

Review of Carbon High Storage Efficient Material in Hydrogen Storage for New Energy Vehicles

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Abstract. With the rapid development of new energy vehicles, research on efficient hydrogen storage materials has become a top priority. This article reviews the hydrogen storage mechanisms, performance, challenges, and future trends of three materials: graphene, carbon nanotubes (CNTs), and metal-organic frameworks (MOFs). Graphene theoretically boasts an extremely high specific surface area and hydrogen storage capacity, but it is limited by the weak van der Waals forces among the material and hydrogen molecules; CNTs achieve ideal functionalities through functionalization at a low cost, but they have poor solubility, and the modified structures may have defects; porous structure of MOFs could be designed, showing great potential as hydrogen storage materials at room temperature, but they have very poor stability and are also expensive. Combining these materials, retaining their advantages while addressing their shortcomings might be an excellent solution.

Keywords: Hydrogen storage, Graphene, CNTs, MOFs.

1. Introduction

Transportation accounts for about 24% of the total global carbon emissions from fossil fuel combustion [1]. Vehicles powered by new energy, such as electric vehicles and hydrogen energy vehicles, play a key role in de-carbonizing the transportation sector and supporting global climate action [2]. However, the cost of hydrogen fuel cars is too high and there is a lack of hydrogen infrastructure, so this project had to be canceled. Since the 1970s, more and more hydrogen energy cars have appeared, and more and more hydrogen energy car companies have emerged on the market. The infrastructure is gradually improving, and the government is paying more and more attention to the development of hydrogen energy cars. The number of electric vehicles has risen from almost zero in 2010 to more than 16 million in 2021 [3]. It is expected that by 2030, the global inventory of electric vehicles will reach 145 million. [4]

As the new energy resources vehicles are developing very rapidly, new energy storage technologies are also booming, driving breakthroughs in various aspects of electric vehicles from multiple perspectives. For Electric vehicles, the position of the battery is very important. For hydrogen energy vehicles, the main concern is the fabrication and storage of Hydrogen. If no changes are made in the use of energy, human greenhouse gas (GHG) emissions will increase by more than double by 2050 [5]. Hydrogen is a multifunctional fuel that could be divided into "gray hydrogen" (hydrogen from natural gas), "blue hydrogen" (hydrogen from NG with CCS), and "green hydrogen" (hydrogen from renewable energy sources) [6]. Among them, "green hydrogen" is mainly used in electrolytes. Hydrogen is becoming increasingly important as a clean energy carrier. According to the generation method, hydrogen that is not green is harmful to the environment in most cases during production. Therefore, the storage of hydrogen is particularly important. This could not only alleviate the problem of cost but also alleviate the problem of the environment. The storage of hydrogen not only determines the mode of transportation but also determines the method of utilization. The storage method of hydrogen gas is constantly improving.

Overall, the energy density of hydrogen fuel is higher than that of conventional fuels [7,8]. For the above reasons, research into hydrogen fuels has been focused and many technologies have been proposed to achieve efficient and safe hydrogen production, storage and utilization, for example, the

hydrolysis of hydrides is an important area for hydrogen production and storage, and in the case of NaBH₄ hydrolysis, Al-Enizi et al. used electrospinning to prepare carbon nanofiber catalysts and nickel-cobalt nanoparticles, achieving a catalyst surface area of up to 650.9 m²/g. Even after five cycles, the catalyst activity still maintains more than 70% of the initial activity [9]. Langmi and others have conducted extensive research on the hydrogen storage capacity of metal-organic frameworks (MOFs), pointing out that their hydrogen capacity at ambient temperature is usually kept below 1 wt%. With hydrogen molecules being trapped in the framework's pores via weak van der Waals forces, physical adsorption is the mechanism by which hydrogen is adsorbed onto MOFs [10]. Liquid Organic Hydrogen Carriers (LOHC) is also an important technology for hydrogen storage, which is safe and efficient. In summary, there has been a lot of research on the production and storage of hydrogen energy, and more and more methods have been invented, and more and more high-quality materials have been used for hydrogen storage. Therefore, the application of hydrogen energy is a very promising undertaking.

At present, there are many hydrogen storage technologies, such as liquefaction and pressurization, but the cost of those approaches is relatively high [11, 12]. The storage density of cryogenic liquid storage is very low, and the losses during the liquefaction of hydrogen gas are very large. In addition, the compression of hydrogen makes it more flammable, and as a result, the method of liquefying and pressurizing is not economically feasible [13, 14]. Hence the importance of hydrogen storage in materials [11]. So far, a number of types of materials for storing hydrogen have been produced [15]. In order to find suitable and highly efficient hydrogen storage materials, carbon-based materials stand out in current research due to their high surface area, low density, high thermal and chemical stability [16, 17]. Notably, the high surface-to-volume ratio of nanomaterials can remarkably boost their hydrogen storage capabilities. Among these, graphene, CNTs, and MOFs are excellent hydrogen storage materials. For instance, their extensive surface areas and robust hydrogen adsorption capacities render them particularly prominent. Moreover, researchers should not underestimate the hydrogen storage potential of MOFs [18].

In this work, the advantages, disadvantages, and future development trends of graphene, CNTs, and MOFs in the field of highly efficient hydrogen storage were comprehensively compared. The reasons why the three have great advantages in hydrogen storage are described in detail from the structural point of view, and their advantages are proved by some experiments. This article consists of these parts: Introduction, Case Description, Analysis & challenge, Summary & suggestions, and Conclusion. The introduction shows the development history and trend of hydrogen energy, the rise of hydrogen energy vehicles, the progress of hydrogen storage technology, and three kinds of nanomaterials used for hydrogen storage. The case description shows the basic introduction of three materials, followed by challenge analysis of the advantages and disadvantages of different materials and experimental detection. Then, according to the above, look forward to the trend of future development. Finally, based on the the paper, the conclusion will summarize the whole article.

2. Case

2.1. Graphene

Composed of a single layer of carbon atoms with sp² hybrid orbitals, graphene forms a two-dimensional honeycomb lattice structure. With a thickness of merely 0.335 nm, it stands as the thinnest known two-dimensional material. Regarded as a future revolutionary material, graphene exhibits exceptional optical, electrical, and mechanical attributes, holding significant application prospects in domains such as materials science, micro-nano fabrication, energy, biomedicine, and drug delivery. The special structure of graphene gives it the characteristics of light weight, large specific surface area, and high stability, so graphene has become the preferred material in hydrogen storage materials. The two-dimensional structure of graphene has selective permeability to protons, but it forms a natural barrier to hydrogen molecules and oxygen molecules. A "sandwich structure" designed based on this feature encapsulates hydrogen produced by photolysis water between

graphene layers, inhibits the reversal of hydrogen oxygen compounding into water, and guarantees hydrogen storage safety.

2.2. Carbon nanotubes (CNT)

Also referred to as barb tubes, Carbon nanotubes (CNTs) are one-dimensional quantum materials featuring a unique structure, with a radial dimension of nanoscale and an axial dimension of microscale, and basically, both extremities of the tube are closed. Consisting primarily of several to tens of concentric cylindrical tubes, carbon nanotubes are constituted by carbon atoms arranged in a hexagonal configuration. With a fixed interlayer spacing of approximately 0.34 nm, carbon nanotubes typically exhibit diameters ranging from 2 to 20 nm. Depending on the distinct axial orientations of the carbon hexagons, these structures can be categorized into helical, zigzag, and armchair configurations. Helical carbon nanotubes possess chirality among these structures, whereas zigzag and armchair carbon nanotubes lack such properties. The hollow tube-like structure and porous surface of carbon nanotubes provide a large number of adsorption sites, and hydrogen molecules can be stored through two mechanisms: physical adsorption (Vanderthal) and chemical adsorption (binding to surface defects or adulterated sites).

2.3. Metal-organic framework (MOF)

MOFs are metal-organic framework compounds, which are a kind of crystalline porous material with periodic network structure formed by self-assembly of inorganic metal centers (metal ions or metal clusters) and bridging organic ligands. MOFs are a kind of organic-inorganic hybrid material, also known as coordination polymer, which is different from both inorganic porous materials and ordinary organic complexes. It has the rigidity of inorganic materials and the flexibility of organic materials. It shows great development potential and attractive development prospects in the field of modern material research. The basic raw materials used to prepare MOFs are metal ion organic ligands, of which the organic ligands are mostly carboxyl-containing organic anion ligands. Nowadays, researchers have synthesized a diverse range of metal-organic framework materials, predominantly featuring carboxyl-containing organic anionic ligands as the core component or incorporating nitrogen-heterocyclic organic neutral ligands. A majority of these metal-organic frameworks exhibit high porosity along with excellent chemical stability. Owing to their pore structure tunability and large specific surface area, MOFs possess more extensive application prospects than other porous materials, including catalysis, magnetic materials, H₂ adsorption separation, and optical materials. In the storage of bulk fuel gases like methane and hydrogen, resources that can furnish convenient energy solutions for upcoming transportation technologies, they demonstrate significant potential as porous materials with extremely low density.

3. Analysis & challenge

3.1. Graphene

Graphene-based solid porous materials have a particularly large surface area (2630m²/g), porous properties, light weight, high chemical and thermal stability, and the possibility of economical and scalable production, showing promising applications in efficient hydrogen storage. According to the prediction, the hydrogen adsorption capacity of graphene under the ideal conditions of low temperature and high pressure can reach 6.6 wt%, but the actual adsorption capacity under environmental conditions is far from the theory, which is mainly due to the weak van der Waals force interaction between hydrogen and graphene. Therefore, Klechikov et al. designed a high specific surface area (SSA) graphene scaffold and optimized the KOH activation procedure [19]. To date, the best method for producing high porosity carbon materials is KOH activation. Theoretically, the carbon produced after KOH activation has a SSA of up to 3000 m²g⁻¹. It is reported that the SSA of the r-Go sample activated by KOH annealing is 3100m²g⁻¹, which is higher than that of other carbon materials. Validating the hydrogen adsorption parameters of KOH-activated r-Go (a-r-Go), which has

a very high specific surface area and pore volume. r-Go is generated by the rapid peeling off of graphite oxide, consisting of single-layer and few-layer graphene sheets with serious defects, but the SSA of r-Go formed in this way is much smaller than that of graphene sheets activated by KOH, which results from the generation of pores within the graphene layers. Within a broad temperature range, the volumetric method determined the hydrogen storage parameters, and the BET method characterized the SSA metrics.

Fig 1a shows the SSA evaluation of r-Go (a-r-Go) activated via nitrogen adsorption isotherms. The isothermal line evaluated via the quenching solid density functional theory (QSDFT) incorporating a slit pore framework yielded a specific magnitude of $2620 \text{ m}^2\text{g}^{-1}$, smaller than the value of 2660 indicated by the non-local density functional theory (NLDFT) using a slit/cylinder pore architecture. Two peaks can be obtained in the a-r-Go aperture distribution simulation using the slit hole QSDFT (Figure 1c), the first peak is around 0.7-0.8 nm, and the second peak is around 1.5 nm. From the figure, it can be seen that the graphene scaffold structure created by the KOH activation process inherently exhibits microporous properties. To substitute the oxygen-containing functional groups with hydrogen, several a-r-GO samples were subjected to additional annealing treatment in a hydrogen atmosphere. After the treatment, when analyzing the pore size distribution of these samples, an extra peak emerged, which corresponded to pores with larger diameters of up to 3 nm. (as detailed in the ESI†). Figure 1b shows a comprehensive analysis of the pore size of the sample with the largest SSA, and the results indicate that a-r-Go is fundamentally a microporous material. The pores within it have a size smaller than 3 nm, accompanied by a maximum BET surface area of $3300 \text{ m}^2/\text{g}$ and a pore volume that can reach up to $2.22 \text{ cm}^3/\text{g}$ [19].

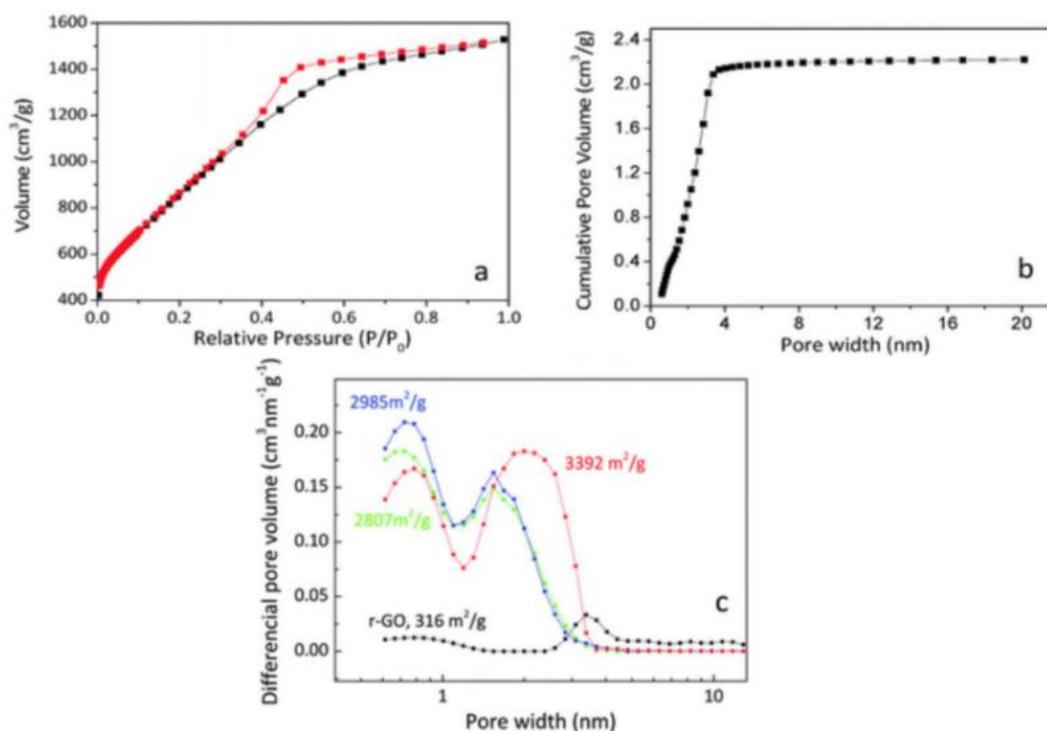


Figure 1. Nitrogen adsorption/desorption isotherms of the a-r-GO sample ($\text{SSA} = 3300 \text{ m}^2\text{g}^{-1}$) (a), along with analysis of simulated isotherms using the Quenched Solid Density Functional Theory (QSDFT) slit pore model: cumulative pore volume plot (b) and pore size distributions for multiple a-r-GO specimens (c) [19].

A few a-r-Go samples were annealed in hydrogen in addition to this to allow hydrogen to replace oxygen-containing functional groups. Essentially, the pore size analysis of the sample with the highest specific surface area (SSA) shows that a-r-Go is a microporous material, and its pore volume is superior to that of other nanoporous materials. Fig 2 shows that under the action of a scanning electron microscope (SEM), 10-micrometer particles form the microstructure of the a-r-Go samples.

According to Figure 2c, the sample can be further described as a strong perforated graphene layer wrapped in a 3D scaffold.

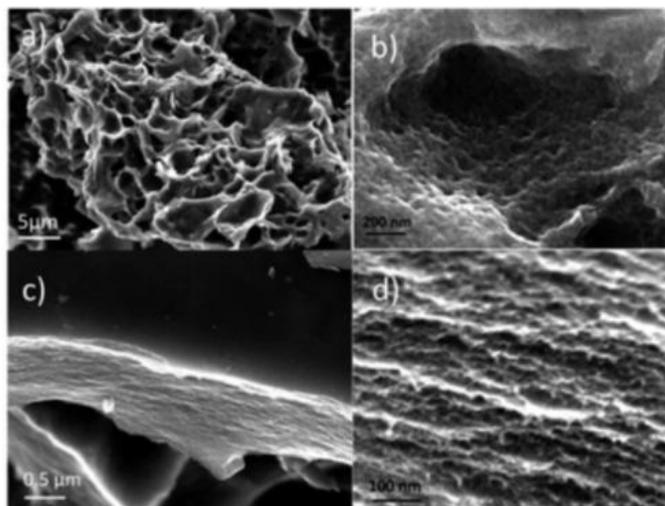


Figure 2. SEM micrographs of a-r-GO specimen with ultrahigh BET surface area ($\sim 3300 \text{ m}^2 \cdot \text{g}^{-1}$): (a, b) hierarchical pore networks; (c, d) layered fracture edges [19].

Using the volumetric method, they evaluated the a-r-Go sample at temperatures of 77K and 296K. At 77K and 40 Bar pressure, the hydrogen adsorption capacity was approximately 7.04 wt%; at 296K and 120 Bar pressure, the hydrogen adsorption capacity reached 1.13 wt%. The isothermal lines of nitrogen measured after annealing before and after the measurement of hydrogen absorption showed unmeasurable changes.

In summary, by adjusting the KOH activation process, it could obtain the structure of high-surface-area graphene scaffold. This structure of SSA is superior to traditional carbon materials and has a stronger hydrogen adsorption ability. However, the cost is still a problem that could not be bypassed, and mass production is also difficult. However, it must be acknowledged that this is a feasible path, and perhaps it is inspired from this experiment by changing the production process of graphene and its derivatives to obtain graphene materials with higher performance and improve the hydrogen storage capacity.

3.2. CNTs

CNTs are also a material that could not be bypassed on the road to hydrogen storage. As a kind of hydrogen storage material, its cost is relatively low, and its production is relatively easy. At present, the research is mainly focused on the purification of CNTs to obtain more high-quality CNTs. Fig. 3 shows two mechanisms related to CNTs [20]. For CNTs, its nitrogen adsorbate adsorbs H_2 and stores it in the top layer, and this adsorption is called physical adsorption. Atomic models and continuum models can show hydrogen storage on CNTs. In the atomic model, the C atom and the H_2 molecule are discrete, and the van der Waals force interaction between C-C and the molecular interaction between C-H and H-H. For large CNTs, hydrogen molecules are represented as internal pressure, with the aim of minimizing the total energy of the model. An elevation in pressure results in a reduction of the distances between $\text{H}_2\text{-H}_2$ and C- H_2 , but an increase in the radius of CNTs, allowing for high hydrogen storage.

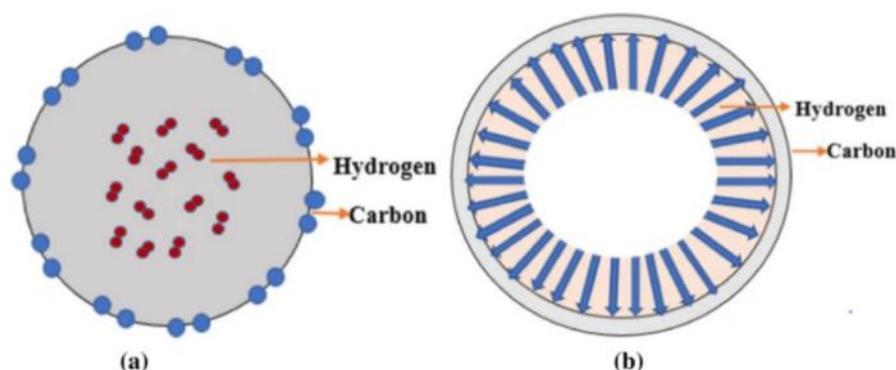


Figure 3. Hydrogen storage mechanisms in CNTs: (a) a representation of hydrogen storage in CNTs at the atomistic level, where both hydrogen and carbon are treated as discrete particles; (b) a continuum representation where CNTs are described by a continuous shell and H₂ by internal pressure [20].

Since CNTs are large molecules and almost insoluble in all solvents, it is necessary to modify the effect of CNTs, that is, to functionalize them, before they can be dissolved in organic solvents. Calisir et al. [21] completed the synthesis of thiol-functionalized carbon nanotubes. Starting from MWCNT (D1-D7), the synthesis of modified MWCNT derivatives functionalized with free hydroxyl (-OH) and thiol (-SH) groups was carried out under both conventional and microwave-assisted reaction environments in their investigation. The porous framework of CNTs enables these voids to adsorb substantial quantities of gas, thereby forming high-density adsorption materials. Widely employed as adsorbents for various materials, CNTs are characterized by large specific surface areas, chemical inertness, hollow geometries, and wettability. At ambient temperature, CNTs with high energy and transitional elasticity act as onboard hydrogen storage media.

In summary, due to its special chemical structure, CNTs are different from graphene in terms of structural design optimization, and the functionalization of CNTs is the mainstream of research. The main direction of research is to obtain the ability to store hydrogen in a special environment by changing the chemical group of CNTs to have a certain chemical property. Similar to graphene, the method of manufacturing defects is still feasible. The CNTs structure has many possibilities for hydrogen storage, and its cost is relatively low, making it a suitable material. However, its practical application is not optimistic, and scientists need to further study the characteristics of CNTs.

3.3. MOFs

At room temperature and low temperature (248-337K), MOFs use their open metal sites and hydrogen exudation mechanism to promote the progress of hydrogen storage technology. Promoting strong adsorption within MOFs is one of the main research directions. In the current research, the study of hydrogen storage in metal-organic frameworks mainly focuses on the low-temperature working temperature. In this environment, these porous materials often have a weak hydrogen absorption mechanism, with a combination enthalpy in the range of -4 to -7KJ / mol, and the absorption amount is about 1.0 wt% with the increase of temperature. The enthalpy required for the combination of MOFs needs to be significantly increased to -15 to -25 KJ/mol. Entropy is also a factor in designing MOFs for hydrogen storage at ambient temperatures. In MOFs, the enthalpy and entropy of H₂ adsorption show a positive correlation. At present, there are three methods to increase the adsorption heat in MOFs to store hydrogen gas: combining open metal sites, alkaline charge-induced interactions, and hydrogen gas overflow. These three methodologies can all elevate the binding enthalpy, thereby augmenting the hydrogen retention capacity at near-ambient temperatures. Table 1 outlines a selection of MOFs incorporating accessible metal sites and registered H₂ adsorption data at near-ambient temperatures [22].

Table 1. Various MOF for H₂ storage [22]

MOF	Temp (K)	Pressure (bar)	Grav (wt%)	BET (m ² /g)	Qst (kJ/mol)	Vol (g/L)
CuI-MFU-4 I	273	1	0.34	-	32	-
Ni ₂ (m-dobdc)	298	100	0.98	1312	12.3	11.9
Ni ₅₀ Co-IRMOF-74	298	100	0.38	1985	-	3.1
V ₂ Cl _{2.8} (bttd)	298	100	1.64	1920	21	10.7
Cu ₂ (BBTA)/	296	100	-	515	32	7.2
Mn ₃ [(Mn ₄ Cl) ₃]	298	90	1.5	2100	10.1	12

An air-stable CuI-MOF (NU-2101) was proposed. It is composed of two different environments, one is a coordinated saturated tetrahedral species, and the other is an unsaturated almost linear two-coordinate CuI species. CuI, which is almost linear, can bind to H₂, and the initial binding enthalpy of this framework is -32 kJ/mol, which is one of the highest reported values to date. However, this initial high binding enthalpy drops sharply at a loading of 0.03 mmol/g, which is only 1% of the linear species of CuI that actively binds to H₂. At 298K and 100Bar, the volume capacity of NU-2100 is 7.2g/L. These examples show that the binding enthalpy in the range of -15 to -25 kJ/mol (or even slightly higher) seems to lead to the highest weight and volume load of hydrogen at near-ambient temperatures reported so far in MOFs. Therefore, it is expected that the next generation of MOFs will target the binding enthalpy in similar systems. In order to further enhance the volume hydrogen storage near the ambient temperature, the density of the open metal field needs to be increased. Each additional open site per unit volume will result in an increase in H₂ per unit volume. This will also help to increase the binding enthalpy per mole of MOFs.

In summary, the advantage of MOFs in hydrogen storage lies in their performance at room temperature. However, a large part of current research focuses on low-temperature conditions. However, research on MOFs for hydrogen storage at room temperature is increasing. There are three main ways to enhance the performance of MOFs, and only the first one is described in detail here: combining open metal sites. The problem of cost is also a major difficulty for this kind of material, and stability is also a key issue for MOFs. Research on it is gradually being carried out, and it has great prospects in the future. When receiving the paper, we assume that the corresponding authors grant us the copyright to use the paper for the book or journal in question. When receiving the paper, we assume that the corresponding authors grant us the copyright to use.

4. Summary

Hydrogen fuel cell vehicles have risen rapidly, making hydrogen storage an important research topic. At the same time, efficient hydrogen storage materials have become a key focus of research. This paper describes three mainstream hydrogen storage materials: graphene, carbon nanotubes, and metal-organic frameworks, analyzing the performance mechanisms, advantages and disadvantages of each material, as well as potential future development directions.

Graphene, a two-dimensional honeycomb nanomaterial, exhibits an extraordinarily large specific surface area, with theoretical values even reaching 2630 m²/g. Under cryogenic and elevated-pressure conditions, its theoretical hydrogen storage capacity can attain 6.6wt%. Experiments demonstrate that the KOH activation process can enhance graphene's porosity, enabling a hydrogen uptake of 7.04 wt% under conditions of 77 K and 40 Bar. However, in real-world applications, graphene's hydrogen uptake significantly lags behind the theoretical predictions. This is mainly due to the weak van der Waals forces between hydrogen molecules and the material. Additionally, the large-scale preparation of graphene is complex and costly, making commercialization challenging. In the future, efforts could focus on mitigating the weak van der Waals forces among materials and hydrogen molecules to design new types of graphene. Alternatively, optimizing graphene processes to integrate graphene manufacturing with other materials could produce multiple products at the same cost.

Carbon nanotubes have a hollow tubular structure with porous surfaces, allowing for hydrogen storage through both physical and chemical adsorption mechanisms. Compared to graphene, carbon

nanotubes are easier to produce and less costly. However, the actual hydrogen storage capacity of carbon nanotubes depends on their functionalized products. For example, thiol-functionalized CNTs can enhance hydrogen adsorption sites, but the hydrogen storage density at room temperature is still insufficient, indicating that further functionalization is needed. Functionalization opens up endless possibilities for carbon nanotubes. Currently, however, issues such as poor solubility and structural defects in CNTs remain. In the future, exploring the functionalization of carbon nanotubes will be essential. By functionalizing different aspects of their properties, more perfect hydrogen storage materials could be designed.

MOFs excel in hydrogen storage at room temperature. They feature a designable pore structure and open metal sites. In the NU-2101 material, hydrogen molecules are bound through these open metal sites, with an initial binding enthalpy of -32 kJ/mol, achieving a volumetric hydrogen storage capacity of 7.2 g/L at 298K and 100 Bar. However, MOFs have poor stability, similar to graphene, and their synthesis costs are also high. MOFs perform better at low temperatures than at room temperature. Boosting the stability of MOFs and enhancing their performance under room-temperature conditions should be the focus of future research. Table 2 reflected the merits and demerits of each material.

Table 2. Comparison of various H₂ storage material

Type	advantages	disadvantages
Graphene	-High specific surface area -High stability -High theoretical hydrogen storage capacity	-Low actual storage capacity -High cost -hard for mass production
CNTs	-Low cost -Easy for functionalization	-in sufficient storage rate at room temperature -the solubility is poor
MOFs	-design-able pore structure	-Poor stability -high cost

5. Conclusion

This paper reviews three mainstream hydrogen storage materials for hydrogen fuel cell vehicles: graphene, carbon nanotubes (CNTs), and metal-organic frameworks (MOFs). Graphene demonstrates exceptional theoretical hydrogen storage capacity (6.6wt% at cryogenic/high-pressure conditions) due to its ultra-high specific surface area (2630 m²/g). KOH-activated graphene achieves 7.04wt% hydrogen uptake at 77 K/40 Bar. However, practical applications are hindered by weak van der Waals interactions with hydrogen, complex large-scale production, and high costs. Future improvements may involve enhancing graphene-hydrogen interactions or integrating graphene with other materials for cost-effective manufacturing. Carbon nanotubes offer advantages in simpler production and lower costs compared to graphene. Their hollow structure supports dual physical/chemical hydrogen adsorption. Functionalization (e.g., thiol groups) enhances adsorption sites, but room-temperature storage density remains inadequate. Challenges include structural defects and poor solubility. Future research should focus on advanced functionalization strategies to optimize performance. MOFs excel in room-temperature hydrogen storage via designable pores and open metal sites. NU-2101 achieves 7.2 g/L volumetric capacity at 298K/100 Bar with a binding enthalpy of -32 kJ/mol. However, MOFs suffer from poor stability and high synthesis costs. Enhancing structural stability and room-temperature efficiency are critical for future development.

In summary, graphene and MOFs face challenges in practical application costs and material stability, while CNTs require functionalization breakthroughs. Cross-material hybridization and structural optimization represent promising pathways for advancing hydrogen storage technologies. Table 2 likely highlights graphene's high capacity vs. scalability issues, CNTs' cost-effectiveness vs. functionalization needs, and MOFs' room-temperature performance vs. stability limitations.

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