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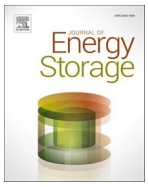

Research papers

Structural and electronic insights into $Na_{1-x}Li_xMgH_3$ hydrides for efficient solid-state hydrogen storage via lithium substitution

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ABSTRACT

This study investigates the hydrogen storage properties of $Na_{1-x}Li_xMgH_3$ hydrides (x = 0 to 1) using Density Functional Theory (DFT). Results show that lithium substitution enhances hydrogen storage, with gravimetric and volumetric capacities increasing from 6.00 wt% and 90 gH_2/l (x = 0) to 8.82 wt% and 214 gH_2/l (x = 1), surpassing U.S. DOE targets. The theoretical specific capacity improves from 1590 to 2331 mAh/g. A phase transition at x = 0.625 (from orthorhombic *Pnma* to trigonal *R3c*) improves hydrogen packing. Among dehydrogenation pathways, Pathway 7 shows the best practical potential, with desorption temperatures dropping from 256 K to 145 K as lithium content increases. Binding and cohesive energy analyses reveal enhanced stability and interactions due to lithium substitution. All compositions are wide-gap insulators, with band gaps increasing from ~3.45 eV to ~6.77 eV as lithium content rises. These findings position $Na_{1-x}Li_xMgH_3$ hydrides as promising candidates for solid-state hydrogen storage.

1. Introduction

Global warming presents a significant threat to humanity, necessitating a comprehensive understanding of its causes to develop effective mitigation strategies. Fossil fuels, which are responsible for approximately 75 % of global greenhouse gas emissions and nearly 90 % of carbon dioxide emissions, are the primary contributors to climate change [1].

The environmental impact of these emissions underscores the need to transition to sustainable energy sources. Advancements in clean, efficient, and renewable alternatives to fossil fuels are critical to ensuring a sustainable energy future. This shift not only mitigates climate change but also fosters energy independence and economic growth. By investing in renewable technologies, such as solar, wind, and hydroelectric power, we can build resilient infrastructure that supports both the environment and society [2]. In this context, renewable energy has emerged as a promising solution to address the global energy crisis [3], with many experts now considering hydrogen as a key energy carrier for the future. Due to its remarkable properties, hydrogen could play a central role in the transition to more sustainable and less polluting energy systems [4,5].

Hydrogen is widely regarded as one of the most promising alternative energy carriers, owing to its abundance, ease of production, exceptionally high energy density per unit mass, and environmentally benign combustion byproducts [6]. However, significant scientific and technological challenges remain, particularly in the areas of hydrogen production, distribution, storage, and utilization in fuel cells. Among these challenges, efficient hydrogen storage is a critical hurdle [7]. Current storage methods, such as compressed gas or cryogenic liquefaction, are costly and pose safety risks [8].

The solid-state storage of hydrogen is considered one of the most practical, efficient, and secure methods for hydrogen storage [9]. Consequently, research in this area is primarily focused on developing advanced materials with optimized properties, particularly those that are lightweight, chemically stable, non-toxic, and economically viable, while also ensuring long-term durability and high hydrogen storage capacity [10]. A recent study by Smruti et al. found a gravimetric hydrogen density of 6.36 wt% at 332.93 K for the yttrium-decorated azatriphenylene COF using VASP code [11]. Light chemical hydrides, such as $B_3N_3H_6$ (7.51 wt%), $C_3N_3H_6$ (7.19 wt%), and $Al_3N_3H_6$ (4.68 wt%), show high hydrogen storage capacities with branched hydrogen chains related to [XH_3NH_3] benzene structures [12]. Alkali metal tetrahydrides like $KAlH_4$ (5.7 wt%) [13], and $LiAlH_4$ (10.5 wt%) [14] are also noteworthy for their high storage capacity and excellent absorption

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