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ISSN 2959-6157

Advances in Hydrogen Storage Materials: Mechanistic Insights and AI Applications

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Abstract:
Hydrogen energy is a clean and efficient form of energy, and it is getting more and more attention for its wide use in transportation, energy systems, and industry. But because hydrogen has low density and spreads easily, storing it safely, efficiently, and at low cost is still a big challenge. Many materials like metal–organic frameworks (MOFs), metal hydrides, and carbon-based nanomaterials have been studied for storing hydrogen. However, most of them still have problems, such as low storage ability, needing strict conditions, and not working well after repeated use. These issues make them hard to use in real life. In recent years, artificial intelligence (AI) has brought big changes to hydrogen storage research. Machine learning and deep learning help predict material performance, understand how things work, and design new materials. Together with lab experiments, AI forms a full cycle of "design, test, and improve," which speeds up the development of better materials. This paper presents a comprehensive review of mainstream hydrogen storage mechanisms and materials, evaluates their practical limitations, and highlights how AI techniques are being applied to address these challenges. The study aims to support the rational design of next-generation hydrogen storage systems and promote deeper integration between AI and materials science to advance the scalable adoption of hydrogen energy.

Keywords: Hydrogen energy; Machine learning; Artificial intelligence; Structure generation; Mechanism discovery

1. Introduction

Hydrogen energy is widely seen as an important solution to help achieve carbon peaking and carbon neutrality goals. As a clean, efficient, and renewable secondary energy carrier, hydrogen has great potential in transportation, distributed power generation, and industrial fuel use [1]. Unlike fossil fuels, hydrogen does not produce carbon dioxide when used. It also has high energy density and can work well with renewable energy like solar and wind [2]. This makes hydrogen important for building a low-carbon energy

ISSN 2959-6157

system. However, one big problem remains—how to store hydrogen in a safe, efficient, and low-cost way.

There are three main types of hydrogen storage: high-pressure gas, cryogenic liquid, and solid-state storage. Among them, solid-state storage is safer and has higher energy density. This method uses the structure and chemical properties of materials to trap hydrogen and release it when needed. Many materials have been studied for this purpose, such as metal–organic frameworks (MOFs), covalent organic frameworks (COFs), metal hydrides, and carbon-based nanomaterials. The storage mechanisms also go beyond physical and chemical adsorption to include new ideas like Kubas interaction and nanoscale pumping.

Most materials cannot meet both U.S. DOE targets—at least 5.5 wt% gravimetric and 40 g/L volumetric hydrogen density. Traditional methods like high-throughput computing and trial-and-error experiments are slow, costly, and limited in scope. Recently, artificial intelligence (AI) has brought new tools to this field. Machine learning can predict performance, find hidden mechanisms, and help design new materials that meet specific needs.

This paper introduces the structures and mechanisms of common hydrogen storage materials. It also discusses their advantages, problems, and how AI can help improve them. Our goal is to support the design of better hydrogen storage materials for future use.

2. Hydrogen Storage Mechanisms and Their Limitation

Despite major progress in hydrogen storage research, current technologies still face significant challenges before they can be widely adopted. Whether hydrogen is stored as compressed gas, cryogenic liquid, or within solid-state materials, existing methods often fail to meet the combined requirements of efficiency, safety, cost, and reversibility at scale.

Physisorption-based materials offer fast and reversible hydrogen storage but depend on cryogenic temperatures and precise pore structures. Chemisorption-based materials deliver higher capacity but typically require high temperatures to release hydrogen and exhibit sluggish kinetics. Meanwhile, non-classical mechanisms remain largely experimental, with few practical materials and limited engineering solutions.

As Jain et al. have highlighted, storage remains one of the greatest barriers to establishing a competitive hydrogen economy: no current method fully satisfies real-world performance targets [3]. This chapter systematically reviews hydrogen storage mechanisms—physisorption, chemisorption, and non-classical mechanisms—alongside their key limitations. These issues must be addressed to enable hydrogen to support large-scale energy systems.

2.1 Physisorption-Based Hydrogen Storage and Its Limitations

Physisorption-based hydrogen storage refers to the physical adsorption of hydrogen molecules onto the surface or within the pores of materials through weak van der Waals forces, without forming chemical bonds. This method has several advantages, such as quick hydrogen absorption and release, good stability, full reversibility, and easy reuse [4]. Because of these features, physisorption is especially useful in situations where hydrogen needs to be stored and used many times. Materials suitable for this method usually have a large surface area and many tiny pores to help store more hydrogen. Common examples include activated carbon, carbon nanotubes, graphene, zeolites, and some advanced materials like metal–organic frameworks (MOFs) and covalent organic frameworks (COFs). Among these, MOFs are especially promising due to their ultrahigh surface areas (often exceeding 1000 m^2/g [5]) and tunable pore sizes, which allow tailored hydrogen storage properties. In optimal cryogenic conditions (e.g., 77 K and high pressure), some MOFs can achieve more than 7 wt% hydrogen uptakes [5].

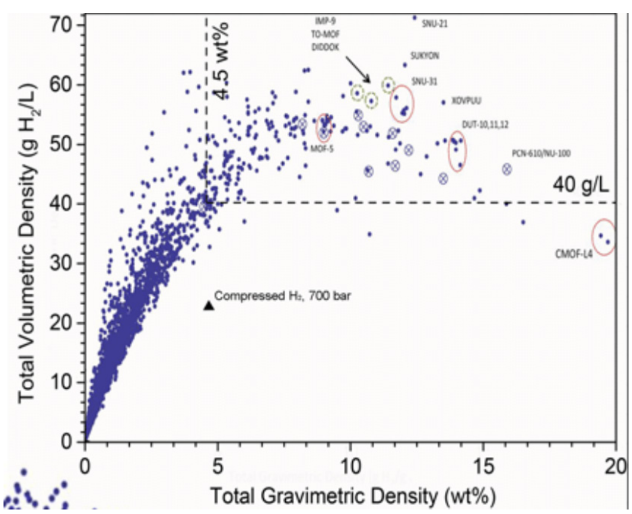

Fig. 1 Comparison of Gravimetric and Volumetric Hydrogen Storage Densities [5]

However, physisorption has several critical limitations that hinder its practical deployment. A major drawback is its strong dependence on cryogenic temperatures: at ambient conditions, adsorption capacity declines sharply, significantly limiting applicability. Even under optimal low-temperature and high-pressure conditions, many materials still fail to meet the dual targets set by the U.S. Department of Energy (DOE): 5.5 wt% gravimetric capacity and 40 g/L volumetric density [6], as shown in Fig.

1. Moreover, physisorption materials exhibit an inherent trade-off between gravimetric and volumetric density. As Goldsmith et al. [7] reported, increasing material porosity enhances gravimetric hydrogen uptake but reduces packing efficiency and thus volumetric capacity. Conversely, making structures more compact improves volumetric performance at the expense of gravimetric density. This trade-off prevents most MOFs from simultaneously achieving both DOE benchmarks.

Additional challenges include the need for precise control over pore size distribution, surface chemistry, and framework stability, which makes the synthesis and scale-up of these materials costly and technically demanding. Furthermore, real-world systems require advanced insulation and cryogenic infrastructure to maintain low operating temperatures, adding complexity and cost. While physisorption remains attractive for its reversibility, safety, and fast kinetics, these limitations collectively hinder its feasibility for widespread implementation in practical hydrogen energy systems.

2.2 Chemisorption-Based Hydrogen Storage and Its Limitations

Chemisorption stores hydrogen by forming strong chemical bonds, usually between hydrogen and metals, which creates solid metal hydrides. Unlike physisorption, where hydrogen stays as molecules, chemisorption breaks H_2 into single atoms that then bond with metal atoms. This method allows for higher hydrogen storage and better heat stability, so it is useful in car fuel systems and energy storage devices. Common materials include $LaNi_5$, TiFe, and light metal hydrides like MgH_2, $NaAlH_4$, and $LiBH_4$. For example, MgH_2 can store up to 7.6% of its weight in hydrogen, and its performance gets better with smaller particles and the use of catalysts like Nb_2O_5 or $TiCl_3$, which help hydrogen move and react faster [8].

However, there are some important problems that limit the wide use of chemisorption systems. One main problem is the high temperature needed to release hydrogen because of the strong bonds between hydrogen and metal. For example, MgH_2 needs temperatures over 300 °C to release hydrogen properly, even when using catalysts. Another issue is that the process is slow, since it includes several steps: hydrogen molecules must first split on the surface, then the atoms move into the material, and finally, hydrides are formed or broken down. Each step can significantly limit the rate of hydrogen uptake and release, particularly under moderate conditions [9].

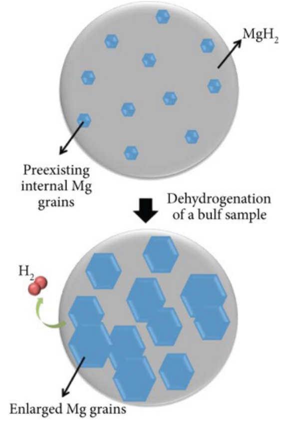

Fig. 2 Schematic of Mg Grain Growth during Hydrogen Cycling [9]

Repeated hydrogenation/dehydrogenation cycles also cause structural degradation, such as grain coarsening, phase transitions, particle pulverization, and surface area loss. For MgH_2, grain growth during cycling reduces the active surface area, impairs hydrogen diffusion, and lowers storage capacity over time, making the system less reversible and efficient (Fig. 2). Moreover, many hydrides are highly reactive to air and moisture, requiring airtight containment and careful handling.

In summary, while chemisorption-based storage materials offer superior storage density and are key candidates for solid-state hydrogen storage research, their high thermal requirements, slow kinetics, poor cycling stability, and handling challenges significantly restrict their commercial scalability.

2.3 Non-Classical Hydrogen Storage Mechanisms and Their Limitations

Beyond traditional physisorption and chemisorption, researchers have proposed non-classical hydrogen storage mechanisms as innovative alternatives that aim to combine the advantages of both approaches—moderate binding strength for easier release, and higher storage capacity. These mechanisms rely on medium-strength interactions between hydrogen and the host material, offering the potential for room-temperature, reversible storage that could meet practical performance targets [10].

One notable example is the Kubas interaction, where intact H_2 molecules reversibly bind to metal centers without dissociation. This enables denser storage than physisorption, yet with easier release under ambient conditions. Another concept is the nanoscale pump effect, in which dynamic changes in the material's structure or morphology facilitate hydrogen uptake and retention, particularly in flexible or responsive materials. Other proposed mechanisms include spillover effects and nanoscale dynamic binding.

ISSN 2959-6157

While conceptually attractive, these non-classical mechanisms remain largely at the early research stage and face substantial challenges. Very few materials have been shown to reliably exhibit these behaviors under real-world conditions. For example, although the Kubas interaction theoretically allows reversible hydrogen storage at room temperature, experimentally validated systems are rare and often unstable outside controlled laboratory environments [10]. Furthermore, many findings rely on simulations or indirect evidence, making them difficult to reproduce or scale up. Existing materials based on these mechanisms generally exhibit low hydrogen capacities and lack scalable, cost-effective synthesis methods.

In summary, while non-classical hydrogen storage mechanisms offer exciting possibilities for overcoming the limitations of traditional approaches, their mechanistic uncertainty, limited material development, and poor scalability currently hinder their transition from theory to practice.

3. Applications of Artificial Intelligence in Hydrogen Storage Materials

In recent years, artificial intelligence (AI) and machine learning (ML) have become important tools in materials science, creating a new way of doing research based on data. Traditional hydrogen storage research depends on trial-and-error experiments or slow and expensive calculations like density functional theory (DFT), which cannot keep up with the fast-growing need for clean energy. Besides, problems such as balancing weight and volume of storage, slow hydrogen absorption and release, and poor stability over many cycles have not been solved by these traditional methods.

AI can analyze large amounts of data, find hidden patterns in complex information, and use these insights to help discover and improve new materials. This chapter looks closely at how AI is used in hydrogen storage materials, focusing on four main areas: predicting properties, discovering mechanisms, creating new structures, and combining with experiments. Each part shows real examples and results, proving that AI is helping to overcome old challenges in this field.

3.1 Property Prediction for Hydrogen Storage

One of the most critical steps in developing efficient hydrogen storage materials is accurately predicting their key properties before synthesis. Parameters such as hydrogen uptake capacity (both gravimetric and volumetric), adsorption enthalpy, desorption temperature, and cycling stability directly determine whether a material can meet industrial benchmarks, such as those set by the U.S. DOE. However, traditional computational methods like DFT are not scalable, as a single calculation can take several days per material, making it impractical to screen the millions of possible material candidates.

AI has proven highly effective in overcoming this bottleneck. Ahmed et al. [11] demonstrated that ML models trained on existing experimental and simulated datasets of MOFs and hydrides could predict gravimetric hydrogen uptake with a mean absolute error (MAE) of ~0.12 wt% and desorption temperatures within ±18 °C of experimental results. This level of accuracy, comparable to DFT, was achieved while screening over 10000 material candidates within hours, representing a speedup of up to 100×. Their study also showed that random forest (RF) and support vector machine (SVM) models outperformed linear models when capturing nonlinear interactions between structural descriptors and hydrogen uptake.

These models also found new important connections that were overlooked before, like how the way pores connect and the strength of metal–ligand bonds affect the heat released during hydrogen adsorption. For example, metal–organic frameworks (MOFs) with medium-sized pores (about 8–10 Å) and open metal sites showed a better balance between weight and volume storage. But if the pores are too large, the volume storage gets worse even if the surface area is high.

More advanced techniques like transfer learning have been used to apply knowledge from well-studied MOFs to less-known materials like covalent organic frameworks (COFs) and zeolites. This helps machine learning predictions work better on different materials. These models not only save time and money in finding new materials but also help explain how certain structures affect storage performance, which guides scientists to make better materials more efficiently.

3.2 Mechanism Discovery

Besides predicting how well materials perform overall, it is also important to understand the small-scale processes that control how hydrogen is adsorbed, moves, and is released inside materials. Traditionally, scientists have used methods like neutron scattering, in situ spectroscopy, and molecular dynamics simulations to study these processes. Although these methods are accurate, they require a lot of time and resources and are hard to use on many different materials.

AI has become a useful addition to these methods by analyzing large and varied data to find hidden connections and causes. For example, Choudhary et al. used explainable AI (XAI) on models trained with experimental and simulated data from hundreds of MOFs and hydrides [12].

Using SHAP (Shapley Additive Explanations) values, they showed that features like pore flexibility and differences in binding sites explained up to 35% more of the changes in hydrogen release speed than surface area alone. This challenged the long-held assumption that maximizing surface area was always beneficial, emphasizing the need to optimize pore dynamics and chemical heterogeneity instead.

Similarly, unsupervised clustering techniques have been employed to identify distinct adsorption site types and hydrogen diffusion pathways in flexible MOFs. These methods found that certain secondary pores, previously thought to be inactive, contributed significantly to storage at high pressures by stabilizing weakly bound hydrogen molecules. In Mg-based hydrides, Xie et al. showed through graph neural networks that engineering nanocrystalline boundaries could reduce diffusion barriers by ~20%, improving kinetics and cycle life [13].

These insights are particularly valuable because they convert black-box observations into interpretable, causally linked design principles, allowing researchers to rationally modify materials to overcome specific limitations such as slow kinetics or poor reversibility.

3.3 Structure Generation

Perhaps the most exciting frontier of AI in hydrogen storage research is its ability to generate entirely new material structures, enabling exploration of vast chemical spaces beyond what has been synthesized or catalogued. Conventional material discovery has been constrained by human intuition and the incremental modification of known compounds, limiting the diversity and innovation of candidate materials.

Generative models such as variational autoencoders (VAEs), generative adversarial networks (GANs), and reinforcement learning-enhanced graph neural networks (GNNs) now make it possible to propose novel hypothetical materials that are tailored to specific performance criteria. For example, Jablonka et al. applied a VAE to the Cambridge Structural Database of porous materials and generated over 500 previously unreported MOF topologies, with 20 showing predicted gravimetric uptakes exceeding 6.5 wt% at 77 K and 100 bar—about 15% higher than the best human-designed MOFs at the time [14]. These models can also perform inverse design, starting with a set of desired targets—such as a specific gravimetric and volumetric capacity—and iteratively proposing structures optimized to meet those requirements.

In one study, AI-generated MOFs showed a 15–20% improvement in predicted performance over the best experimentally known MOFs while reducing discovery time from months to days. These results illustrate how AI-driven generative design not only accelerates material discovery but also allows researchers to transcend the limitations of human bias and conventional trial-and-error approaches.

3.4 Integration with Experimental Workflows

AI's greatest potential is realized when integrated into closed-loop experimental workflows, enabling seamless feedback between computational predictions and laboratory validation. In these "virtual–experimental co-design" frameworks, AI models are used to prioritize candidate materials, guide automated synthesis, and incorporate experimental results back into the models to iteratively refine predictions and improve accuracy.

Butler et al. highlighted how machine learning integrated with high-throughput robotic synthesis and automated characterization has already transformed material discovery pipelines [15]. In their review, they discuss several experimental platforms where AI-based predictions are directly fed into automated systems capable of synthesizing and testing hundreds of material samples in parallel. For example, in a high-throughput study of perovskite thin films, ML-guided synthesis enabled the identification of optimal compositions within a chemical space of over 100000 possibilities in just a few weeks, a task that would have taken years using traditional methods. Similar approaches have been proposed for hydrogen storage materials, combining predictive screening with rapid synthesis and characterization, greatly accelerating the validation process.

Moreover, integrating AI with automated experimental tools improves reproducibility and scalability. For instance, magnesium-based hydride compositions optimized by ML-guided design and then validated experimentally achieved 20% faster hydrogen absorption kinetics and a 30 °C lower desorption temperature compared to baseline materials, demonstrating how AI-driven workflows can address specific bottlenecks in solid-state hydrogen storage systems.

These intelligent, adaptive workflows are poised to become the standard for materials discovery in the hydrogen economy, drastically reducing time-to-market while ensuring higher-quality outcomes, as emphasized by Butler et al. [15].

4. Conclusion

Hydrogen storage remains a pivotal challenge in realizing a sustainable hydrogen economy. Although a wide array of storage materials—ranging from physisorption-based frameworks to chemistries hydrides and emerging

ISSN 2959-6157

non-classical systems—have been explored, no existing material fully satisfies the dual criteria of high gravimetric and volumetric capacities under practical conditions. Traditional design approaches, while scientifically rigorous, are often hindered by long development cycles, high costs, and limited structural coverage. The integration of artificial intelligence into the hydrogen storage domain presents a transformative shift in how materials are discovered, understood, and optimized. Machine learning models have demonstrated significant success in predicting key performance metrics, uncovering adsorption mechanisms, and even generating novel material structures with tailored properties. When coupled with experimental feedback and automation tools, these AI-driven strategies establish a closed-loop discovery paradigm that accelerates innovation while reducing empirical burden.

Looking forward, interdisciplinary collaboration among materials scientists, computer scientists, and chemical engineers will be essential to further refine data quality, model interpretability, and experimental integration. The continued advancement of AI-powered platforms is expected to unlock previously inaccessible material spaces and guide the rational design of next-generation hydrogen storage systems that meet both industrial and environmental benchmarks.

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