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Development and Research Status of Magnesium-Based Hydrogen Storage Materials

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Abstract: MgH_2 has a theoretical gravimetric hydrogen-storage capacity of approximately 7.6 wt.%; however, the stable Mg-H bond and the surface oxide layer that hinders hydrogen dissociation and diffusion lead to slow activation and high hydrogen-release temperatures in pure Mg systems. In recent years, research on Mg-based hydrogen storage materials has shifted from a single emphasis on capacity enhancement to the coupled regulation of thermodynamics, interfacial catalysis, cycling microstructure and heat exchange in material beds. In Mg-Ni systems, Mg_2Ni/Mg_2NiH_4 pathways improve hydrogen dissociation and diffusion; rare-earth elements often stabilize microstructures through LPSO phases, rare-earth hydrides and multiphase interfaces; and MXenes, high-entropy oxides and transition-metal sulfides mainly reduce the energy barriers for Mg-H bond cleavage and H migration. Reported samples can release approximately 5 wt.% hydrogen at 573 K or achieve rapid dehydrogenation at 250 ℃. Nevertheless, when powder-level performance is translated into devices, it remains constrained by cost, capacity dilution, bed-temperature gradients and cyclic pulverization. From an engineering perspective, Mg-based materials should not be evaluated only by peak temperature and single-cycle capacity; hydrogen pressure, cycle number, bed-temperature gradient and the mass of heat-exchange structures should also be reported.

1. Introduction

With the expansion of renewable-power-driven hydrogen production, the safety, energy consumption and system mass associated with hydrogen storage have become practical constraints. High-pressure gaseous storage provides fast response, but the vessels are heavy and require costly safety protection. Liquid hydrogen offers high volumetric density, yet it depends on cryogenic liquefaction and continuous thermal insulation. Metal hydrides store hydrogen through reversible reactions at relatively low operating pressures, making them more suitable for stationary energy storage, distributed hydrogen supply and integration with industrial waste heat.

The appeal of Mg-based materials arises from both capacity and resource advantages: metallic Mg is lightweight and abundant, and MgH_2 has a theoretical gravimetric hydrogen-storage capacity of approximately 7.6 wt.%. However, the reaction enthalpy of the Mg/MgH_2 system is approximately 74–76 kJ mol^{-1} H_2, and hydrogen release from pure Mg systems usually requires temperatures above 300 ℃ [1,2]. The critical question is therefore not whether hydrogen can be stored, but whether reversible capacity can be retained under acceptable temperature, pressure and cycle-number conditions. Recent review and application-oriented studies have therefore shifted attention from powder-level capacity toward alloy design, catalytic/nanostructural modification and reactor-level heat management [3,4].

2. Overview of Mg-Based Hydrogen Storage Materials

2.1. Development History of Mg-Based Hydrogen Storage Materials

Early studies mainly compared two reaction pathways, Mg-H and Mg-Ni-H. MgH_2 has a high capacity but is difficult to activate before hydrogen absorption. Mg_2Ni can form Mg_2NiH_4, with a theoretical capacity of about 3.6 wt.%; although this is lower than that of MgH_2, it provides faster hydrogen dissociation and transport channels. Mechanical ball milling, rapid solidification and powder metallurgy were subsequently used to refine grains and introduce defects, and multiphase interfaces together with short-range diffusion pathways gradually became central to kinetic regulation.

Since the beginning of the twenty-first century, rare-earth elements, transition metals, carbon materials and oxide catalysts have been introduced into Mg-based systems. Rare-earth elements can form LPSO phases, eutectic structures or rare-earth hydrides; Ni, Ti, Fe and their compounds mainly accelerate hydrogen dissociation and Mg-H bond cleavage; and recent reviews summarize catalytic and nanostructuring strategies involving high-entropy oxides, MXenes and transition-metal sulfides [5,6]. The research focus has also expanded from powder PCT curves to shaped materials, heat exchangers and tonne-scale storage and transport devices. The development route is summarized in Figure 1. Overall, the development of Mg-based hydrogen storage materials has moved from proving reversible hydrogen storage to controlling the coupled behavior of thermodynamics, kinetics and microstructure. Early work focused on whether Mg or Mg-Ni phases could absorb and release hydrogen reversibly, whereas recent studies increasingly emphasize how phase boundaries, catalytic interfaces and material-bed structures work together under repeated cycling.

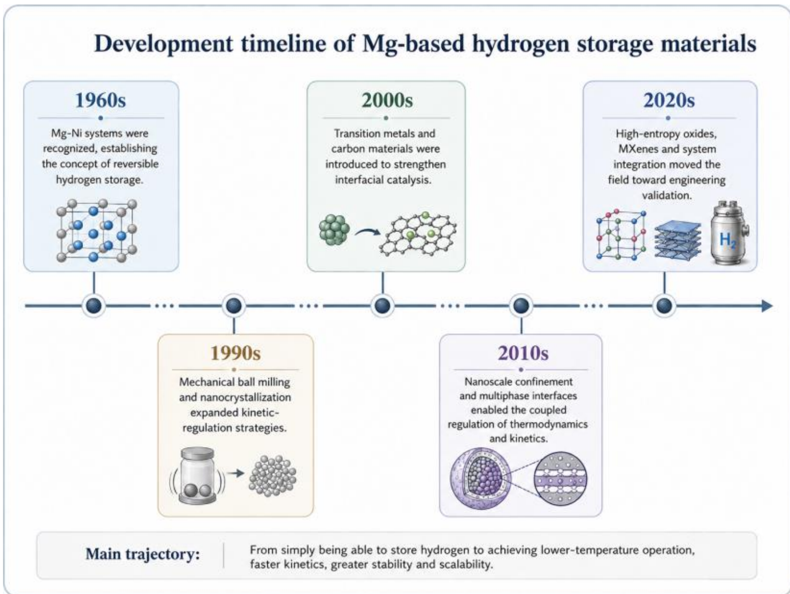

Figure 1. Development timeline of Mg-based hydrogen storage materials.

2.2. Hydrogen-storage mechanism of Mg/MgH_2

The reversible Mg/MgH_2 reaction can be written as $Mg + H_2 \rightleftharpoons MgH_2$. Hydrogen absorption involves H_2 adsorption and dissociation at the surface, diffusion of H atoms along grain boundaries or defects, and nucleation and growth of MgH_2. Hydrogen desorption proceeds through Mg-H bond cleavage, H-atom migration, H_2 recombination and desorption. Heat effects and mass-transfer pathways jointly control the reaction rate; no single elementary step is sufficient to explain the complete absorption/desorption curve.

The surface oxide layer on pure Mg impedes H_2 dissociation, while the MgH_2 layer that forms afterward further extends the solid-state diffusion path. The exothermic absorption process can cause local heating in the bed, whereas the endothermic desorption process is easily limited by insufficient heat supply. The four stages shown in Figure 2 correspond to surface reaction, bulk diffusion, phase-boundary migration and heat transfer. Catalysts mainly address the first two kinetic resistances, alloying changes hydride stability and nucleation sites, and reactor heat exchange determines whether powder performance can be converted into output capability at the material-bed level.

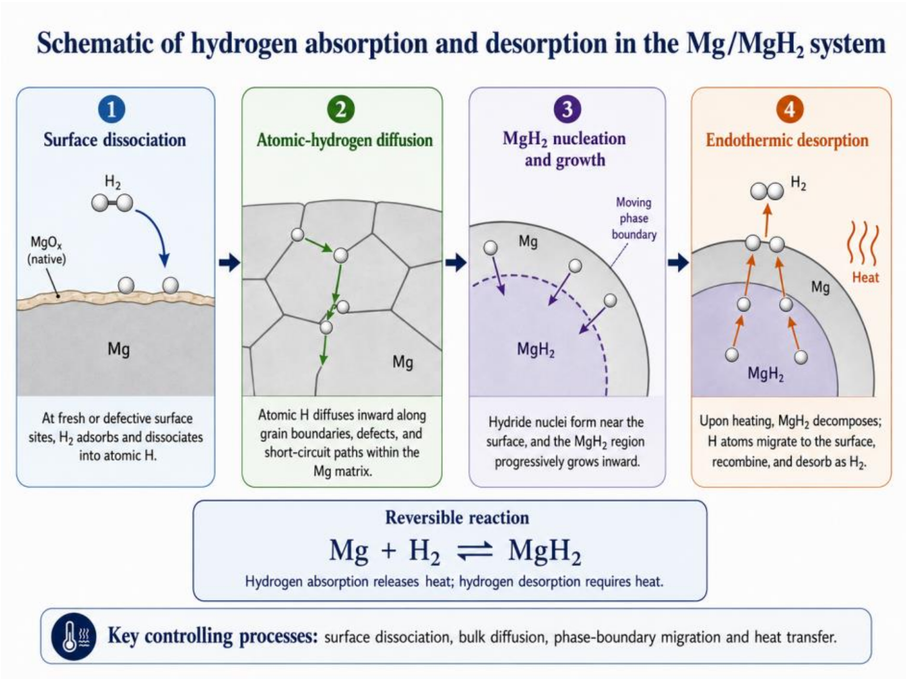

Figure 2. Schematic of hydrogen absorption and desorption in the Mg/MgH_2 system.

2.3. Advantages and limitations of Mg-based hydrogen storage materials

Compared with metal hydrides such as $LaNi_5$ and TiFe, MgH_2 has a higher theoretical gravimetric capacity; compared with lightweight complex hydrides, the Mg/MgH_2 reaction path is relatively simple. Mg-Ni and Mg-RE multiphase alloys sacrifice part of the capacity, but they can provide hydrogen dissociation and diffusion pathways at interfaces. Table 1 lists the capacities and limitations of several representative systems. These data are suitable for materials screening, but they should not be directly equated with system-level hydrogen storage density after containers, heat exchangers and safety margins are included.

The drawbacks of Mg-based materials are concentrated at three levels: thermodynamics, kinetics and engineering. The stability of MgH_2 makes low-temperature dehydrogenation difficult; oxide films, MgH_2 shells and particle agglomeration extend diffusion paths; and the low thermal conductivity of powder beds readily produces temperature gradients during hydrogen absorption and desorption. Nanostructuring and catalysts can increase the initial rate, but they also introduce oxidation, pyrophoric risk, catalyst agglomeration and capacity dilution.

Table 1. Major performance characteristics of representative Mg-based hydrogen storage materials.

Material system	Capacity or representative index	Main functional features	Main limitations
Mg/MgH_2	Theoretical capacity about 7.6 wt.%; dehydrogenation usually requires temperatures above 300 ℃	High capacity, clear reaction pathway and suitable for thermally coupled hydrogen storage	Stable MgH_2, difficult activation and poor thermal conductivity of powder beds
Mg_2Ni/Mg_2NiH_4	Theoretical capacity about 3.6 wt.%	Ni promotes H_2 dissociation and kinetics are usually better than those of pure Mg	Capacity is markedly lower than MgH_2 and medium-to-high-temperature heating is still required
$Mg_{85}Ni_{13}Y_2$	Absorbs about 4.96 wt.% H_2 within 5 min at 613 K and 3 MPa; capacity retention >97% after 20 cycles	Y induces multiphase interfaces and rare-earth hydrides, improving desorption rate and cycling stability	Rare-earth addition increases cost and dilutes gravimetric capacity
Mg-Ni-Y/TiO_2@C	Absorbs about 5.32 wt.% H_2 at 200 ℃ and 30 bar	LPSO phase and TiO_2@C catalytic interfaces act synergistically, enabling rapid initial absorption	Composite preparation is complex and long-term stability of catalytic phases requires verification
MgH_2/high-entropy oxide	Releases about 5.56 wt.% H_2 within 20 min at 250 ℃	Multielement oxides form composite active interfaces during cycling and lower the activation energy	High additive loading may reduce effective capacity

3. Research Progress in Modification Technologies for Mg-Based Hydrogen Storage Materials

3.1. Alloying modification

Alloying affects hydrogen-storage behavior by changing phase constitution, electronic structure and phase-boundary distribution. Ni can form Mg_2Ni/Mg_2NiH_4 channels and promote H_2 dissociation. Rare-earth elements such as Y, Gd and Ce can induce LPSO phases, eutectic structures or rare-earth hydride interfaces. Elements such as Al, Ti and V may alter Mg-H bond stability through solid solution or second-phase formation. The key issue in Mg-Ni systems is not simply to increase the Ni content, but to control phase boundaries, defects and the distribution of catalytic phases [7].

Recent data on Mg-Ni-RE alloys show that rare-earth and microalloying elements are particularly sensitive to the activation process. Na-microalloyed Mg-Ni-Gd-Y-Zn-Cu alloys can shorten the initial activation process [8]. $Mg_{85}Ni_{13}Y_2$ absorbs approximately 4.96 wt.% hydrogen within 5 min at 613 K and 3 MPa H_2, releases approximately 5.00 wt.% hydrogen within 7 min at 573 K, and retains more than 97% of its capacity after 20 cycles [9]. It should also be noted that reducing energy barriers by alloying inevitably introduces non-hydrogen-storage constituents, while complex alloys increase the difficulty of melting, compositional segregation control and cost management. Thus, alloying should be regarded as a balance between destabilization, interface construction and capacity retention, rather than as a simple search for lower dehydrogenation temperature.

3.2. Catalyst modification

Catalytic modification mainly addresses H_2 dissociation, H-atom migration and Mg-H bond cleavage. Ni, Ti, Fe and their oxides or sulfides can provide active sites, while carbon materials, graphite sheets and MXenes can improve catalyst dispersion and suppress particle agglomeration. In situ growth of NiS_2 on graphite sheets [10], high-entropy-oxide catalysis and TiO_2@C loading follow the same logic: active phases are anchored at relatively stable interfaces to reduce agglomeration and deactivation during cycling.

High-entropy oxides and composite catalysts show particularly strong low-temperature kinetic performance. For example, MgH_2 catalyzed with 10 wt.% Cr1:1 high-entropy oxide can release approximately 5.56 wt.% hydrogen within 20 min at 250 ℃ and reabsorb approximately 6.0 wt.% hydrogen within 8 min at 150 ℃ [11]. Mg-Ni-Y/TiO_2@C can absorb approximately 5.32 wt.% hydrogen at 200 °C and 30 bar H_2 [12]. Importantly, catalysts do not directly change the equilibrium thermodynamics of MgH_2. A lower peak temperature, a higher isothermal rate and cycling capacity retention should therefore be evaluated separately, and excessive catalyst loading can reduce the overall gravimetric capacity.

3.3. Nanostructuring modification

Nanostructuring improves MgH_2 nucleation and decomposition by shortening diffusion distances and increasing the density of grain boundaries and defects. Mechanical ball milling can rapidly refine particles and introduce dislocations; rapid solidification can generate fine multiphase microstructures; and nanoscale confinement can use carbon frameworks, oxide pores or two-dimensional materials to suppress Mg/MgH_2 grain growth. Size reduction alone usually cannot solve low-temperature kinetics and cycling stability simultaneously, so it is often combined with catalysts [6].

MXenes combine two-dimensional interfaces with tunable surface functional groups. They can host active components such as Ni, Ti and V and may also participate directly in electron transfer [13]. Their advantages lie in dispersing catalytic phases and increasing H-atom migration channels. Their limitations are equally direct: as powder specific surface area increases, oxidation becomes easier; after pelletization or packing, reduced porosity changes gas permeation and heat-transfer conditions. If the nanostructure cannot be retained during cycling, the initial performance advantage decays rapidly.

3.4. Composite modification technologies

Composite modification is not simply the superposition of alloying elements, catalysts and nanoscale carriers. Instead, it assigns functions according to the rate-limiting steps: alloying regulates hydride stability and phase boundaries, catalysts lower the barriers for H_2 dissociation and Mg-H bond cleavage, and carbon materials or MXenes improve dispersion and heat conduction. Reported composite systems involving high-entropy oxides, TiO_2@C, MXenes and transition-metal/rare-earth co-catalysts illustrate this division of functions [11-14].

The main difficulty in composite systems is mechanistic attribution. If a catalyst transforms into a mixed metal, oxide or hydride phase during the first absorption/desorption process, the initial composition is not the same as the active phase during cycling. Evaluation should therefore report additive amount, effective capacity, temperature/pressure conditions, cycle number and phase-structure evolution at the same time. Figure 3 summarizes the technological routes for modification, Table 2 compares the targets and limitations of different routes, and Figure 4 presents the corresponding performance-enhancement mechanisms.

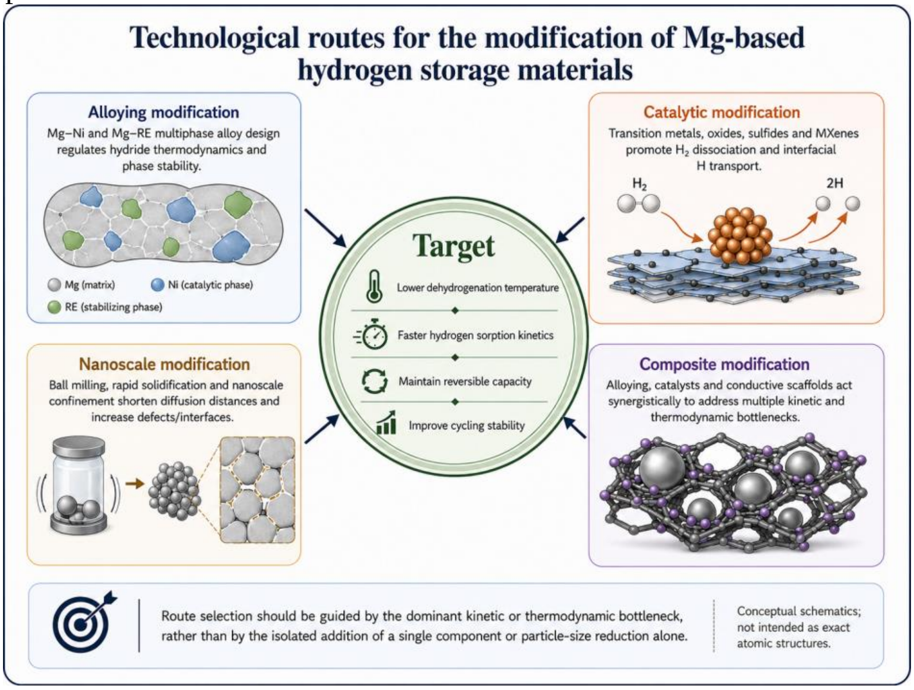

Figure 3. Technological routes for the modification of Mg-based hydrogen storage materials.

Table 2. Comparison of major modification technologies.

Modification technology	**Representative objects**	**Mode of performance improvement**	**Limitations**
Alloying	Mg-Ni, Mg-RE and LPSO phases	Constructs phase boundaries and reversible channels and reduces diffusion resistance; Y addition enables rapid desorption of $Mg_{85}Ni_{13}Y_2$ at 573 K	Non-hydrogen-storage elements dilute capacity and alloy composition must be tightly controlled
Catalyst modification	NiS_2, TiO_2@C and high-entropy oxides	Provides sites for H_2 dissociation and Mg-H bond cleavage; high-entropy oxides can reduce dehydrogenation activation energy to approximately 70 kJ mol^{-1}	Catalytic phases may agglomerate or transform, and excessive loading lowers effective capacity
Nanostructuring	Ball-milled powders, nanoscale confinement and MXene carriers	Shortens diffusion distance, increases grain boundaries and defects, and improves initial activation	Powders oxidize easily; porosity and heat-transfer conditions change after compaction
Composite modification	Alloying + catalyst + thermally conductive carrier	Simultaneously regulates thermodynamics, kinetics and heat transfer, making it suitable for material-bed design	Mechanistic attribution is complex and consistency in scale-up preparation is difficult to control

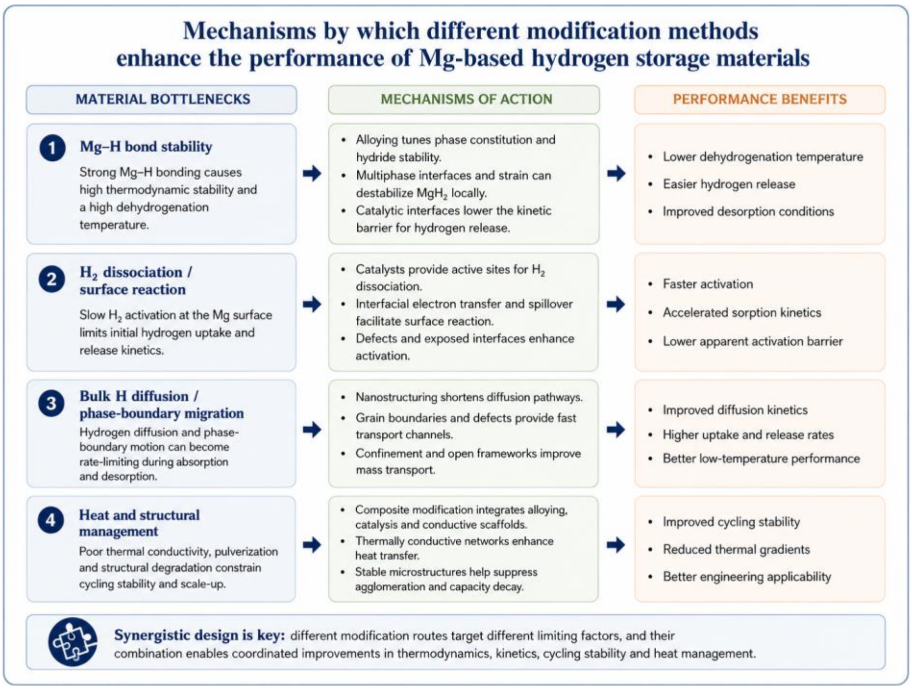

Figure 4. Mechanisms by which different modification methods enhance performance.

4. Analysis of Domestic and International Research Status

4.1. Domestic research progress

Domestic studies have emphasized the microstructural regulation of Mg-Ni and Mg-RE alloys, while also introducing material beds and storage/transport devices into the discussion at an early stage [3,4,7]. The advantage of this route is that capacity, thermal management and device mass can be evaluated within the same framework, rather than remaining limited to powder PCT curves. At the material level, Mg-Ni phase-boundary regulation and dispersion of catalysts such as NiS_2 address diffusion paths and interfacial contact, respectively [7,10].

The remaining weakness is insufficient horizontal comparability. Slight differences in sample mass, heating rate, hydrogen pressure, ball-milling time and cycle number can make absorption/desorption data difficult to compare directly. Compared with laboratory results, engineering samples must also withstand compaction, thermal-cycling stress and impurity-containing atmospheres, yet these factors are often weakened in reported data.

4.2. International research progress

International studies place greater emphasis on interfacial chemistry and in situ structural evolution. Discussions of high-entropy oxides, MXenes and carbon-based composite catalysts have shifted from asking which catalyst is effective to analyzing electron transfer, surface functional groups, two-dimensional confinement and active-phase reconstruction during cycling [5,11,13,14]. First-principles calculations are often used to estimate the influence of dopants on Mg-H bond strength, H diffusion barriers and interfacial adsorption energy, but the results still need verification by PCT, DSC, in situ XRD or TEM.

Application-oriented research treats heat exchange in material beds as an independent variable. Reactor simulations by Shi et al. [15] indicate that the heat released during hydrogen absorption can raise the reaction-zone temperature to approximately 350 ℃, and that the arrangement of heat-exchange tubes and fins directly affects bed-temperature differences and absorption/desorption response. This shows that low-temperature catalysis cannot replace heat-exchanger design; the two address problems at different length scales.

4.3. Comparison of representative results

Representative results should not be ranked only by capacity. $Mg_{85}Ni_{13}Y_2$, high-entropy-oxide-catalyzed MgH_2 and Mg-Ni-Y/TiO_2@C represent three routes, namely alloy phase boundaries, in situ catalytic interfaces and composite carriers [9,11,12]. The first shows good cycling stability, the second retains relatively high rehydrogenation capacity, and the third enables rapid initial hydrogen absorption at lower temperature. These differences show that capacity, rate and lifetime still involve trade-offs.

From an engineering perspective, stationary energy storage can tolerate a certain mass of material bed and heat exchanger, making cycling stability and temperature matching more important. Mobile hydrogen storage, however, is quickly penalized at the system level by the mass of catalysts, rare-earth elements and heat-exchange structures. The fast kinetics of milligram-scale powders cannot be directly extrapolated to kilogram-scale material beds, which are also controlled by thermal resistance, gas permeability and powder compaction state.

5. Existing Problems and Development Trends

5.1. Current research bottlenecks

The contradiction between thermodynamic stability and reversible capacity has not yet been eliminated. Reducing the stability of MgH_2 usually introduces non-hydrogen-storage elements or stable intermediate phases; as the dehydrogenation temperature decreases, gravimetric capacity is lost. Catalysts can lower kinetic barriers, but they are unlikely to change the equilibrium plateau pressure. Nanostructuring shortens diffusion distances, but it increases the risk of oxidation and grain growth. Current debate mainly concerns whether low-temperature performance should be attributed to thermodynamic modification or kinetic acceleration; the two are often mixed in temperature-programmed curves.

5.2. Challenges for engineering application

Engineering application is constrained by factors more complex than material metrics, including batch preparation, oxidation-resistant transfer, shaping and encapsulation, gas permeability, thermal-conduction enhancement and cycling-stress control. After powder packing, heat released during hydrogen absorption can cause local temperature rise, whereas hydrogen desorption may become unstable because of insufficient heat supply. Pulverization changes porosity and increases local stress. Heat-exchange structures must match material kinetics; otherwise, the rate of powder samples cannot be reproduced in material beds [15]. Rare-earth elements, MXenes and high-entropy oxides also increase cost, and their mass and price penalties must be offset by longer cycle life, higher heat-exchange efficiency and improved system capacity.

5.3. Future development directions

Future evaluation should shift from single dehydrogenation peak temperature to parallel reporting of reversible capacity, isothermal rate, cycling retention and thermal-management conditions. First-principles calculations can be used to pre-screen a small number of efficient catalytic phases, but the screening results must be linked with PCT, DSC and in situ characterization. Nanoscale powders also need to be transformed into shapeable porous bodies or thermally conductive composites so that gas channels, mechanical strength and heat-exchange structures can be controlled simultaneously.

A more standardized testing framework is also needed. For Mg-based materials, reporting only a peak desorption temperature or a maximum hydrogen capacity is insufficient. Sample mass, heating rate, hydrogen pressure, catalyst loading, cycle number, pellet density and heat-transfer boundary conditions should be reported together, so that different modification routes can be compared on the same basis.

On the application side, scenarios with stable heat sources are more suitable starting points. If industrial waste heat or heat-pump heating is used, the relatively high dehydrogenation temperature of MgH_2 can be treated as a boundary condition for thermal management. The heat released during hydrogen absorption can also be recovered to reduce system energy consumption. Material-bed tests should incorporate hydrogen purity, temperature fluctuation and start-stop cycling, and low-cost preparation, oxidation-resistant encapsulation and heat-exchanger mass should be evaluated together with dehydrogenation temperature.

6. Conclusions

The theoretical capacity of MgH_2, approximately 7.6 wt.%, and the high stability of the Mg-H bond define the fundamental contradiction of Mg-based hydrogen storage materials: the capacity advantage originates from the Mg/MgH_2 reaction, whereas application resistance arises from hydrogen-release temperature, kinetics and bed-level heat transfer. Mg-Ni systems improve hydrogen dissociation and diffusion. Rare-earth elements enhance the stability of cycling microstructures through LPSO phases, rare-earth hydrides and multiphase interfaces. MXenes, high-entropy oxides and sulfides mainly reduce kinetic barriers. In our view, engineering application of Mg-based materials is not limited by the absence of a single highly efficient catalyst; rather, it lacks coordinated evaluation among the material, the shaped body and the heat exchanger. If future data do not simultaneously report capacity, temperature, pressure, cycle number, cost and heat-exchange parameters, powder performance will still be insufficient to describe the operating capability of a material bed.

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