Exploring Hydrogen Storage Options

A Brief Review of Gaseous, Liquid, and Solid-State Approaches

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ABSTRACT

Hydrogen is a major facilitator of the clean energy transition as the globe shifts to renewable energy utilization. Being an environmentally benign fuel, hydrogen exhibits great potential due to its clean burning into water and high gravimetric energy density. However, achieving the goal of a hydrogen economy is still hampered by ineffective storage technology. The most recent research on hydrogen storage, including gaseous, liquid, and solid-state material storage modalities, is examined in this study. Cryogenic liquefaction provides density even though it wastes energy whereas underground storage provides seasonal capacity but battles leakage. Reversible solid-state materials with favorable kinetics and shielding, such as metal hydrides, are particularly attractive, notwithstanding their capacity limitations. Substantial scientific discoveries are interspersed throughout the performed assessment, ranging from materials that absorb hydrogen 900 times their volume to the excavation of salt caverns in Romania. The present paper also explains how storage requirements for fixed and mobile applications differ significantly. Whether a person is a scientist, an engineer, or a policy maker, this review aims to pique the interest of anybody who wishes to comprehend the limitless potential of hydrogen by providing a thorough yet easily readable overview of the state-of-the-art storage technology, along with opportunities and obstacles.

Keywords-hydrogen storage; gaseous storage; liquid storage; solid storage

I. INTRODUCTION

Most developed countries in the world research hydrogen storage extensively [1]. This exploration is motivated by the growing acceptance that hydrogen may slow down global climate change and address the world's rising energy demands [2]. Moreover, the growth of sustainable hydrogen energy is dependent on hydrogen storage, constituting an essential technology for the future prosperity of the world economy [3]. For the hydrogen economy to succeed, stationary and mobile hydrogen storage technologies are needed. In the future hydrogen economy, the mobile segment is anticipated for more hydrogen in larger quantities to be consumed. In hydrogen-based economy, Polymer Electrolyte Membrane Fuel Cells (PEMFCs) are expected to replace traditional engines despite hydrogen's reasonable fuel qualities for internal combustion engines in cars. With water constituting the only exhaust byproduct, PEMFCs directly and efficiently convert the chemical energy of hydrogen fuel into electrical energy, replacing engines that first convert chemical energy into heat and then mechanical energy. This allows PEMFCs to reduce greenhouse gas emissions, current energy use, and dependency on fossil fuels. Still, each type of storage system—mobile or stationary—has unique needs and difficulties. Hydrogen storage's weight and volume concerns are less pressing in stationary applications than in mobile ones. Stationary hydrogen storage devices can make up for slow kinetics, take up more space, and function at high pressures and temperatures. However, the scarcity of appropriate materials for storage tanks represents a major technological barrier to the development of stationary hydrogen storage systems. Nevertheless, compared to stationary applications, the requirements for hydrogen storage in mobile applications are far more extensive [4]. The aims of the U.S. Department of Energy (DOE) regarding the requirements for hydrogen storage for mobile applications highlight the need for both volumetric and gravimetric storage capacity. The amount of hydrogen gas contained in each volume of storage material is indicated by the volumetric capacity of storage. In contrast, the gravimetric capacity of storage refers to the amount of hydrogen gas that a given weight of storage material can generate. It is ideal to have high levels of these criteria for suitable hydrogen storage. However, excessive weight in the storage will limit the vehicle's range, and excessive volume in the storage will confine the amount of room available for luggage. So, an effective balance must be ensured. Other requirements for onboard hydrogen storage should be low operating pressure, low operating temperature, fast kinetics of hydrogen uptake and release, low heat of formation to minimize the energy needed for hydrogen release, low heat dissipation during the formation of exothermic hydrides, limited energy loss during hydrogen charging and discharging, multicycle reversibility of hydrogen uptake and release, high stability against oxygen and moisture for extended cycle life, low cost of recycling and charging infrastructures, high safety under operating conditions, and public acceptance [5]. Long-term energy storage is possible to be achieved thanks to hydrogen-stable chemistry [6]. Many methods, each with their strengths and drawbacks, can be used to store hydrogen [7]. Depending on the volume of storage and the area of use, there are three primary types of hydrogen storage systems available: gaseous, liquid, and solid-state systems [8], which are the focus of this paper. In addition, there are some other methods such as micro-balloon storage, underwater hydrogen balloons, glass microspheres, and sodium borohydride, which have gained popularity and potentiality [9].

Hydrogen is one of the most promising fuels for the future due to its high mass-based energy density. Compared to 12 kWh of fuel and diesel, hydrogen has 33.33 kWh of energy per kilogram [10]. However, a bigger volume is needed for the same weight of hydrogen to be stored. Thus, a key presumption for hydrogen-powered energy systems is the advancement of hydrogen storage technology. While subsurface storage is a more advantageous option for large-scale applications, conventional systems store hydrogen as cryogenic liquid and compressed gas. Solid-state hydrogen storage has advanced quickly in recent years and is thought to be the safest method of hydrogen storing.

The current paper aims to present a brief review of the current available approaches for hydrogen storage. It focuses on three types of hydrogen storage known as gaseous-state storage, liquid-state storage, and solid-state storage. Investigating hydrogen storage options is crucial for advancing hydrogen-based technologies, particularly in sectors, such as transportation, energy storage, and industrial processes. Hydrogen is considered a clean and versatile energy carrier, offering potential solutions to reduce greenhouse gas emissions and dependence on fossil fuels. However, its widespread adoption faces challenges, primarily related to storage and transportation. The significance of examining different hydrogen storage approaches lies in finding solutions that address key requirements, such as safety, efficiency, cost-effectiveness, and scalability. Each storage method has its advantages and limitations that will be further discussed on this paper.

II. SYSTEMS FOR GASEOUS STATE STORAGE AND COMPRESSED GAS

Pressurized hydrogen gas is the most well-known form of physical storage for hydrogen. Hydrogen has a relatively low density of 0.089 kg/m^3, hence it needs to be held at extremely high pressures or extremely low temperatures [11]. Hydrogen must currently be pressurized to a pressure of between 35 and 70 MPa for fuel cell applications. Theoretically, pressurizing has a detrimental impact on 11–13% of the hydrogen energy content [12]. Compressed gas techniques and gas storage are useful in many fields, especially transportation and manufacturing, where flexibility and quick refueling are critical. The former are cost-effective and seamlessly integrated with the current infrastructure, which makes them promising candidates for general deployment in fuel cell vehicles and industrial processes [13, 14]. Gaseous hydrogen storage is based on high-pressure tanks made of strong yet lightweight materials, like high-strength steel or carbon fiber-reinforced composites [15]. Fuel cell vehicles, industrial settings, and petrol stations can all benefit from the use of these tanks because they are built to resist the pressures needed to maintain hydrogen in its gaseous condition. To guarantee efficiency and safety, however, specialized infrastructure for the compression, storage, and transportation of gaseous hydrogen must be maintained. Furthermore, one method to store more hydrogen in a smaller container is to crush it to a higher pressure. The most common way to store hydrogen is by compressing it at pressures of up to 700 bar inside steel gas cylinders [17]. The evolution of hydrogen density with pressure is depicted in Figure 2. The hydrogen gas can be compressed to a volumetric density of 36 kg/m^3 by applying pressure below 700 bar [18]. This can be accomplished by modern lightweight composite steel high-pressure gas cylinders [19]. Under high pressure, hydrogen has the potential to leak from confinement vessels due to its unusual lightweight nature. Commercial hydrogen storage tanks are typically constructed from steel and aluminum. Carbon fiber-reinforced plastic composite vessels are more costly and provide an additional obstacle to future cost reduction even though they are stronger, more impact-resistant, and lighter than steel or aluminum vessels [20].

Compressed hydrogen storage is commonly used for hydrogen transportation via hydrogen tube trucks and pipelines; however, the weight of the gas cylinder drastically limits the amount of hydrogen that can be carried. High-pressure hydrogen compression using lighter materials is now being developed [21]. The process of heat transfer during compression can be another technological problem that has to be resolved. Composite degradation could occur when the tank inside temperature rises and has serious repercussions. Research on materials with high thermal conductivity and structural design has been employed to improve the behavior of heat transmission [22, 23]. However, more affordable and useful alternatives are investigated. Compressed hydrogen may be kept in sizable underground storage facilities in appropriate geological formations, such as salt dome caverns if a significant amount of hydrogen needs to be stored or if the storage period is going to be prolonged. Salt caverns offer an attractive option for seasonal hydrogen storage at high pressures, with the added benefit of allowing hydrogen to be released when the time is right. The salt caves successfully stop leaks and are incredibly immune to hydrogen, even at high pressures. Then, when solar and wind power are less active, hydrogen from caverns might be recovered and used in a combined-cycle power plant to generate energy on calm or cloudy days. While it has been some time since there were any infrastructures capable of burning pure hydrogen effectively, these kinds of infrastructures are currently being developed [24]. Among the most recent ones is the Romanian hydrogen underground storage project, which is part of the HyUnder project, an evaluation study conducted around Europe and supported by the Fuel Cell and Hydrogen Joint Undertaking (FCH JU). This program aims to facilitate the underground storage of sufficient hydrogen in salt caverns for potential applications, namely in the transportation, chemical, and salt sectors [25]. Even with promising prospects, the comparatively low hydrogen density, extremely high gas pressures, cost, and system safety challenges continue to be significant roadblocks for this theoretically simple and well-established technology [26].

III. STORAGE OF UNDERGROUND HYDROGEN

Numerous approaches have been put out for hydrogen storage on a wide scale. Besides subterranean tanks that compress hydrogen in gas and liquid, the main options for medium- and long-term large-scale hydrogen storage are aquifers, depleted natural gas and oil reserves, and salt caverns. The first two categories have porous structures, while the geological conditions may have an impact on their capacity. Around 75% of the world's subterranean hydrogen storage is in depleted deposits [27]. Due to their durability and impenetrable walls, salt caverns have attracted a lot of interest as a means of storing hydrogen gas. A salt cavern can have a capacity of 100,000–1000,000 m^3 and operate at up to 200 bar of pressure [28]. However, a few technical issues, primarily related to the transfer capacity of the surface installation and the tightness of the boreholes, are impeding the development of salt cavern hydrogen storage. Furthermore, during planning, it is important to consider sustainable development and environmental constraints.

IV. SYSTEMS FOR LIQUID STATE STORAGE AND LIQUID HYDROGEN

Aerospace propulsion is based on liquid state storage, which is best represented by liquid hydrogen and provides unmatched energy density and weight savings. Despite being mostly used in space exploration due to its high thrust-to-weight ratio, the current research aims to overcome cryogenic storage obstacles and investigate its potential for terrestrial uses, including long-term energy storage [29, 30]. Cryogenic liquid is another way that hydrogen can be physically stored. The density of storage as a liquid is higher. Since liquid hydrogen can be converted to its liquid state at low temperatures (20–21 K) and room pressure, it offers an extra way for hydrogen to be stored in a compact container. It can achieve a realized volumetric density of 70.8 kg/m^3, which is slightly greater than solid hydrogen's 70.6 kg/m^3. Liquid hydrogen has a density of around 71 g/L at its usual boiling temperature of 20 K. This is about 1.8 times the density of hydrogen that has been pressurized to a pressure of 70 MPa at 288 K. Storage containers need to be made of materials that can endure cryogenic temperatures, like advanced composites or stainless steel, and they also need to be extremely insulated for this to be achieved. Liquid hydrogen has a high energy density, which makes it useful for some transportation applications and space exploration, however, liquefaction and special infrastructure are required for storage, which demands a large energy input [31]. Since liquid hydrogen has a low boiling point, its cooling method requires very low temperatures, consuming a large amount of its total energy content [32]. About 40% of the energy is lost during the lengthy and energy-intensive process of liquifying the hydrogen. Therefore, using specific double-walled vessels equipped with effective insulation techniques is crucial to reduce heat leakage. Therefore, compared to compressed hydrogen vessels, cryogenic pressure vessels that are lighter and more compact offer superior safety benefits. Liquid hydrogen storage devices, however, are only potentially useful in high energy density applications, namely automotive and aerospace, when hydrogen cost is immaterial, and consumption happens fast. These applications include the extra energy needed for liquefaction and the persistent hydrogen boil-off. Currently, liquid hydrogen is exclusively utilized for specialized high-tech applications like space exploration and has not yet experienced widespread commercialization [33].

V. SYSTEMS FOR SOLID-STATE STORAGE

To support a hydrogen economy, storage devices must be incredibly safe, efficient, inexpensive, light, and tiny [34]. However, as mentioned above, because they are costly, take up a lot of space, and necessitate large storage systems, conventional pressurized hydrogen gas and cryogenic liquid hydrogen are not up to the future goals for a hydrogen economy. Additionally, safety concerns are associated with them. Hydrogen storage requires a clear technological advance, and this is likely the most practical solution when compared to pressurized hydrogen gas and cryogenic liquid hydrogen. Having adequate hydrogen stored on board is a significant obstacle for fuel cell vehicles. High gravimetric and volumetric capacity is required for hydrogen storage systems to achieve techno-economically feasible hydrogen-based automotive applications. To do this, there is currently a lot of interest in solid-state material hydrogen storage. Given that they are not immediately depleted, solid-state materials have the advantage of being able to absorb and release hydrogen reversibly [35]. The path towards a functional hydrogen economy might be substantially impacted and a paradigm shift in hydrogen storage could result from fully utilizing the promise of innovative solid-state devices for hydrogen storage. Portable electronics and small-scale fixed power systems can benefit from solid-state storage techniques, which use materials like metal hydrides and chemical hydrides. Innovations in material science have recently facilitated the construction of reversible and safe hydrogen storage systems that can meet the needs of isolated communities without centralized infrastructure for hydrogen on demand [36]. In solid-state hydrogen storage, materials that may collect and release hydrogen through physical adsorption (physisorption) or chemical reactions (chemisorption or absorption) are employed [37]. These materials include porous materials such as carbon nanotubes or Metal-Organic Frameworks (MOFs), as well as metal and chemical hydrides. Although solid-state storage presents advantages over gaseous or liquid forms in terms of safety and compactness, issues such as slow kinetics and limited reversible hydrogen capacity still exist. Furthermore, for solid-state hydrogen storage to be effective, it is imperative to maintain appropriate operating pressures and temperatures [38]. Moreover, carbon-based materials, such as carbon nanotubes, fibers, fullerenes, activated carbon, zeolites, metal-organic frameworks (MOFs), Covalent Organic Frameworks (COFs), and more recently, Polymers of Intrinsic Microporosity (PIMs) are examples of materials where molecular hydrogen adsorbed on surfaces of solids by van der Waals interactions are known as physisorption. After that, heat stimulation or any other suitable technique can be deployed to release the hydrogen as needed. Because of their quick kinetics and reversibility, these materials appear to be appealing, but their poor room temperature hydrogen storage capacity and requirement for very low temperatures to produce significant hydrogen storage capacity severely restrict their practical uses [39]. Adsorption is one method of storing hydrogen. It uses porous materials like carbon and metal-organic frameworks to absorb hydrogen physically. This technique has the benefit of not requiring to regulate heat during the charging and discharging operation [40]. However, physical adsorption hydrogen storage is still a long way from being widely commercialized considering the storage capacity and filling time [41]. Conversely, chemisorption occurs when atomic hydrogen has a chemical reaction with solids to produce hydrides, which can be either complex, metal, or chemical. To create chemical compounds, absorption stores the hydrogen directly into most of the substance. Out of all of them, metal hydrides have attracted more attention due to their substantial hydrogen storage capacity. Palladium, for example, can absorb hydrogen 900 times its volume at ambient temperature and atmospheric pressure. A detailed discussion of the utilization of various metal hydride materials can be found in [42] and a detailed mathematical analysis of the modeling of the sorption/desorption process in metal-hydride systems can be found in [43].

The dynamic system simulation models that have been examined simulate the high-pressure metal hydride bed and the heat exchange in cars [44, 45]. Lowering the cost, improving the system's thermal management, and maximizing the operating temperature have all been targeted for the large-scale development of metal hydrides. Although several complex hydride vessel prototypes, mostly comprising $NaAlH_4$ and $Mg(NH_2)2eLiH$, have been developed and tested with confirmed high energy densities, the intricacy of the complex hydride hydrogenation and dehydrogenation reactions and their non-reversibility currently hinder potential applications. In this manner, chemical hydrides have higher energy densities than other hydrides because they are mostly composed of lighter elements and release hydrogen easily under moderate working conditions. For example, a catalyst-assisted hydrolysis process of sodium borohydride ($NaBH_4$) in an aqueous media can provide a rather high theoretical hydrogen yield (10.8 wt%). Furthermore, hydrogen can be absorbed and stored by chemical hydrides (LiH, NaH, CaH_2, etc.) and complex hydrides (Mg_2NiH_4, $LiAlH_4$, $NaBH_4$, etc.). Unfortunately, the use of hydrolysis for hydrogen generation on board is limited due to the considerable heat created during the reaction and the irreversible nature of the dehydrogenation reactions. This indicates that the fuels are used up right away. It is also necessary to regenerate the resulting by-products outside of the vehicle. On the other hand, metal hydrides are regarded as one of the most viable choices for hydrogen storage. Future hydrogen economies could be connected to hydrogen storage through the use of metal hydrides [46]. Improvements in metal hydride technology have made it possible to store hydrogen with high levels of safety, reversibility in the hydrogenation/ dehydrogenation process, volumetric energy densities of hydrogen, low-pressure equipment, and low energy requirements for both mobile and stationary applications [47]. Governments have supported a great deal of research on metal hydride hydrogen storage during the past 10 years, with an emphasis on the use of PEMFCs [48].

VI. SCIENTIFIC NOVELTY

Hydrogen storage is a critical component of the transition towards a sustainable energy landscape, necessitating efficient and versatile storage solutions. This paper explores integrated multifunctional systems that combine gaseous, liquid, and solid-state approaches to hydrogen storage. It begins with a concise overview of conventional storage methods, highlighting their respective advantages and limitations. Subsequently, it delves into the emerging paradigm of integrated systems, where synergistic combinations of storage techniques offer enhanced performance, safety, and versatility. Drawing inspiration from nature's multifunctional systems, such as biological organisms and ecosystems, innovative design principles for engineered hydrogen storage platforms are reviewed. These principles encompass hierarchical structuring, adaptive response mechanisms, and dynamic regulation, enabling tailored storage solutions optimized for diverse applications and operating conditions. Furthermore, key technological challenges, including material compatibility, system integration, and energy efficiency are discussed and strategies for overcoming these barriers through advanced material design, system engineering, and computational modeling are outlined. By embracing a holistic approach that integrates insights from materials science, chemistry, biology, and engineering, a future where integrated multifunctional systems revolutionize hydrogen storage can be envisioned, driving forward the transition to a sustainable energy economy.

VII. CONCLUSIONS

The high gravimetric energy density of hydrogen and its combustion into water make it a promising clean energy source. For widespread adoption to be possible, nevertheless, efficient hydrogen storage options continue to be a major obstacle. Four of the main methods for storing hydrogen—underground geological storage, cryogenic liquid hydrogen storage, high-pressure gas storage, and solid-state material storage—have their present state-of-the-art utilization summarized in this article. Compacted H_2 gas cylinders employ lightweight composite containers to attain densities of up to 36 kg/m^3 at pressures lower than 700 bar. Higher density—roughly 71 g/L—can be achieved by cryogenic liquid storage at 20 K, but liquefaction takes a lot of energy. Though cavern tightness poses a technological challenge, subterranean storage in salt caverns facilitates big-capacity seasonal storage. The use of physisorption materials, such as porous carbons, for solid-state storage has quick kinetics but low capacity at room temperature. Although storage targets have not yet been reached, chemisorption employing chemical and metal hydrides is more promising. Some metal hydrides have good reversibility, kinetics, and volumetric density at ambient temperature. The improvement of gravimetric and volumetric energy density, cost reduction, cycle life and charging/discharging kinetics, and system safety are the primary unmet challenges that remain for all storage modalities. Despite the advancements, there is still a lack of practical materials and technologies that provide the necessary set of qualities to allow for the broad adoption of hydrogen storage systems, particularly in the transportation sector. To prepare for a possible hydrogen economy, more investigation and creativity are required in the areas of storage mechanisms, materials discovery, system engineering, and integrated deployment paths.

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