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Electronic structure, mechanical and optical properties of hydrogen storage alkaline amides XNH_2 (X = Li, Na) compounds

T. Saadi [a,b], H. Baaziz [a,b,*], T. Ghellab [a,b], H. Latelli [a,b], Ahmad Telfah [c,d], Z. Charifi [a,b,**]

[a] Department of Physics, Faculty of Science, University of M'sila, 28000, M'sila, Algeria
[b] Laboratory of Physics and Chemistry of Materials, University of M'sila, Algeria
[c] Fachhochschule Dortmund University of Applied Sciences and Arts, 44139, Dortmund, Germany
[d] Department of Physics, Yarmouk University (YU), Irbid, 21163, Jordan

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ABSTRACT

Alkaline amides XNH_2 (X = Li, Na) were studied to assess their potential for hydrogen storage applications using first-principles calculations. Structural analyses revealed that $LiNH_2$ crystallizes in a tetragonal structure (space group I-4), while $NaNH_2$ adopts an orthorhombic structure (space group *Fddd*). The electronic band structure, calculated using the Generalized Gradient Approximation (GGA), Local Density Approximation (LDA), and Engel-Vosko Generalized Gradient Approximation (EV-GGA), shows that both materials are wide-bandgap semiconductors with bandgap values of 4.45 eV for $LiNH_2$ and 3.97 eV for $NaNH_2$. The valence bands are dominated by $[NH_2]^-$ states, which play a critical role in hydrogen storage. The mechanical stability of both compounds was confirmed by elastic constants, with $LiNH_2$ exhibiting superior mechanical strength compared to $NaNH_2$. Phonon dispersion analysis verified the dynamic stability of both materials. Optical properties, such as refractive index, reflectivity, and absorption coefficient, were evaluated, revealing high optical contrast, making these materials promising for optoelectronic applications. Thermal behavior analysis indicated that increasing temperature leads to higher entropy and internal energy, and lower free energy, favoring hydrogen desorption. The gravimetric hydrogen storage capacities were calculated as 8.78 wt% for $LiNH_2$ and 5.17 wt% for $NaNH_2$, highlighting their potential for energy storage. This study provides novel insights into the structural, electronic, mechanical, optical, and thermal properties of XNH_2, positioning $LiNH_2$ as a promising candidate for hydrogen storage and optoelectronic applications.

1. Introduction

The development of efficient and cost-effective methods for hydrogen storage and transport remains a critical goal in advancing hydrogen-based energy systems [1,2]. Hydrogen (H) is recognized as a highly desirable green energy source. Consequently, hydrogen energy and hydrogen storage materials have been the focus of extensive research in recent years due to their advantages in terms of high efficiency and stability for electrocatalytic hydrogen production [3]. Among the materials under investigation, alkali metal amides (XNH_2, where X can be Li, Na, or K) have attracted significant attention due to their high gravimetric hydrogen densities, which make them promising candidates for hydrogen storage. Metal hydrides are widely regarded as promising hydrogen storage materials due to their enhanced absorption and desorption kinetics [4]. The discovery of alkali metal amides dates back to the early 19th century, as first documented by Gay-Lussac et al. [5]. In recent years, interest in these materials has been renewed, largely due to their potential in both hydrogen storage and transport. Key research efforts have focused on understanding their hydrogenation and dehydrogenation mechanisms, crystallographic structures, reaction products, and physicochemical properties [6–8]. These topics have been extensively reviewed in the literature [9]. And it has been done XAlSiH (X = Sr, Ca, and Ba) compounds have been studied for hydrogen storage since the arrangement of hydrogen atoms in the crystal structure of these compounds can provide unique sites for hydrogen storage [10]. Also, compounds such as SrGaSiH, CaGaSiH, and BaGaSiH have been very

* Corresponding author. Department of Physics, Faculty of Science, University of M'sila, 28000, M'sila, Algeria.
** Corresponding author. Department of Physics, Faculty of Science, University of M'sila, 28000, M'sila, Algeria.
E-mail addresses: hakim.baaziz@univ-msila.dz (H. Baaziz), zoulikha.charifi@univ-msila.dz (Z. Charifi).

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