption-based. Large pore volume and surface area characterize these physisorption-based storage materials [89]. But, the practical application of these physisorbents is restricted by the necessity of maintaining an ultra-low temperatures (77 K) at 1 atm pressure though they exhibit substantial reversible storage capacity and excellent hydrogen uptake and release kinetics [90]. One type of chemisorbent that provides exceptional hydrogen storage properties over an extended pressure and temperature range is metal hydrides (MH) [91]. Depending upon the hydrogen bonding nature, these MH materials are clearly classified into three groups: chemical, intermetallic hydrides, and complex [92]. DOE sets a system-level hydrogen storage targets for light-duty automotives, aiming for 40 gH_2/L volumetric and 55 $g\text{-}H_2/kg$ gravimetric storage by 2025.

5.6 kg is the entire usable hydrogen capacity, and it has to be refueled in three to five minutes. Because interstitial metal hydrides include heavy elements, their gravimetric storage capabilities (wt.% <3%) are limited. Although the amount of hydrogen stored can be increased by partially substituting these interstitial hydrides with appropriate elements, it remains behind the necessary DOE targets. A small number of elemental hydrides (such as MgH_2), chemical hydrides, and complex hydrides, among other SSHS materials, have the potential to theoretically fulfill the storage density targets; however, these materials don't meet the on-board refueling requirements, exhibit irreversible or thermally stable reaction kinetics, or are too sluggish [93]. Many techniques are employed to address the suitability of complex hydrides for onboard applications, including thermal destabilization, catalyst addition, and nano-confinement. Chemical hydrides, on the other hand, are irreversibly done. Proven methods to reduce the heat reaction and influence the kinetics of magnesium-based hydrides include catalyst doping and surface modification [94].

8. PHYSICAL SORPTION

Hydrogen is stored on solid surfaces through the process of adsorption of hydrogen molecules. With 4–10 kJ/mol of binding energy, the adsorption creates weak Van der Waals bonds, making the physisorption reversible and straightforward to manage lossless and fast adsorption-desorption kinetics [95]. Large surface area materials are usually preferred for hydrogen storage. These materials include covalent organic frameworks (COF), metal-organic frameworks (MOF), zeolites, polymers of intrinsic microporosity (PIM), and porous carbon-based material. The operating conditions, pore volume, and surface area all affect the storage capacity that is provided. A variety of uses, such as phototherapy, photocatalysts, supercapacitors, batteries, and fuel storage, are made possible by the physisorption.

The typical problem with physisorption-based storage solutions is that they cannot attain reasonable storage capacities below 77 K, which demands expensive and energy-intensive chilling processes. Since hydrogen has a low binding energy, cryogenic conditions become necessary [18]. The constraints of carrier material, such as low weight and storage capacity, make it harder to be used for practical purposes [96]. Moreover, the necessity of tank insulation and thermal management systems significantly lowers the performance of this storage method [97].

8.1. Carbon Structures

Carbon structures like benzene but with distinct configurations are being investigated for their potential to store hydrogen and their affordability for usage in cars and portable electronics. These structures fall into two categories: irregular structures, such as carbon aerogels (CA) and activated carbon (AC), and those that have longer carbon hexagons, such as carbon nanofibers (CNF) and carbon nanotubes (CNT). Other carbon compounds, such as graphene and templated carbon, have been investigated for hydrogen storage. Two methods can be used to increase the adsorption of carbon materials: doping carbon with light elements, which can increase specific surface area by pore geometry optimization, and carbon network polarity by introducing electron deficiency.

Single-walled (SWCNT) and multi-walled (MWCNT) are two types of carbon nanotubes (CNT) that may be categorized according to the wall structure [98]. Since their discovery in 1991, CNTs' capacity to store hydrogen has been extensively studied [99]. Under various experimental settings, storage capacities using CNT methods are obtained between 0.25 wt% to 56 wt% [100]–[111]. Nonetheless, up to 6 wt% average storage capacities are still thought to be beneficial [112]. The various capacities obtained in the literature are mostly due to the variations in the processes used to produce (synthesis) CNTs. At room temperature and 77 K, the specific pore volume or surface area of microporous carbon materials determines their capabilities of hydrogen storage [113], [114]. These materials have strong cyclic stability and are expensive, too.

Activated carbons are inexpensive synthetic carbons having pore diameters in between micropores to macropores and a specific area of up to 3000 m^2g^{-1}, which makes it a viable choice for physisorption. The majority of AC's hydrogen uptake value is approximately 2.5 wt%, although it can range from 0.2 wt% to 5.5 wt% [115]–[117]. The greatest capacity at ambient temperature and 70 MPa pressure is reported as 1 wt% [118]. CA exhibits a linear connection between adsorption and surface area, with maximum hydrogen capacity recorded as 5.3 wt% and surface area up to 3200 m^2g^{-1} [119].

Chemical deposition method creates CNFs by breaking down carbon-bearing gases from stacking sheets on catalyst surfaces that are one on top of the other [120]. At room temperature and pressure, its capacity ranges from 2.2 wt% to 6.5 wt% [121], [122]. The disadvantages of this method are the expense and slowness of the process. However, doping with different elements had a very small impact.

Through the process of templating, porous materials are converted into porous carbon materials. Depending on the synthesis techniques, tunable hydrogen capacities containing microporous and mesoporous carbons can be produced [123]–[126]. At cryogenic temperatures, the literature reports hydrogen adsorption capacities range from 7.8 wt% to 1.78 wt% [127]–[132]. Honeycomb-structured carbon foam is a possible alternative because of its natural mechanical and structural qualities [133]. Another technology with a large capacity for storing hydrogen is graphene. Hydrogen atoms can be added to it to create graphene, which can then be heated to 450°C to release stored hydrogen [134].

8.2. *Zeolites*

Zeolites are a kind of component having three-dimensional crystalline aluminosilicates that possess consistent pore diameters, an ordered structure, and a large interior surface area of 1000 m^2g^{-1} [133], [134], [141]. At high temperatures and pressures, encapsulation drives hydrogen into the zeolite pores, where it is trapped at ambient temperature [135]. At room temperature, it has capacities of below 0.1 wt% [136]. Zeolites have been shown to have a capacity of up to 2.55 wt% when the temperature decreases to 77K [98], [137]–[140]. Doping the structure with metals [141] and bridging and spillover mechanisms [142] can both increase the capacity.

8.3. *Metal-Organic Framework (MOF)*

This crystalline material contains pores in it to store hydrogen by diffusing through those crystals [143], [144]. It also contains inorganic metal oxide units and organic linkers and has specific surface areas of 4800 m^2g^{-1} [145]. Their enthalpy is higher, and their density is lower [146]. Only a few seconds are needed for adsorption [147], and the rate is determined by crystal size and the hydrogen diffusivity of the MOF [148]. According to several studies, MOFs can provide capabilities of up to 7.5 wt% at 78 K and 1 wt% at room temperature [139], [149]–[151]. The MOFs' ability to store hydrogen was enhanced by the addition of platinum and palladium nanoparticles [152]. It is expected that metal ions like Mg^{2+}, Cu^{2+}, and Li^{+} will enhance MOFs' ability to store hydrogen under ambient circumstances [153].

To charge and discharge the hydrogen for vehicle applications, two designs, namely, the modular adsorption tank insert (MATI) and the HexCell, have been developed which control thermal kinetics. Each design can store up to 5.6 kg of hydrogen [154]. Type 1 aluminum welded tank of the Savannah River National Lab-modified HexCell system weighs 96 kg in total, excluding hydrogen, where the weight of the MOF assembly is 32 kg. For hydrogen discharge, it is electrically heated, and for adsorption, liquid nitrogen cools it. The internal heat transmission mechanism and MOF of the MATI system are different from those of the HexCell. The powdered MOF is compressed into discs measuring 40 cm in diameter and 5 cm in thickness. 41 kg of MOF is needed for the MATI system, having a total of 102 kg weight.

The limitation of this material is that they have very low thermal conductivity, which means that managing MOF-based storage systems requires a lot of resources [155]. The industrial penetration of MOF is hampered by the initial expensive capital investment and high raw ingredients costs; this includes metal salts and organic ligands [156].

8.4. *Organic Polymers*

Low cost, high stability, high specific surface area, very light atomic composition, and regulated porosity of H, N, C, and O are the principal causes for the popularity of organic polymers. They also have the benefit of polysynthetic functionalization and changeable structure. The capacity of organic polymers to hold hydrogen is covered in detail [157]. The term "organic polymers" refers to crystalline nanoporous organic polymers based on carbon (COF), porous aromatic frameworks (PAF), conjugated microporous polymers (CMP), hyper-crosslinked polymers (HCP), and PIM. Low densities, large surface areas, and strong mechanical and thermal stabilities characterize the PAF and COF. In the range of 3.5 wt%–10.0 wt%, COF has good storage capabilities at cryogenic temperatures (77 K) and high pressures (80 bars–100 bars); however, in high-temperature environments, the capacity sharply decreases. Similarly, at high pressure and room temperature, PAFs can provide increased capacity ranging from 7.0 wt% to 16.03 wt%. The doping with metals Li, Ca, and Mg produced increased capacity values. Although COF and PAF exhibit larger capacities, their use is restricted by their poor processability, high cost, and mechanical qualities because they are in powder form. Besides, low surface area restricts the storage capacity of CMP, PIM, and HCP despite their low cost and excellent processability. The HCP offers less than 0.5 wt% at room temperature and less than 4 wt% hydrogen absorption capacity at cryogenic temperatures. In atmospheric pressure settings and at 77 K, CMP revealed values less than 3 wt%. However, 6.1 wt% was seen when lithium was added, whereas 3.7 wt%–4.1 wt% is demonstrated by PIM.

9. CHEMICAL SORPTION

This process works by breaking the hydrogen molecules into atoms and forming a chemical connection that diffuses the atoms. Hydrides can be classified as covalent, ionic, or metal (metallic) bonding based on the type of bond in which they are bonded. Complex hydrides are another name for the latter two [14]. The appropriateness of hydrides as absorption thermodynamic characteristics and kinetics determines a storage medium. Compared to solid hydrides, which need to be heated at least 120°C to release hydrogen, liquid hydrides are more straightforward to use under room temperature.

9.1. Metal Hydrides

Magnesium and transition metals are components of simple metal hydrides, which are created at high temperatures by combining hydrogen with metals and their alloys [14]. Because of the benefits such as low-pressure change requirement, high storage efficiency, and high volumetric density for both hydrogen adsorption and desorption, metal hydrides become the best choice for mobile power devices [158], [159]. Solid hydrides are being examined as a solution to the engineering and safety issues that arise when developing cars with physical hydrogen storage containers, and they achieve a volumetric density of about 115 kgm^3. The storage was volume efficient and safe due to metal hydrides since they can hold more hydrogen atoms compared to liquid or gaseous [91], [160].

As illustrated in Fig. 6, metal hydrides are created when hydrogen gas comes into contact with metals, causing the adsorption of hydrogen gas molecules at metal's surface. Following their dissociation, the hydrogen molecules become atoms, which diffuse into the metal structure and occupy the metal crystal lattice's interstitial spaces. The term "α-phase" refers to form of a solid solution having less than 0.1 hydrogen-to-atomic metal ratio. More energy added to the system causes the ratio to rise and the hydride phase to nucleate and expand, referred to as β-phase. The pressure-composition isotherm displays a flat plateau when both phases coexist, and the plateau length is the determinator of how much hydrogen can be stored [90], [161]. The hydrogen-to-atomic metal ratio in metallic hydrides is typically 2 [21].

At normal temperature and pressure, the reversible metal hydrides are created by common transition metals like actinides, lanthanides, and members from vanadium and titanium groups [18], [21]. Obtained performance from transition metals is less than 3 wt% since they often have large atomic numbers.

Surface structure, purity, and morphology all affect a metal's capacity to dissociate hydrogen [134]. Metals typically have oxide layers that vary in thickness. Because the oxide layer prevents hydrogen from chemisorbing, high temperatures and pressures are required to activate the materials in order to use them in practical applications [162], [163]. The inclusion of nanoconfinement, nanostructuring, alloying, and catalysis are some of the techniques utilized to enhance hydrogen absorption and desorption kinetics [23], [162], [164]–[166].

Researchers are trying to find novel light elements for MH storage that will perfectly suit stationary applications such as storage systems or power productions. They can also be adapted for mobile applications by using hydrogen for heating and energy storage [160]. Military and underwater systems, including submarines and autonomous underwater vehicles, have employed metal hydrides. Furthermore, they have been thoroughly investigated for usage in railway applications, as well as in heavy-duty utility trucks and light-weight passenger cars [167].

9.2. Magnesium Hydride

High volumetric capacity, gravimetric storage capacity, lightweight, low cost [168], and abundance [169] make magnesium hydride a desirable option. Its applicability is restricted by the poor sorption, desorption kinetics, and high decomposition temperature above 300°C (at 1 bar) [91], [161], [165], [170]–[173]. The sorption kinetics are improved by producing micron and nano-sized particles, known as ball milling, and by adding nano-metal additions, including ruthenium, palladium, and platinum [174]. Magnesium hydride raises additional safety concerns since it can catch fire in the presence of water or air.

9.3. Complex Hydrides

The light metals from the periodic table under groups I, II, and III, like Li, B, and Al, typically form different types of metal-hydrogen complexes having large capacities. According to article [21], complex hydrides exhibit the tetrahedral structure with B or Al in the core and hydrogen at the corners. Because of their low weight, amides, alanates, and borohydrides have recently caught the interest of researchers [90], [161]. High gravimetric capacity is provided by ionic hydrides. They can only be reversed, though, in situations with high pressure and temperature. While it produces byproducts, the hydrolysis reaction is beneficial in producing hydrogen.

9.4. Boron-Based Materials

Despite the large capacities of Borates, they must also get past kinetic obstacles to offer useful solutions. The composite LiBH4 has a large gravimetric capacity of 18 wt% but can release hydrogen only around 0.3 wt% at room temperature [175]. An elevated temperature of 400°C is necessary for full breakdown. While boron nitride fullerenes have a storage capacity of 3 wt% [176], boron nitride nanostructures show up to 2.6 wt% capacity [177]. The high hydrogen storage capacities of sodium amidoboranes and lithium are 7.5 wt% and 10.9 wt%, respectively, at 90°C dehydrogenation temperature. According to the article, at room temperature and pressure, they are non-explosive, non-flammable, ecologically friendly, and stable solids. They are not readily reversible, though. A technique called "nanoconfinement," in which hydrides are confined within the nano-porous material [178], is used and explored to enhance the kinetics of hydrides [179]–[182]. Elements having atomic numbers less than 20 are extremely susceptible to moisture and air and are improved by the synthesis of lightweight hybrid hydrides [183]. All boron-based complex hydrides require high temperatures in order to release hydrogen, with the exception of $Al(BH_4)_3$, which has a 17 wt% capacity and is liquid at ambient temperature. Furthermore, due to their hygroscopic nature, they are challenging to handle and need a moisture-free atmosphere for storage [90].

9.5. Alanates

Higher hydrogen capacities are found in Alanates, which can reach 5.6 wt% for $NaAlH_4$ and 10.6 wt% for $LiAlH_4$. Compared to $LiAlH_4$, which is thermodynamically unstable and needs 400°C temperature to liberate hydrogen, $NaAlH_4$ has more manageable operating conditions. Ti or V-based catalysts can be used to reduce the release temperature of $LiAlH_4$, and doping with Ti or Zr-based materials can improve the sluggish kinetics of $NaAlH_4$ [30]. Alanates have limited use since they require specific storage conditions and cannot be reversed under practical circumstances.

9.6. Amides and Imides

High volumes of hydrogen are stored by Amides and Imides materials at low temperatures. Lithium amide has maximum hydrogen storage capacity recorded as 10.4 wt% [184], while imide has maximum capacity of 7.0 wt%. But, these materials cannot be used in practical applications since they require a high temperature (over 400°C) at normal pressure in the way of releasing almost allabsorbed hydrogen. Increased storage capacities are made possible by introducing Mg or Ca, which also lowers storage temperatures and raises desorption pressure [185], [186].

10. Large Scale Hydrogen Storage

Sometimes, hydrogen must be kept in stationary off-board reservoirs before being used as fuel in a power conversion device. This is necessary to fulfill the need to refuel the onboard system, such as the gasoline tanks of a car or a power-generating facility. The refilling station can be located inside the regular fuel station under the national hydrogen network prior to vehicle demand, or it can be close to the hydrogen production system. Storage requirements will vary if hydrogen is stored in this manner prior to usage as onboard fuel because it occasionally needs to be kept in one place for long periods of time before meeting the demand. Therefore, while planning and constructing large-scale hydrogen storage, some key factors, such as structural performance, energy expenditure, fuel boil-off, fuel venting losses, and life safety, need to be considered. Given these specifications for large-scale hydrogen storage, compressed gas storage is frequently the recommended option for minimizing costs, volumetric footprint, and fuel energy content loss. When hydrogen needs to be kept for an extended amount of time without fuel offtake, technologies, except high-pressure tanks, become appealing possibilities. This is in contrast to storage within a power generation unit or on board a vehicle. The storing of compressed hydrogen in sizable subterranean geological formations, like salt domes, is one such viable technological solution. Salt domes successfully stop leaks and are incredibly immune to hydrogen, even at high pressures [187]. Recent studies have looked into the technological viability of utilizing salt domes for large-scale storage [188]. The total technological storage potential is calculated to be 84.8 PW for Europe, accounting for 27% only for offshore areas.

Large-scale off-board liquid storage systems are required more in the aircraft industry due to the growing demand for liquid hydrogen. Therefore, rather than concerns about tank weight, an adequate liquid hydrogen storage technique is required, which is different from onboard liquid tanks. Additionally, a suitable venting mechanism is required to avoid excessive pressurization, which can lead to the collapse of holding structures as a result of the accumulation of hydrogen vapor during delivery, transfer, and refueling operations. The utilization of triple-jacketed ground storage Dewar tanks is one such technological possibility for off-board liquid hydrogen storage [32]. Liquid hydrogens are contained within the inner shells of these Dewar's containers. Next, an outer shell, another vacuum space, a radiation shield cooled by liquid nitrogen, and a vacuum space around the inner shell. This construction needs to withstand a complete vacuum, at least on the outside wall, in addition to having many shells. These constructions are bigger than the standard lightweight composite cryotank designs that are utilized in onboard storage purposes [25], but they fit for long-term liquid storage because of their much lower tank inactivity pressure and boil-off rates.

11. Result and Discussion

Hydrogen gas offers the highest energy content per unit among fuels, making it highly efficient for storing and distributing energy. Its energy density (120 MJ/kg) allows significant energy storage in small amounts, and its stable chemical properties ensure long-term energy retention. Table II represents the energy content of different hydrocarbons, including hydrogen.

TABLE II: Classification and Key Points of Every Type of Pressure Vessel

Fuel	(J/kg)
Gasoline	43×10^6
Methane	50×10^6
Methanol	20×10^6
Ethanol	27×10^6
Coal	34×10^6
Paper	17×10^6
Fruit loops	16×10^6
Hydrogen	120×10^6

12. Conclusion

Hydrogen storage is a crucial enabler for broader utilization as a key component of the future energy landscape. While existing storage techniques such as liquid hydrogen and compressed gas provide viable options, they face challenges in terms of cost, safety, and efficiency. Emerging technologies, including solid-state storage and advanced materials, offer promising alternatives with potential improvements in energy density and system integration. However, further research and development are essential for overcoming current limitations and achieving commercial viability. The successful deployment of hydrogen storage solutions will be instrumental in realizing hydrogen's full potential as a clean, sustainable energy carrier, contributing to global efforts to decarbonize energy systems and achieve climate goals. As technological advancements continue, hydrogen storage is poised to play a vital role in the transition to a low-carbon economy, supporting a diverse range of applications from renewable energy integration to transportation and industrial processes.

Conflict of Interest

Authors declare that they do not have any conflict of interest.

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