INTERNATIONAL JOURNAL OF AUTOMOTIVE AND MECHANICAL ENGINEERING
ISSN: 2229-8649 e-ISSN: 2180-1606
VOLUME 22, ISSUE 2, 2025, 12528 - 12546
DOI: https://doi.org/10.15282/ijame.22.2.2025.20.0957

REVIEW ARTICLE

Hydrogen as a Clean Fuel: Storage Technologies and Associated Thermal Challenges

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ABSTRACT – Hydrogen has emerged as a clean, renewable, and non-toxic alternative energy carrier with the potential to support a sustainable energy future. Despite its advantages, one of the major obstacles to widespread adoption is the challenge of efficient and safe storage. This review critically examines hydrogen storage technologies—including gaseous, liquid, and solid-state methods—focusing on the thermal management issues associated with each. The study methodology involved a comprehensive review of current scientific literature, government reports, and technical papers, particularly focusing on studies addressing heat transfer limitations and enhancement techniques in hydrogen storage systems. The analysis reveals that gaseous hydrogen storage suffers from high compression heat and limited density; liquid hydrogen faces challenges related to boil-off and cryogenic insulation, while solid-state storage—though safer and denser—encounters difficulties due to low thermal conductivity and slow kinetics. Among the examined techniques, solid-state storage appears to be the most promising for practical use, especially when integrated with thermal enhancement strategies such as fins, metal foams, and expanded graphite. The review highlights the need for hybrid approaches that combine multiple heat transfer augmentation methods and identifies nanoparticle-enhanced fluids as a promising direction for future research. Overall, this work provides an integrated perspective on the thermal barriers in hydrogen storage technologies and outlines potential pathways for performance improvement, supporting ongoing research and development in this field.

ARTICLE HISTORY

Received	:	06th Sept. 2024
Revised	:	25th Mar. 2025
Accepted	:	04th June 2025
Published	:	27th June 2025

KEYWORDS
Hydrogen storage
Metal hydride
Heat and mass transfer
Nanofluid

1. INTRODUCTION

Energy storage and transfer are essential and fascinating aspects of our daily lives. Since 1950, the global population and the desire for an improved standard of living have resulted in an exponential rise in energy demand. This demand is projected to peak around 2035, based on historical and forecasted global primary energy consumption trends reported in World Energy 2014–2050 [1]. It highlights the historical dominance of fossil fuels and the projected rise of renewable energy sources such as wind, solar, and biofuels. The shift toward cleaner energy emphasises the growing need for efficient storage solutions, like hydrogen, aligning with global decarbonisation goals. The primary challenge to address is the imbalance between energy demand and supply. Additionally, the finite nature of non-renewable energy sources like oil, coal, and natural gas has raised concerns about energy security in many countries. Although the world has ample natural gas and coal reserves, they will eventually be depleted. With the current consumption rate, natural gas and coal resources are projected to last for around 70 and 200 years, respectively, while oil reserves will be depleted even sooner [2,3].

During the last few decades, the extensive use of fossil fuels for energy production, which emit greenhouse gases, has led to a change in climate conditions and a negative impact on human health [4,5]. As a result, alternative energy sources are required to meet future energy needs, especially in the transportation sector, which accounts for nearly 60% of the world's energy consumption [6]. One of the most significant challenges today is developing energy systems that can meet energy needs using renewable and sustainable sources. These systems must be feasible, cost-effective, reliable, and environmentally friendly, causing minimal or no harm to the ecosystem. Various potential alternatives, including solar, nuclear, wind, hydro, tidal, geothermal energy and biofuels, have been proposed. However, hydrogen appears to be the most suitable candidate due to its highest energy density per unit mass (120 MJ/kg), negligible environmental impact, and abundant presence in the universe [7–9]. In recent years, there has been growing interest in hydrogen fuel-based technologies. Key aspects of a hydrogen-driven economy include its production, purification, transportation, storage, and utilisation. Among these, hydrogen storage has been a central research focus for many years [10]. Presently, extensive research is underway to assess the requirements, impacts, opportunities, and challenges associated with implementing a hydrogen-based economy. One of the most significant obstacles in using hydrogen as a fuel is enhancing the methods for its practical storage.

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Currently, numerous studies have been conducted, and various hydrogen storage methods have been proposed; however, only a few meet the latest targets set by the U.S. Department of Energy (DOE). Each method comes with its own set of limitations. This field of research is still evolving, with ongoing efforts focused on improving existing technologies. The hydrogen storage approaches discussed in this paper include compressed hydrogen, liquid hydrogen, and solid-state storage (involving metallic and complex hydrides). All these methods involve highly interconnected transport phenomena such as chemical reactions, heat transfer, and mass transfer. A comprehensive understanding of these processes and their influence on system performance is essential for the effective design and optimisation of hydrogen storage systems.

Apart from individual studies, a considerable number of review articles are available that have broadly summarised the latest developments in several facets of hydrogen storage technologies, such as heat transfer along with associated energy-competence issues, realistic reactor design considering geometrical as well as operative parameters, reaction kinetics, hydride bed material, fuelling tactics, thermal management techniques, optimisation perceptions, and application perspectives. Zhang et al. [11] addressed heat transfer and associated energy-efficiency issues for H_2 storage technologies, which had not been focused on earlier. Sakintuna et al. [12] presented the state-of-the-art materials for solid-state H_2 storage. Yang et al. [13] reviewed technical issues related to realistic and optimised MH reactor design to perform well. Murthy [14] presented studies on the thermodynamic aspect of solid-state H_2 storage, including the effect of thermo-physical and kinetic properties on system performance. Several storage materials with the necessary sorption characteristics were proposed. Mazzucco et al. [15] presented state-of-the-art advances for effective thermal management systems used in metal hydride storage tanks for light-duty vehicles. Also, several fuelling tactics and optimisation criteria were discussed. Mohammadshahi et al. [16] reviewed the characteristics of mathematical models of metal hydride tanks. The progress of the tank geometries, as well as arrangements of cooling systems, along with factors affecting MH tank performance, were also discussed. Shafiee and McCay [17] presented comparative studies on different reactor and heat exchanger arrangements for metal hydride H_2 storage. Afzal et al. [18] summarised the progress, effectiveness and classification of different heat exchange techniques for metal hydride H_2 storage. Chanchetti et al. [19] analysed research articles published from 2000 to 2015, focusing on hydrogen storage materials. The findings indicated that metal-organic frameworks and simple hydrides emerged as highly investigated materials in the field. Yao et al. [20] provide a comprehensive review of the various operation modes of hydrolysis reactions and their practical applications. The different approaches and their effectiveness are examined and compared to identify the most suitable option for practical applications that require high energy density.

While previous studies have addressed individual hydrogen storage technologies, many of them do not provide a consolidated analysis of the heat and mass transfer challenges across all three storage forms—gaseous, liquid, and solid. This review aims to bridge that gap by offering a comparative evaluation of the thermal management issues encountered in each method, along with potential strategies to address them. The paper also highlights recent advancements in the field, particularly innovative approaches for improving heat transfer performance. Furthermore, it identifies the integration of nanoparticles into heat transfer fluids used in metal hydride reactors as a promising area for future investigation. The review is based on an extensive survey of academic publications, technical documents from the U.S. Department of Energy, and pertinent industry literature.

2. HYDROGEN FUNDAMENTALS

2.1 Hydrogen as a Clean Fuel

Hydrogen, in its natural state, is a colourless, odourless, and tasteless gas. Although abundantly available, it is typically found in compound forms such as water (H_2O). One of hydrogen's key advantages is that it can be stored and converted into usable energy, releasing only water vapour upon combustion, which classifies it as a clean fuel. It is also renewable and widely available in nature [21–23]. Hydrogen's potential for storage in portable and transportable systems makes it a strong candidate for use in electric and fuel-cell vehicles. Compared to fossil fuels, hydrogen has the highest energy content per unit mass but a significantly lower energy content per unit volume [24–27], which poses a storage challenge. Table 1 presents a technical comparison of hydrogen with conventional fuels. It is the lightest element and has a higher diffusivity in air, allowing it to disperse quickly if leaked, thereby reducing the risk of explosion in open environments. Its very low boiling point (20.3 K) demands advanced cryogenic systems for handling liquid hydrogen. While the low density of hydrogen gives it a favourable energy-to-weight ratio, the energy-to-volume ratio remains a drawback.

Hydrogen also has a wide flammability range and a high flame velocity compared to other fuels. However, it requires a relatively high temperature for auto-ignition, making it less prone to accidental ignition without an external trigger. The ignition energy of hydrogen is extremely low—only about 0.02 MJ is required to ignite a hydrogen-air mixture, much lower than that for methane. Despite these characteristics, hydrogen can be considered relatively safe when handled properly. Its quick dispersion reduces the likelihood of accumulation in open spaces, and with proper ventilation, sensors, leak detectors, and flame arrestors, the risks can be effectively mitigated. Thus, while hydrogen's flammability demands careful system design and operational controls, its behaviour in leaks and combustion scenarios—along with ongoing advances in safety technologies—supports its viability as a clean and safe fuel. This understanding is essential before examining the challenges related to its storage, discussed in the subsequent sections.

Table 1. Technical evaluation of H_2 with other fuels [28]

Parameter	Unit	Hydrogen (H_2)	Petroleum (C_nH_{2n})	Methane (CH_4)	Methanol (CH_3OH)	Ammonia (NH_3)	Propane (C_3H_8)
Boiling point	k	20.3	350−400	111.7	337	240	230.8
Liquid density at NTP	kg/m^3	70.8	702	425	797	771	507
Gas density at NTP	kg/m^3	0.0899	---	0.718	---	0.77	2.01
Heat of vaporisation	KJ/kg	444	302	577	1168	1377	388
Heat value (High−Low)	MJ/kg	141.9−120	46.7−44.38	55.5−50	23.3−20.1	22.5−18.6	48.5−46.4
Lower heating value (liquid)	MJ/m^3	8520	31170	21250	16020	14350	23520
Diffusivity in air at NTP	cm^2/s	0.63	0.08	0.2	0.16	0.2	0.1
Flammability limit (Low−High)	vol% (in air)	4−75	1−6	5−15	7−36	15−28	2−10
Auto-ignition temperature	0C	585	222	534	385	561	466
Ignition energy	MJ	0.02	0.25	0.3	---	---	0.25
Flame velocity	cm/s	270	30	34	---	---	38

2.2 Hydrogen Storage Methods

The storage of H_2 is a crucial step towards its extensive use [29–31]. The hydrogen economy necessitates both movable and stationary storage systems for hydrogen. However, there are typical desires and differences for both storage systems. Hydrogen storage can be categorised into two primary methods: physical-based and chemical-based. Physical storage encompasses compressed hydrogen stored in gaseous form, liquefied storage, and cryo-compressed storage. On the other hand, chemical storage involves storing hydrogen in various materials such as simple and complex metal hydrides, carbon-based materials, and chemical hydrides [32,33]. This classification is depicted in Figure 1.

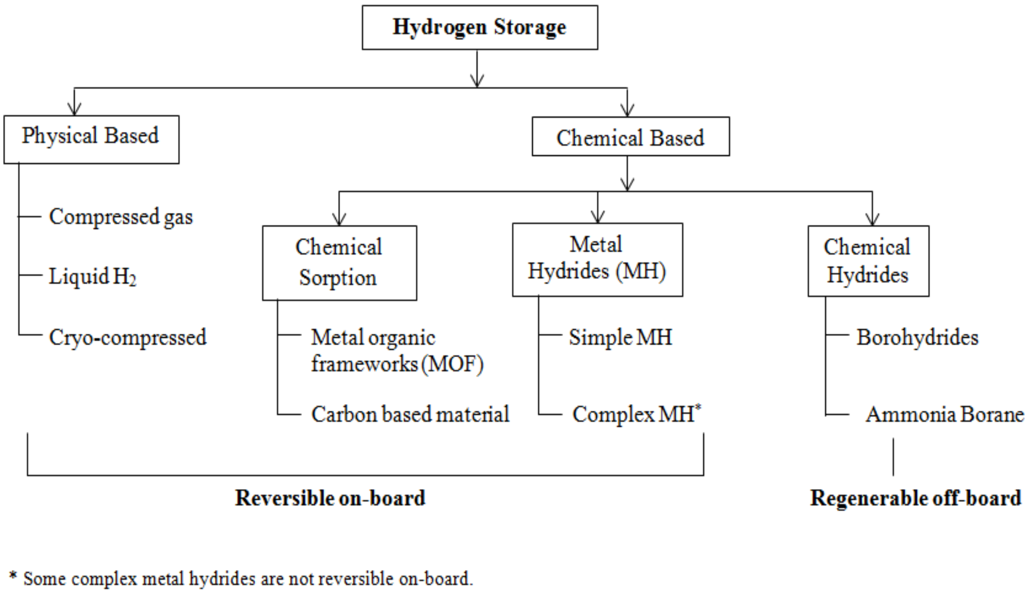

Figure 1. Hydrogen storage methods

Storing hydrogen at high pressure in its gaseous state is the most common and straightforward approach. However, this approach has limitations in terms of volume due to the low density of hydrogen at high-pressure values. It also incurs high material costs due to the large volume required. An alternative method involves using Metal-Organic Framework (MOF) hydrides, which use recyclable aromatic cycloalkane pairs as hydrogen storage materials. These hydrides provide a high storage capacity, typically between 5% and 10% by weight, and are economical and efficient. Additionally, MOF hydrides are suitable for transporting hydrogen over long distances and for storing it over extended periods, such as between seasons. However, the equipment needed for catalytic hydrogenation and dehydrogenation is expensive, and the associated technologies are complex, resulting in low dehydrogenation efficiency. On the other hand, carbon materials possess variable microholes and highly effective specific surface areas, making them suitable for hydrogen storage with advantages such as low cost, high safety, suitable adsorption and desorption conditions, and long life. The hydrogen storage capacity of carbon materials ranges from 0.2% to 10% by mass. While they show good reversible performance, carbon materials are insufficient for meeting the hydrogen storage requirements of automotive applications.

Compared to liquid and gaseous storage methods, metal hydrides offer a safer and more efficient option for onboard vehicle hydrogen storage, as they can retain hydrogen in solid form under moderate pressure and temperature conditions. They also provide high volumetric density, with hydrogen storage densities of 6.5 atoms/cm³ for MgH_2, 4.2 atoms/cm³ for liquid hydrogen, and 0.99 atoms/cm³ for hydrogen gas. The storage capacity of metal hydrides is affected by several factors, including phase distribution, cell volume, absorption/desorption kinetics, grain size, and crystalline structure [12,34]. When assessing hydrogen storage technologies, key criteria include volumetric storage density, reversibility, hydrogen uptake and release rates, lifecycle durability, and safety performance. It is noted that high volumetric storage densities, decent reversibility, low absorption-desorption temperatures, as well as rapid reaction rates are the key characteristics of ideal hydrogen storage materials. For improvement in gravimetric densities of parent materials, several new materials, such as complex hydrides, metal frameworks, carbon forms, etc., are proposed. To design a larger hydrogen storage capacity device with faster reaction kinetics, extensive work on heat and mass transfer analysis is necessary.

2.3 Department of Energy Targets

Various research institutions and companies have proposed standards for hydrogen storage, such as the U.S. Department of Energy (DOE), the World Energy Network, and the International Energy Agency. The DOE's published standard is widely recognised as relatively authoritative. Table 2 shows targets given by the U.S. Department of Energy (DOE) for onboard hydrogen storage. These technical performance benchmarks serve as a framework for research on onboard hydrogen storage and establish a long-term vision for its practical applications. But it has been recognised to be tough to satisfy all these targets at the same time. It is desirable for any system for hydrogen storage to have high volumetric and gravimetric storage capacity. The ultimate goal for hydrogen storage systems is to attain a gravimetric density of 6.5 wt.% and a volumetric density of 5.0 wt.%. Additionally, there is a target to reduce the cost of hydrogen to $266 per kilogram.

Table 2. DOE targets for onboard storage of hydrogen [35–37]

Storage variables	Unit	2020	2025	Ultimate
Gravimetric capacity (Material-based)	kWh/kg	1.5	1.8	2.2
Gravimetric capacity (System-based)	kgH_2/kg system	0.045	0.055	0.065
Volumetric capacity (Material-based)	kWh/L	1	1.3	1.7
Volumetric capacity (System-based)	kgH_2/L system	0.03	0.04	0.05
Cost of storage system	$/kWh net	10	9	8
Cost of fuel	$/kgH_2$	333	300	266
Atmospheric temperature	0C	-40/60 (sun)	-40/60 (sun)	-40/60 (sun)
Delivery temperature (Min/Max)	0C	-40/85	-40/85	-40/85
Cycle life (1/4 tank to full)	cycles	1500	1500	1500
Min. delivery pressure from a tank	bar	5	5	5
Mix. delivery pressure from a tank	bar	12	12	12
Charging/Discharging rates	min	3-5	3-5	3-5

3. COMPRESSED HYDROGEN STORAGE

High-pressure compression of hydrogen is the prevailing storage method, offering well-established technology and rapid filling and release rates of hydrogen. A notable advantage is that energy is not needed for hydrogen release. To compete with fossil fuel-based systems, it is generally necessary to store around 4 to 7 kg of hydrogen onboard. Given hydrogen's low density, achieving this storage amount requires compressing it to high pressures. In fuel cell applications, hydrogen is typically compressed to pressures between 350 and 700 bar. At room temperature, hydrogen has a density of approximately 23 kg/m³ at 350 bar and about 40 kg/m³ at 700 bar. Despite this high compression, the resulting density remains relatively low, presenting a significant challenge [38].

3.1 Principle of Compression

At atmospheric temperature, the behaviour of H_2 is similar to that of an ideal gas and satisfies the ideal gas law. The process of compression from ambient pressure to 700 bar requires an enormous amount of energy. The minimum work required for the reversible constant temperature compression process is calculated as follows:

$$W = mRT_1 ln\frac{P_2}{P_1} \tag{1}$$

where the 1 and 2 suffixes indicate parameters before and after compression, respectively.

Compressing hydrogen results in an energy loss of approximately 11 to 13% of its total energy content. Although, in theory, compressing hydrogen to 800 bar at a constant temperature requires about 2.21 kWh/kg of energy, the actual process typically consumes significantly more. In practice, despite using multistage intercooling, hydrogen's temperature rises notably during compression. Consequently, the compression process is more accurately described as polytropic. The energy requirement for such a polytropic process can be expressed as:

$$W = \frac{n}{n-1} R T_1 \left[\left(\frac{P_2}{P_1} \right)^{\frac{n-1}{n}} - 1 \right] \tag{2}$$

where n = Polytropic index. Equations (1) and (2) demonstrate that lowering the inlet temperature of hydrogen can proportionally reduce the compression work. This is the fundamental principle behind the design of intercooling systems.

3.2 Compressed Hydrogen Storage Vessel

Metal or composite cylinders/tanks of various sizes and capacities are widely employed for hydrogen storage, representing the prevailing method. Due to the impracticality of spherical containers, cylindrical-shaped containers are employed for onboard hydrogen storage. However, the small size of hydrogen molecules makes them more susceptible to leakage through certain materials, cracks, or inadequate joints in storage tanks than other gases at similar pressures [39]. It is crucial to select a vessel material that is both lightweight and cost-effective while being capable of withstanding high pressures. The tanks used for hydrogen storage are engineered to meet a maximum operational pressure, and the minimum thickness of their walls is determined by the tensile strength of the material employed [40]. The most desirable material for storing hydrogen should possess a low density, high tensile strength, and must be non-reactive to hydrogen while preventing hydrogen diffusion. Based on these criteria, four types of vessels that are commonly used for the storage and transportation of compressed hydrogen gas are compared in Table 3.

Table 3. Categorisation and comparison of various types of compressed hydrogen vessels

Type	Description	Material	Gravimetric density (wt%)	Working pressure (MPa)	Application area
I	Completely metallic pressure vessel	steel or aluminium	1.7 [41]	15–30 [42]	H_2 is stored as industrial gas [43]
II	Seamless metallic pressure tank hoop-wrapped with fiber-resin composite	steel or aluminium	2.1 [41]	10–95 [42]	Stationary applications, e.g. high-pressure H_2 storage at H_2 refuelling station (HRS) [44]
III	Completely composite pressure tank wrap with metal liner	seamless or welded aluminium liners	4.2 [41]	30–70 [42]	Onboard H_2storage e.g. H_2trailer, [42] H_2 tube trailer [43]
IV	Fully composite	High-density polyethylene (non-metallic) inner with glass/carbon fibre	5.7 [41]	30–70 [42]	On-board H_2 storage e.g. H_2trailer, [42] H_2 tube trailer [43] Stationary hydrogen storage low-pressure at HRS [6]

Type I tanks refer to seamless containers constructed from steel or aluminium, which are characterised by their heavy weight and thick walls. These options are more cost-effective and commonly available. The maximum operating pressure for type I tanks is typically set at 30 MPa. While type I containers offer good safety and strength properties, their weight remains a significant drawback. To address this issue, steel containers have been substituted with composite containers that incorporate a thin inner metal liner to ensure gas tightness. Type I tanks are offered in various net volumes ranging from 2.5 to 50 L. However, due to their relatively low gravimetric density, they are primarily suitable for stationary applications, particularly for on-site storage of hydrogen in industrial settings [45]. Type II tanks are constructed with seamless metallic tanks wrapped with a fibre-resin composite in a hoop configuration. They are capable of withstanding pressures up to 95 MPa. These tanks are commonly utilised as high-pressure buffers at hydrogen refuelling stations. Compared to Type I tanks, Type II tanks are lighter, weighing approximately 30-40% less. However, they are also more expensive, costing around 50% more. The competitive cost of Type II tanks is attributed to the relatively low quantity of fibres used in their construction. However, neither Type I nor Type II tanks are suitable for automotive uses because of their heavy weights and large sizes [46].

When the metallic or polymeric inner layers are fully enveloped with fibre material, the resulting pressure vessels experience significant weight reduction, making them suitable for meeting the requirements of gravimetric density for onboard hydrogen storage. Type III tanks feature seamless or welded aluminium liners entirely wrapped with fibre resin composite, and the chosen materials are less susceptible to hydrogen embrittlement. However, an issue that may arise with Type III tanks is their low thermal conductivity, which can lead to a slow release of heat during hydrogen compression [47]. On the other hand, Type IV tanks consist of non-metallic liners embedded within a fibre/epoxy matrix. The fibre wrapping provides the strength to contain pressurised hydrogen, while the liner acts primarily as a barrier against permeation [46]. While these cylinders are lighter than those with all-metal liners, they tend to be more expensive compared to other metal liner cylinders. Type III and Type IV pressure vessels are frequently used in tube trailers and modular containers for transporting hydrogen. Type IV vessels, in particular, are becoming increasingly popular for low-pressure, stationary hydrogen storage at refuelling stations. Although Type I vessels can hold an equivalent amount of hydrogen with comparable investment costs, they occupy significantly more space [44].

3.3 Heat and Mass Transfer Issues

Two types of compressors, namely reciprocating piston compressors and diaphragm compressors, are extensively used to compress the hydrogen. These compressors are to be utilised to increase the hydrogen pressure up to a minimum of 150 bar. Generally, a diaphragm compressor has a comparatively huge surface area for the compression chamber; as a result, it removes heat more efficiently than piston compressors. However, an increase in the temperature of the gas throughout compression controls the compression ratio in the case of diaphragm compressors. The compressor performance can be improved by extending the number of cooling circlets and adding micro/mini passages. Heat transfer modelling and design for piston compressors is quite challenging due to the enormous speed as well as the cyclic form of the compression process. Conventionally, lubricant oil is used to cool such compressors. However, this method may not be practicable in hydrogen gas since a fuel cell requires highly pure hydrogen gas. Yong [48] settled on a cooling method that uses a water spray on the inner side of the cylinder to lower the temperature of the gas through evaporation. The use of a greater stroke length, along with the slow motion of the piston and the use of water for cooling the cylinder is an alternative tactic for reciprocating piston compressors. However, this method is subject to small flow rates.

To store hydrogen as compressed gas, a significant heat transfer concern is the increase in temperature of the filling tank [49,50]. At ambient temperature, hydrogen shows a reverse Joule-Thomson effect. By this effect, hydrogen is subjected to heating instead of cooling, resulting from the throttling process. The rise in temperature of the inner side of the tank is about 85°C during fast filling (around 1 kg of hydrogen per minute) [51,52]. As a result of increasing temperature, there is an adverse impact on the composite tank, and its capacity to store hydrogen (at a given pressure) decreases. It is reported that pre-cooling of the filling gas is a possible remedy for overheating [53,54], but such a method would necessitate extra energy for cooling. Improvement in internal and external transfer of heat and optimisation of the throttling process are energy-effective solutions. The use of a cascade system might be effective in order to reduce losses due to the throttling process as well as subsequent heating throughout the tank filling processes.

Storing hydrogen in compressed gaseous form offers several practical advantages, including relatively simple technology and rapid tank filling and discharge times [55]. As such, it is widely used in prototype fuel-cell vehicles. However, a key limitation lies in the energy densities—both gravimetric and volumetric—achievable with this method. While increasing the storage pressure does lead to higher hydrogen density, the relationship is nonlinear due to the compressibility of hydrogen at high pressures [56]. Unlike an ideal gas, hydrogen does not continue to compress efficiently beyond a certain threshold (typically around 350–700 bar), meaning that significant increases in pressure result in only marginal gains in stored energy. This makes the system bulky and less efficient for vehicular use, where space and weight are critical. Moreover, as hydrogen is the lightest element, it is highly diffused and can potentially leak from pressurised containers. The safety of storing hydrogen at such high pressures is a primary concern, especially in densely populated areas where the consequences of tank rupture or leakage could be severe.

4. LIQUID HYDROGEN STORAGE

The second method commercially used for hydrogen storage is to store it as a liquid at cryogenic temperature. When the hydrogen gas at normal conditions gets cooled below 20K, it converts into a liquid state at the critical point temperature and pressure (33K, 12.9 bar) [57,58]. Hydrogen storage in a liquid state offers high volumetric storage density at comparatively low pressures. Thus, per unit volume, it is possible to store high energy. As compared with compressed gaseous hydrogen ($\mathrm{CGH_2}$) at 700 bar, 288K, the liquid hydrogen ($\mathrm{LH_2}$) has almost double mass density (approximately 71 kg/m^3) at normal boiling point (20K) [59]. The operating pressures for $\mathrm{LH_2}$ vary between 1 and 3.5 bar [60]. The energy density of $\mathrm{CGH_2}$ is 4400 MJ/m^3, whereas for $\mathrm{LH_2}$, it is up to 8400 MJ/m^3 [61,62].

4.1 Liquefaction Process

Thermodynamically, liquefaction of hydrogen comprises three heat transfer steps: (i) removal of sensible heat (approximately 4000 kJ/kg) to lower the temperature from atmospheric conditions to 20 K; (ii) removal of latent heat (approximately 450 kJ/kg) for condensation of hydrogen gas at the same temperature; and (iii) ortho-para conversion, which removes heat (approximately 703 kJ/kg) [63]. Hence, a liquefaction cycle should remove a total heat of 5153 kJ/kg for hydrogen liquefaction. The Joule-Thompson cycle is a modest form that can be utilised for this purpose. For the liquefaction process, the heat exchangers used must have an effectiveness above 0.85 to produce cryogenic liquid [64]. The effectiveness of the heat exchanger can be increased by comprising minor temperature differences between inlet and exit streams, a higher surface area to volume ratio, and bigger heat transfer coefficients [65]. Storage of hydrogen as a liquid suffers from obstacles such as the efficiency of the liquefaction process and boil-off of the liquid.

4.2 Liquid Hydrogen Storage Tank and Associated Heat and Mass Transfer Issues

Storing hydrogen in its liquid state requires specially designed double-walled containers with advanced thermal insulation systems. The heat may transfer from the surroundings to the tank by three basic modes that are conduction, convection and radiation. Heat transfer by conduction mode can be reduced by making storage tank walls with low thermal conductivity materials. The space in between the inside and outside vessel is usually subjected to high vacuum (approximately 1.3×10^{-8} bar) in order to minimise convection [66]. In the vacuum superinsulation technique, special insulating materials are used to enclose the coats near the vacuum. Heat transfer at the inner surface of the tank due to radiation can be diminished by placing multi-layered insulation (MLI) [67]. MLI consists of 30 to 50 sheets of radiation shields having low emissivity, e.g., Al foil with $\varepsilon = 0.05$ [68]. These sheets are separated by spacers having low conductivity (e.g., glass wool or fibreglass paper). Heat conduction can also be diminished using thermal insulation with the vacuum MLI [69,70].

Microsphere insulation can be used as an alternative to MLI. Its thermal conductivity is isotropic [71–73]. The tank loses more hydrogen when coated with microsphere insulation as compared to MLI. However, the transfer of heat will be less in the case of a sphere, as it has the lowest ratio of surface area to volume. It is desirable to have a cylindrical shape for a tank storing LH_2, as it offers many advantages when compared with a spherical shape, such as ease in manufacturing, economical and easy onboard installation. Also, many heat transfer issues are involved in the design of LH_2 transfer lines. Most of the time LH_2 transfer line experiences mixed-phase flow due to heat transfer. This induces a negative impact on the carrying capacity of the LH_2 transfer line. The remedy for this is proper insulation of the LH_2 transfer line so that heat transfer from outside can be reduced. However, as no insulation is perfect even with utmost care, the transfer of heat cannot be zero, and hence, venting losses exist. Hence, the goal of insulation is only to postpone the venting process.

4.2.1 Boil-off

The key issue in LH_2 storage is boil-off caused by heat inflow. It refers to the phenomenon that as a result of a heat transfer from outside, some fraction of the liquid hydrogen boils, becomes gaseous, and leaks [74]. This leads to hydrogen loss with no benefit. The ortho-para conversion of hydrogen, which is an exothermic reaction, acts as a heat source for the gasification. The conversion heat (approximately 520 kJ/kg at 77 K) is more than the latent heat of vaporisation (451.9 kJ/kg) of normal hydrogen at the normal boiling point [75]. There is an increase in pressure inside the vessel due to boil-off. When the pressure rises above the recommended value, it is necessary to remove the vapours of hydrogen outside the tank. Boil-off can be reduced by using thermally designed insulation. Another factor affecting heat transfer is tank size. Boil-off is inversely proportional to tank size since an increase in tank size reduces evaporation rate, as shown in Table 4.

Table 4. Boil-off losses for double-walled vacuum-insulated tank [39]

Losses per day (%)	Tank volume (m^3)
0.40	50
0.20	100
0.06	20000

During running conditions, the vehicle engine consumes hydrogen more rapidly as compared to the boil-off effect, so venting may not be necessary. However, when a vehicle is in stationary mode, continuous boil-off causes an increase in tank pressure [76]. The venting process must be utilised to control this pressure. The time measured from keeping the vehicle stationary and venting hydrogen as a gas is known as ‘dormancy’. The key factors for the design of LH_2 tanks are the temperature of hydrogen, insulation thickness and operating pressure. Multistage cryo-coolers with many shields can be used for high efficiency in the case of long-term storage of LH_2.

4.2.2 Remedies other than insulation

Liquid or vapour shields can be employed to reduce boil-off rates. In such an arrangement, low-temperature vapours of hydrogen are routed in such a way that they could absorb some part of the sensible heat, which would otherwise increase the temperature of LH_2 [77]. However, such an arrangement increases the volume and weight of the storage tank. While searching for another solution for boil-off, some tank designs have been suggested where storage vessels are used for CGH_2 could be adapted for the storage of LH_2 with slight reforms, such as the addition of an insulating layer.

4.2.3 Thermal stratification and sloshing

Thermal stratification within cryogenic hydrogen tanks occurs when heat from the surroundings leads to uneven temperature distribution, forming a warmer upper layer. This warmer hydrogen layer evaporates more readily, accelerating the boil-off rate and causing a rise in tank pressure up to saturation conditions [78]. Consequently, the interval between successive venting cycles shortens. The buildup of a thermally stratified hot layer is the primary contributor to this pressure increase. While effective insulation remains the first line of defence, high-conductivity metal rods may be used in small vehicle tanks to help redistribute internal heat and mitigate stratification [79].

In addition to stratification, sloshing—the movement of liquid hydrogen within the tank due to vehicle motion—further intensifies thermal effects. As the cryogenic fluid moves, mechanical energy from motion is partially converted into thermal energy through viscous friction, turbulence, and wave breaking at the liquid-vapour interface. These effects increase localised temperatures and can disturb the thermal equilibrium, leading to additional vapour generation and pressure rise. This is particularly problematic in urban driving scenarios, where frequent starts and stops cause more intense fluid motion. While anti-slosh baffles can dampen these effects, they reduce usable hydrogen mass, thus negatively affecting the system’s gravimetric efficiency.

5. SOLID STATE HYDROGEN STORAGE

5.1 Metal Hydride

A key stage towards the prevalent usage of hydrogen (H_2) is reliable and safe storage. Storage of hydrogen as CGH_2 demands extremely high pressures, whereas the storage of LH_2 requires low temperatures in the cryogenic range. To store in solid form, hydrogen is inserted reversibly in a solid medium called 'Metal Hydride' (MH). Such metal hydrides offer many constructive features. Many scientists have obtained greater volumetric densities for solid-state hydrogen storage as compared to those possible with CGH_2 or LH_2 storage. However, future targets for volumetric densities are far away from present values.

5.1.1 Formation and working of metal hydride bed

A metal hydride is a concentrated chemical compound consisting of a metal or metal alloy combined with hydrogen. Metal hydrides are created through the direct reaction between gaseous hydrogen and metals, metallic alloys, or intermetallics. When a hydride-forming metal or metal alloy comes into contact with gaseous hydrogen, it undergoes a reaction to form the corresponding metal hydride. The process of formation of MH can be written as:

$$M + \frac{x}{2}H_2 \leftrightarrow MH_x + Heat \tag{3}$$

Metal hydride consists of element (A), which has the highest affinity for hydrogen to form hydride, and element (B), which has a weak affinity for hydrogen to form hydride. The element (A), which tends to build a stable hydride, is a rare earth or alkaline metal such as La, Mg, Ti, or Zr. The element (B), which has a tendency to build unstable hydrides, is a transition metal such as Ni, Mn, or Fe. Also, it acts as a promoter in the process of hydrogen dissociation [80].

While numerous metals, alloys, and intermetallics can form reversible hydrides, it is primarily the materials that readily produce metal hydrides under moderate pressures and temperatures that are of practical significance. The hydrogen is allowed to react with specific materials to build hydrides. In the hydriding process, the molecules of hydrogen gas (H_2) disintegrate into hydrogen atoms (H), which make a connection with the surface of the metal by chemical reaction to form metal hydride (MH). This hydride is subjected to a reaction for hydrogen extraction as required. During the hydriding process (also called the charging or absorption process), heat is released since the reaction is exothermic, whereas, during the dehydriding process (also called the discharging or desorption process), heat input is required since the reaction is endothermic. Primarily, the sorption of H_2 relies upon the transfer of heat mass, reaction kinetics, and the geometrical configuration of the MH bed. α and β-phases can be used to study thermodynamic characteristics of MH development. The α-phase is a solid solution of hydrogen in the metallic matrix which, after reaching some saturation concentration, can transform into the β-phase (known to be hydride) with higher H_2 concentration. Usually, the MH bed is in the form of powder with a grain size of 50 to 100 μm. The powder decomposes to an equilibrium grain size of about 1 μm after approx. 10 to 100 charging-discharging cycles [81]. Such fine MH powder generally has a low effective thermal conductivity (~0.1 W/m-K).

5.1.2 Types of intermetallics compounds

Not all, but many metals and alloys for which reaction with hydrogen is reversible can form hydrides [82]. Earlier analysis has focused on typical hydrides formed by an alloy and an intermetallic compound. In general, such intermetallic compounds are classified as $AB, A_2B, AB_2, AB_3, AB_5$. Some classic intermetallic compounds are $LaNi_5$, ZrV_2, $ZrMn_2$, $TiFe$, $ZrNi$, Mg_2Ni and Ti_2Ni. Being lightweight, the best options among metal hydrides are A_2B and AB_2 type compounds. Magnesium containing compounds have excellent capacity for hydrogen storage but too slow reaction kinetics. The existence of heavy transition metals limits the amount of hydrogen that can be absorbed. Table 5 shows some prototypes and hydrides of intermetallic compounds.

Table 5. Some prototypes and hydrides of intermetallic compounds [83,84]

Intermetallic compound	Prototype	Hydrides
AB_5	$LaNi_5$	$LaNi_5H_6$
AB_2	ZrV_2, $ZrMn_2$, $TiMn_2$	$ZrV_2H_{5.5}$
AB3	$CeNi_3$, YFe_3,	$CeNi_3H_4$
A_2B_7	$Y2Ni_7$, $Th2Fe_7$	$Y_2Ni_7H_3$
A_6B_{23}	Y_6Fe_{23}	$Y_6Fe_{23}H_{12}$
AB	TiFe	$TiFeH_2$
A_2B	Mg_2Ni, Ti_2Ni	Mg_2NiH_4

5.1.3 Design of the reactor

The design of the MH reactor plays a significant part in terms of the amount of hydrogen stored and heat-mass transfer characteristics. The process of porous flow, reaction kinetics and exchange of heat and mass in the reactor is extremely transitory and complex. Therefore, the design configuration of the metal hydride reactor is a key parameter. Several configurations of reactors have been proposed until now, but the basic configurations of metal hydride reactors can be classified according to geometry, as shown in Table 6.

Table 6. Basic configuration of the reactor

Sr. No.	Name	Features	Ref.
1	Tubular reactor	• the first type, which was developed and studied • somewhat simple to manufacture and maintain • somewhat easy to escalate by increasing the number of units • better sealing, as well as tremendous bearing pressure • central artery is for the flow of hydrogen, MH material filled in the hollow circular region amid the artery and the wall, and heat exchange through the external surface of the tube wall. • the aspect ratio (ratio of cylinder length to diameter) must be larger than 10 • cross sections of the tube may be circular or elliptical.	[85–87]
2	Disc reactor	• chosen mostly for kinetic measurement • flat shape reaction bed • the axial flow of hydrogen • comparatively large area is available for heat exchange results in higher reaction kinetics • comparatively hold a minimal amount of MH material	[88]
3	Tank or chamber reactor	• primarily implemented in storage-related equipment • bed is in the form of a large cylinder or cube • comparatively, hold a large amount of MH material • heat exchange can be done via embedded elements, such as heat pipes, spiral coils and ordinary tube bundles • dehouche et al. proposed a micro-channel reactor, which is a compact configuration of a tank reactor	[89–91]

5.1.4 Challenges for metal hydride systems

The selection of suitable material for hydride, faster reaction kinetics during the hydriding and dehydriding processes, low construction and operational costs, etc., are significant challenges in the development of MH systems. However, an inherent drawback to MH is its poor heat transfer kinetics, which demands special attention to its thermal design. An additional severe problem is related to low hydriding and dehydriding rates, which depend to a certain extent on thermal properties. The rate of chemical reaction in an MH reactor greatly depends on the rate of addition or extraction of heat from the metal hydride bed [92]. During hydriding, the process may slow down or stop due to insufficient heat removal, which results in a high temperature of the bed. In contrast, during the dehydriding process, the same may occur due to poor heat supply to the bed. This may impact the storage capacity of the bed. Hence, greater internal heat transfer is necessary to increase the performance of the system as well as to retain its reliability. Another area for study includes the kinetics behind hydriding-dehydriding and retaining cycling capacity. One more objective of hydride research is to construct MH storage systems with significant volumetric densities.

5.1.5 Heat transfer enhancement techniques

Many researchers have studied various factors affecting the process of heat transfer in hydride, which includes studies on hydride properties, heat and mass transfer [93–95] and system simulation [96,97]. Figure 2 shows different heat transfer improvement techniques. Several attempts are reported to enhance the heat conductivity of the MH bed, such as putting aluminium foam, adding a copper wire net structure, using a multilayer waved sheet for packing, compacting metal hydride powder with expanded natural graphite (ENG), and micro-encapsulating the MH compact. One of the techniques to enhance heat transfer is increasing the area for heat transfer, as evidenced by Fourier's law for heat conduction. The area available for transfer of heat in the case of a hydride system can be increased by two approaches, namely: hydrogen storage device having a heat exchanger with multiple tubes and fins. It must be noted that increasing the area for heat transfer and enhancing the thermal conductivity of the MH bed are two dissimilar approaches to deal with heat transfer problems. On the other hand, the use of heat pipes, phase change material, micro/mini channel heat exchangers, etc., is reported as non-conventional techniques for heat transfer improvement. It has been seen that an integrated design approach for MH reactors is not available. However, many researchers have been attempting to improve the design for particular conditions. It should be kept in mind that the design of a reactor integrating different types of tools for heat transfer appears to perform better than a design having a single tool.

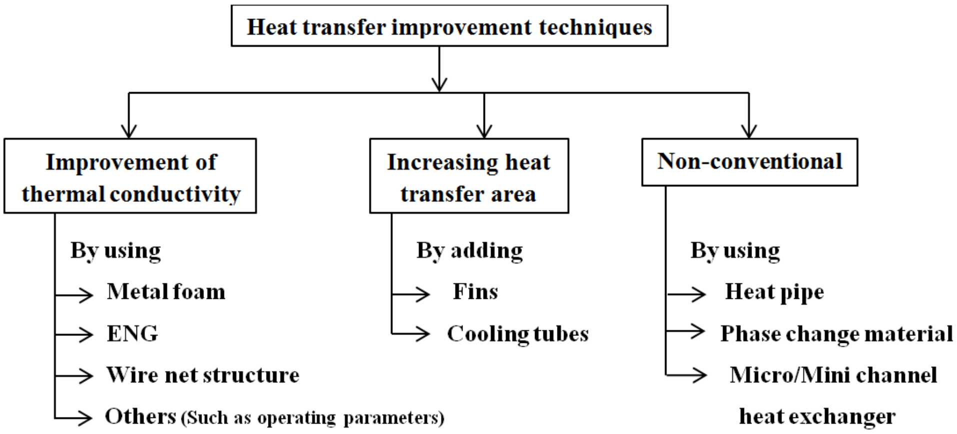

Figure 2. Heat transfer improvement techniques

5.1.6 Summary of studies

Before the 1990s, it had been recognised that the transfer of heat to and from the hydride bed had a significant influence on the system's performance; hence, efforts taken during this time were primarily focused on studying the reaction kinetics of hydrogen sorption. However, during this time, the behaviour of the system was not completely understood. Because of the accessibility of computational amenities, researchers had primarily focused on the modelling and simulation of the hydrogen storage system during the late 1990s and early 2000s. A remarkable investigation has been made possible because of the accessibility of sophisticated modelling software from the late 2000s. As a result, researchers have focused chiefly on modelling different kinds of reactor configurations, which would have been nearly impossible by actual experimentation.

Figure 3 shows a statistical comparison of the heat transfer techniques studied. Among the published heat transfer enhancement techniques, it is observed that researchers have primarily focused on fins, cooling tubes, and jackets. They reported that enhancing any single parameter, such as thermal conductivity, surface area, or heat transfer coefficient, may not be sufficient to fully resolve heat transfer issues. It is important to note that heat transfer enhancement methods improve the rates of hydrogen absorption and desorption reactions but do not increase the actual storage capacity. However, heat transfer intensification might be detrimental to portable and mobile hydrogen storage uses. There is no effect of reactor geometry on the development process of hydride; however, it affects the reaction speed. The hydrogen storage capacity can be improved by upgrading the configuration of the reactor and the design of the heat exchanger.

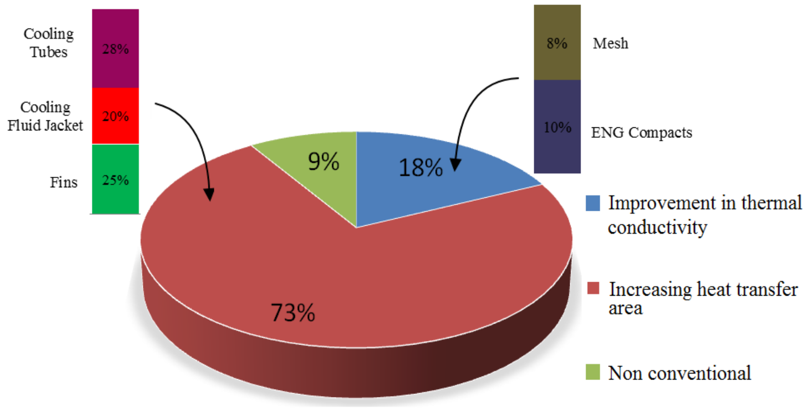

Figure 3. Statistical comparison of heat transfer techniques studied

5.2 Complex Hydride

A recent focus of research in solid-state hydrogen storage involves using complex metal hydrides, which are comparatively lightweight. These materials offer high theoretical hydrogen storage capacities and densities, operate at elevated desorption pressures and temperatures, and are relatively cost-effective. The major drawback of conventional metal hydrides is their small hydrogen storage capacities [98–100]. Conventional metal hydrides mainly consist of heavy elements from the periodic table, and metals like Lithium (Li), Sodium (Na) and Beryllium (Be) are lighter elements than Mg, which have the capacity to make solid-form compounds with hydrogen. Unlike conventional metal hydrides, the reaction is completed in more than one step in the case of complex hydrides. In such hydrides, the hydrogen is usually situated at the corners of a tetrahedron and forms covalent bonds with central atoms. The generalised formula for complex hydrides is $A_xMe_yH_z$, where A = Group I or II elements from the periodic table and Me = mostly B or Al. In many instances, there are two hydrogen atoms associated with each metal atom. Table 7 shows the hydrogen storage capacities of complex hydrides. This kind of complex hydride has the highest volumetric density [e.g., 150 kg/m^3 in Mg_2FeH_6 and $Al(BH_4)_3$] as well as the highest gravimetric density [e.g., 18 wt% in $LiBH_4$].

Table 7. Hydrogen storage capacities of complex hydride [14]

Hydride	H_2 (wt. %)
$LiBH_4$	18.0
$Al(BH_4)_3$	17.0
$Be(AlH_4)_2$	11.3
$LiAlH_4$	10.5
$NaBH_4$	10.4
$Mg(AlH_4)_2$	9.3
$Ti(AlH_4)_4$	9.3
$Ca(AlH_4)_2$	7.7
$NaAlH_4$	7.5
$KAlH_4$	5.7

5.2.1 Types of complex hydrides

Alanates

Alanates consist of H_2 & Al and are well-known as ‘complex alumino hydrides or complex aluminium hydrides’. In the case of hydrides, Al is well-known as a destabilising element [101]; moreover, alanates lose hydrogen when they come in contact with water [102]. The alanates are easy to produce. This constellation consists of magnesium alanate, sodium alanate, potassium alanate, lithium alanate, calcium alanate and others. Table 8 shows the characteristics of alanates. $NaAlH_4$ is the most famous material among Alanates.

Table 8. Characteristics of alanates [103]

Alanates	Hydrogen capacity (wt %)	Dehydrogenation temperature (^0C)		Dissociation enthalpy (kJ/mol H_2)	
		First step	Second step	First step	Second step
$Mg(AlH_4)_2$	9.3	110-200	240-380	41	76
$LiAlH_4$	7.9	160-180	180-220	-10	25
$Ca(AlH_4)_2$	5.9	127	250	-7	28
$KAlH_4$	5.7	300	340	55	70
$NaAlH_4$	5.6	210-220	>250	37	47
Na_2LiAlH_6	3.2	245		53.5±1.2	
K_2NaAlH_6	2.8	325		98	

In a generalised form, alanates can be shown as $MAlH_4$, where M = Na, Li or K. Alanates have been studied extensively as decent possible aspirants for hydrogen storage as they have large theoretical hydrogen storage capacities [104–106]. The dehydrogenation process for alanates is given by equations (4) to (6).

$$MAlH_4\ (s) \rightarrow MAlH_4(l) \tag{4}$$

$$MAlH_4\ (l) \rightarrow \frac{1}{3}M_3AlH_6 + \frac{2}{3}Al + H_2 \tag{5}$$

$$\frac{1}{3}M_3AlH_6 \rightarrow MH + \frac{1}{3}Al + \frac{1}{2}H_2 \tag{6}$$

With the help of an appropriate catalyst, it could be possible to lower the decomposition temperature for such hydrides to atmospheric temperature. With extensive efforts, it could be possible to manufacture alanate with good thermodynamic and kinetic properties. For example, $TiCl_3$ can be added as a catalyst in order to enhance reversibility and to achieve balanced kinetic as well as thermodynamic properties of $NaAlH_4$. However, because of the high cost of the $TiCl_3$, it could be substituted by TiF_3 which has comparable catalytic performance. Also, it is reported that adding dopant materials to the base material decreases the reversible hydrogen storage capacities of the complex metal hydrides.

Borohydrides

Borohydrides, likewise known as tetrahydroborate, have considerable appeal to researchers because of their higher volumetric and gravimetric hydrogen storage capacity and tunable characteristics [107–109]. Numerous borohydrides exist, which include sodium borohydride, calcium borohydride, lithium borohydride, magnesium borohydride, and other borohydrides having a generalised formula $M(BH_4)_n$ where n = 1, 2, 3, 4 and M = Mn, Zn, Dy, Al, Be, Y, Rb, Gd, Zr, Cs [110]. Table 9 shows the characteristics of borohydrides. Though borohydrides hold a large number of hydrogen, their dehydrogenation needs a high temperature, except $Al(BH_4)_3$.

Table 9. Characteristics of borohydrides [111]

Borohydrides	Hydrogen capacity (wt %)	Dehydrogenation Temperature (^{0}C)	Dissociation enthalpy (kJ/mol)
$NaBH_4$	10.8	400	-216.7 to -272.4
$LiBH_4$	18.5	>300	-75.0
$Mg(BH_4)_2$	13.7	260-400	-393.0 to -57.0
$Ca(BH_4)_2$	9.6	350	32.0

The decomposition reactions of borohydrides are given below.

$$M(BH_4)_n \rightarrow nMH + nB + \frac{3}{2}nH_2 \tag{7}$$

$$nMH + nB \rightarrow nB + nM + \frac{1}{2}H_2 \tag{8}$$

The borohydrides with higher storage densities (gravimetric and volumetric), such as $LiBH_4, NaBH_4, Mg(BH_4)_2, Ca(BH_4)_2$ proved to be high-potential hydrogen storage materials among all other borohydrides [112,113]. On the other hand, these borohydrides have great thermodynamic stability and poor kinetics. However, the thermodynamics and kinetics of such borohydrides can be enhanced by applying destabilisation techniques or doping.

Nitrides (Amides, Imides and Mixtures)

At last, amides $[M(NH_2)_x]$ and imides $[M(NH)_x]$ are the next group that appealed to researchers was because of their high storage capacity. On the other hand, their practical application is limited due to low absorption kinetics [114]. It mainly includes lithium, calcium, and magnesium. Table 10 shows the characteristics of nitrides.

Table 10. Characteristics of nitrides [115]

Materials	Start of decomposition (^{0}C)	Melting point (^{0}C)	Molecular Weight (g/mol)	Hydrogen Content (mass %)
$LiNH_2$	120	390	22.9600	8.7
Mixture of LiA(NH_2) and 4LiH	130	---	---	More than 5
$NaNH_2$	400-500	410	39.0100	5.1
Li_2NH	600	---	28.8970	3.5
$Mg(NH_2)_2$	---	---	72.1232	5.5
$Mg(NH_2)_2$	---	633	56.3502	7.2

The decomposition reactions for different nitrides are

$$LiNH_2 + LiH \rightarrow Li_2NH + H_2 \tag{9}$$

$$Ca(NH_2)_2 + CaH_2 \rightarrow 2CaNH + 2H_2 \tag{10}$$

$$Mg(NH_2)_2 + 2MgH_2 \rightarrow Mg_3N_2 + 4H_2 \tag{11}$$

For absorption/desorption of hydrogen molecules and to form chemical bonds, it requires the supply or removal of the heat of the reaction.

5.2.2 Limitations

A fundamental understanding of atomic structure and the thermodynamics involved in the hydrogen charging and discharging reactions is crucial for advancing complex hydrides. Unlike metallic hydrides, complex hydrides release hydrogen through a series of chemical reactions in multiple steps, each requiring specific conditions. As a result, this process is not fully reversible in the same way as traditional metallic hydride systems. So, there is a significant gap between the theoretical and realistically achievable hydrogen storage capacities. It is seen that the maximum hydrogen storage capacity of 9.1 (wt%) can be achieved for lithium borohydride ($LiBH_4$) [111]. Also, it has slow kinetics, excellent thermodynamic stability, and a high discharge temperature. On the other hand, thermal properties fluctuate considerably between the absorption and desorption of hydrogen, as well as depend intensely on the size of the particle and its density. Consequently, the thermal characteristics of wholly and partly discharged reactors might vary considerably, and heat transfer systems need to be flexible for all such situations. As a result, many of these hydrides fail to achieve the US-DOE targets. The thermal simulation of this metal hydride type is more complicated than that of simple metal hydrides. However, it can be possible to overcome such difficulties to a definite level by thermodynamic destabilisation, control over the particle size of a hydride or by adding a dopant or catalyst. Such techniques can tune complex hydride materials to achieve good conditions.

6. COMPARISON OF HYDROGEN STORAGE TECHNOLOGIES

Three hydrogen storage technologies are reviewed in this literature. Each of these technologies has its own merits and demerits. Table 11 shows the current scenario and challenges related to hydrogen storage technologies.

Table 11. Current scenario and challenges related to hydrogen storage technologies

Storage Technology	Compressed Hydrogen	Liquid Hydrogen	Hydrides
Availability Status	Commercially available	Commercially available	Moderately developed, however, not yet commercially available
Energy efficiency	More than 90%	Less than 70%	Approx. 90%
Suitability	Stationary as well as on-road vehicles	Mostly space vehicles	Aimed at on-road vehicles
Economic Considerations	Less expensive	Expensive	Too expensive
Safety Concerns	Usual pressure vessel provisions and monitoring, Need for ventilation	Along with provisions for pressure vessels, caution is required to avoid the mixing of condensed air and hydrogen.	Comparatively safe
Other benefits	• Low weight • Some previously existing setup • Easy linking with fuel cells	• Low weight • Less operating pressure • Small volume • Good storage density can be possible	• Low weight • Less operating pressure • Hydrides can be made • into various shapes
Disadvantages	Storage, as well as fuel energy densities at high pressure, are as good as liquid form hydrogen, however still less than fossil fuels	• Required super insulation because of very low temperatures • Some hydrogen • lost through ventilation • The energy stored is not as good as liquid fossil fuels	Hydrides can degrade with time
Challenges	• Safety • Heat and mass transfer • High volume energy required for compression	• Energy-competence issues • Heat and mass transfer (dormant-boil off)	• Weight • Heat and mass transfer ✓ Higher desorption temperatures ✓ Slower desorption kinetics ✓ Unfavourable thermodynamics ✓ Thermal management • Tank filling/emptying time and pressure • Pyrophoric • Cycle life • Tank compatibility • Optimisation • High cost

7. POTENTIAL FUTURE RESEARCH DIRECTION

The thermal conductivity of the heat-exchanging fluid plays a crucial part in the improvement of the heat-exchange process. Earlier, water, oil and ethylene glycol were utilised as heat-exchanging fluids. All these fluids have low thermal conductivity, which is the key restraint for developing such equipment. Also, all the methods used to enhance the heat transfer, such as producing turbulence, enhancing the area, etc., do not yield much effect. Water has the largest thermal conductivity (~0.6 W/m-K) among all these conventional fluids. However, this value is much lower when compared with most of the metals. Hence, the improvement of thermal conductivity is significant for improving the execution of the systems [116–119]. The solids have larger thermal conductivity than liquids; a suspension of solid particles in a fluid gives many possible ways of heat transfer enhancement [120]. In general, most metallic and non-metallic solids and polymeric particles have much greater thermal conductivity than conventional heat transfer fluids. Hence, an inventive method is to suspend such particles in the base fluids to improve their thermal characteristics. Much investigation has been conducted to improve the thermal characteristics of base fluids by the addition of a minor amount of a highly thermal conductive solid.

However, micron-sized or larger particles can create difficulties such as poor suspension stability, sedimentation and, hence, channel clogging in the flow path of the fluid. In this case, nanotechnology offers a sophisticated answer to such difficulties. Nanoparticles (<100 nm) have a higher surface-to-volume ratio and also exhibit adequate suspension characteristics. Nanoparticles are evenly suspended in the base fluid and do not block the micro-channels; hence, they avoid sedimentation. Choi and Eastman [120] first used the term nanofluids, denoting fluids having discrete nano-sized particles with much greater thermal conductivity. Nanofluids show greater thermal attributes when compared with ordinary heat exchange fluids. The concept of nanofluids for heat transfer has been known for the last two decades. During this period, a substantial number of research publications reported an improvement in the thermal conductivity of ordinary fluids using nanoparticles. Other than the thermal conductivity of the fluid, heat capacity and flow rate are also very important for achieving a good heat exchange between the MH bed and the heating/cooling accessories. However, it has been observed that there is a lack of separate studies presented on the effect of these properties on the performance of the MH reactor.

8. CONCLUSIONS

In this review paper, hydrogen as a fuel and the exhaustive studies on thermal issues associated with its storage techniques are presented. The review included the storage of hydrogen in all three states and also covered nanoparticles and newer complex hydrides. As stated, hydrogen is a clean, renewable, and non-toxic alternative source of energy. However, many challenges are present that require extreme work to realise as well as fully uncover the hydrogen economy. Hydrogen production, purification, transportation, storage, and utilisation are all essential components that drive a hydrogen-based economy. On the other hand, the greatest challenge is to construct safe, compact, portable and economical hydrogen storage systems.

The specific concluding remarks on the different techniques are as follows:

i) It is observed that properties such as heating value, boiling point, heat of vaporisation, density, diffusivity, ignition energy, and flame velocity are better for hydrogen than conventional fuels. Because of all such distinct properties, hydrogen can be treated as a unique fuel. Hydrogen has the highest energy capacity per unit mass when compared with fossil fuels, but it has a low energy capacity per unit volume, which is an actual challenge for its storage.

ii) The prior study shows that each storage technology has considerable heat transfer problems. In some instances, such problems are so critical that they create feasibility questions about storage methods. The heat transfer problems and possible solutions related to hydrogen storage methods are elaborated.

iii) Enhanced heat transfer is essential for a compressed hydrogen storage system during the compression, intercooling and tank filling processes to minimise the work input. Despite being compressed to pressures as high as 700 bar, hydrogen still maintains a relatively low density, posing a significant challenge. Additionally, the compression process results in an energy loss of approximately 11 to 13% of hydrogen's total energy content. Notably, a substantial temperature increase occurs during compression, even when multistage intercooling is applied. Based on desirable material properties, four types of vessels are commonly used for the storage and transportation of compressed hydrogen gas, out of which type IV pressure vessels made up of fully composite material can be utilised for both stationary and onboard hydrogen storage at very high pressure (~700 bar).

iv) In the case of liquid hydrogen storage, improved heat transfer is crucial, as there is a necessity to retain very low temperatures for a long time. Due to its extremely low boiling point of 20.3 K, the preparation and maintenance of liquid hydrogen requires sophisticated technology. Also, cryogenic tanks should have better thermal insulation to reduce the boil-off effect. However, as no insulation is perfect even with utmost care, the transfer of heat cannot be zero, and hence, venting losses exist. It is desirable to have a cylindrical shape for a tank storing LH_2, as it offers many advantages when compared with a spherical shape, such as ease in manufacturing, economical and easy onboard installation.

v) Heavy metal hydrides are limited by their low gravimetric density and relatively high absorption-desorption temperatures. In contrast, light metal hydrides face challenges due to the significant energy required for hydrogen release. Important design considerations of solid-state hydrogen storage devices are gravimetric hydrogen storage capacity and absorption-desorption rates of the device. Improved heat transfer for metal hydride systems is needed because of the low effective thermal conductivity of particle beds and for quick hydrogenation and dehydrogenation.

vi) The heat transfer intensification techniques mainly utilised ENG, metal foam, fins, water jackets, cooling tubes, etc. It was observed that the use of ENG is a more efficient technique for heat transfer enhancement as compared to metal foam and fins. From the published heat transfer enhancement techniques, it is observed that researchers were focused more on fins, cooling tubes and jackets and reported that enhancement in any one of the parameters, such as thermal conductivity, surface area and heat transfer coefficient, may not be able to sort out the problem of heat transfer completely. Hence, it is suggested to use more than one technique at a time.

vii) Complex hydrides are promising alternatives for solid-state hydrogen storage due to their advantages over traditional metal hydrides. However, many of these materials face challenges such as slow reaction kinetics and high thermodynamic stability. These issues can be partially addressed through catalysts and thermodynamic destabilisation techniques.

viii) Each technology has its issues, the solution for which may or may not be applied to others. All methods have their own merits as well as demerits. Compared with other states of hydrogen storage, the storage in solid-state compounds seems to be the best possible answer.

ix) In order to improve heat transfer for hydride systems, one more aspect, i.e., the use of nanoparticles to enhance the thermal conductivity of the cooling fluid, is revealed as a potential future research direction. Other than the thermal conductivity of the fluid, heat capacity and flow rate are also very important for achieving a good heat exchange between the MH bed and the heating/cooling accessories. However, it has been observed that there is a lack of separate studies presented on the effect of these properties on the performance of the MH reactor.

The review indicates that no single hydrogen storage method currently fulfils all the targets set by the US-DOE. It is anticipated that this paper will serve as a valuable resource for the research community working on hydrogen storage materials by providing a comprehensive overview of existing studies.

ACKNOWLEDGEMENT

The authors would like to express their gratitude to everyone who contributed indirectly to the development of this work.

CONFLICT OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

AUTHORS CONTRIBUTION

Dr. Rahul Uday Urunkar: Conceptualization, Data curation, Writing - original draft.

Dr. Sharad Dattatray Patil: Supervision, Methodology, Writing - review & editing.

Dr. Trupti Uday Urunkar: Formal analysis, Writing - review & editing.

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NOMENCLATURE

H_2	Hydrogen	K	Thermal conductivity, W/m-K
Abs	Absorption	h	Heat transfer coefficient (W/m^{2}K)
Des	Desorption	MH	Metal Hydride
ΔH	Enthalpy of formation, J/mol H_2	HE	Heat exchanger
ΔS	Entropy of formation, J/mol H_2-K	P	Pressure, bar
C	Hydrogen concentration	R	Ideal gas constant, J/mol-K
C_p	Specific heat, J/kg-K	T	Temperature, K
E_a	Activation energy, J/mol H_2	V	Volume, m^3