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Review

Recent advances in additive-enhanced magnesium hydride for hydrogen storage☆

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ABSTRACT

The discovery of new hydrogen storage materials has greatly driven the entire hydrogen storage technology forward in the past decades. Magnesium hydride, which has a high hydrogen capacity and low cost, has been considered as one of the most promising candidates for hydrogen storage. Unfortunately, extensive efforts are still needed to better improve its hydrogen storage performance, since MgH_2 suffers from high operation temperature, poor dehydrogenation kinetic, and unsatisfactory thermal management. In this paper, we present an overview of recent progress in improving the hydrogenation/de-hydrogenation performance of MgH_2, with special emphases on the additive-enhanced MgH_2 composites. Other widely used strategies (e. g. alloying, nanoscaling, nanoconfinement) in tuning the kinetics and thermodynamics of MgH_2 are also presented. A realistic perspective regarding to the challenges and opportunities for further researches in MgH_2 is proposed.

1. Introduction

What hampers our step towards a hydrogen-based energy system? Although plenty of issues, such as hydrogen generation, hydrogen transportation, hydrogen application, must be solved in our way to fulfil the worldwide commercial usage of hydrogen, it is the lack of ideal hydrogen storage material that primarily blocks the progress of hydrogen technology. The discovery of each novel hydrogen storage material (alloys, carbon materials, metal organic frameworks (MOFs), organic liquids, metal alanates, metal hydrides et al. [1–7]) had greatly promoted the revolution of hydrogen storage technology.

An ideal hydrogen storage material should meet the following standards: (1) cost-affordable, (2) high hydrogen storage capacity (5.5 wt% and 40 g L^{-1} hydrogen capacity by 2020, U.S. Department of Energy [8]), (3) mild operation temperature and easy to absorb/de-absorb hydrogen, (4) long life-span. Fig. 1 showed some typical hydrogen storage technologies and compared their operation conditions [9]. Apparently, amongst all these strategies, metal hydrides exhibit huge potential for commercial storage of hydrogen. Especially, MgH_2 has been considered as one of the most promising candidates, due to its high hydrogen storage capacity (7.6 wt%, 110 g L^{-1} H_2), abundance in deposit, and low cost.

MgH_2 is an ionic compound and its charge distribution is $Mg^{1.91+}H^{0.26-}$. This structure results a high thermodynamic stability of MgH_2, which shows an enthalpy value of 74.7 kJ mol^{-1}, and an entropy value of 130 J K^{-1} mol^{-1}. The high stability of MgH_2 leading to a high operation temperature (> 350 °C) which is not good for practical application. Another drawback of MgH_2 is its sluggish sorption kinetics, causing by the following reasons: (1) existence of oxide on its surface, (2) slow diffusion rate of hydrogen in the bulk MgH_2/Mg, (3) poor decomposition of hydrogen on the Mg surface [10]. Additionally, the unsatisfactory heat management of MgH_2/Mg also affects its hydrogenation/de-hydrogenation behaviours. All these problems above have hampered the worldwide application of MgH_2/Mg. To date, numerous efforts have been carried out to overcome these disadvantages, and huge improvements have been achieved. For example, Xia et al. synthesized monodisperse MgH_2 nanoparticles (NPs) that anchored on graphene sheet, and confirmed its impressive hydrogen storage properties, which released 5.4 wt% hydrogen within 30 min and showed an ultra-long cycle-life of 100 times [11].

In this paper, we summarized a wide range of promising strategies to tuning the hydrogen storage performance of MgH_2/Mg, with special focus on the advantages and challenges of each method. To better present the research progress of MgH_2/Mg, we classified the technologies into four categories: alloying, nanoscaling, nanoconfinement, and additive-addition. Rather than detail all outcomes of each classic

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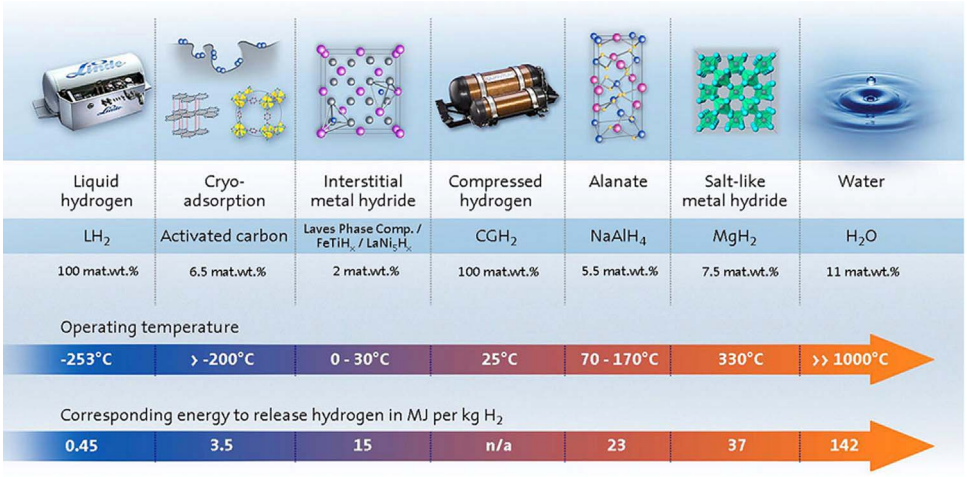

Fig. 1. Comparison of different hydrogen storage strategies and their operation conditions [8].

method, we pay especial attention on the additive-enhanced MgH_2 systems. By reviewing these data, we hope this paper can help more researchers better understanding MgH_2, and shed light on further works.

2. Overview of tuning strategies

The hydrogenation/de-hydrogenation process of MgH_2/Mg is illustrated in Fig. 2. Apparently, decreasing the energy barrier and/or changing the reaction enthalpy of MgH_2/Mg are fundamental ways to improve its hydrogen storage performance.

2.1. Alloying

One of the most effective methods to reduce the thermodynamic barrier of MgH_2/Mg is alloying. Instead of direct reaction between MgH_2 and Mg, the formation of Mg-alloys changes its sorption path. By forming thermodynamic more stable alloys, the operation temperature of MgH_2/Mg can be reduced. Various elements had been used to alloy with Mg, including rare earth elements, transition metals, and partial main group elements [12–24]. Table 1 summarizes the basic properties of some commonly investigated Mg-based hydrogen storage alloys.

Mg_2NiH_4, which has a lower enthalpy of 64 kJ mol^{-1}, is one of the most investigated Mg-alloys [37–39]. Kumar et al. reported that the hydrogen sorption of nanocrystalline Mg_2NiH_4 alloy started at 200 °C. However, the Mg_2NiH_4 alloys exhibited a low hydrogen capacity of only 3.6 wt%. Meanwhile, Mg_2FeH_6, which has a higher theoretical hydrogen capacity of 5.5 wt%, shows an even high enthalpy of 95 kJ mol^{-1} H_2[40–43]. Chen et al. reported the hydrogen storage performance of

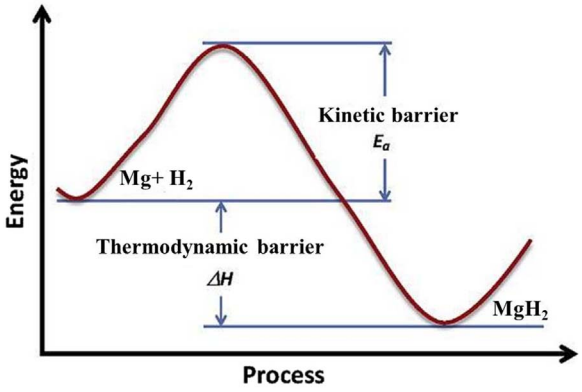

Fig. 2. Illustration for the hydrogenation/de-hydrogenation process of MgH_2/Mg.

Table 1
Fundamental information of some Mg-based alloys [25].

Name	E_a (kJ mol^{-1})	ΔH (kJ $mol^{-1}H_2$)	Capacity (wt%)	T (°C)	Ref.
Mg	–	74.5	7.6	300	Stampfer [26]
Mg (2–7 nm)	–	71.2	7.6	276	Paskevicius [27]
$Mg_{90}Ce_{10}Ni_{10}$	109.2	77.9	5.4	284	Lin et al.[28]
Mg_2Ni	–	64.5	3.6	254	Reilly [29]
$Mg_3LaNi_{0.1}$	–	81	2.73	284	Ouyang [30]
Mg_3Cd	69	65.5	2.8	–	Skripnyuk.[31]
MgH_2-Ti	30.8	75.2	6.7	278	Cui [32]
$Mg_{0.95}In_{0.05}$	–	68.1	5.3	–	Zhong [33]
Mg_3Ag	–	68.2	2.1	–	Si [34]
Mg_2Si	–	36.4	5.0	–	Vajo [35]
Mg_5Ga_2	149	68.7	5.7	300	Ouyang [36]

Mg_2FeH_6@MgH_2 core-shell, which released 5.0 wt% hydrogen within 50 min at 280 °C [14]. In 2012, Liu et al. reported the hydrogen storage properties of Mg@$Mg_{17}Al_{12}$ particles, which delivered 6.0 wt% hydrogen with 30 min at 350 °C, and absorbed 7.0 wt% hydrogen at 400 °C [44]. As shown in Fig. 3, the $Mg_{17}Al_{12}$ shell enclosed Mg core and effectively resisted the formation of MgO. The shell grown with the increased content of Al, but over high Al content would broke this core-shell structure.

Fig. 4 showed the de-hydrogenation/hydrogenation performance of $LaMg_{11}Ni$+x wt% Ni alloys [15]. Zhang et al. found that milling time and Ni content were both essential factors for enhancing the kinetics of $LaMg_{11}Ni$+x wt% Ni alloys. Good improvement was achieved for the $LaMg_{11}Ni$+200 wt% Ni sample, which absorbed 6.41 wt% hydrogen within 18 min, and reduced E_a to only 68.5 kJ mol^{-1}. Additionally, the $Mg_{6-7}TMH_{12-16}$ (TM=Ti, Zr, Hf, V, Nb), which have a similar structure with CaF_2, also gained huge attention. The $Mg_{6-7}TiH_{12-16}$ releases 4.7 wt% hydrogen at 330 °C, but its re-hydrogenation process is rather tough.

Generally, Mg-base hydrogen storage alloys can effective decrease the operation temperature, but the introduction of heavy metals also result a decreased hydrogen capacity in the system, and most of these Mg-base alloys still suffered from poor reversibility. Detail information about these Mg-based hydrogen storage alloys can refer to a recent review [24].

2.2. Nanoscaling

Nanoscaling has proven its possibility in optimizing the thermodynamics and kinetics of MgH_2. Theoretical calculation (based on the first principle theory) has predict that when reducing the grain size of

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