

Research on the progress of hydrogen storage technology and its commercialization path

Zhuokai Yuan*

Shanghai pinghe school, Shanghai, 201206, China

Abstract. Nowadays, global warming and energy scarcity have prompted mankind to develop new energy technologies. Given that new energy generation technologies such as solar and wind energy are subject to climatic conditions with factors such as unstable power generation, the storage process of electrical energy is particularly important. Therefore, the importance of hydrogen energy and its storage technology has received increasing attention from researchers based on the advantages of its wide distribution, high calorific value, and lack of greenhouse gas production. This paper summarizes the current research status of various hydrogen storage technologies, and at the same time assesses and compares the gap between each hydrogen storage technology and the commercialization standard. Based on the analysis, two key routes for the future development of hydrogen storage technologies are proposed.

1. Background of hydrogen energy utilization

1.1 Global warming and the increasing decrease of fossil fuels have prompted the development of new energy.

Nowadays, global warming and the increasing decrease of fossil fuels have prompted mankind to develop new energy technologies. Since the industrial revolution, human consumption of fossil fuels such as coal, oil and natural gas has increased dramatically, leading to a sharp increase in the content of greenhouse gases in the atmosphere. These greenhouse gases, by reducing the heat dissipation from the sun to outer space, have been increasing the temperature of the earth [1]. It is highly likely that human activities are the main cause of global warming over the past 50 years [2]. Global warming can bring about many serious problems, such as the melting of glaciers in the polar regions, rising sea levels, and damage to marine ecosystems [3]. At the same time, there are many problems with fossil energy sources. At the same time, there are many problems with fossil energy sources such as limited reserves and uneven distribution. These problems make it necessary to develop new energy sources to replace fossil energy to meet human demand for energy.

1.2 A variety of new energy technologies have their own advantages and disadvantages and applicable fields

At present, new energy sources such as solar, wind, biomass, and nuclear are widely developed and used by

humans, but these new energy sources all have storage problems. For example, solar and wind energy need to meet the appropriate weather conditions to generate electricity, and direct access to the grid will make the grid fluctuate too much. Therefore, a better solution is to store the electricity generated by new energy sources in storage media, and then re-generate electricity and connect it to the grid at the right time, so as to solve the disadvantages of transient electricity and improve the stability, controllability and security of the grid.

Hydrogen energy, which can be used to store energy and at the right time participate in reactions without polluting, can be used to guarantee grid stability through hydrogen technology.

2. Advantages and applications of hydrogen energy utilization

2.1 Advantages of hydrogen energy

Hydrogen is considered as a promising alternative to fossil energy in the future because it has the following advantages.

(1) Hydrogen is widely distributed, with hydrogen atoms making up 90% of all atoms.

(2) Hydrogen has the highest calorific value and gives off the most heat for equal mass combustion.

(3) The only product of the reaction between hydrogen and oxygen as a fuel is water, which does not produce greenhouse gases and other environmental hazards.

*Corresponding author's e-mail: yuanzhuokai@shphschool.com

2.2 Application of hydrogen energy

The use of hydrogen energy is attracting attention both at home and abroad. The International Hydrogen Energy Commission predicts that the demand for hydrogen energy in 2050 will reach 10 times the current level and account for more than 15% of the end-use energy consumption.

In November 2002, the U.S. Department of Energy issued the "National Hydrogen Energy Roadmap," which provides a comprehensive and systematic description of hydrogen energy technologies such as hydrogen preparation, storage and transportation, conversion and application. In 2017, Japan released the "Basic Strategy for Hydrogen Energy," which makes specific development goals in terms of hydrogen production, storage and utilization.[4] The "China Hydrogen Energy Industry Development Report 2022" [5] points out that



Figure 1 Dawood et al. proposed a four-party model for hydrogen energy research

3. Research status of hydrogen storage development

At present, hydrogen storage is divided into three forms according to the state in which hydrogen is stored, namely gaseous hydrogen storage, liquid hydrogen storage and solid hydrogen storage.

3.1 Gaseous hydrogen storage

Gaseous hydrogen storage is the storage of high-pressure gaseous hydrogen in a specific container by high-pressure compression above the critical temperature of hydrogen. The density of hydrogen is not linearly related to the pressure, especially in the high-pressure stage, where the density of hydrogen is $20\text{kg} \cdot \text{m}^{-3}$ at 30 MPa and $40\text{kg} \cdot \text{m}^{-3}$ at 70 MPa [7]. Its main advantages are low-energy consumption during storage, low cost in a low pressure environment, fast charging and discharging of hydrogen, easy release at room temperature and less influence by the temperature of the working environment.

However, high-pressure gaseous hydrogen storage also has the following disadvantages: low bulk hydrogen storage density, poor safety performance, risk of hydrogen embrittlement in long-term operation, and

China is the world's largest hydrogen producer, with a national output of 33 million tons of hydrogen in 2021, of which about 12 million tons meet industrial standards.

2.3 Hydrogen energy application in various aspects

To realize the large-scale application of hydrogen energy, Dawood et al. proposed a four-party model of hydrogen energy research as shown in the figure, pointing out that the research of hydrogen energy can be divided into four aspects: production, storage, safety and utilization, and to achieve proper production must be perfected in the other three aspects. For transportation, a typical vehicle needs to store hydrogen for more than 500 km, which means it must carry 4-8 kg of hydrogen [6]. Thus, the storage of hydrogen is the most important part to make hydrogen energy available and commercialized.

possible leakage of hydrogen from the gap of the vessel resulting in reduced efficiency.

High-pressure gaseous hydrogen storage is a more mature method of hydrogen storage and is most widely used today. On-board hydrogen storage containers are often multi-layer steel hydrogen storage high pressure containers (Type III hydrogen storage bottle) and full composite plastic hydrogen storage containers (Type IV hydrogen storage bottle) instead of pure steel hydrogen storage bottles (Type I hydrogen storage bottle) and steel lined fiber wound bottles (Type II).

Type I hydrogen storage bottle consists of only one layer of pressure-resistant steel. Type I hydrogen storage bottles appeared around 1870, generally stored at a gas pressure of 15 to 30MPa, with mass storage hydrogen density of about 1%.

The main body of Type II hydrogen storage bottle is made of the same pressure-resistant steel as Type I hydrogen storage bottle, while the outside of the bottle is wrapped with fiber-resin composite material to enhance its pressure resistance. The hydrogen storage density of Type II bottle can reach about 1.5% by mass.

Type III hydrogen storage bottle uses a reinforced structure formed by winding a variety of fibers cured outside the metal liner. The fiber winding layer can be chosen from carbon fiber, aramid fiber and glass fiber, etc. For example, type III hydrogen storage bottle often uses T700 carbon fiber due to its heat resistance, erosion,

high toughness and other characteristics. Common hydrogen storage tools in automobiles are 35MPa & 70MPa type III hydrogen storage bottles, and the hydrogen storage density is generally around 5%.

The IV type hydrogen storage bottle uses a plastic liner with fiber winding to reduce mass and increase strength. The inner liner of the hydrogen bottle mainly plays the role of sealing hydrogen, while the composite

layer mainly plays the role of carrying pressure. The advantages of Type IV hydrogen storage bottles are light weight, high pressure carrying capacity, high hydrogen storage density and long life. For example, Toyota Mirai is equipped with the latest generation of Type IV hydrogen storage bottle, which has a storage density of 5.7wt%, 15% less weight than the previous generation, and a maximum pressure of 70MPa.

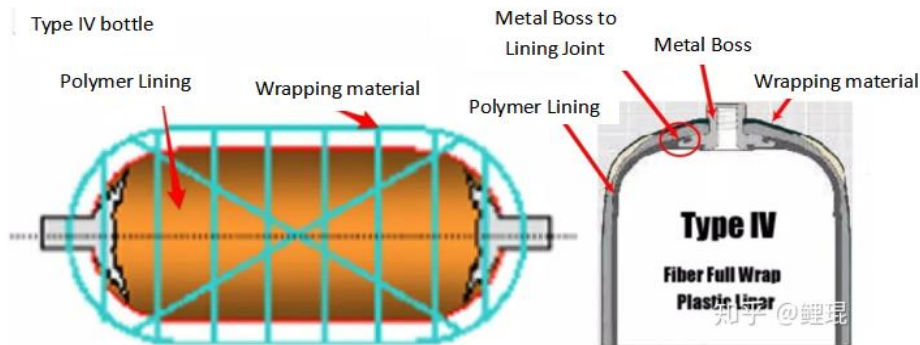


Figure 2 Structural description of type IV hydrogen cylinder

Table 1 Comparison of technical parameters and main materials of various types of hydrogen storage bottles

Types of hydrogen storage bottles	Pressure range (MPa)	Hydrogen storage density (wt.%)	Main Materials
I	15-30	1	Pressure-resistant steel
II	>30	1.5	Pressure-resistant steel + fiber hoops
III	35/70	5	Metal lined with wound fibers
IV	70	5.7	Plastic liner + fiber winding

3.2 Liquid hydrogen storage

Liquid hydrogen storage is divided into two ways: liquefied hydrogen storage and organic liquid hydrogen storage.

3.2.1 Hydrogen storage by liquefied hydrogen

The principle of liquefying hydrogen to store hydrogen is to reduce the pressure of hydrogen to its boiling point to make it liquid for convenient storage. Liquefied hydrogen has the following advantages over other hydrogen storage methods: its relatively high mass hydrogen storage density means that liquid hydrogen per unit mass is the medium that can carry the most hydrogen in the hydrogen carrier.

However, there are problems with liquefied hydrogen: firstly, the container in which it is stored needs to be well insulated to slow down the evaporation losses of the liquid hydrogen. Second, liquefied hydrogen requires a large amount of energy, even 30% of the energy that can be released from the hydrogen itself [8].

At present, liquid hydrogen is mainly used in the aerospace field, but some of them have started to turn to commercial use for large-scale transportation. In

September 2021, the first set of hydrogen liquefaction system independently developed by China has been successfully commissioned with a production capacity of tons, and has achieved continuous and stable production. In the civil sector, such as automobiles, the US company General Motors has recently launched the "Hydrogen3" car. The liquid hydrogen tank weighs 90 kg and can store 68 L liquid hydrogen. The weight hydrogen storage density is 5.1 % and the volume hydrogen storage density is 36.6 kg / m³. The hydrogen storage density of liquid hydrogen technology for civil use is close to 5wt. % of the commercial standard, but the cooling of liquid hydrogen, the high energy cost of production, the preservation of liquid hydrogen at room temperature and the vaporization of liquid hydrogen are still problems that cannot be ignored.

3.2.2 Hydrogen storage in organic liquid

Organic liquid hydrogen storage refers to the use of reversible hydrogenation and dehydrogenation of unsaturated organic liquids to store hydrogen. In recent years, the study of organic liquid hydrogen storage media has been carried out on many liquids such as cyclohexane, benzene, carbazole and other media. The

organic liquid hydrogen storage works in the following way: after hydrogenation of hydrogen carrier, through the pipeline, the storage equipment stores and transports the hydrogenated hydride. After being transported to a dehydrogenation unit and catalytically dehydrogenated, hydrogen is released to supply the user and the hydrogen medium can be catalytically hydrogenated again by piping back to the hydrogenation station [9].

There are many advantages of organic liquid hydrogen storage. First of all, the hydrogen storage density of organic liquid is high, for example, the mass hydrogen storage density of cyclohexane is up to 7.19%, and the theoretical mass hydrogen storage density of ethyl carbazole ($C_{14}H_{13}N$) reaches 5.8%, which is much higher than the hydrogen storage density of metal alloy hydrogen storage. Cyclohexane and methylcyclohexane are liquid at room temperature and can be transported not only convenient but also over long distances and in large quantities using pipeline equipment. In addition, the dehydrogenation and hydrogenation reactions of organic hydrogen storage media are reversible, so the media can be recycled to save costs.

Hydrogen energy demonstration projects using organic liquid hydrogen storage have already started in Europe. 2020 Germany has proposed the GETE2 polarization and is committed to gradually building a hydrogen energy infrastructure covering the whole of Germany, in which a storage and transportation system for hydrogen storage with organic liquids is planned. Hydrogenous Technologies (HT) will build the world's largest hydrogen storage plant in Dormagen, Germany's largest industrial park, using dibenzyltoluene as a liquid hydrogen storage carrier and putting it into operation in 2023.

However, there are many problems with organic liquid storage: the temperature is still relatively high, for example, the temperature required for cyclohexane dehydrogenation is greater than 250 degrees Celsius, which is difficult to use in fuel cells, and the high temperature required for the reaction can damage the catalyst structure and lead to deactivation, while the volatile impurity components produced by the hydrogen release process can poison the fuel cell.

3.3 Solid-state hydrogen storage technology

Solid-state hydrogen storage technology mainly includes physical solid-state hydrogen storage technology and chemical solid-state hydrogen storage technology.

3.3.1 Physical solid state hydrogen storage technology

The principle of hydrogen storage in physical hydrogen storage materials is that hydrogen molecules are adsorbed on the surface of solid materials by van der Waals forces, and storage is achieved without the destruction of hydrogen molecules.

Physical hydrogen storage materials include inorganic porous materials and metal-organic skeletal compounds. Inorganic porous materials are inorganic

porous materials with nano-pore channels, such as zeolite, seafoam, etc. However, these inorganic porous materials are often not ideal for hydrogen storage because of their large masses. A study on ZSM-5 zeolite by Nijikamp et al [10], showed that its hydrogen storage capacity was only 0.7 wt% at $-196^{\circ}C$ and 100 kPa. Metal-organic skeletal compounds are a class of materials with great specific surface area. However, the mass density of hydrogen storage at acceptable temperatures is very low for this class of materials.

Physical hydrogen storage materials have excellent kinetic properties of hydrogen uptake and release with good cyclic stability, but the process of hydrogen uptake is due to van der Waals forces acting between hydrogen molecules and the material and the interaction forces are weak, so physical hydrogen storage must be at low temperature and relatively low hydrogen storage capacity in order to achieve a suitable hydrogen storage density. For example, the composite material of carbon nanotubes prepared by Yang has a maximum hydrogen storage capacity of 1.52wt% at 77k, but it decreases to a maximum of 0.6wt% at 298k [11]. Therefore, physical hydrogen storage technology is difficult to adapt to fuel cells because the reaction in fuel cells often occurs at about 393k, and the ambient temperature is too high, resulting in extremely low hydrogen storage density of physical hydrogen storage materials.

3.3.2 Chemical solid-state hydrogen storage technology

Chemical hydrogen storage technologies are mainly divided into two categories: metal hydrides and coordination hydrides.

(1) Metal hydride

Metals or alloys can react with hydrogen to form metal hydrides, which in turn can release hydrogen through the reaction when heated for the purpose of storing hydrogen. Metal hydride is a direct connection between hydrogen atoms and metal atoms, and the particles are connected in the form of covalent bonds to store hydrogen.

Metal hydrides mainly include $AB_5(LaNi_5)$, $AB(TiFe)$, $AB_2(TiMn_2)$, $A_2B(Mg_2Ni)$, $BCC(TiCrMnV)$ and other types. These hydrides have many advantages: their excellent kinetic properties of hydrogen absorption and discharge, reversibility of the reaction, and suitable temperature and pressure of hydrogen discharge, so they have been more studied. However, the practical application of metal alloy hydrides (e.g. $LaNi_5H_6$) is limited by the fact that the hydrogen storage density of these hydrides tends to be low, around 2wt%, much lower than the 5.5wt% given by the US Department of Energy, making it difficult to use them commercially. For light metal hydrides, their hydrogen storage density is relatively high, for example, MgH_2 has a theoretical hydrogen storage capacity of 7.6wt% [12], and the reversibility of the reaction is high. However, due to the small atomic radius of its light metals, the strong bonding of metal atoms with hydrogen atoms leads to its high thermodynamic stability and high hydrogen release

temperature. Its rate of hydrogenation and dehydrogenation in the reaction with hydrogen is very slow and its activation energy is relatively high.

There are many ways in which metal hydrides can be modified. The addition of a catalyst can make the reaction thermodynamically less stable to react at low temperatures. Catalysts can accelerate the reaction by promoting the dissociation of hydrogen molecules and the assembly of hydrogen atoms with the substrate. Common catalysts include transition metals and transition metal oxides, non-metals, etc. Zhang et al. prepared iron nanocatalysts, which significantly reduced the reaction temperature from 375.3 °C to 222.6 °C after mixing with MgH₂, and the activation energy of the reaction was also significantly reduced. For MgH₂, the addition of other elements such as Si to form an alloy can also reduce the hydrogen absorption and release temperature and the enthalpy change of the reaction, but the corresponding hydrogen storage will be lower. For example, the reaction enthalpy change of Mg₂Ni alloy formed by adding Ni is reduced by 12 KJ/mol, but its hydrogen storage density is reduced to 3.8 wt%.

(2) Compound hydrides

In metal coordination compounds, the hydrogen atom and the central atom are covalently bonded to form a complex anion, and this complex anion forms a structure of coordination hydride with the metal cation. The molecular formula of metal coordination hydrides can be expressed as AxMe_yH_z, where A is often an alkali metal or alkaline earth element and Me usually refers to elements such as B, Al or N [13]. Hydrogen gas can be stored in the coordination bonds of such coordination compounds and is released when conditions are right.

Metal coordination hydrides have very high weight hydrogen storage density and strong hydrogen release performance, for example, Ye et al. investigated that lithium borohydride has a weight hydrogen capacity of 18.5 wt% and can decompose to LiH and B under heating, releasing 13.9 wt% of hydrogen [8]. However, the reaction temperature of coordination hydrides is high, for example, the decomposition of LiBH₄ in the former reaction generally requires a temperature of more than 400 degrees Celsius, which is not suitable for commercial use. Due to the relative stability of the B-B bond during hydrogenation, it also needs to be hydrogenated at 600°C and >35 MPa hydrogen pressure, which limits the hydrogen absorption and discharge performance of LiBH₄ and makes it difficult to obtain high theoretical reversible hydrogen storage capacity of LiBH₄. Scientists have made many researches to make metal coordination hydride modified destabilization, for example, Kang mixed lithium borohydride with aluminum by ball milling method with a molar ratio of 2:1, and the performance of hydrogen uptake and release was greatly improved with a small amount of TiF₃ as catalyst [14].

3.4 Methods to improve hydrogen storage performance

3.4.1 Modification of substances by nanotechnology

Nanomaterials can achieve a very high surface area through the arrangement of atoms at the nanoscale and therefore can bind more hydrogen molecules by van der Waals forces to achieve a higher mass hydrogen storage density. For carbon nanomaterials, the higher the surface area, the more hydrogen can be stored, and experiments have shown that the hydrogen storage density is only 1.5 wt.% at a surface area of 1000 m²g⁻¹ [15], while it reaches 7% wt at a surface area of 3200 m²g⁻¹. Experimenters have used zeolites with silicon as inorganic substrates to prepare hydrogen-enhancing adsorbents. Walker et al synthesized zeolite-like microporous carbon using EMC-2 zeolite as a template. Under the conditions of 2MPa and 469K, the total hydrogen storage capacity of carbon with a high degree of zeolite-like order increased to 6wt. % [16].

For chemical hydrogen storage, the atoms of nanomaterials will be more reactive. The tendency is to react with more hydrogen to form bonds. These properties also have an effect on the kinetic properties of the nanomaterials, allowing the reaction to proceed more easily and at lower temperatures. Activated carbons produced with KOH as an activator have a high surface area suitable for hydrogen adsorption. Wang proposed a reaction method with physical activation with CO₂ followed by chemical activation with KOH [17]. At 469 K and 2 MPaH₂, the hydrogen adsorption of activated carbons can reach 7.08 wt.%. Jean et al. prepared nano-loaded PMMA with a diameter of 4 nm, which is air stable and can have a reversible hydrogen storage mass of 4 wt% at 200 degrees C.

3.4.2 Addition of dopants

Research on the addition of functional groups or dopants to promote the assembly of hydrogen with materials has progressed rapidly in recent years. The use of light atoms such as fluorine, boron, etc. can lead to electron deficiencies and increase the polarity of the carbon framework to enhance hydrogen storage capacity. For example, multi-walled carbon nanotubes with a hydrogen storage density of 1.2 wt.% have a reversible hydrogen storage capacity of 3.9 wt.% at 77k after doping with lithium [18]. Also, doping with metal atoms can increase the carbon reversible storage capacity. The material undergoes a spillover effect at room temperature, allowing hydrogen molecules to unwind from the carbon framework.

Graphene is an excellent single-layer carbon nanostructure, and research on graphene has been carried out for a long time but it has now been shown that single-layer graphene cannot be used as a stand-alone hydrogen storage material [19]. The addition of H₂ to multilayer graphene can improve the hydrogen storage capacity of the material. For a graphene layer with a gap of 6 Å, a layer of hydrogen molecules can be added in between to increase the hydrogen storage density by at least 2% wt. The addition of transition metal and alkali

metal catalysts can also increase the adsorption capacity of graphene. Graphene materials with the addition of titanium atoms can achieve a hydrogen storage density of 4.9 wt%, while graphene with the addition of lighter metals also shows a significant increase in hydrogen storage capacity [20].

4. Prospects for the commercialization of each hydrogen storage technology

Currently, commercial hydrogen storage technologies must meet three conditions simultaneously.

(1) Relatively high hydrogen storage density such as greater than 5.5 wt%.

(2) A low reaction temperature such as 393K to suit the fuel cell reaction.

(3) Stable and reversible hydrogen absorption and discharge performance.

A comparison of the above technologies in terms of hydrogen storage density and reaction temperature is shown in the figure below.

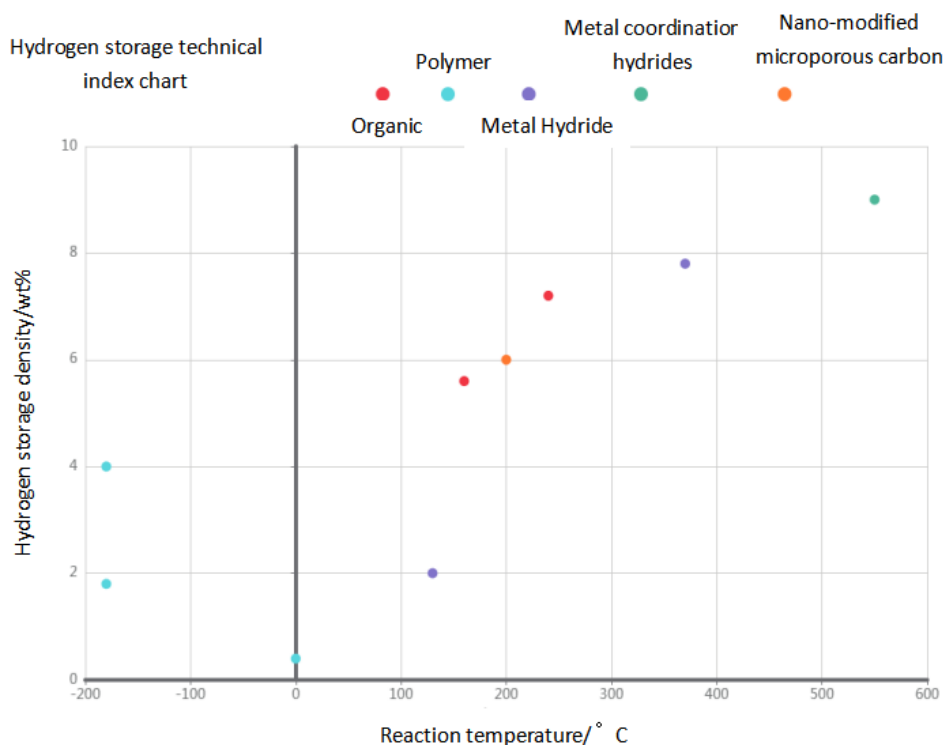


Figure 3 Comparison chart of each technical index

It can be seen that the current hydrogen storage methods are not able to satisfy the above three conditions at the same time. Metal coordination hydrides such as lithium borohydride, organic liquid hydrogen storage materials such as dibenzyltoluene, and physisorption materials such as nano-modified zeolite-like microporous carbon are close to commercial standards in terms of reaction temperature and hydrogen storage density with Type IV hydrogen storage bottles.

However, each of these techniques still has its own problems. Metal coordination hydrides have poor kinetic properties for hydrogen uptake and release, and require effective catalysts to facilitate the reaction at room temperature. Organic liquid hydrogen storage materials can release toxic substances when reacting, which can damage the reactor and human body. Physical adsorption materials require low temperatures for better hydrogen storage, while high-pressure hydrogen storage cylinders have large masses and are difficult to use in vehicles and other locations.

The development of suitable hydrogen storage materials to achieve reversible reactions, suitable reaction temperatures and sufficiently high hydrogen storage density are three factors that still face technical

challenges and constrain the commercialization of hydrogen energy today. Although scientists have made some progress in the research of various hydrogen storage materials, the mainstream hydrogen storage materials have different degrees of applicability problems to hinder their commercialization. Future research in hydrogen storage technology should focus on the weaknesses of various hydrogen storage materials and find ways to modify them to improve their performance or to find more suitable materials for hydrogen storage.

5. Summary and Prospect

At present, the background of global warming and energy scarcity has prompted mankind to develop new energy sources. Since solar and wind energy are subject to climatic conditions and problems such as unstable power generation, the storage of electrical energy is particularly important. Therefore, based on the advantages of hydrogen energy itself, the importance of hydrogen energy and the storage technology of hydrogen

energy has received more and more attention from researchers.

This paper summarizes the current research status of various hydrogen storage technologies. Hydrogen storage is divided into three forms according to the state in which hydrogen is stored, namely gaseous, liquid and solid hydrogen storage, and analyzes the improvement of hydrogen storage performance by nanotechnology and the addition of dopant technology. At the same time, the gap between each hydrogen storage technology and the commercialization standard is evaluated and compared in this paper.

However, each hydrogen storage technology has not yet fully reached the commercialization standard. Looking into the future, hydrogen storage carriers with high reversibility, high hydrogen storage density and low reaction temperature are the future trend of hydrogen storage materials development. The focus of future research on hydrogen storage technology should focus on the weaknesses of various hydrogen storage materials and find ways to modify them to improve their performance, or find more suitable materials for hydrogen storage, both of which will be the way to achieve the development of hydrogen energy.

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