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Combination of nanosizing and interfacial effect: Future perspective for designing Mg-based nanomaterials for hydrogen storage

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ABSTRACT

Hydrogen storage is now the “bottle neck” to realize application of hydrogen as the renewable energy. The breakthrough in hydrogen storage is quite urgent. Magnesium is a promising candidate for hydrogen storage that attracts tremendous interest in last a few decades and significant progress has been made in recent years. Accordingly, in this article, we comprehensively reviewed different strategies to overcome the key barriers of high desorption temperature and low kinetics, especially on the recent approaches of nanosizing and interfacial confinement. We also try to give our own point of view on the future perspectives of research in Mg for hydrogen storage.

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List of abbreviations: DOE, Department of Energy; Er, reaction energy; PVD, physical vapor deposition; MOFs, metal–organic frameworks; SSA, specific surface area; SWNTs, single-walled carbon nanotubes; G, graphite; AC, activated carbon; CB, carbon black; BNNTs, Boron nitride nanotubes; ACF, activated carbon fiber; MgH₂, magnesium hydride; MgBu₂, dibutylmagnesium; PMMA, poly(methyl methacrylate); NC, nanoporous carbon; OMC, ordered mesoporous carbon; XRD, X-ray diffraction; SEM, scanning electron microscopy; TEM, transmission electron microscopy; HRTEM, high resolution transmission electron microscopy; STEM, scanning transmission electron microscopy; DF, dark field; HAADF, high angle annular dark field; EDX, energy dispersive X-ray spectroscopy; SAED, selected area electron diffraction; DSC, differential scanning calorimetry; TDMS, thermal desorption mass spectra; DFT, density functional theory

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1. Introduction

With the rapid population growth and economic development, fossil fuel reserves are largely depleted and the environmental pollution is further aggravated [1]. Exploring sustainable clean energy sources and environmental technology to satisfy the world's growing demand are emerging as hot research fields and one of the foremost challenges for this century. Among renewable energy resources, hydrogen as the new energy carrier is alternative to fossil fuels. Specially, it is potential for implementation in a carbon-free emission cycle and high efficiency when combined with oxygen in the electrochemical reaction of a fuel cell to produce electricity [2–6]. However, a main obstacle impeding anticipated future hydrogen economy is the lack of safe, efficient and economical on-board hydrogen storage. In response to this challenge, the U.S. Department of Energy (DOE) has set technical targets for new hydrogen storage technologies to meet [2,3]. These include the gravimetric and volumetric densities, ease of charge and discharge, system and fuel costs and operability ranges. Some of the key targets, which were revised in 2012, are summarized in Table 1. Unfortunately, none of the current hydrogen storage technologies can fulfill these benchmarks. Therefore, developing reliable media and technologies to efficiently storage hydrogen in high density has been the focus of research efforts for several decades.

Hydrogen storage strategies can be basically classified into two categories: physical and chemical routes. Currently, gaseous hydrogen compression and hydrogen liquefaction are the most mature technologies [7]. However, they are still associated with drawbacks such as manufacturing cylinders capable of withstanding ultrahigh pressure (up to 70 MPa) with light weight, energy consuming process for both gaseous hydrogen compression and liquefaction, permeation and safety issues. Physisorption as the alternative way in physical hydrogen storage routes absorbs hydrogen in highly porous solid state adsorbents such as nanoporous carbon and metal–organic frameworks (MOFs). However, significant hydrogen adsorptive storage capacities in physisorption need to be reached at liquid-nitrogen temperatures (77 K) and pressures of several MPa. This is attributed to weak binding energy between molecular hydrogen and the surface of sorbents in the range of 2–5 kJ mol⁻¹ H₂ [8–12]. Generally, the hydrogen storage properties of physisorption appear to be limited by specific surface area (SSA), pore structures and pore size distributions, surface functionality and the bulk density of the adsorbents. More details on these aspects have been summarized in previous review

articles about carbon/MOFs for physical hydrogen storage [13–15]. On the other hand, distinct from physisorption for hydrogen storage using nanoporous adsorbents described above, hydrides of some metals and non-metals form a different class of materials for hydrogen storage characterized by their ability to store larger amounts of H₂ through the formation of chemical bonds. For instance, metal hydrides for hydrogen storage starts with dissociation of hydrogen molecules, migration and chemical bonding of the hydrogen atoms by integration in the lattice of metals with a high binding energy (> 50 kJ mol⁻¹ H₂), therefore storage capacities and the operating temperatures are elevated [12,16–19]. However, the main shortcoming in chemisorption is usually requiring high temperatures to absorb/release the hydrogen due to high binding energy.

Fig. 1a shows the range of binding energies targeted by DOE, suggesting that the ideal materials for hydrogen storage would achieve a compromise between physisorption and chemisorption within the range of 10–60 kJ mol⁻¹ H₂, which is strong enough to hold H₂ but weak enough to allow for quick desorption [20–23].

Table 1
2012 Revised DOE hydrogen storage targets [3].

Target	Gravimetric capacity (kg H ₂ /kg system)	Volumetric capacity (kg H ₂ /L system)
2010	0.045	0.028
2017	0.055	0.040
Ultimate	0.075	0.070

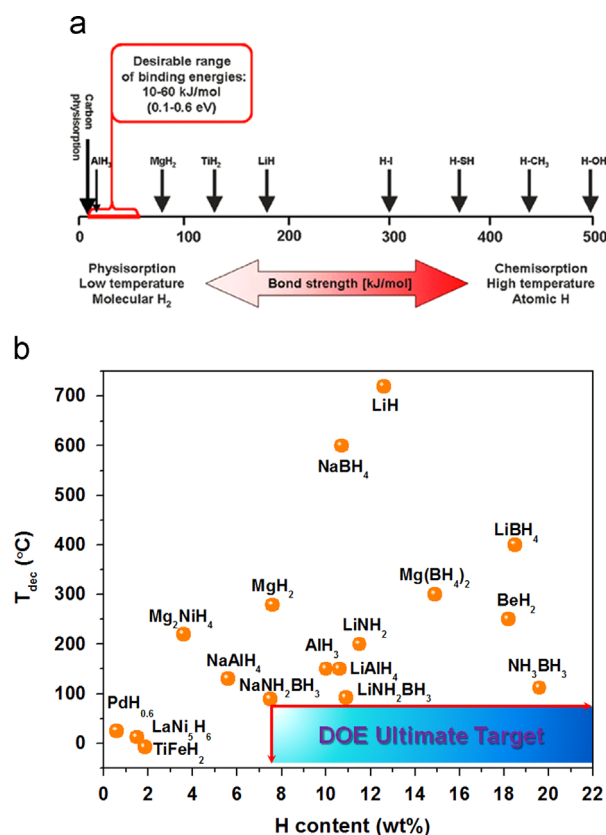


Fig. 1. (a) Targeted range of bond strengths that allow hydrogen release around room temperature [26]. (b) Overview of various solid-state hydrides, plotting the decomposition temperatures (under 1 bar H₂ pressure) as function of the gravimetric hydrogen content. The DOE ultimate target is shown in shadow bar.

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