

# Improved Hydrogen Storage Performance of Novel Metal Hydrides and Their Composites

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To date, the majority of energy supply is still generated by non-renewable sources, which places a dramatic burden on our environment. Although renewable energies have gained increasing attention in recent years, such CO<sub>2</sub>-free energy sources are not yet competitive with fossil fuels [1]. Nevertheless, a promising candidate that can be introduced into energy grids as a secondary energy carrier is hydrogen since its energy density per unit mass (120–140 MJ/kg) is the largest among the elements [2], and its use does not create any harmful byproducts when applied in a fuel cell to generate electrical energy for transportation and off-board applications [3]. A large number of current research studies have been dedicated to improving the hydrogen storage performance of various solid-state systems, including metal hydrides [4]. Here, a brief summary is presented on some selected review and research papers in the field of solid-state hydrogen storage of metal hydrides and their composites published recently in *Energies*.

A comprehensive review by Yuchen Liu et al. summarizes the development of A<sub>2</sub>B, AB, AB<sub>2</sub>, AB<sub>3</sub> and A<sub>2</sub>B<sub>7</sub> intermetallic compounds, where “A” is a hydride-forming metal, such as Mg, Ti, V or Zr, and “B” is a non-hydride-forming element, such as Cr, Fe, Mn and Ni [5]. Commonly, these compounds can be produced via mechanical alloying and, due to their large volumetric H storage capacity, they are at the forefront of intensive research. During mechanical alloying, both powder particle size and crystallite size are significantly reduced; this is coupled with an increase in the specific surface area, which promotes faster hydrogen diffusion. As a highly non-equilibrium technique, mechanical alloying by ball milling not only induces microstructural refinement but can also promote the formation of unique intermetallic compounds made of immiscible elements.

Among metallic hydrides, MgH<sub>2</sub> is the best candidate due to its large storage capacity (7.6 wt.% gravimetric and 0.11 kg H/L volumetric capacity), low density, high abundance and low cost. Several studies have undoubtedly proven that high-energy ball milling is a promising technique to improve the H-sorption properties of Mg-based nanocomposite materials via the creation of different lattice defects, such as dislocations, grain boundaries and stacking faults. A review paper by Révész and Gajdics presents recent progress on the hydrogenation/dehydrogenation of ball-milled magnesium-based materials catalyzed by different additives, including transition metal-based catalysts, intermetallic compounds, catalysts generated during in situ reactions, multiple catalysts with synergetic performance, and catalysts with an irregular shape and morphology [6]. The effect of these different additives is discussed in detail, focusing on their catalytic mechanisms. In the last section of the paper, the authors provide some directions for future research.

The review by Pasquini takes a broader look at the design principles for the development of new classes of nanomaterials for hydrogen storage [7], including bulk-like mechanochemically processed hydride nanomaterials; thin films and multilayers; nano-objects with composite architectures, such as core-shell and composite nanoparticles; and nanoparticles on porous or graphene support. In this work, the author also sheds light on the effect of microstructure on the H-kinetic properties of these different types of nanostructures.



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A recent review paper by Ahmad et al. targets  $\text{Mg}(\text{BH}_4)_2$ , which is the best candidate of the borohydride family owing to its very high gravimetric H storage capacity (14.9 wt.%) [8]. As discussed in the paper, doping  $\text{Mg}(\text{BH}_4)_2$  with different additives and catalysts via ball milling not only refines the microstructure but also reduces the activation energy of hydrogen release.

The latest on-going research on the H storage performance of different systems processed via a bulk deformation method, high-pressure torsion, is systematically reviewed by Révész and Gajdics [9]. Their review shows that the high-pressure torsion procedure can considerably improve the hydrogenation/dehydrogenation kinetics of various hydride composites by means of crystallite size refinement and generation of high-angle grain boundaries and texture. Aside from enhancing the sorption behavior of severely deformed materials, new compounds and metastable phases can also be synthesized via high-pressure torsion. In a research article by Gajdics et al., ball milling and subsequent high-pressure torsion deformation procedures were applied on Mg powders catalyzed by  $\text{TiO}_2$  nanotubes [10]. It was established that the processing route positively alters the sorption properties, i.e., the composite material produced with longer co-milling of  $\text{TiO}_2$  nanotubes and Mg powder promotes better overall hydrogenation performance. This observation can be attributed to the more homogeneously dispersed and partially damaged  $\text{TiO}_2$  sections.

Another severe plastic deformation technique, fast forging, was successfully carried out on Mg-Ni powder compacts. In a research article, de Rango et al. demonstrated that forging at elevated temperatures not only promotes microstructural refinement but also induces solid-state reaction, which leads to the formation of  $\text{Mg}_2\text{Ni}$  intermetallic compound [11]. According to this research, fast forging can be applied as a safe and cost-efficient process to produce large amounts of Mg-based materials for hydrogen storage.

An excellent work of Jiahui Tan et al. describes a heat and mass transfer model for the hydrogen absorption reaction of metal hydrides in a tank [12]. Their numerical simulation results were validated by existing sorption experiments. These results confirm that the overall absorption reaction rate is fast inside the storage tank at the initial stage, and the reaction first takes place on the wall of the tank and then extends to the interior regions.

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