

Catalytic Enhancement of Hydrogen Storage in MgH_2 Using Metal Oxide Catalysts: Recent Advances and Future Perspectives

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Received: April 22, 2026 | Review began: April 27, 2026 | Review ended: May 08, 2026 | Published: May 28, 2026

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Abstract

In recent decades, significant efforts have been directed toward developing efficient hydrogen storage materials for sustainable energy applications. Magnesium hydride (MgH_2) is considered one of the most promising solid-state hydrogen storage materials due to its high theoretical hydrogen capacity (~7.6 wt%), abundance, low cost, and good reversibility. However, its practical application is limited by strong thermodynamic stability and sluggish hydrogen absorption/desorption kinetics, typically requiring temperatures above 300°C and exhibiting activation energies of ~200-210 kJ/mol.

Among various strategies, catalyst incorporation has proven to be an effective approach to enhance hydrogen storage performance. In particular, metal oxide catalysts have attracted considerable attention due to their stability, availability, and strong catalytic activity. This review highlights recent advances in the catalytic enhancement of MgH_2 using metal oxide catalysts, with emphasis on transition metal oxides such as TiO_2 , Fe_2O_3 , NiO , Co_3O_4 , and V_2O_5 . The incorporation of these catalysts significantly improves hydrogen sorption behavior, reducing activation energy to ~114-136 kJ/mol (~30-45% reduction) and lowering desorption temperatures by ~50-150°C. Optimized systems demonstrate hydrogen release capacities of ~4-6 wt% within 10-15 minutes at 250-300°C, while some catalysts (e.g., TiO_2 -based systems) enable rapid hydrogen uptake of ~5.5 wt% within 5 minutes at 250°C.

The key mechanisms, including enhanced hydrogen diffusion, defect formation, oxygen vacancy generation, and interfacial catalytic effects, are discussed. Furthermore, synthesis and modification strategies such as high-energy ball milling, nanostructuring, defect engineering, and hybrid catalyst design are examined in relation to their impact on performance. Recent developments in nanostructured, defect-rich, and multi-component catalyst systems demonstrate substantial improvements in kinetics, reversibility, and cycling stability.

Despite these advancements, challenges related to scalability, cost, and long-term durability remain. Future research should focus on achieving hydrogen desorption below 200°C, reducing activation energy below 100 kJ/mol, and developing scalable synthesis routes, along with integration into renewable energy systems for practical MgH_2 -based hydrogen storage applications.

Categories: Advanced Materials, Catalysis and Reaction Engineering, Energy Efficiency and Conservation

Keywords: hydrogen storage, MgH_2 , thermodynamic, kinetics, catalysts, metal oxides

How to cite this article:

Sharma S (May 28, 2026) Catalytic Enhancement of Hydrogen Storage in MgH_2 Using Metal Oxide Catalysts: Recent Advances and Future Perspectives . Cureus J Eng 3 : es44388-026-00087-6. DOI <https://doi.org/10.7759/s44388-026-00087-6>

Introduction And Background

The increasing depletion of traditional energy resources, including petroleum, natural gas, and other fossil fuels, has emerged as a major global challenge, prompting intensive efforts to identify sustainable and alternative energy solutions. Hydrogen is widely recognized as a promising energy carrier with significant potential to support the transition away from fossil fuel-dependent systems. However, its environmental benefits depend strongly on the method of production, as hydrogen derived from renewable energy sources (green hydrogen) is considered clean, whereas the majority of current global hydrogen production relies on fossil fuels, resulting in substantial CO_2 emissions. Despite these challenges, owing to its high energy efficiency and versatility, hydrogen remains a key component of future sustainable energy systems [1]. It is important to note that hydrogen does not exist as a primary energy source; rather, it functions as a carrier capable of storing and delivering energy. Unlike conventional hydrocarbon fuels such as gasoline and diesel, hydrogen conversion produces only water as a by-product, thereby eliminating carbon emissions at the point of use.

Hydrogen is characterized by a high gravimetric energy density of $119.93 \text{ MJ kg}^{-1}$ (lower heating value, LHV), nearly three times that of conventional fuels such as gasoline ($\sim 44 \text{ MJ kg}^{-1}$). However, its very low volumetric energy density ($\sim 0.0838 \text{ kg m}^{-3}$ or $\sim 10.05 \text{ MJ m}^{-3}$ at ambient conditions) poses a major challenge for efficient storage and transportation [2]. Traditional storage techniques, such as high-pressure gaseous storage (typically up to 5000-10,000 psi) and cryogenic liquid storage, involve considerable technical and economic challenges. These methods require either robust high-pressure containment systems or extremely low temperatures, leading to concerns related to safety, cost, and energy consumption. For example, Liquid hydrogen storage in cryogenic tanks provides higher volumetric density ($\sim 70.8 \text{ kg m}^{-3}$) than compressed hydrogen, but still requires nearly four times more storage volume than gasoline for equivalent energy output. In contrast, compressed hydrogen storage at pressures up to 77 MPa offers lower volumetric density ($\sim 40 \text{ kg m}^{-3}$) and requires even larger storage space. Despite their technological maturity, both storage methods are limited by high energy consumption, high cost, extreme pressure or low-temperature requirements, and safety concerns, which restrict their large-scale commercial application.

To overcome these limitations, solid-state hydrogen storage has been extensively investigated as a safer and more efficient alternative. In this approach, hydrogen is stored within materials through mechanisms such as physical adsorption or chemical bonding, enabling operation under comparatively moderate conditions while improving storage density. Over recent decades, considerable research efforts have been directed toward identifying materials that can meet the performance targets established by organizations such as the U.S. Department of Energy (DOE), which has proposed operating temperatures in the range of $40\text{--}85^\circ\text{C}$ and material-level hydrogen storage capacities exceeding 8 wt% for solid-state hydrogen storage systems [3-5].

Among the various materials explored, magnesium hydride (MgH_2) has attracted significant attention due to its high theoretical hydrogen storage capacity ($\sim 7.6 \text{ wt\%}$), along with advantages such as abundance, low cost, recyclability, and favorable thermodynamic stability. Despite these benefits, its practical utilization remains limited due to several intrinsic drawbacks, including slow hydrogen absorption and desorption kinetics, elevated desorption temperatures, and sensitivity to oxidation upon exposure to air, where the formation of thermodynamically stable MgO/Mg(OH)_2 surface layers suppresses hydrogen dissociation and diffusion, thereby deteriorating the hydrogen sorption kinetics and reversible storage capacity [6]. These limitations are primarily associated with its strong thermodynamic stability and restricted hydrogen diffusion.

To enhance the hydrogen storage performance of MgH_2 , several modification strategies have been explored, including nanostructuring to shorten hydrogen diffusion pathways and increase surface area, alloy formation to tailor thermodynamic stability and lower desorption temperature, nanoconfinement to suppress particle agglomeration and improve reaction kinetics, and catalytic doping to reduce activation energy barriers for hydrogen dissociation and recombination. Among these approaches, the use of catalysts has proven particularly effective in improving hydrogen

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storage characteristics. Catalysts facilitate surface reactions, promote hydrogen dissociation and recombination, and reduce activation energy barriers, thereby accelerating overall kinetics. A wide range of catalytic materials, including transition metals, metal oxides, metal halides, and carbon-based compounds, have been explored in this context.

Notably, metal oxide catalysts have demonstrated considerable potential due to their ability to significantly enhance hydrogen sorption behavior, decrease desorption temperatures, and improve cyclic stability. These materials contribute to microstructural refinement, introduce defects, and create active sites that facilitate hydrogen transport and reaction processes.

In light of these developments, the present review aims to systematically examine the role of metal oxide catalysts in improving the hydrogen storage performance of MgH_2 . It provides a critical overview of recent advancements, mechanistic insights, existing challenges, and future research directions for the development of efficient solid-state hydrogen storage systems.

AI/LLM tools were used only for minor language editing and paraphrasing, and no scientific content was generated by these tools.

Methodology of review

The present review compiles and analyzes recent research on the catalytic enhancement of MgH_2 using metal oxide catalysts. A comprehensive literature search was conducted using databases including ScienceDirect, Scopus, Web of Science, and Google Scholar, covering publications primarily from the last two decades. The search strategy employed specific keywords such as " MgH_2 hydrogen storage," "metal oxide catalysts," "transition metal oxides," "hydrogen absorption/desorption kinetics," and "nanostructured catalysts," combined with appropriate Boolean operators to refine the results.

The selection of studies was based on clearly defined inclusion criteria, including experimental investigations on MgH_2 -based hydrogen storage systems reporting quantitative performance metrics such as hydrogen storage capacity (wt%), activation energy (kJ mol^{-1}), hydrogen desorption temperature ($^{\circ}\text{C}$), and kinetic improvement data. Studies lacking sufficient experimental or quantitative data were excluded. Peer-reviewed research articles published in English were included, while review articles and duplicate reports were considered only when they provided critical comparative or mechanistic insights supported by experimental evidence.

Both recent advances and foundational studies were included to ensure a balanced perspective. The selected literature was systematically analyzed to evaluate the influence of catalyst type, synthesis route, and structural modifications on hydrogen storage performance.

Fundamentals of hydrogen storage in MgH_2

Hydrogen storage in magnesium hydride (MgH_2) is governed by its thermodynamic and kinetic properties, which play a crucial role in determining its suitability for practical applications. A clear understanding of these fundamental aspects is essential for improving its hydrogen storage performance.

Thermodynamics

The thermodynamic behavior of MgH_2 determines the equilibrium conditions for hydrogen absorption and desorption. MgH_2 possesses a relatively high enthalpy of formation ($\approx -75 \text{ kJ}\cdot\text{mol}^{-1} \text{ H}_2$), indicating strong Mg-H bonding and high thermodynamic stability. While this stability ensures safe hydrogen storage, it also leads to high hydrogen desorption temperatures, typically above 300°C under ambient pressure conditions.

Such high temperatures are not suitable for practical applications, especially when compared to the target operating range of $40\text{--}85^{\circ}\text{C}$ set by the U.S. DOE. Therefore, modifying the thermodynamic properties of MgH_2 without significantly compromising its high storage capacity ($\sim 7.6 \text{ wt}\%$) remains a major research challenge.

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Kinetics

In addition to thermodynamics, the hydrogen storage performance of MgH_2 is strongly influenced by kinetic factors. The hydrogen absorption/desorption process involves multiple steps, including dissociation of hydrogen molecules, diffusion of hydrogen atoms into the bulk, nucleation, and phase transformation. However, MgH_2 exhibits inherently slow kinetics due to several limitations such as slow diffusion of hydrogen atoms within the MgH_2 lattice, limited active sites for nucleation and growth of phases, high activation energy barrier for hydrogen dissociation and recombination. These factors result in sluggish hydrogen uptake and release, significantly limiting its practical applicability. Enhancing the hydrogen sorption kinetics is therefore essential and is commonly achieved through mechanical ball milling, nanostructuring, and catalytic modification, which collectively reduce particle size, introduce lattice defects, increase surface area, and lower the activation energy for hydrogen dissociation and diffusion [7-17].

Metal oxide catalysts

Metal oxide catalysts play a crucial role in enhancing the hydrogen storage performance of MgH_2 by improving sorption kinetics and, in some cases, slightly modifying thermodynamic behavior. Their effectiveness arises from their ability to facilitate hydrogen-related reactions and induce favorable microstructural changes within the MgH_2 matrix.

Mechanism of catalysis

The catalytic enhancement of hydrogen storage in MgH_2 by metal oxides is primarily governed by their ability to modify both surface reactions and bulk diffusion processes. Metal oxide catalysts reduce the activation energy required for hydrogen absorption and desorption by providing alternative reaction pathways with lower energy barriers, thereby enabling faster kinetics and lower operating temperatures. During hydrogen uptake, these catalysts facilitate the dissociation of hydrogen molecules into atomic hydrogen on their surfaces, while during desorption they promote the recombination of hydrogen atoms into molecular hydrogen, significantly accelerating the overall sorption process. Additionally, strong interfacial interactions between MgH_2 and the dispersed metal oxide particles play a crucial role in improving performance. These interfaces act as active sites for hydrogen transfer and enable the spillover of hydrogen atoms from the catalyst surface into the MgH_2 lattice. Furthermore, the presence of metal oxides induces structural defects, grain boundaries, and lattice distortions within MgH_2 , which enhance hydrogen diffusion and provide nucleation sites for phase transformation. In some cases, partial reduction of metal oxides during cycling leads to the formation of metallic species that further boost catalytic activity. Collectively, these effects contribute to improved hydrogen sorption kinetics and reduced desorption temperatures in MgH_2 -based systems [18-20]. Figure 7 illustrates the catalytic mechanism of hydrogen absorption and desorption in MgH_2 .

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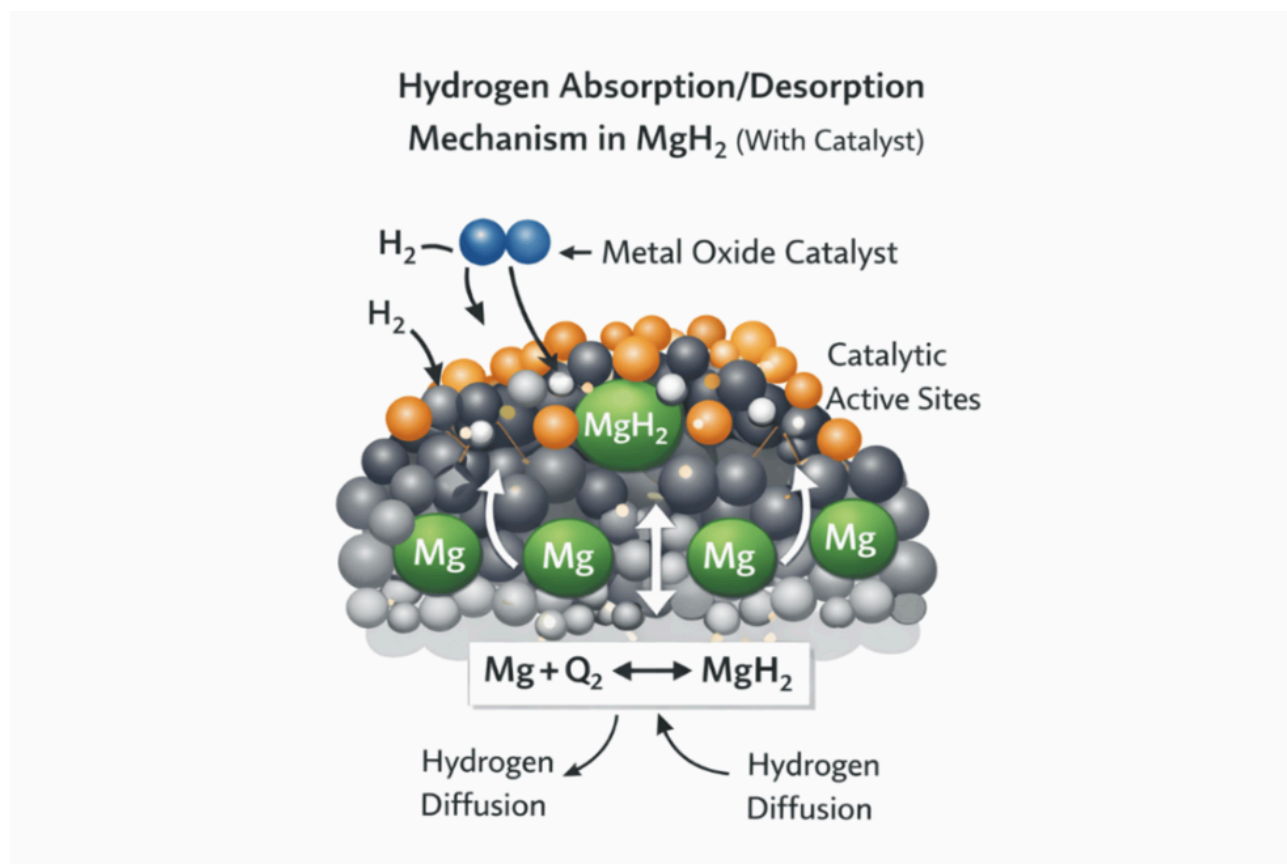


FIGURE 1: Hydrogen absorption/desorption mechanism in MgH_2 with catalyst

Review

Metal oxides

Various metal oxides have been investigated as catalysts for MgH_2 , each contributing differently to performance enhancement.

Transition Metal Oxides

Transition metal oxides are among the most widely studied catalysts owing to their high catalytic activity and ability to undergo reversible redox transformations during hydrogen sorption reactions. Common examples include TiO_2 , Fe_2O_3 , V_2O_5 , NiO , and Co_3O_4 . During cycling, these oxides may undergo partial reduction, resulting in the formation of catalytically active sub-stoichiometric oxide species and, in certain cases, metallic phases that contribute to enhanced hydrogen sorption kinetics. Among these catalysts, Ti-based oxides have demonstrated particularly significant effects in improving hydrogen absorption/desorption kinetics and lowering desorption temperatures [21-25].

Rare-Earth Oxides

Rare-earth oxides such as CeO_2 , La_2O_3 , and Y_2O_3 have also attracted considerable attention as catalysts for MgH_2 due to their distinct electronic structures and defect chemistry. CeO_2 is particularly effective because of its reversible $\text{Ce}^{4+}/\text{Ce}^{3+}$ redox behavior and oxygen vacancy formation, which facilitate hydrogen dissociation and diffusion. In contrast, La_2O_3

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and Y_2O_3 mainly improve hydrogen sorption through defect generation, lattice distortion, and enhanced interfacial diffusion kinetics. These oxides have also been reported to improve the thermal stability and cycling durability of MgH_2 systems [26-30].

Mixed Oxides

Mixed metal oxides, including spinel-type oxides such as NiFe_2O_4 and CoFe_2O_4 and perovskite-type oxides such as LaFeO_3 and LaNiO_3 , combine the advantages of multiple metal components and often exhibit synergistic catalytic effects. These materials can provide enhanced surface activity, improved catalyst dispersion, accelerated hydrogen diffusion, and superior structural stability during repeated hydrogenation/dehydrogenation cycles. Consequently, mixed metal oxides are increasingly being explored as advanced catalysts for next-generation MgH_2 -based hydrogen storage systems [31].

Recent advances: metal oxides and their catalytic effect on MgH_2

Recent studies have demonstrated that metal oxide catalysts significantly enhance the hydrogen storage performance of MgH_2 by improving sorption kinetics, lowering desorption temperatures, and promoting reversible hydrogen uptake. These improvements are mainly achieved through nanoscale design, defect engineering, and synergistic multi-component systems.

Catalyst addition is considered one of the most effective strategies to enhance the hydrogen sorption kinetics of MgH_2 , as it provides more active sites for hydrogen absorption/desorption reactions and accelerates reaction rates. Among various catalysts, metal oxides are the most widely used due to their strong catalytic activity and structural stability. During hydrogenation/dehydrogenation, these oxides may undergo partial reduction to lower valence states, forming intermediate species that facilitate electron transfer between Mg^{2+} and hydrogen. Additionally, metal oxides can disrupt the surface MgO layer, create diffusion pathways for hydrogen, and introduce grain boundaries and defects during processes such as ball milling, thereby significantly improving hydrogen sorption kinetics [32].

When different oxides such as Nb_2O_5 , TiO_2 , V_2O_5 , Cr_2O_3 , CuO , and Fe_3O_4 were added to MgH_2 ; Nb_2O_5 was found superior in comparison to other oxides in terms of improved kinetics. It provides a pathway for the sorption of hydrogen in metal-based materials and delivers fast sorption kinetics [33]. Barkhordarian et al. (2006) investigated the catalytic efficiency of Nb_2O_5 in MgH_2 and reported significantly enhanced hydrogenation/dehydrogenation kinetics compared with several other metal oxide catalysts [34], as illustrated in Figure 2. The figure demonstrates the strong catalytic influence of Nb_2O_5 on the desorption rate of MgH_2 , which has been mainly attributed to its ability to reduce activation energy and promote hydrogen diffusion. However, despite its excellent kinetic performance, the relatively high cost and limited abundance of Nb may pose challenges for large-scale commercial applications.

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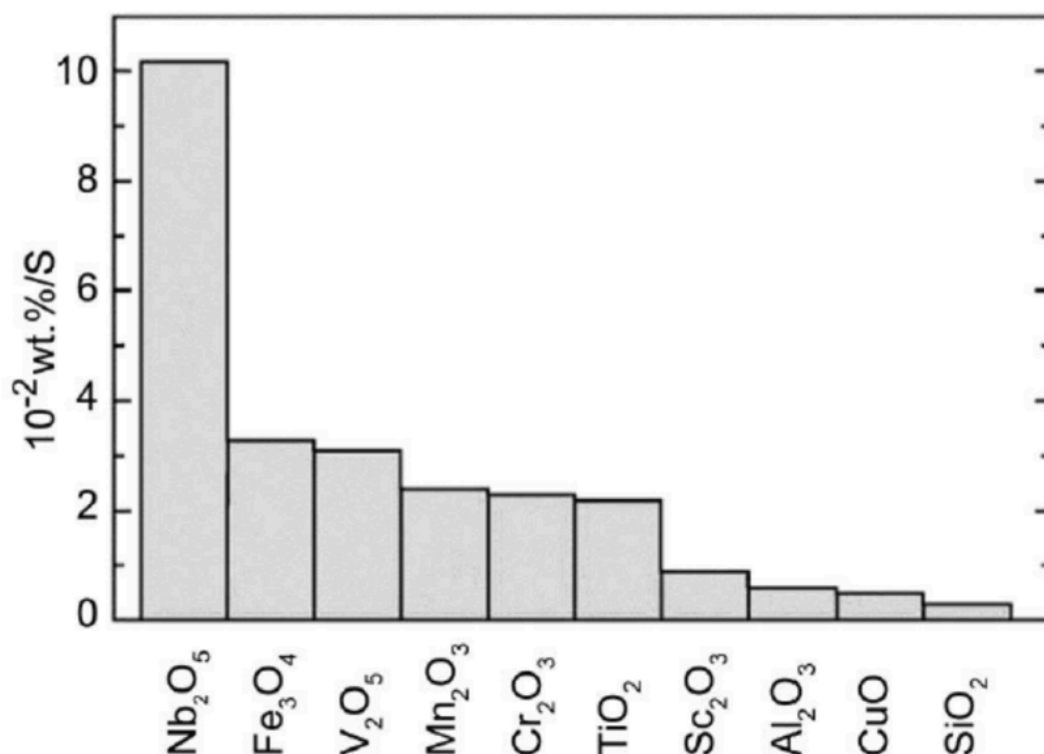


FIGURE 2: Numerous transition-metal oxides and their catalytic effect on the hydrogen desorption reaction rate of magnesium hydride

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Patah et al. [35] investigated the combined catalytic influence of Nb_2O_5 with a minor addition of Cr_2O_3 on MgH_2 and reported a marked enhancement in hydrogen sorption behavior. The co-doped system exhibited a substantial reduction in activation energy to ~ 136 kJ/mol, compared to ~ 206 kJ/mol for pristine MgH_2 . This improvement can be attributed to a synergistic catalytic mechanism in which Nb_2O_5 undergoes partial reduction during ball milling and cycling to form lower-valence Nb species ($\text{Nb}^{4+}/\text{Nb}^{3+}$) or NbO_x phases. These species act as active centers for hydrogen dissociation. Simultaneously, Cr_2O_3 contributes to defect formation and promotes charge transfer at the MgH_2 -catalyst interface, facilitating hydrogen diffusion and weakening the Mg-H bond, thereby accelerating both absorption and desorption processes.

Oelerich et al. [36] systematically studied a range of metal oxides, including TiO_2 , Sc_2O_3 , Cr_2O_3 , V_2O_5 , Fe_3O_4 , Mn_2O_3 , CuO , SiO_2 , and Al_2O_3 , and demonstrated that oxide additives significantly improve hydrogen sorption kinetics. The enhancement mechanism is generally associated with the formation of catalytically active interfaces and defect-rich regions. Transition metal oxides such as TiO_2 and V_2O_5 can undergo partial reduction under hydrogenation conditions, generating oxygen vacancies and lower oxidation states that act as active sites for H_2 dissociation. Cr_2O_3 , which showed the fastest absorption (~ 4.7 wt% at 300°C), is believed to promote rapid hydrogen uptake through defect-induced pathways and enhanced surface reactivity. In contrast, oxides such as V_2O_5 and Fe_3O_4 facilitate hydrogen desorption by weakening the Mg-H bond via electronic interactions and by providing fast diffusion pathways for hydrogen atoms through their partially reduced structures.

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Conceição et al. [37] reported improved hydrogen storage performance for MgH_2 doped with Nb_2O_5 , with absorption and desorption capacities of ~5.2 wt% and ~6.0 wt% at 300 °C. The catalytic effect of Nb_2O_5 is often linked to its in situ transformation into Nb hydride or NbO_x species during cycling, which enhances hydrogen spillover. In this mechanism, hydrogen molecules dissociate on the catalyst surface and migrate as atomic hydrogen onto the Mg matrix, thereby improving overall kinetics. Similarly, Aguey-Zinsou et al. [38] demonstrated that prolonged ball milling (up to 200 h) with a high Nb_2O_5 content (~17 wt%) leads to significant refinement of particle size and increased interfacial contact. This not only shortens hydrogen diffusion distances but also increases the density of active catalytic sites, resulting in faster sorption kinetics.

Overall, Nb_2O_5 -based systems are considered highly effective due to their ability to simultaneously provide multiple catalytic functions, including hydrogen dissociation, defect generation, and facilitation of hydrogen diffusion, making them superior among many oxide catalysts for MgH_2 .

ZrO_2 has also been explored as a catalyst for improving MgH_2 kinetics [39]. The MgH_2 - ZrO_2 system demonstrated hydrogen absorption of ~5.7 wt% at 150°C and retained ~2.9 wt% even at ambient temperature (298 K). The catalytic role of ZrO_2 is primarily associated with its ability to stabilize nanostructured MgH_2 and generate interfacial strain. This strain modifies the electronic environment of Mg-H bonds, reducing their stability and enabling hydrogen release at lower temperatures. Additionally, oxygen vacancies in ZrO_2 can act as trapping and diffusion sites for hydrogen atoms.

Yang et al. [40] compared TiO_2 , Al_2O_3 , and Fe_2O_3 as catalysts in MgH_2 systems and identified TiO_2 as the most effective, enabling hydrogen uptake of ~5.5 wt% within 5 minutes at 250°C. The superior performance of TiO_2 is attributed to its ability to form Ti^{3+} species and oxygen-deficient structures during processing. These features enhance electron transfer and create active sites for hydrogen dissociation. Furthermore, Ti-based species may form transient Ti-H bonds, which facilitate reversible hydrogen exchange and promote rapid kinetics. In contrast, Al_2O_3 , being relatively inert, primarily acts as a dispersing agent, while Fe_2O_3 contributes through redox cycling between $\text{Fe}^{3+}/\text{Fe}^{2+}$ states, albeit less effectively than TiO_2 .

Mustafa and Ismail [41] demonstrated that the addition of 5 wt% CeO_2 significantly improves the hydrogen sorption behavior of MgH_2 (Figure 3). The CeO_2 -doped MgH_2 sample released approximately 3.6 wt% H_2 within 30 min, whereas pristine MgH_2 desorbed only ~1.0 wt% H_2 under similar experimental conditions. The enhanced performance was attributed to the reversible $\text{Ce}^{4+}/\text{Ce}^{3+}$ redox behavior of CeO_2 , which promotes oxygen vacancy formation and facilitates hydrogen dissociation and diffusion. In addition, the CeO_2 surface may promote hydrogen spillover and weaken Mg-H bonding through electronic interactions, thereby improving desorption kinetics. The authors also reported a reduction in the apparent activation energy of hydrogen desorption from approximately 133 kJ mol⁻¹ for pure MgH_2 to about 108 kJ mol⁻¹ for the CeO_2 -catalyzed system. Although the reported low-temperature hydrogen sorption performance is highly promising, near-ambient-temperature hydrogen storage in MgH_2 -based systems remains a significant scientific challenge. Furthermore, the study provided limited long-term cycling stability analysis and mechanistic discussion; therefore, the reproducibility and practical applicability of the reported results require further comprehensive investigation and independent validation.

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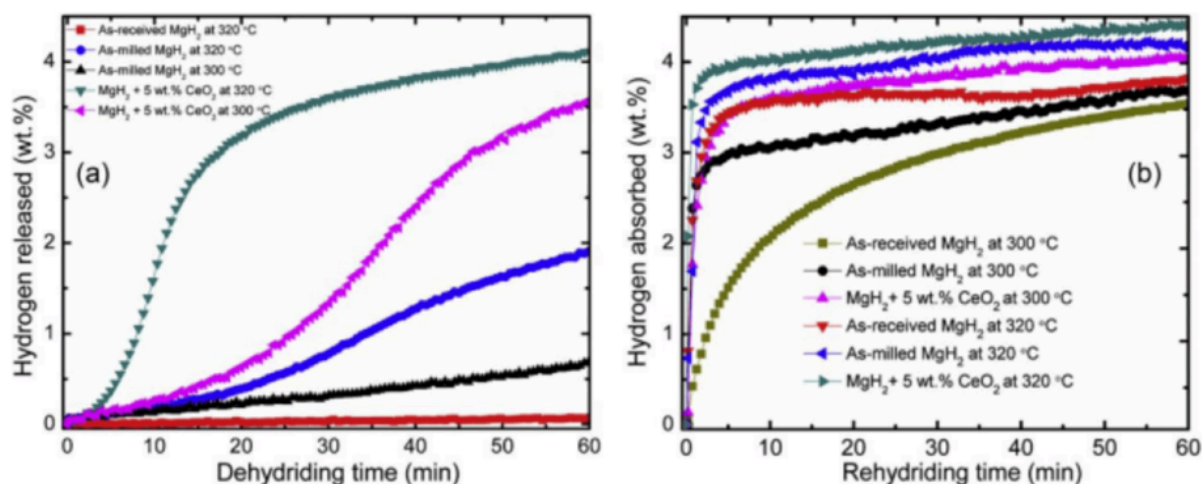


FIGURE 3: Isothermal dehydrogenation kinetics (a) and hydrogenation kinetics (b) of the as-milled MgH_2 and MgH_2 doped with 5 wt% CeO_2 at 320°C and 300°C

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Dan et al. [42] investigated the effect of MoO_3 incorporation into MgH_2 via mechanical milling and reported a significant enhancement in hydrogen desorption kinetics. The modified system exhibited rapid hydrogen release, achieving approximately 5 wt% within 900 s at 300°C. Additionally, the activation energy for dehydrogenation was markedly reduced to 114.7 kJ/mol. This improvement can be attributed to the partial reduction of MoO_3 during cycling, leading to the formation of lower-valence Mo species (e.g., $\text{Mo}^{5+}/\text{Mo}^{4+}$) and oxygen-deficient structures. These features provide active sites for hydrogen dissociation and facilitate electron transfer, thereby weakening the Mg-H bond and accelerating hydrogen release.

In recent years (2020-2026), significant progress has been made in the design of advanced metal oxide catalysts, particularly focusing on nanostructured [43-47], defect-engineered, and multi-component systems [48-55] with enhanced catalytic efficiency. In addition to the conventional oxide catalysts discussed above, recent studies have increasingly focused on the development of advanced composite and heterostructured oxide systems to further enhance the hydrogen storage performance of MgH_2 . In particular, nanostructured oxides have demonstrated superior catalytic behavior due to their high surface-to-volume ratio, uniform dispersion, and increased density of active sites. These systems combine multiple catalytic functionalities, resulting in significantly improved kinetics and reduced operating temperatures. Nanoscale catalysts such as TiO_2 , CeO_2 , and Fe_3O_4 , especially when integrated with conductive supports like carbon matrices or graphene, promote faster hydrogen transport and reduce kinetic barriers. At the atomic level, these systems enhance hydrogen spillover, where dissociated hydrogen atoms migrate from the catalyst surface to the MgH_2 matrix, resulting in improved sorption kinetics and cycling stability [56]. Furthermore, advanced morphologies such as two-dimensional nanosheets and porous oxide frameworks provide larger interfacial areas and inhibit particle agglomeration during repeated hydrogenation/dehydrogenation cycles. For instance, $\text{Nb}_2\text{O}_5\text{-TiO}_2$ -based composite catalysts have demonstrated hydrogen desorption capacities of ~6 wt% within 5-10 min at 300°C, along with activation energy reductions exceeding 40% compared to pristine MgH_2 . Similarly, heterostructured $\text{TiO}_2@\text{Nb}_2\text{O}_5$ systems have been reported to lower the initial desorption temperature to ~160-180°C, representing a reduction of more than 100°C, thereby indicating substantial progress toward low-temperature hydrogen storage. Recently, composite catalysts such as $\text{Nb}_2\text{O}_5@\text{TiO}_2\text{-C}$ have demonstrated exceptional performance, enabling hydrogen release of >6 wt% within 5 min at 300°C and rapid absorption of ~6 wt% within 1 min at 200°C, along with ~42% reduction in activation energy [57].

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Another key strategy involves the deliberate introduction of oxygen vacancies within metal oxides to enhance catalytic activity. Oxygen-deficient materials such as TiO_{2-x} , CeO_{2-x} , and $\text{Co}_3\text{O}_{4-x}$ exhibit a high concentration of defect sites that serve as active centers for hydrogen molecule dissociation and recombination [58]. These vacancies alter the electronic structure of the catalyst, often inducing mixed valence states and facilitating charge transfer between the catalyst and MgH_2 . As a result, the Mg-H bond strength is reduced, lowering the desorption temperature and improving overall reaction kinetics. Additionally, vacancy-rich structures support the formation of intermediate species during hydrogen cycling, contributing to enhanced reversibility and long-term stability compared to fully stoichiometric oxides.

More recently, MXene-derived metal oxides have emerged as promising next-generation catalysts for MgH_2 -based systems. MXenes, particularly titanium-based compositions, possess layered structures and high electrical conductivity, enabling uniform dispersion of catalytic species. Upon oxidation or thermal treatment, these materials transform into defect-rich oxide phases with multiple valence states, which enhance catalytic reactivity and hydrogen diffusion pathways [59-61]. The strong interfacial interaction between MXene-derived oxides and MgH_2 further facilitates efficient charge transfer and accelerates hydrogen sorption processes, resulting in improved kinetics and reduced operating temperatures. In particular, advanced MXene-derived $\text{Nb}_2\text{O}_5\text{-C/Ti}_3\text{C}_2\text{T}_x$ catalysts have been reported to reduce the activation energy to as low as $\sim 62.5 \text{ kJ mol}^{-1}$ while maintaining excellent cycling stability ($\sim 99.3\%$ capacity retention after 50 cycles). Although these results demonstrate the significant potential of MXene-based catalysts for enhancing MgH_2 performance, the long term structural stability, large-scale synthesis feasibility, oxidation sensitivity of MXenes, and cost-effectiveness of such complex catalyst systems still require further investigation before practical implementation can be fully realized [62].

In addition, hybrid and multi-component catalyst systems have demonstrated substantial improvements due to synergistic interactions among their constituents. Composites incorporating metal oxides with transition metals, carbon-based materials, or other oxides-such as $\text{Ni/TiO}_2/\text{graphene}$, Co_3O_4 -based hybrids, and oxide-MXene composites-enhance hydrogen storage performance through multiple complementary mechanisms [63]. These systems typically achieve hydrogen absorption capacities of $\sim 5.5\text{-}6.5 \text{ wt}\%$ within a few minutes at temperatures of $200\text{-}300^\circ\text{C}$, owing to enhanced hydrogen spillover and improved electronic conductivity. The presence of carbon supports also prevents catalyst agglomeration and stabilizes nanostructured MgH_2 during repeated cycling.

Another important advancement involves defect-engineered oxides with a high concentration of oxygen vacancies. Materials such as TiO_{2-x} and CeO_{2-x} exhibit significantly enhanced catalytic activity due to the presence of unsaturated coordination sites, which facilitate hydrogen dissociation and recombination. These systems have demonstrated activation energy reductions to below $\sim 100 \text{ kJ/mol}$ in optimized cases, approaching the threshold required for practical hydrogen storage applications.

In addition, recent experimental and theoretical investigations have explored the role of bimetallic and ternary oxide systems, where synergistic interactions between multiple metal species create highly active catalytic interfaces. For example, V-Nb oxide systems and NiO-based hybrid catalysts have shown improved hydrogen diffusion and reduced thermodynamic barriers through multi-valence redox behavior and enhanced charge transfer. Some of these systems exhibit partial hydrogen absorption even at near-ambient temperatures, highlighting their potential for next-generation applications.

As summarized in Table 7, transition metal oxides such as TiO_2 and Fe_3O_4 primarily enhance hydrogen storage performance by reducing activation energy barriers and facilitating hydrogen diffusion. In contrast, rare-earth oxides like CeO_2 contribute through oxygen vacancy-mediated mechanisms and redox flexibility. Furthermore, binary and ternary oxide systems (e.g., $\text{Sc}_2\text{O}_3/\text{TiO}_2$) exhibit strong synergistic effects, resulting in improved kinetics and increased hydrogen storage capacity. Composite catalysts, such as $\text{Co/CeO}_2@\text{C}$, further demonstrate that coupling metal oxides with conductive supports significantly enhances catalytic efficiency through combined effects of hydrogen spillover, electron transfer, and structural stabilization.

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Metal Oxide Catalyst	Type/Structure	Key Catalytic Effects on MgH_2	Performance Improvement	Mechanism	Reference
TiO_2	Transition metal oxide/nanosheets	Reduces desorption temperature and activation energy	Onset desorption ↓ to $\sim 196^\circ\text{C}$; E_a ↓ to ~ 75.7 kJ/mol	Oxygen vacancies + multivalent states enhance H diffusion and active sites	[56]
CeO_2	Rare-earth oxide	Improves hydrogen diffusion and cycling stability	E_a reduced from ~ 145 to ~ 93 kJ/mol (with composites)	Oxygen vacancies + $\text{Ce}^{3+}/\text{Ce}^{4+}$ redox improve hydrogen spillover	[64]
Fe_3O_4	Transition metal oxide / heterojunction	Enhances sorption kinetics and hydrogen transfer	Faster absorption/desorption kinetics	Formation of heterointerface promotes electron transfer and catalytic activity	[65]
$\text{Sc}_2\text{O}_3/\text{TiO}_2$	Binary oxide (bimetallic)	Significant improvement in hydrogen storage capacity and kinetics	~ 6.08 wt% H_2 absorption at 200°C ; rapid desorption	Synergistic catalytic effect and uniform dispersion of active sites	[56]
$\text{Co}/\text{CeO}_2@\text{C}$	Composite oxide with carbon support	Strong enhancement in kinetics and stability	5.15 wt% H_2 desorbed at 300°C ; E_a ↓ to ~ 93 kJ/mol	Triple synergy: hydrogen spillover + electron transfer + confinement stabilization	[64]
Nb-doped TiO_2 ($\text{Ti}/\text{Nb}_{0-15}\text{O}_2$)	Doped oxide/nanosheets	Improves kinetics and cycling stability	6.3 wt% H_2 released in 7 min at 300°C	Doping creates defects and suppresses agglomeration	[56]
Mixed/Ternary Oxides	Multi-component oxides	Superior catalytic activity compared to single oxides	Lower activation energy + faster kinetics	Synergistic effects, defect engineering, improved dispersion	[66]

TABLE 1: Catalytic role of metal oxides in MgH_2

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Overall, these recent developments, supported by substantial quantitative improvements in activation energy (down to ~75-100 kJ/mol), hydrogen capacity (~5-6.5 wt%), and reduced operating temperatures (<200-300°C), highlight the critical role of advanced catalyst design strategies in overcoming the kinetic limitations of MgH₂ and advancing its practical application as an efficient hydrogen storage material.

Comparative performance of metal oxide catalysts

The performance of metal oxide catalysts in MgH₂ systems is typically evaluated based on desorption temperature, activation energy, hydrogen capacity, and cycling stability. The addition of catalysts significantly enhances hydrogen sorption kinetics and reduces the energy barrier compared to pure MgH₂.

Transition metal oxides such as TiO₂, Fe₃O₄, and Nb₂O₅ effectively lower desorption temperature and activation energy, while rare-earth oxides like CeO₂ improve cycling stability and reversibility. Overall, most catalysts maintain high hydrogen capacity (~6 wt%) while mainly improving kinetic properties. A comparative summary of these effects is presented in Table 2.

Catalyst System	Desorption Temp (°C)	Activation Energy (kJ/mol)	Hydrogen Capacity (wt%)	Cycling Stability	Reference
Pure MgH ₂	~300–350	~160	~7.6	Poor	[6]
MgH ₂ + TiO ₂	~200–250	~75–90	~6.5–7.0	Good	[56]
MgH ₂ + CeO ₂	~250	~90–110	~6.0–6.8	Excellent	[64]
MgH ₂ + Fe ₃ O ₄	~220–270	~85–100	~6.5	Good	[65]
MgH ₂ + Nb ₂ O ₅	~200	~70–80	~6.8	Very good	[56]

TABLE 2: Performance comparison of MgH₂ with different catalysts

Table 2 clearly demonstrates that the incorporation of metal oxide catalysts significantly enhances the hydrogen storage performance of MgH₂ compared to the pristine material. In particular, catalysts such as TiO₂ and Nb₂O₅ effectively reduce activation energy and desorption temperature, while maintaining high hydrogen storage capacity. These improvements highlight the importance of catalyst selection in optimizing MgH₂-based systems.

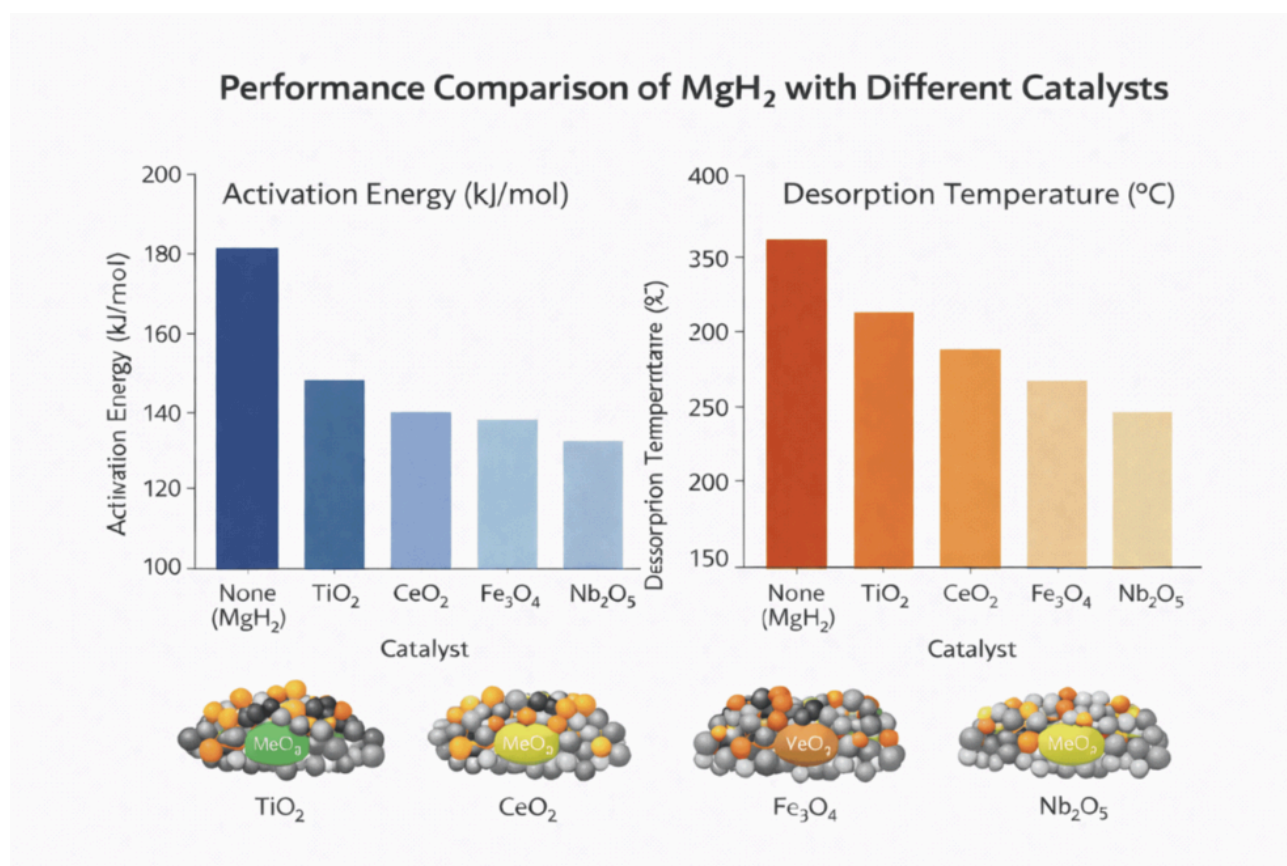


FIGURE 4: Comparison of MgH_2 with different metal oxide catalysts

Figure 4 compares the activation energy and desorption temperature of MgH_2 with various metal oxide catalysts. The addition of catalysts significantly reduces both activation energy and desorption temperature compared to pure MgH_2 , indicating enhanced hydrogen sorption kinetics and improved storage performance.

Challenges and limitations

Despite significant progress in enhancing the hydrogen storage performance of MgH_2 using metal oxide catalysts, several challenges and limitations still hinder their large-scale practical application. One of the primary issues is the agglomeration of catalysts during repeated hydrogenation and dehydrogenation cycles, especially under high-temperature conditions. This leads to a reduction in active surface area and catalytic efficiency over time. Additionally, although many advanced catalysts such as nanostructured oxides and hybrid systems exhibit excellent performance at the laboratory scale, cost and scalability remain critical concerns. The synthesis of highly engineered nanomaterials, including defect-rich oxides and MXene-derived catalysts, often involves complex, energy-intensive, and expensive processes, which limit their commercial viability. Furthermore, long-term degradation and cycling stability pose serious challenges, as catalysts may undergo structural changes, oxidation, sintering, or loss of active sites during prolonged use. These factors can deteriorate hydrogen storage capacity and kinetics, making it essential to develop more robust and economically feasible catalytic systems for real-world applications.

How to cite this article:

Sharma S (May 28, 2026) Catalytic Enhancement of Hydrogen Storage in MgH_2 Using Metal Oxide Catalysts: Recent Advances and Future Perspectives . Cureus J Eng 3 : es44388-026-00087-6. DOI <https://doi.org/10.7759/s44388-026-00087-6>

Conclusions

In conclusion, metal oxide catalysts have demonstrated remarkable potential in overcoming the kinetic limitations of MgH_2 , significantly advancing its viability for solid-state hydrogen storage. Across the reviewed studies, catalytic modification has been shown to significantly reduce the activation energy of hydrogen desorption in MgH_2 . While pristine MgH_2 generally exhibits activation energies of approximately $200\text{--}210\text{ kJ mol}^{-1}$, catalyst-modified systems commonly display reduced values in the range of $\sim 114\text{--}136\text{ kJ mol}^{-1}$, corresponding to an overall reduction of about 30–45%, depending on the catalyst type and processing conditions employed. In addition, catalytic modification can lower the hydrogen desorption temperature of MgH_2 by approximately $50\text{--}150^\circ\text{C}$, while optimized systems have demonstrated hydrogen release capacities of $\sim 5\text{--}6.5\text{ wt\%}$ within 10–15 min at temperatures below $\sim 200\text{--}300^\circ\text{C}$, depending on the catalyst composition and operating conditions. Some advanced systems, such as TiO_2 - and ZrO_2 -based composites, further demonstrate hydrogen uptake of $\sim 5.5\text{--}5.7\text{ wt\%}$ within minutes at temperatures as low as $150\text{--}250^\circ\text{C}$. These improvements are enabled through strategies such as nanostructuring, oxygen vacancy engineering, and the development of multi-component and MXene-derived catalyst systems, which enhance hydrogen dissociation, diffusion, and reversibility.

Metal oxides are particularly effective due to their strong catalytic activity, structural stability, and ability to form intermediate species and defect-rich interfaces that facilitate hydrogen transport. Among the various catalysts, Nb_2O_5 and TiO_2 based systems have demonstrated particularly significant improvements in hydrogen sorption performance due to their ability to reduce activation energy, facilitate defect generation, and enhance hydrogen diffusion/spillover processes. Overall, these catalytic advances have brought MgH_2 -based materials significantly closer to practical hydrogen storage applications.

Looking ahead, future research should focus on addressing key challenges related to catalyst agglomeration, cost, scalability, and long-term cycling stability. A major objective is to further reduce the desorption temperature below 200°C and achieve activation energies below 100 kJ/mol for practical deployment. The integration of artificial intelligence and machine learning in materials discovery is expected to accelerate the identification of optimized catalyst compositions with tailored electronic and structural properties. Additionally, the design of advanced nanostructures, including hierarchical, defect-engineered, and hybrid systems, will play a crucial role in enhancing catalytic efficiency and durability.

Emphasis should also be placed on scalable synthesis routes and improving material robustness under realistic operating conditions. Importantly, the integration of MgH_2 -based hydrogen storage systems with renewable energy sources such as solar and wind energy presents a sustainable pathway for clean energy storage and utilization. Furthermore, magnesium-based energy storage technologies, including emerging Mg batteries, offer the potential for higher energy densities and improved safety compared to conventional lithium-based systems, further highlighting the importance of continued research in magnesium-based materials. Overall, sustained innovation in catalyst design, quantitative performance optimization, and system-level integration will be essential for realizing the full potential of MgH_2 in future hydrogen-based energy systems.

Additional Information

Author Contributions

All authors have reviewed the final version to be published and agreed to be accountable for all aspects of the work.

Concept and design: Shashi Sharma

Acquisition, analysis, or interpretation of data: Shashi Sharma

Drafting of the manuscript: Shashi Sharma

Critical review of the manuscript for important intellectual content: Shashi Sharma

How to cite this article:

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Disclosures

Conflicts of interest: In compliance with the ICMJE uniform disclosure form, all authors declare the following: **Payment/services info:** All authors have declared that no financial support was received from any organization for the submitted work. **Financial relationships:** All authors have declared that they have no financial relationships at present or within the previous three years with any organizations that might have an interest in the submitted work. **Other relationships:** All authors have declared that there are no other relationships or activities that could appear to have influenced the submitted work.

Data Availability Statements

Data sharing is not applicable. This work is theoretical; no datasets were generated or analyzed.

Acknowledgements

The author sincerely thanks the International Conference on Renewable Energy (ICRE) for providing a platform to present this work and for the valuable feedback that helped improve this study.

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