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Physical and Chemical Adsorption of Hydrogen Storage Nanoporous Materials: Characteristics, Modifications and Applications

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Abstract:

This article reviews the application and research progress of nanoporous materials in the field of hydrogen storage. Hydrogen energy is a type of clean energy source, hydrogen energy's efficient storage is the key to achieving hydrogen economy. The article first outlines the main kinds of adsorption of hydrogen storage technology, covering different methods such as physical adsorption technology, chemical adsorption technology and composite material adsorption technology. Subsequently, these technologies are further categorized and the materials that utilize these technologies are introduced, especially the performance characteristics and practical application of physical adsorption hydrogen storage materials (such as some carbon-based nanomaterials) and chemical porous materials (such as metal hydrides) were analyzed. It is especially pointed out in the article that the hydrogen storage capacity of nanomaterials can experience a significant improvement through microstructure optimization design and collaborative construction of composite materials. Finally, it is emphasized that reducing the working temperature and optimizing the pore size distribution are important ways to change the performance of hydrogen storage, and the future development prospects of the hydrogen economy are forecasted. Meanwhile, the practical application cases of physical adsorption materials, chemical adsorption materials and various hydrogen storage materials that have been optimized for treatment are listed in detail.

Keywords: Material; adsorption; nanoparticles; hydrogen storage; organic framework.

1. Introduction

The development of new energy has become a mainstream trend around the world. As an important representative of clean energy, hydrogen energy technology is attracting the attention of scientific research institutions in various countries. At present, the hydrogen production process has become more and more perfect, and the market demand for hydrogen continues to rise, which makes hydrogen storage technology a key link to restrain the evolution of the hydrogen energy industry. Especially in the field of on-board hydrogen storage, various hydrogen storage devices have been applied in practice, but the technical requirements in this field are extremely strict. At present, hydrogen storage technology has become a major technical barrier that hinders the commercialization and promotion of hydrogen fuel cell vehicles [1]. Safety and hydrogen storage efficiency have become key problems that must be broken through in the current research and development of the hydrogen energy industry. Traditional hydrogen storage materials include metal hydrides and high-pressure gases, which have obvious defects in weight, volume and safety. Nanoscale hydrogen storage materials optimize the storage kinetic characteristics and upper capacity of hydrogen by reducing particle size and greatly improving the specific surface area and reactivity. In particular, porous nanomaterials, such as metal organic skeleton compounds and carbon nanotubes, can achieve efficient storage of hydrogen molecules through physical adsorption with their super-large specific surface area and special pore structure.At present, hydrogen storage technology mainly covers different methods such as low-temperature liquid, organic liquid and solid metal hydrogen storage [2]. These nanomaterials can effectively adsorb hydrogen molecules by building porous structures, which is a kind of solid hydrogen storage technology. When hydrogen comes into contact with the adsorbed material, it will adhere to the surface of the material or enter the internal pores to achieve the purpose of hydrogen storage. When hydrogen needs to be released, you only need to adjust the temperature or pressure to detach the hydrogen from the adsorption material.

Materials commonly used for adsorption of hydrogen storage include metal hydrides, carbon-based materials and metal organic skeleton materials. Porous adsorption technology can classified into two types: physical adsorption and chemical adsorption. As the current mainstream hydrogen storage method, how to improve hydrogen storage performance from both physical and chemical dimensions has become an important research direction in this field.

2. Physical adsorption materials

The basic mechanism of hydrogen storage in physical adsorption materials comes from its huge specific surface area and micro-hole structure characteristics. Hydrogen molecules are attracted to the surface or pores of the material by the Van der Waals force, forming a physical adsorption state. This hydrogen storage method can be completed under mild conditions without chemical reactions. It has strong reversibility and high selectivity, and is very suitable for efficient storage and controlled release of hydrogen energy. Differences in pore size distribution can significantly affect the hydrogen storage structure and performance. Metal organic frame materials (MOFs) and carbon nanotubes(CNT) are widely used due to their excellent physical adsorption properties.

2.1 Carbon Nanomaterials

Carbon-based nanomaterials: Such materials include CNT, graphene and their derivatives, and have special physical properties that make them ideal hydrogen storage media. The primary advantage is that it has an ultra-high specific surface area, which supplys a lot of adsorption sites to hydrogen molecules, thus significantly improving the hydrogen storage capacity. Secondly, the excellent electrical conductivity and thermal stability of this material help to maintain structural stability during hydrogen storage and improve the adsorption efficiency of hydrogen molecules [3]. In addition, the nanometer-scale size and porous structural characteristics further accelerate the diffusion rate and adsorption dynamics of hydrogen molecules, enabling hydrogen molecules to enter and exit the material more efficiently, and optimizing the properties of hydrogen storage and hydrogen release [4]. In 2002, the Simonya research team systematically evaluated carbon nanotubes as hydrogen storage materials, but most of the samples failed to reach the 6 wt% standard made by the U.S. Department of Energy for transportation [5]. Based on this, materials scientists have proposed through in-depth research that parameter regulation is the core way to optimize the hydrogen storage performance of nanomaterials, which is mainly achieved through three mechanisms: surface chemical functionalization, atomic doping and defect site engineering. Through chemical functionalization, silicon carbon nanotubes (SiC-NT) have been developed, in which silicon atoms and carbon atoms are replaced by point charges. More than half of the charge continues to transfer from silicon to carbon, which significantly improves the practical application efficiency [6]. In the study of embedding heteroatoms into the skeleton of carbon-based materials, oxygen-rich microporous carbon materials were successfully prepared. Its specific surface

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area is as high as about 3,800 m^2/g, and it shows excellent weight hydrogen storage performance at low temperature conditions of 77 K.

2.2 Metal-organic framework materials

MOF is a porous crystal material formed by the combination of metal ions or ion clusters with organic ligands through coordination bonds. The core mechanism lies in the synergy between the metal center and the organic ligand to build a highly ordered porous network structure to achieve efficient capture and storage of gas molecules. This kind of material has the outstanding characteristics of ultra-high specific surface area, adjustable aperture size and pore configuration, rich chemical functional characteristics, and excellent thermal and chemical stability. These characteristics make MOFs show a broad application prospect in gas storage, separation and purification, catalytic transformation and other fields. After specific modification treatment, this kind of material can capture hydrogen molecules through chemical adsorption and become a promising candidate for hydrogen storage at r.m.t [7]. In terms of nanoscale research, some MOF materials can control particle size within the range of 1-100 nm through precise regulation of synthesis processes, which are typical nanomaterials [8]. This kind of material mainly relies on the action of Van der Waals force to achieve the physical adsorption of hydrogen with its huge specific surface area and accurately adjustable aperture. Table 1 and Figure 1 show some experimental data on the specific surface area, porosity and hydrogen storage performance of different materials respectively.

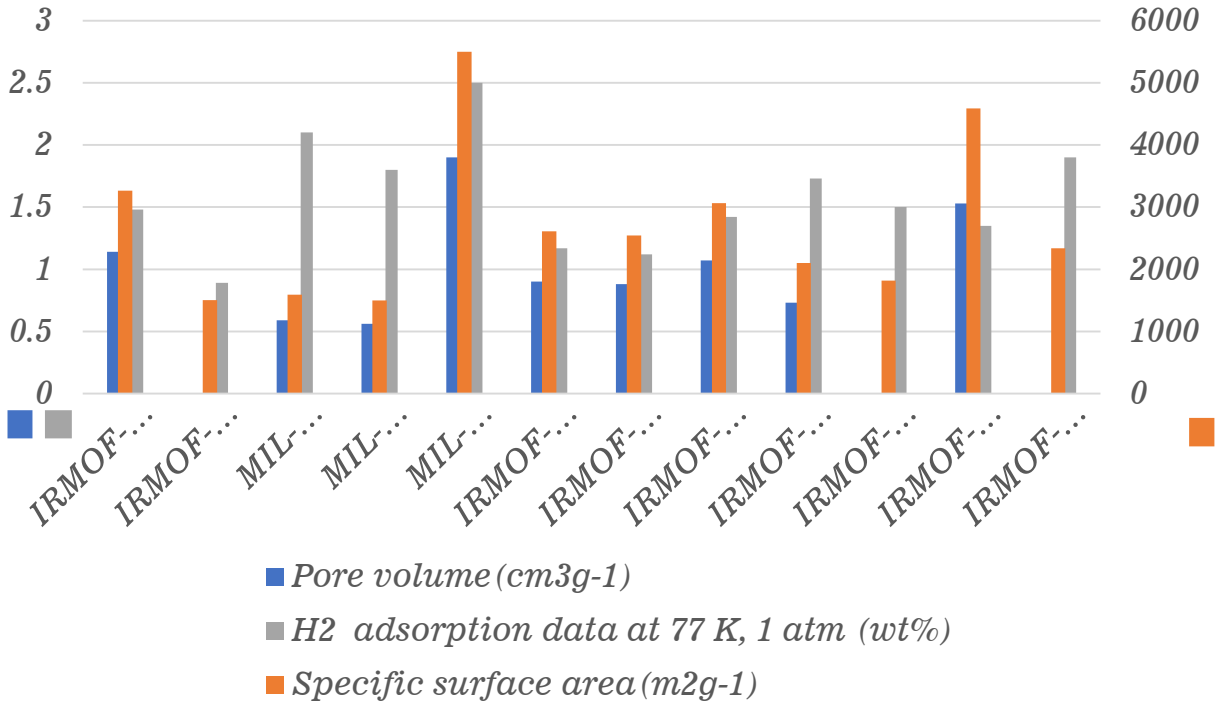

Fig. 1 The comparison of hydrogen storage capacity based on the pore size volume and specific surface area of different substances

Table 1. The pore volume, specific surface area and hydrogen adsorption data for MOFs

Material	Pore volume(cm^3g^{-1})	Specific surface area(m^2g^{-1})	H_2 adsorption data at 77 K, 1 atm (wt%)
IRMOF-6, $Zn_4O(cbbdc)_3$ [8, 9]	1.14	3263	1.48
IRMOF-18, $Zn_4O(tmbdc)_3$ [10]	——	1501	0.89
MIL-53(Al), Al(OH)(bdc) [8, 11]	0.59	1590	2.1
MIL-53(Cr), Cr(OH)(bdc) [8, 11]	0.56	1500	1.8
MIL-101, $Cr_3OF(bdc)$ [8]	1.9	5500	2.5

IRMOF-9, $Zn_4O(bpdc)_3$ [9]	0.9	2613	1.17
IRMOF-2, $Zn_4O(bbdc)_2$ [12]	0.88	2544	1.12
IRMOF-3, $Zn_4O(abdc)_3$ [9]	1.07	3062	1.42
IRMOF-13, $Zn_4O(pydc)_4$ [9]	0.73	2100	1.73
IRMOF-8, $Zn_4O(ndc)_3$ [9, 12]	——	1818	1.5
IRMOF-20, $Zn_4O(ttdc)_3$ [9, 12]	1.53	4590	1.35
IRMOF-11, $Zn_4O(hpdc)_3$ [9, 12]	——	2340	1.9

The key role of specific surface area can be clearly seen from the experimental data: to achieve high hydrogen storage capacity, the material must have a large specific surface area (more than 3,000 m^2/g), but at the same time, it needs to be optimized and adjusted with the aperture size [5]. The aperture parameters also have a decisive impact: when the aperture is maintained at about 0.6 nm, the van der Waals force is maximized through the superposition of the potential field. In a low-pressure environment, materials with smaller apertures tend to show better performance [1]. Take the MIL-101 ($Cr_3OF(bdc)$) shown in the bar chart as an example, which obtains the highest hydrogen storage by virtue of the largest specific surface area. The second-ranked hydrogen storage performance in the figure is MIL-53(Al)(Al(OH)(bdc)). Although its specific surface area is not as good as the former, its pore volume is closer to the best range of 0.6-1 nm. It can be seen that the pore volume parameters also have an important impact on the performance of the material. These rules provide a clear theoretical basis and optimization path for the design of new high-efficiency MOFs hydrogen storage materials [13].

Although metal organic frame materials show excellent hydrogen storage potential at r.m.t, the research on their practical application still needs to be explored in depth. According to the current research results, researchers are committed to adjusting the aperture size and pore structure, while increasing the distribution density of low-cost transition metal sites, in order to develop new MOFs materials with suitable hydrogen binding anthalpy and optimal hydrogen storage properties at room temperature.

2.3 Covalent Organic Frameworks

In the process of exploring MOFs, researchers have a strong interest in other adsorption materials in the area of hydrogen storage, especially covalent organic frameworks (COF). The reason why this kind of material is suitable for hydrogen storage is mainly due to its excellent specific surface area characteristics and rich pore structure. It is worth noting that unlike the metal frame containing heavy metal elements, COF is composed entirely of lightweight elements, and this unique composition enables it to form a special material with extremely low density. This is an improvement for MOF materials. Finally, although experimental and computational results indicate that covalent organic framework materials have potential hydrogen storage capabilities, their practical applications have not yet been realized [5]. Despite still facing issues such as low temperature and high pressure dependence, insufficient stability, and difficulty in achieving large-scale production, the technology of COFs in gas separation has become more mature. For instance, the 10-meter-long COF-LZU1 membrane has now been commercialized and is used for H_2/CH_4 separation [14].

3. Chemical adsorption materials

Chemical porous hydrogen storage involves using chemical bonding between the material and hydrogen to store hydrogen. These materials include metal hydrides, MXene materials and single-atom catalyst materials, they are suitable for high-temperature environments, enhancing storage density and efficiency.

Metal hydrides are one of the most important categories of chemically porous hydrogen storage materials. The researchers combined the porous hydrogen storage characteristics of AlH_3, $NaBH_4$ and MgH_2 with the advantages of nano-hydrogen storage, and crushed the particles of MgH_2 and other metal hydrides into nanocrystalline or microphase scales through mechanical grinding. This reduces the desorption activation energy and further optimizes their hydrogen storage performance. Mechanical grinding of metal hydrides doped with nano-catalysts is carried out, such as altering the model of MgH_2 nano-clusters and the distribution of nano-catalysts at the active sites, in order to achieve efficient on-board hydrogen storage [4]. Aristizabal et al. have demonstrated that nanotube materials can absorb 5.5 wt% of hydrogen at room temperature [15].

Meanwhile, metal hydrides are also applied in the aerospace field. NASA uses the hydrolysis system of $NaBH_4$ (with catalyst: Co-B nanoparticles) as a space propulsion agent.It can produce hydrogen at an instantaneous rate of 5 L/min per gram at r.m.t, with a high specific impulse advantage (280 s), enabling it to replace traditional hydrazine fuel [16].

MXene materials are formed through partial etching with hydrofluoric acid to create a multi-layer structure, and they utilize highly active surface sites to achieve rapid

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and reversible hydrogen storage.The single-atom catalyst material is a system of Ca single atoms supported on dual-doped graphene. Through charge induction, it enhances the hydrogen adsorption heat (-22.7 to -43.6 kJ/mol), and its capacity exceeds that of traditional carbon-based materials. These two materials both have great potential for energy storage in the future [17].

4. Optimization methods

With the advancement of technology, people are no longer satisfied with the existing hydrogen storage materials. Therefore, they began to optimize these materials. The types of optimization mainly include: structural design, composite material co-design and surface treatment.

4.1 Structural Design

Structural design is divided into two major categories:
Materials suitable for optimization through pore regulation mainly include categories with adjustable pore size and pore structure, such as MOFs and COFs. Such substances have significant application potential due to their designable porosity characteristics. These materials can have their pore size and distribution precisely controlled through chemical synthesis methods, thereby optimizing their adsorption and separation performance for specific molecules. For example, MOFs can adjust their pore diameters by selecting different organic ligands and metal nodes to achieve selective adsorption of molecules of different sizes.
The materials suitable for optimization using Dimension Engineering mainly include those that can improve performance by changing dimensions, such as nanowires and nanosheets. By controlling the growth conditions and post-treatment processes of these materials, their dimensions can be precisely regulated, thereby optimizing their electrical, optical, and catalytic properties.

4.1.1 Pore Control

Pore regulation overcomes the hydrogen storage bottleneck through three major approaches: enhancing adsorption energy through quantum confinement, accelerating mass transfer through graded pores, and activating sites through surface chemical modification. Based on pore size, they can be classified as ultra-small pores (<1 nm), slit pores (in layered materials), mesopores (2-50 nm), and macropores (>50 nm). Nanomaterials with different pore sizes have different functions: Ultra-microscopic pores have strong adsorption capacity; slit pores are often found in activated carbon and MOFs; mesopores provide a rapid diffusion path for H_2 molecules, reducing mass transfer resistance and increasing diffusion rate; large pores act as a buffer layer to prevent micro-pores from clogging, improving cycling stability, such as graphene aerogel [18, 19]. Because the diameter of a hydrogen molecule is near 0.29 nm [20], and the author summarizes the relationship between pore sizes and their performances are shown in Table 2:

Table 2. The influence of different pore sizes on the hydrogen storage mechanism and performance

Range	Hydrogen storage mechanism	Advantage	Boundedness
<0.6 nm	Quantum sieving effect	Ultra-high adsorption enthalpy (> 10 kJ/mol) [21]	The diffusion resistance is high and the kinetics is slow.
0.6–1.0 nm	Van der Waals force superposition	The hydrogen density is close to that of a liquid (> 60 g/L) [21]	Limited specific surface area
1.0–2.0 nm	High specific surface area adsorption	High theoretical capacity	The adsorption capacity at room temperature is low (< 1 wt%) [22]
>2.0 nm	Weak physical adsorption	Easy to desorb, with good recyclability	The adsorption capacity has sharply decreased [22]

For hydrogen storage due to the quantum confinement effect and the superposition of van der Waals forces, the optimal aperture value ranges from 0.5 to 0.7 nm [23]. Within this range, the highest volumetric/mass hydrogen storage density can be achieved at low temperature (77K).

4.1.2 Dimension Engineering

Dimension engineering is a technology that optimizes the performance of materials by precisely controlling their dimensions and structures. In applications such as energy storage and hydrogen production, dimension control is widely employed in the design and preparation of high-performance nanomaterials to achieve higher energy density and faster reaction kinetics. The research dimension engineering involves the following methods. Utilizing 2D nanosheets (such as MoS2): exposing more edge active sites. And using 3D frameworks (such as COF-102): increasing the density of adsorption sites per unit volume [24].

4.2 Composite Material Co-design

The advantages and disadvantages of composite materials coexist. They can increase the hydrogen storage capacity, optimize the kinetics (reducing the hydrogen diffusion path from the micrometer scale to the nanometer scale), and enhance the cycling stability. However, composite materials also encounter several limitations such as high complexity in production, performance degradation (short cycle life), and high production costs [1, 2, 6, 7].

The change of hydrogen storage performance also involves the composite application of metal organic frame materials and carbon-based materials. Researchers have made remarkable progress by combining the high specific surface area characteristics of MOFs with the electrical conductivity of carbon materials. Relevant literature shows that Yang et al. have developed an acidic multi-walled carbon nanotube composite system to improve material stability [25]. While the Yoo team proposed a method of adding surfactants to the drying process. These surfactants will be absorbed by MOF-5 materials, making their surface hydrophobic, thus effectively improving the water resistance of the material.

Furthermore, some researchers have incorporated metal hydrides/nanoparticles and utilized the catalytic hydrogenation reaction kinetics of the nanoparticles. The experiments also achieved satisfactory results.

Recently, the YongGao research team found through experimental verification that the hydrogen storage performance of the double-doped system is significantly better than the performance of the single-doped system, and its hydrogen storage capacity has exceeded the performance limit of existing carbon-based hydrogen storage materials. The team used a systematic screening method to finally determine the optimal hydrogen storage configuration of calcium monoatoms on the double-doped graphene carrier [26].

In summary, materials with large open pore volume and concentrated pore distribution are the key elements to achieve efficient hydrogen storage performance [22].

4.3 Surface Treatment

Surface treatment technology refers to changing the characteristics of the surface layer of a material by physical, chemical or biological means to improve its performance or give new functions. In terms of improving hydrogen storage capacity, the core of surface treatment is to change the chemical composition, physical structure or surface energy state of the surface of the material, so as to optimize its adsorption and storage efficiency of hydrogen.

For example, the deposit of precious metal nanoparticles (such as platinum, palladium, etc.) on the surface of metal oxides can significantly improve the adsorption and desorption rate of hydrogen, thus improve the capacity of hydrogen storage. This surface modification method utilizes the high catalytic activity of precious metals and can promote the process of hydrogen molecular dissociation and adsorption [7].

5. Applications

For hydrogen storage, physical adsorption and chemical adsorption each show unique application value and technical advantages. The applications of physical adsorption are mainly based on the intermolecular van der Wals force, which is especially suitable for hydrogen energy storage in a low-temperature environment. Take the activated metal organic skeleton material as an example. With its huge specific surface area and rich porous structural characteristics, it can efficiently complete the hydrogen adsorption process [18].

Chemical adsorption fixes hydrogen molecules by establishing chemical bonds, which allows it to show excellent hydrogen storage performance under high temperature conditions. Metal hydrides are a typical example of chemical adsorption materials, which can react with hydrogen at high temperatures to form stable hydrides, achieving high-density hydrogen storage [7].

However, both physical adsorption and chemical adsorption have their own limitations. To overcome the limitations, researchers are dedicated to developing optimized materials. This kind of material combines the dual advantages of physical adsorption and chemical adsorption. Through specific structural design and synergy between composite materials, it significantly improves the capacity and efficiency of hydrogen storage. For example, some studies enhance the chemical adsorption capacity by introducing metal nanoparticles or modifying the surface in MOFs, while retaining the high specific surface area advantage of physical adsorption [17].

In addition, some research focuses on developing new porous materials, such as COFs and porous organic polymers (POPs). Through fine molecular construction and synthesis processes, these substances can provide more effective adsorption sites while maintaining a large specific surface area, thus significantly improving hydrogen storage efficiency.

6. Conclusion

As concerns of people about atmospheric changes and non-sustainable energy consumption continue to grow, compared to other energy sources, the visualized hydrogen economy remains a viable option. This paper systematically discusses the characteristics and applications of a variety of solid hydrogen storage materials. To achieve the hydrogen energy economy, hydrogen storage technology remains a key research area. Only by solving the problem

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of hydrogen storage can hydrogen energy be more widely applied in all aspects of life. This paper deeply discusses the research progress and practical application of nanomaterials in the area of hydrogen storage with their porous properties and adsorption properties. It concludes that a low operating temperature is the ideal choice for hydrogen absorption by porous materials.However, to improve the system performance, relevant scholars have studied adjusting the pore size and volume of nano-porous materials, or developing composite materials. But all these actions are aimed at developing better hydrogen storage materials, which can enable clean energy represented by hydrogen to replace traditional energy, thereby realizing the vision of environmental optimization and protection.

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