



Innovative Materials and Techniques for Enhancing Hydrogen Storage: A Comprehensive Review of Damage Detection and Preventive Strategies

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Hydrogen is a promising alternative energy resource, but an improvement of secure and efficient storage solutions must be developed for its increased use. This review will investigate efforts to improve the storage of hydrogen using Solid-State methods such as Activated Carbon, Carbon Nanotubes, Metal-Organic Framework, and Metal Hydrides in comparison with traditional liquid and gaseous storage methods. Solid-state methods rely on the temporary trapping or chemical bonding of the hydrogen atoms and molecules to reduce the reactivity and explosivity of the hydrogen and improve safety and equipment sustainability. To support the research into storage methods and improve the industrial infrastructure for an increase in hydrogen use, several methods for detecting hydrogen are explored, including Acoustic Emissions Testing, Scanning Kelvin Probe Testing, and Digital Image Correlation. Lastly, various preventative measures used to improve the performance of material used in Hydrogen environments are researched, including Laser Shock Peening, Hydrogen Recombination Coatings, Hydrogen Diffusion Barriers, Hydrogen Getter Coating, and Microstructure Testing. [DOI: 10.1115/1.4065360]

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1 Introduction

Hydrogen has the potential to be an excellent alternative to traditional energy sources because it has a gravimetric density, is easy to generate, and produces only water vapor when burned in oxygen [1]. To support the replacement of fossil fuels with cleaner options, such as hydrogen and other renewables, research into how hydrogen interacts with material is becoming an essential area of research [1,2]. Due to its small atomic size, hydrogen can penetrate through materials, possibly changing their structure and properties [3]. For hydrogen to be used as a valuable renewable energy carrier, scientists must find ways to store and distribute it safely. Hydrogen-based systems require storage materials, a subject of extensive research in material science, physics, chemistry, and engineering. The search for reliable hydrogen-storage materials and their breakdown processes is growing in modern times. Topics of research include searching for new materials with higher hydrogen storage capacity, improving the

strength and durability of existing materials, and studying damage mechanisms that can happen during hydrogen absorption and desorption. There have been plenty of published reviews on hydrogen storage. However, the existing reviews focus solely on storage materials or the overall hydrogen economy. This work explicitly investigates ways to identify and mitigate damage to hydrogen storage materials. It also comprehensively analyzes several storage materials, including their advantages, disadvantages, and current endeavors to enhance their performance. It gives important information about the current state-of-the-art hydrogen research and the making of safe and effective hydrogen storage systems in the future. This study thoroughly reviews scholarly articles and conference records to demonstrate the detection and prevention of damage to storage materials. The review diligently aims to highlight unresolved problems and possibilities for additional investigation in hydrogen storage.

2 Research Progress on Hydrogen Storage Materials

In recent decades, there has been an increasing focus on developing hydrogen storage materials that are both economical and

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efficient. Scholars have investigated diverse materials and technologies to store and utilize hydrogen securely. Below is a chronological representation of the advancements in research on hydrogen storage materials over several decades.

During the 1970s, researchers investigated diverse materials such as metals, alloys, and hydrides to explore their potential as hydrogen storage mediums [2]. The fundamental hydrogen spillover concepts were established in the late 1960s and 1970s [3]. One of the initial investigations revealed that metal hydrides, including magnesium hydrides, exhibit a high capacity for hydrogen absorption [4]. During the 1980s, there was a notable expansion in research on hydrogen storage materials primarily focused on investigating the thermodynamics and kinematics of hydrogen absorption and desorption for diverse materials, for example, the development of commercial hydrogen storage electrodes and rechargeable nickel hydride batteries [5].

During the 1990s, the advancement of nanotechnology presented novel opportunities for hydrogen storage materials [6]. Research indicated that nanoparticles of specific metals exhibited superior hydrogen absorption and release capabilities compared to their bulk counterparts. Scientists investigated chemical hydrides, which can release hydrogen via a chemical reaction with water or other substances. The research emphasis transitioned towards exploring the potential of utilizing complex hydrides, such as metal borohydrides and metal amides, for hydrogen storage [7].

During the 2000s, research on hydrogen storage materials shifted toward applications. The focus was identifying materials that could be utilized in hydrogen fuel cells for vehicle transportation and other related purposes [8]. Additional research endeavors were conducted to explore novel materials, including composites of metal hydrides, and to investigate the potential of reversible chemical reactions for hydrogen storage and release. Researchers found that porous materials, such as metal-organic frameworks and covalent organic frameworks, possess substantial surface areas and can store hydrogen [9]. During the 2010s, significant progress was made in materials science and engineering, resulting in novel hydrogen storage materials that exhibit superior properties, including enhanced storage capacity, accelerated absorption and desorption rates, and reduced operating temperatures. Researchers also investigated novel techniques for hydrogen storage, including cryo-adsorption and hydrogen physisorption [10]. Metal hydride systems were developed in the 2010s [11] for hydrogen storage in emergency or backup power units, i.e., for stationary applications.

Ongoing research in the 2020s is dedicated to developing hydrogen storage materials to enhance their efficiency, safety, and practicality for diverse applications. Current research is investigating novel materials, including metal-organic polyhedra (MOPs) [12] and advanced carbon materials [13], as well as innovative techniques for hydrogen storage, such as solid-state hydrogen storage and liquid organic hydrogen carriers.

3 Hydrogen Storage Materials

Various techniques are available for hydrogen storage, such as compressed gas, liquid-state storage, and solid-state storage [11]. Compressed hydrogen gas storage systems require less specialized equipment and infrastructure than alternative methods, such as complex metal hydrides or liquid hydrogen storage. Liquid hydrogen exhibits a greater energy density per unit volume than compressed hydrogen gas, enabling a higher storage capacity within a smaller volume. The storage of liquid hydrogen is a desirable alternative for space and aviation applications due to its advantageous characteristics in terms of weight and volume [14]. Storing hydrogen in a solid material, commonly known as solid-state hydrogen storage, presents an appealing alternative owing to its potential for high storage capacity, safety, and convenient transport.

3.1 Solid-State Hydrogen Storage. Using chemical compounds for hydrogen storage through absorption or adsorption on

carbon materials offers distinct benefits in terms of safety. Considerable research efforts have been dedicated to developing solid hydrogen storage systems encompassing Metal-Organic Frameworks (MOFs), Zeolites, Metal Hydrides, Metal Nitrides, Metal Imides, and carbon-based materials [15]. Hydrogen is bonded in solid-state storage through physical means, such as using MOFs and carbon-based materials, or bonded by chemical forces, such as hydrides, imides, and nitrides. Aerogels like silica and carbon offer unique properties, such as high surface area and low density, which facilitate hydrogen adsorption and efficient diffusion. They can be functionalized to enhance hydrogen binding affinity and storage capacity. Carbon aerogels, on the other hand, offer excellent mechanical strength, thermal stability, and high specific surface area, making them suitable for both physical and chemical hydrogen storage mechanisms. Physisorption is a viable method for storing hydrogen in porous materials. Physisorption and chemisorption are two distinct ways gases interact with solid surfaces. Chemisorption involves hydrogen molecules binding to solid materials through strong chemical bonds, resulting in high storage capacities. It requires materials with suitable chemisorption sites like metal hydrides or complex metal-organic frameworks. Physisorption, on the other hand, involves hydrogen molecules adsorbing onto a material's surface through weak van der Waals interactions, offering lower storage capacities but reversible processes and milder conditions. Porous materials like activated carbons and zeolites are commonly investigated for physisorption-based hydrogen storage applications [16]. The physisorption of hydrogen in materials is of particular interest for various technological applications due to its rapid kinetics, complete reversibility, short refueling time, and high cycle life [9]. On the contrary, chemisorption exhibits a higher degree of gas adsorption, albeit occasionally accompanied by irreversibility. The desorption of the adsorbed gas necessitates an elevated temperature.

3.1.1 Activated Carbon. Activated carbon is a synthetic carbon material modified to possess high porosity, consisting of crystallized graphite and amorphous carbon with a high surface area [17]. Activated carbon's hydrogen storage rate and capacity can be impacted by its morphology and shape, specifically in powder, fiber, and granular forms. The rate of hydrogen adsorption in fibrous form exhibits a significantly higher value, ranging from 2 to 50 times faster under optimal conditions [18]. The hydrogen absorption in conventional activated carbon is directly proportional to its surface area and pore volume. Based on monolayer adsorption, the Langmuir isotherm model can accurately describe it. Under conditions of low temperature and high pressure, a significant increase in adsorption capacity is observed. Extensive research has been conducted on hydrogen adsorption on various commercial and modified activated carbon products [19]. Empirical findings indicate that products possessing micropore volumes exceeding 1 mL/g can retain approximately 2.2 wt% of hydrogen through physisorption. Refining the adsorbent and sorption conditions can enhance the storage capacity to 4.5–5.3 wt%. Activated carbon, which has undergone mechanical milling, exhibits a nanostructure that is characterized by defects, thereby leading to an increase in its specific surface area. Research shows that hydrogen storage capacity increases from 0.9 wt% to approximately 1.7 wt% after 10 h of milling [20]. Research has indicated that the adsorption capacity can be enhanced by loading precious metals, such as Platinum (Pt), onto activated carbon [21]. The confluence of chemisorption occurring on the Pt surface and physisorption on the carbon surface results in a noteworthy quantity of hydrogen spillover (Table 1).

3.1.2 Carbon Nanotubes. In the late 1990s, carbon nanotubes and other carbon nanostructures were investigated as promising hydrogen storage solutions [27–29]. Nanocarbon materials possess porous structures that serve as trapping sites, potentially enabling enhanced storage capacity [20]. Dillon et al. uncovered the hydrogen storage potential of carbon nanotubes and

Table 1 Hydrogen storage capacity of activated carbon

Adsorbent	H ₂ Storage (wt%)	Temp, Pressure	Ref.
AC	0.67	303 K, 10 MPa	[22]
AC	1.4	77 K, 0.1 MPa	[23]
AC	1.6	296 K, 13 MPa	[24]
AC	4.5	77.4 K	[25]
AC	5.7	77 K, 3 MPa	[22]
AC (KOH treated)	6.6	77 K, 4 MPa	[26]
AC (Ni doped)	1.8	77 K, 0.1 MPa	[23]
AC (Pt doped)	2.3	298 K, 10 MPa	[27]
AC (Pd doped)	5.5	298 K, 8 MPa	[28]

Table 2 Hydrogen storage capacity of carbon nanotubes

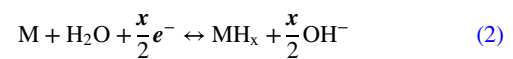
Adsorbent	H ₂ storage (wt%)	Temp, Pressure	Ref.
SWNT	5–10	133 K, 0.04 MPa	[32]
SWNT	3.50–4.50	298 K, 0.067 MPa	[33]
SWNT	1–15	77–300 K, 8 MPa	[34]
SWNT	8.50	80 K, 7 Mpa	[35]
SWNT	1.20	298 K, 4.8 MPa	[36]
MWNT	56	298 K, 12.16 MPa	[37]
MWNT	0.25	300–700 K, 0.1 MPa	[38]
MWNT	0.68	298 K, 10 MPa	[39]
MWNT	13.80	300 K, 1 MPa	[40]
MWNT	0.70–0.80	300 k, 7 Mpa	[41]

demonstrated encouraging outcomes. The two primary species of carbon nanotubes are distinguished by the composition of their walls, namely the Single-Walled Nanotubes (SWNT) and the Multi-Walled Nanotubes (MWNT) [30]. Diverse experimental conditions applied to these materials have yielded hydrogen storage values that exhibit significant variation, ranging from 0.25 to 56 wt%. As per the technical report published by the International Energy Agency, it is widely agreed that achieving very high hydrogen storage capacities, approximately 30–60 wt%, as reported a few years ago, is highly unlikely due to potential measurement errors. The storage capability, which can reach up to 6 wt%, remains advantageous under cryogenic conditions and in carbons with exceptionally high surface areas [31] (Table 2).

3.1.3 Metal-Organic Frameworks. Hydrogen storage within porous materials is a promising approach for transportation applications. The data related to hydrogen storage using MOF-5 (Table 3), which was first reported in 2003, indicated a significant surge in hydrogen absorption under external pressures, with a peak of 4.5 wt% observed at temperatures of 77 K and pressures below 1 bar [32], and an average of 1.3 wt% at a pressure of 1 bar [33]. After this, further advancements in diverse MOFs were documented, exhibiting elevated hydrogen adsorption capacities at 77 K and increased pressures. This can be attributed to these innovative frameworks' substantial pore volumes and notable specific surface areas [33,34]. In a theoretical investigation by Ahmed

et al. [36], approximately 500,000 MOFs were screened, revealing a maximum usable volumetric capacity of approximately 40 g H₂ L⁻¹ was found.

3.1.4 Metal Hydrides. Metal hydrides represent a class of materials with considerable potential to meet the benchmarks for hydrogen storage. Basic metal hydrides such as MgH₂ and complex metal hydrides like NaAlH₄ and LaNi₅H₆ have been evaluated for this application. The energy density by volume of these materials is commendable; however, their energy density by weight is frequently inferior to that of hydrocarbons. Solid hydrides, which have the potential to be shaped into pellets or granules, require an approximate temperature of 120 °C to release the stored hydrogen. For example, the hydrogen storage capacity of MgH₂ has been observed to reach up to 7.60 wt%. The hydrogen storage density of MgH₂ is comparatively higher than that of compressed hydrogen gas and liquid hydrogen, with a value of 6.5 hydrogen atoms per cm³ for MgH₂, 0.99 hydrogen atoms per cm³ for compressed hydrogen gas, and 4.2 hydrogen atoms per cm³ for liquid hydrogen. As a result, metal hydride storage has been identified as a safer and more space-efficient option for on-board applications. Two potential methods for synthesizing metal hydrides are direct dissociative chemisorption and electrochemical water splitting. The ensuing reactions are as follows: M denotes the metal, while x signifies its valence.



Light metals, e.g., lithium, beryllium, sodium, magnesium, boron, and aluminum-based storage materials, are popular substances used for storage. The different hydrogen capacities of some selected metal hydrides are shown in Fig. 1.

3.2 Liquid State Hydrogen Storage. Liquid hydrogen storage is the most common method, but it is expensive and complicated due to the need for extremely low temperatures (20 K) for liquefaction (Fig. 2). As a result, alternative liquid carriers with more reasonable storage conditions are being considered. The focus of attention, especially under the International Energy Agency Task 32, has primarily been on two specific carriers: Ammonia and Liquid Organic Hydrogen Carriers (LOHCs) [41–45]. Both have more moderate storage requirements than liquid hydrogen, reducing estimated storage and transportation expenses [44]. Most proposed uses for liquid carriers do not need on-board dehydrogenation in fuel cell vehicles. Instead, they involve centralized dehydrogenation at ports or other facilities to distribute hydrogen gas locally or at recharging stations.

3.2.1 Cryogenic and Compressed Storage. Cryogenic storage of hydrogen offers high energy density; however, their storage necessitates intricate systems to limit boil-off. These liquid tanks' complexity, size, and weight increase capital costs. In addition, costly liquefaction equipment is also necessary. Furthermore, the high energy requirements (30–33% of the total energy) for

Table 3 Measured and calculated crystallographic properties of high-capacity MOFs [37]

MOF	Surface Area			
	Gravimetric (m ² /g) Expt./Calc.	Volumetric (m ² cm ⁻³) Calc.	Pore Volume (cm ³ g ⁻¹) Calc.	Void Fraction Calc.
MOF-5	3512/3563	2172	1.36	0.81
IRMOF-20	4073/4127	2000	1.65	0.84
SNU-70	4944/4756	1905	2.14	0.86
UMCM-9	5039/4847	1805	2.31	0.86
PCN-610	6050/5777	1603	3.17	0.88

Hydrogen Storage of Some Metal Hydrides

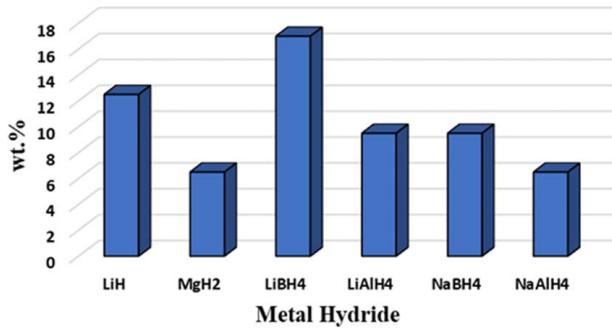


Fig. 1 Hydrogen storage performance of some selected metal hydrides [38]

hydrogen liquefying and economic considerations impede its development. Even the best-insulated containers can still result in hydrogen boil-off, energy efficiency, and security issues. At 1 bar and 20 K, the density of liquid hydrogen is 70 g/L. Although there is theoretical potential for greater hydrogen storage capacity, the practical efficiency of the liquid hydrogen tank limits its applicability. Because liquid hydrogen has a low boiling point (20 K), maintaining the cryogenic temperature requires a specifically made metallic double-walled container with an excellent insulating system. Even the best-insulated containers can still result in hydrogen evaporation, increased container pressure, energy efficiency, and security issues [45]. Cryo-compressed hydrogen storage requires comparatively low pressure and does not need a costly Carbon Fiber Reinforced Plastic (CFRP) tank. It can also increase energy efficiency and reduce hydrogen boil-off. At 276 bar and 20 K, the density of hydrogen increases to about 87 g/L in the cryo-compressed storage technique. The cryo-compressed container's hydrogen storage capacity rises to 0.058 kg H₂/kg system and 0.043 kg/L, meeting the target of the DOE 2025 on-board automotive storage values [46] (Table 4).

3.2.2 Liquid Organic Hydrogen Carriers. LOHCs are an efficient means of chemically bonding elemental hydrogen, providing possibilities for safely storing energy on a large scale [48]. The LOHC compounds such as cyclohexane, methylcyclohexane, and decalin can release up to 6 wt% hydrogen. The dehydrated form of LOHC undergoes a hydrogenation reaction to assimilate hydrogen. Hydrogenation releases heat energy and is conducted under high pressures, often ranging from 30 to 50 bar, and at temperatures around 150–200 °C in the presence of a catalyst. Once the hydrogen is required again, the LOHC undergoes dehydrogenation, resulting

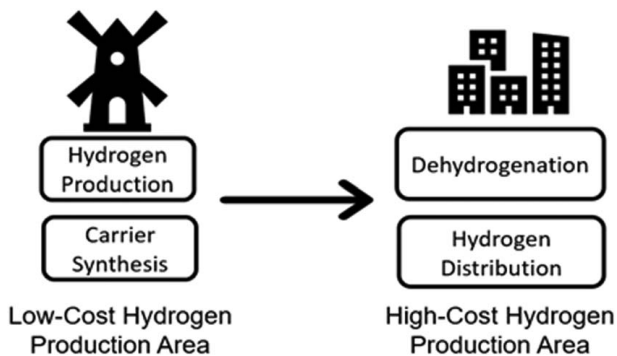


Fig. 2 Liquid hydrogen carriers to store energy generated in regions with affordable hydrogen for later usage in regions with more costly hydrogen

Table 4 Estimated performance for liquid hydrogen storage systems [47]

Storage system	Temp, Pressure	Hydrogen density (g/L)	Gravimetric density (Kg H ₂ /Kg System)
Cryogenic	20 K, 1 bar	70	0.05
Cryo-compressed	20 K, 276 bars	87	0.058

in the release of hydrogen (Fig. 3). The reaction occurs at high temperatures (250–320 °C) under the influence of a catalyst [49].

The dehydrogenation process to produce the corresponding aromatic compounds was predominantly carried out using platinum-based catalysts at temperatures ranging from 300 °C to 350 °C [43,46]. The elevated dehydrogenation temperature and the necessary costly catalysts impeded the commercial application of the LOHC processes. Scholarly investigations into these carriers have centered on reducing dehydrogenation temperature by modifying the LOHC molecule, thereby reducing the enthalpy of the reaction. The quest for catalysts that exhibit enhanced activity and stability and are composed of readily available earth-abundant materials for dehydrogenation is underway. Efforts to alter the enthalpy of dehydrogenation have predominantly focused on incorporating heteroatoms, specifically nitrogen, within the cyclic architecture [48–50]. Some materials, such as N-ethyl carbazole, have demonstrated better hydrogen storage and release properties than an unsaturated system [51,52]. The present study reports the synthesis of crystalline lithium amines through ball-milling primary amines with LiH. The results indicate that this method exhibits mild endothermic dehydrogenation and improved selectivity toward hydrogen release [51–53]. For catalyst support, certain materials have been discovered to enhance the hydrogenation rate, such as palladium nanoparticles, which showed increased activity when supported on a covalent triazine framework [54].

3.2.3 Ammonia. Due to the limited options available for transporting liquid hydrogen, there is significant interest in utilizing ammonia for hydrogen transportation [49]. Using ammonia as a hydrogen carrier offers a great opportunity for a cost-efficient and secure approach to storing and transporting zero-carbon energy (Fig. 4). Ammonia, with its dense concentration of hydrogen, has been widely produced for manufacturing fertilizers, leading to the creation of large-scale storage and transportation infrastructure. Ammonia can decompose into nitrogen and hydrogen, which can be used as fuel through cracking. The cracker consists of three primary elements: an evaporator, which vaporizes the liquid ammonia; a preheater, which raises the temperature of the ammonia gas to 400 °C; and a reactor, which contains the catalyst

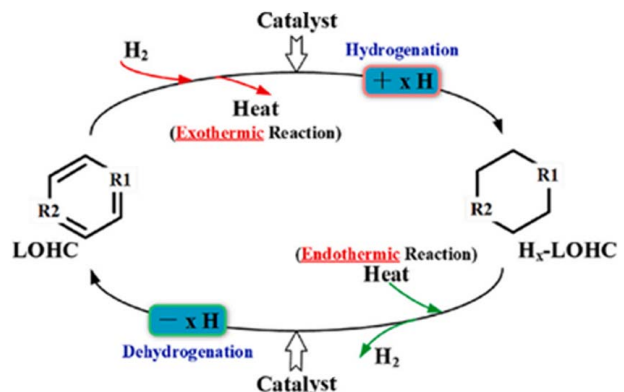


Fig. 3 Hydrogenation–dehydrogenation process [50]

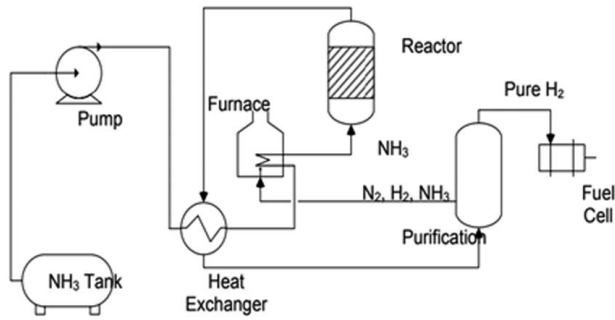


Fig. 4 Ammonia as a hydrogen carrier [55]

for the cracking reaction. A separator is necessary to achieve pure hydrogen extraction from the cracked gas due to the presence of a hydrogen and nitrogen combination.

Numerous research endeavors are underway to enhance the cost-effectiveness and efficacy of hydrogen storage associated with ammonia. The technology for hydrogen release from ammonia is not widely established, as it does not constitute a substantial component of the current ammonia markets. The catalysis of ammonia decomposition has been thoroughly investigated in transition metal systems to evaluate catalytic performance for ammonia synthesis [56,57]. Ruthenium has been identified as the metal with the highest level of reactivity. However, comparable performance levels can be attained by utilizing metals abundant in the earth's crust, such as iron, cobalt, and molybdenum [58–60]. The impact of temperature is a crucial factor to consider, as the attainment of complete hydrogen conversion necessitates elevated temperatures (>450 °C) even for the most dynamic Ruthenium-based catalysts. The development of novel catalysts is necessary to achieve a substantial reduction in operational temperatures. Metal-nitrogen-hydrogen materials, specifically light metal amides and imides, have successfully mediated ammonia decomposition. These materials have exhibited catalytic performance comparable to that of supported Ruthenium catalysts [61–63]. Sorption materials can store ammonia while exhibiting a considerably decreased vapor pressure, as evidenced by sources [43,64–66]. The ammine complexes of metal halides exhibit ammonia densities comparable to liquid ammonia. Research has demonstrated that ammonia release can be customized to suit a specific application by manipulating metal ion combinations within a ternary metal halide system. The decomposition pathways may be significant in customizing gas release for specific purposes, such as the incineration of ammonia, where hydrogen is a valuable co-fuel.

3.3 Gaseous State Hydrogen Storage. Gaseous storage of hydrogen under high pressure is a commonly used technique. To accomplish the high-pressure storage (350 bar to 700 bar), a tank material is required, which must have high tensile strength, low density, non-reactivity with hydrogen, and low diffusivity and be economical. Using low prices and high thermic conductivity, some steel- and aluminum-type materials are utilized to construct storage tanks. These metal materials are nondurable, heavy-weight, and have safety concerns [67,68]. In contrast to metals, CFRP [68–70], which is lightweight, has sufficient strength, and has durable properties, is a more promising material for pressurized

Table 5 Materials for high-pressure hydrogen storage tanks [72]

Material	Tensile strength (MPa)	Density (kg/m ³)
Steel alloys	703	8160
Titanium alloys	924	4430
Carbon composites	2070	1900

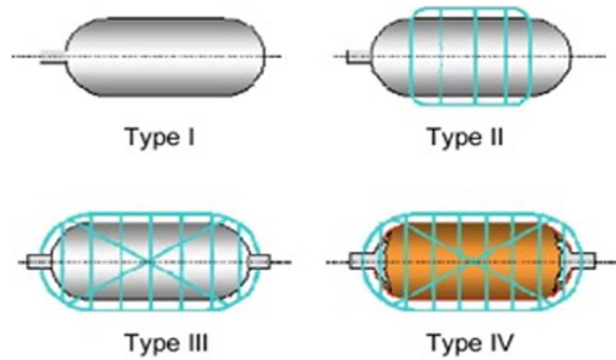


Fig. 5 A schematic illustration of the four types of pressure vessels [78]

gas vessels. At the same time, the low thermal conductivity and high-price issues need to be solved before the CFRP can be extensively used. Underground salt caverns can store high-pressure gas and are applied to stationary store-pressure hydrogen gas [69–71]. It is a feasible option for compressed hydrogen gas storage, with adjustable storage capacity, high-pressure storage ability, adaptable operating pressure, and minimized hydrogen leakage (Table 5).

Novel composite cylinders with reduced weight, capable of enduring up to 80 MPa pressures, have been developed. This allows hydrogen to attain a volumetric density of 36 kg/m³, half that of its liquid state at the standard boiling point [73]. The gravimetric hydrogen density negatively correlates with pressure due to the augmented thickness of the pressure cylinder walls. Pressure vessels are widely utilized in various fields, such as industrial, automotive, commercial, and aerospace applications, ranging from small bottles to large storage tanks. The pressure vessels presently employed in hydrogen storage systems for automobiles and buses are distinguished by a pressure of 35 MPa or 70 MPa. However, the preference for 70 MPa is rising to enhance storage density. The categorization of pressure vessels is based on four distinct types: Types I, II, III, and IV (Fig. 5; Table 6).

Type I refers to a vessel made solely of metal, usually steel. As a result, it is the heaviest type of vessel. It is commonly used in industries for stationary purposes. Type I vessels have a hydrogen storage capacity of approximately 1 wt% at pressures ranging from 200 to 300 bar [75].

Type II is a metal liner hoop-wrapped composite cylinder with a lower weight than Type I. Type I and Type II vessels are unsuitable for vehicle applications due to their inadequate hydrogen storage density resulting from their substantial weights and the challenges posed by hydrogen embrittlement.

Type III vessels consist of a completely wrapped composite cylinder with a metal liner to prevent hydrogen permeation. The metal liner is composed of aluminum, effectively addressing the embrittlement issue and enhancing mechanical strength by over 5%. Type III vessels offer a 25–75% mass gain over Types I and II, making them more suitable and expensive for vehicle applications. Type III vessels have also been shown to be reliable at pressures up

Table 6 Types of Pressure Vessels [74]

Type	Materials	Typical pressure (bar)	Gravimetric density (wt%)
I	All-metal construction	300	1.7
II	Mostly metal, composite overwrap in the hoop direction	200	2.1
III	Metal liner, full composite overwrap	700	4.2
IV	All-composite construction	700	5.7

Table 7 Benefits and drawbacks of different hydrogen storage methods

Hydrogen storage methods	Benefits	Drawbacks
Activated carbon	Abundant availability, ease of fabrication, cost-effectiveness, and non-toxicity [79]	Moderate storage capacities, slow adsorption and desorption, and susceptibility to contamination [9]
Carbon Nanotubes	Lightweight, thermal stability and mechanical strength, Compatibility with existing infrastructure [80]	Slow hydrogen adsorption and desorption kinetics, difficulty of synthesis, challenges in controlling nanotube size and diameter, and susceptibility to structural damage [81]
Metal-Organic Frameworks	Open pore structure, Low operating pressures, highly reversible, kinetics of hydrogen adsorption and desorption is fast [9]	Operation at low temperatures, Moisture sensitivity, Synthesis challenges, Stability issues at high temperatures or under high-pressure conditions [82]
Metal Hydrides	Low-pressure operation, Safe option, High volumetric and gravimetric hydrogen storage capacities, Efficient for compact storage systems [72]	High cost, slow Kinetics during hydrogen absorption and desorption, Issues related to high operating temperatures, and harder interfacing with fuel cells [72]
Cryogenic and Compressed Storage	Commercial availability, High volumetric capacity, denser packing of hydrogen molecules at lower temperatures. Easier transportation and storage [83]	High liquefaction demands energy-intensive cooling, affecting overall system efficiency, and the required insulation for maintaining low temperatures increases system complexity and cost, and hydrogen boil-off losses [72]
Liquid Organic Hydrogen Carriers	High volumetric hydrogen density can store much hydrogen in a relatively small space and is safe and easy to handle, even in large quantities. Closed carbon cycle [73]	Require energy to convert hydrogen to and from the LOHCs. This energy can be a significant cost, especially for large-scale applications, short life cycles, and difficulty in developing dehydrogenation catalysts [73]
Ammonia	Well-established industrial chemicals with existing production and distribution infrastructure flexibility in the release of hydrogen, fast kinetics, high hydrogen storage capability, and low cost [42]	It can harm human health if inhaled as it is toxic. Ammonia is a flammable gas, which means that it can be dangerous. It is a corrosive gas that can damage materials like metals and plastics and involves high temperatures, which can be energy-intensive, and the problems related to trace amounts of ammonia in the hydrogen after decomposition [75]
Gaseous State Storage	Commercially available, lightweight, suitable for fuel cell vehicles, easier integration into existing infrastructure, can be refueled quickly, rapid turnaround times [42]	A large volume and hydrogen embrittlement are required on the wall. Pressure vessels can leak hydrogen, permeation through walls, energy loss—high pressure, high cost of materials, and a required safety factor of more than 2.25 [42]

to 450 bar, but there are still challenges associated with pressure cycling tests at 700 bar.

Type IV pressure vessels are like Type III vessels, except that they feature a non-load-bearing polymer liner instead of a metallic liner. In specific applications, polymer liners in an “all-composite” construction may present superior cost and performance benefits compared to vessels that incorporate metal liners [76]. Type IV vessels are the lightest of the pressure vessels, making them most suitable for vehicle applications, and they can endure high pressures of up to 1000 bar. However, they are too costly due to the considerable cost contribution of carbon fibers. Cost projections show that the carbon fiber cost constitutes about 75% of the storage vessel cost, considering the high production volumes [77].

3.4 Comparison of Different Hydrogen Storage Techniques. The benefits and drawbacks of different hydrogen storage systems and their comparisons are shown in Tables 7 and 8, respectively.

4 Testing Methods to Detect Hydrogen Damages

Detecting and understanding hydrogen-induced damage in various materials is crucial for ensuring the integrity and reliability of critical structures related to the storage of hydrogen. As the detrimental effects of hydrogen damage continue to pose significant challenges to the safety and performance of these systems, the need for advanced testing methods becomes increasingly paramount. This section explores the efficacy of three innovative techniques—Acoustic Emission Testing (AET), Scanning Kelvin Probe Testing (SKPT), and Digital Image Correlation (DIC)—in detecting and characterizing hydrogen-induced damages. By examining the principles, applications, and limitations of each method, this study aims to provide a comprehensive overview of their potential

contributions to the field of materials science and engineering. Through a comparative analysis of these techniques, this research endeavors to enhance the current understanding of hydrogen-induced damage mechanisms and facilitate the development of more robust mitigation strategies for ensuring the structural integrity of critical components.

4.1 Acoustic Emission Testing. Acoustic emission testing involves attaching piezoelectric sensors to a test specimen to listen for high-frequency sound waves released when the material undergoes internal damage like cracking. As damage accumulates, it abruptly releases stored energy in the form of stress waves that propagate through the material and are detected by the sensors. Parameters of the emission signal, like amplitude, frequency, and hits, are analyzed to characterize the damage source event and understand the progressive damage mechanism in real time. Advanced analysis can also pinpoint the origin location of the damaging acoustic emissions. A signal was noticed after reaching the pitting potential. Used acoustic emission to monitor crack initiation and growth in high-strength martensitic steels during hydrogen charging and finite element method (FEM) in slow strain-rate tensile (SSRT) tests and welding tests [77]. Acoustic sensors detected pressure waves from hydrogen-induced crack advancement. Kaige Wu used the potentiodynamic method to conduct an acoustic examination of the hydrogen bubbles on the counter electrode during the pitting corrosion of 304 stainless steel [78]. There was a brief lag before an Acoustic Emission (AE) (Fig. 6).

4.2 Scanning Kelvin Probe Testing. The scanning Kelvin probe uses a vibrating reference probe to measure the Volta potential difference between the probe and points on the specimen surface. Scanning the probe across the sample surface maps local variations in Volta potential. Changes in subsurface properties

Table 8 Comparison of different hydrogen storage methods [76]

S/N	Hydrogen storage methods	Storage type	Energy density (KWh · m ⁻³)	Lifetime	Cost
1	Metal hydrides	Chemical absorption	244	Less than 2000 h	9.01–19.67 USD/kg (Ni metal hydride)
2	Liquid hydrogen storage	Physical	2359.30	300–500 days	3.66 USD/kg
3	CGH ₂ for Type IV Vessel	Comp. + Cooling	3.3–1320 (depends on compression strength)	Over 20 years	Starts from 466 USD/kg
4	Underground hydrogen storage	Physical	Can reach about 300,244	20–40 years	1.61 USD/kg

like hydrogen ingress that alter the local Volta potential are detected. The data produces a voltage map correlating surface potentials to internal material degradation. Kelvin probes provide high sensitivity to early-stage hydrogen damage through localized Volta potential changes well before cracking. The advanced analysis also reveals mechanisms and diffusion behavior based on voltage distribution. Celine Larignon demonstrated how Kelvin probe force microscopy (KFM) can find and identify hydrogen in damaged 2024 aluminum alloys [84]. Flavien et al. used Kelvin probe mapping to image hydrogen-induced cracking in high-strength alloys. The investigation of hydrogen penetration through high-strength DP1180 steel was conducted using X-ray photoelectron spectroscopy (XPS) and the Scanning Kelvin Probe (SKP) [85]. Crack initiation sites were correlated to Volta potential variations (Fig. 7).

4.3 Digital Image Correlation. Digital Image Correlation (DIC) is a non-contact optical technique used to measure material deformation and strain. It works by capturing a series of images

of a surface, typically before and after deformation. DIC software then tracks small subsets of pixels, called “correlation windows,” between the images. DIC calculates displacements and strains by analyzing how these subsets move relative to each other, providing valuable insights into material behavior and structural integrity.

In an electrochemical hydrogen (H) charging environment, David Martelo explained how to comprehend the fracture events that occurred during a slow strain rate test (SSRT) of a Ni-based alloy, UNS N07716, and how to link these events with in-situ and offline failure analysis methodologies [86]. Using a high-resolution digital image correlation, the in-situ techniques entailed continuous monitoring of fracture occurrences during an SSRT. Horikawa developed an advanced analytical system to dynamically and quantitatively monitor the environmental hydrogen embrittlement of aluminum alloys based on Al, Zn, and Mg at atmospheric air pressure. The system consists of a digital image correlation step, a slow strain rate testing equipment, and gas chromatography with a SnO₂-based semiconductor hydrogen sensor. It was shown that hydrogen atoms are produced when airborne water vapor reacts chemically with the alloy surface in the absence of oxide layers, leading to plastic deformation of Al–Zn–Mg alloys. Digital image correlation also made it clear that the specimen surface had multiple localized grain boundary cracks induced by the created hydrogen atoms, which ultimately led to a localized grain boundary fracture (Fig. 8 and Table 9) [87].

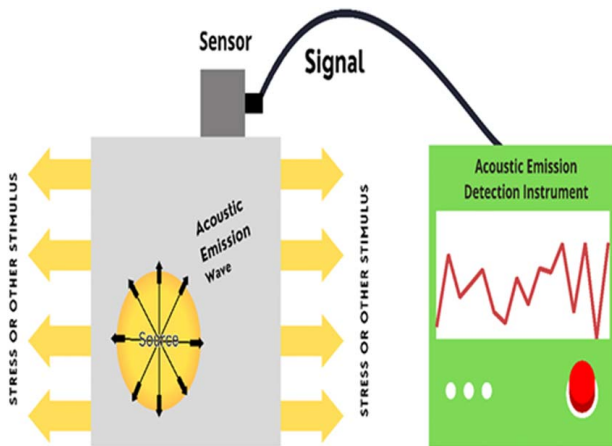


Fig. 6 Acoustic emission testing

5 Preventive Measures for Hydrogen Damages

Hydrogen-induced damage has emerged as a critical concern in various industrial sectors, including aerospace, energy, and transportation. To mitigate the detrimental effects of hydrogen on structural materials, several preventive measures have been proposed and researched. First, material selection is crucial, emphasizing the use of alloys with improved resistance to hydrogen embrittlement and corrosion. Additionally, the implementation of effective coatings, such as polymer films or metallic layers, serves as a protective barrier, shielding the material from direct hydrogen exposure. Furthermore, the application of strict operating and maintenance protocols, including controlled pressure and temperature

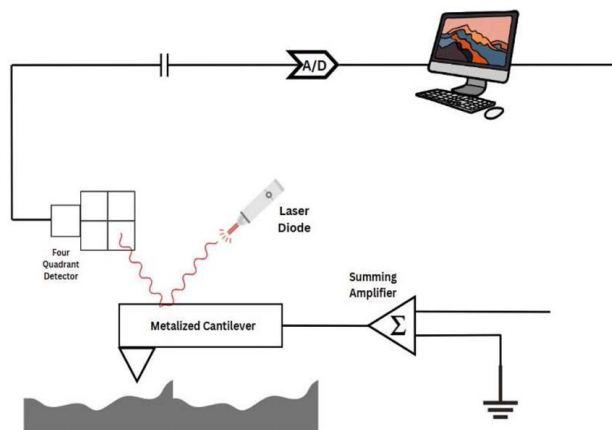


Fig. 7 Scanning Kelvin probe testing

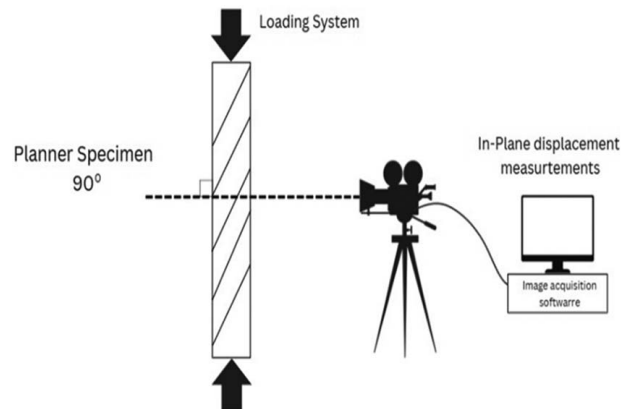


Fig. 8 Digital image correlation

Table 9 Advantages and disadvantages of different damage detection methods

Testing method	Advantages	Limitations
Acoustic Emission Testing	Early detection of defects and damage, Real-time monitoring of structural integrity, Non-destructive testing method, Detection of active defect propagation, Sensitivity to dynamic loading conditions, Applicable to a wide range of materials, Provides information on defect location and severity, Can be used for in-service monitoring and preventive maintenance	Limited depth penetration for defect detection, Directionality and challenges in pinpointing defect location, Signal attenuation and absorption in certain materials, Potential for background noise and false signals, Sensitivity to defect size and energy release, Influence of material properties and structural geometry, Environmental factors affecting testing conditions, Frequency range and signal variability based on defect type, Calibration and validation requirements for reliable results
Scanning Kelvin Probe Testing	The advantages of SKP testing for hydrogen damage detection include its Sensitivity, quantitative measurement capabilities, non-destructive nature, high spatial resolution, versatility, real-time monitoring capability, and complementarity with other analytical techniques	Surface sensitivity without depth profiling, Surface preparation requirements for accurate measurements, Susceptibility to environmental influences, Limited ability to detect subsurface or bulk damage, Complexity of data analysis and interpretation, Dependence on sample condition and homogeneity, Sensitivity to noise and artifacts
Digital Image Correlation	The high spatial resolution, non-contact nature, quantitative analysis capabilities, and versatility make it a promising tool for studying the effects of hydrogen exposure on materials and structures	DIC primarily measures surface deformation and strain. It may not effectively capture subsurface damage caused by hydrogen embrittlement, such as micro-cracks or hydrogen-induced material degradation that occurs beneath the surface. Surface preparation, including cleaning, coating, and roughening, is critical to ensure proper image acquisition and analysis. DIC data analysis involves complex algorithms and image processing techniques to extract deformation and strain information from image pairs

conditions and regular inspections, has proven instrumental in minimizing the likelihood of hydrogen-related failures. Laser shock peening, a surface treatment technique, has demonstrated promising results in enhancing material resistance to hydrogen-induced degradation by inducing compressive residual stresses and improving fatigue life. Hydrogen recombination coatings provide a passive layer that facilitates the recombination of hydrogen atoms into molecular hydrogen, minimizing their detrimental effects on the material's mechanical properties. Additionally, hydrogen diffusion barriers act as a protective shield, restricting the ingress of hydrogen into the material's bulk. Complementary to these measures, hydrogen getter coatings can effectively trap and remove hydrogen atoms from the material surface, preventing their accumulation and subsequent embrittlement. Lastly, microstructure tailoring, through techniques such as grain refinement and phase manipulation, plays a pivotal role in controlling hydrogen embrittlement by optimizing material properties and enhancing resistance to hydrogen-induced damage. Collectively, these preventive strategies offer a comprehensive approach to safeguarding structural integrity and ensuring the reliable performance of materials exposed to hydrogen environments (Table 10).

6 Future Direction of Research

Even with the noteworthy advancements achieved in the domain of hydrogen storage materials, there remains the requirement to develop hydrogen storage materials that are both efficient and economical. Subsequent investigations should prioritize the synthesis of substances that exhibit superior hydrogen storage capabilities, rapid hydrogen absorption and release rates, reduced operating temperatures and pressures, and exceptional thermal and cycling durability. Potential future research areas can be methods to enhance the storage capacity, optimize the kinetics of hydrogen adsorption and release, and enhance the stability and durability of activated carbon and carbon nanotubes. Research on the impact of temperature, pressure, and cycling on the performance of MOFs, as well as the development of cost-effective synthesis and production methods and strategies for integrating MOFs into current hydrogen storage and delivery systems, are required. Research on the development of new metal hydrides with improved properties, including the kinetics of hydrogen adsorption and desorption and heat management, can be promising. Innovative materials with improved

mechanical properties, cryogenic insulation technologies, and efficient liquefaction processes should be explored to reduce energy consumption and system costs. An investigation opportunity can be found in the large-scale production of LOHCs, which is essential for the commercial use of liquid hydrogen storage. A research gap remains on cost-effective and environmentally friendly catalysts that need to be developed, and new catalysts should be explored for their stability, selectivity, and recyclability. Another research area is the safety profile of ammonia-based systems, whose synergies with renewable energy sources are to be explored. Possible research opportunities include new materials that can withstand higher pressures and new manufacturing techniques to produce pressure vessels at lower costs and with improved safety features.

A promising avenue lies in the development of novel nanocomposite materials with enhanced hydrogen storage capacities and improved structural integrity. By integrating advanced nanotechnology and material science techniques, researchers can focus on designing multifunctional materials that not only exhibit superior hydrogen storage capabilities but also possess self-healing properties for damage detection and prevention. Exploring the synergy between nanomaterials and smart sensors, along with incorporating advanced predictive modeling and machine learning algorithms, would enable the creation of highly efficient and durable materials capable of withstanding harsh environmental conditions, thereby ensuring the safety and reliability of hydrogen storage systems in various applications, such as fuel cells and energy storage devices.

Major research gaps exist around hydrogen damage mechanisms and kinetics, especially under complex loading conditions. The composite layer design is one of the main factors that directly influence the bearing pressure and fatigue performances of composite hydrogen storage vessels [97]. Advanced multi-modal characterization techniques are required to capture hydrogen-defect interactions across multiple length scales. The utilization of hydrogen patents, publications, and standards as evaluation indicators for the hydrogen technology innovation system was pioneered by Dingran et al. When they analyzed hydrogen standards using bibliometric techniques, the findings also emphasized the significance of hydrogen safety [98]. Chemical spot testing, scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), X-Ray diffraction (XRD) and predictive computational models informed by micromechanical testing need further development and validation to enable damage quantification [99]. Surface peening is a potential method of suppressing hydrogen embrittlement. Wang

Table 10 Damage prevention techniques

Preventive techniques	Mechanisms
Laser Shock Peening	Song and his team examined the resistance of 2205 duplex stainless steel (DSS) to hydrogen embrittlement (HE) after Laser Peening (LP) treatment at varying laser power densities [88]. The findings demonstrate how LP alters the distribution and morphologies of the ferrite and austenitic phases, altering the hydrogen's mode of diffusion and transportation. More tortuous grain boundaries are produced by LP-induced grain refinement, making it harder for hydrogen atoms to pass through them
Hydrogen Recombination Coatings	Under simulated nuclear waste disposal conditions of the Beishan area, the hydrogen permeation and corrosion behavior of Cu-coated carbon steel have been studied [89]. When 5 and 10 μm thick Cu coatings were electrodeposited on the steel, the permeation current (with $-500 \mu\text{A} \cdot \text{cm}^{-2}$ charging current) dropped from 3778 to 141 and 47 nA.cm ⁻² , respectively. S. Fite developed the pulsed-DC magnetron sputtering technique to create CrN layers on gadolinium substrates, demonstrating the potential of CrN coatings [90]. When exposed to hydrogen, the coated samples showed no signs of damage from hydrogen, as determined by SEM, XRD, and GD-OES, while the untreated samples showed signs of a severe hydrogen attack
Hydrogen Diffusion Barriers	Nianwang Ke applied Silicon oxy-carbide as an effective barrier coating to reduce hydrogen permeation rates. SiOC coatings were applied to an X70 pipeline steel substrate using a straightforward low-temperature dipping method. Their effectiveness in preventing hydrogen penetration was investigated using an electrochemical hydrogen permeation experiment, yielding a low hydrogen diffusion coefficient of $8.20 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$, nearly three orders of magnitude lower than the X70 steel's $3.58 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ [91] In this study by Y. Lei, twelve different commercially available coatings were examined, including bisphenol A di glycidyl ether (DGEBA)/polyether amine (D-400) epoxy coatings, poly (vinyl chloride), and crosslinked poly (vinyl alcohol) (PVA) made in house. The hydrogen permeability of films made from two commercial epoxies was 0.40 Barrer and 0.35 Barrer, respectively, indicating their potential as coating materials. A crosslinked poly (vinyl alcohol) coating was found to have a hydrogen permeability of 0.0084 Barrer, indicating that it has the highest potential of all the coatings examined. Then, it was modeled to observe the unsteady-state hydrogen diffusion via coated steel to assess how the coating film affects hydrogen embrittlement reduction [92]
Hydrogen Getter Coatings	This work used the electroless plating approach to deposit Pd–Ag coating on several Zr-based Lave phase alloys, such as ZrV ₂ , Zr _{0.9} Ti _{0.1} V ₂ , and Zr _{1.57} V _{3.6} Fe ₇ . An attempt is made to demonstrate how Pd–Ag coating affects Zr-based alloys' ability to hydrogenate against gaseous contaminants [93]. The ZrH _{1.8} substrate was anodized in a constant current mode to increase the anodic oxide layer's density and hydrogen barrier effectiveness on the ZrH _{1.8} surface [94]
Microstructure Tailoring	By hydrogen pre-charging and then performing slow strain rate tensile tests, the hydrogen embrittlement behaviors of a high-Mn TWIP (twinning-induced plasticity) steel with different grain sizes—from coarse grains to ultra-fine grains—were investigated [95]. The yield strength and tensile strength were not affected by hydrogen charging, according to the findings of the tensile tests; nevertheless, the total elongation decreased in the coarse-grained specimen but remained unchanged in the ultrafine-grained specimen Two austenite-ferrite medium Mn steel samples with significantly distinct phase properties were studied by Binhun Sun. The first one has a high dislocation density ($\sim 10^{14} \text{ m}^{-2}$) in ferrite and an embedded austenite in a ferritic matrix ($\sim 74 \text{ vol}\%$ ferrite). The second one features an embedded ferrite and an austenitic matrix ($\sim 59 \text{ vol}\%$ austenite) with a well-recrystallized microstructure. Because the hydrogen embrittlement microprocesses operating in the two types of microstructures differ fundamentally [96]

et al. showed the numerical analysis of hydrogen transport into PSB1080 high-strength steel after shot peening (SP), focusing on the combined effect of residual compressive stress and SP plastic deformation on diffusion [100]. Detection and monitoring technologies must mature to provide automated, real-time assessment of hydrogen damage progression. With concerted efforts in microstructural engineering guided by mechanics-based models, hydrogen compatibility could be significantly improved for structural materials. Ultimately, cross-disciplinary research fusing experiments, theory, and data science will be key to unlocking the full potential of hydrogen as a sustainable energy vector.

7 Conclusion

The article provides an in-depth comparative analysis of materials and techniques for hydrogen storage, with a particular focus on damage detection and preventive measures. The study critically examines a variety of materials including metal hydrides, carbon-based materials, and composites, highlighting their respective strengths and weaknesses in relation to hydrogen storage. It underscores the importance of early detection of potential hazards through advanced testing methods like Acoustic Emission Testing, Scanning Kelvin Probe, and Digital Image Correlation. The article also explores various preventive strategies such as material coatings, microstructural tailoring, and strict operating protocols to minimize hydrogen-related failures. The conclusion emphasizes the necessity of ongoing research and innovation in understanding material properties, improving storage capacities,

and refining detection methods. This is pivotal for advancing the use of hydrogen as a clean and efficient energy source, aligning with the global pursuit of sustainable and renewable energy solutions.

Conflict of Interest

There are no conflicts of interest. This article does not include research in which human participants were involved. Informed consent not applicable. This article does not include any research in which animal participants were involved.

Data Availability Statement

No data, models, or code were generated or used for this paper.

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