



Structural evolution and thermodynamic tuning of $\text{Sr}_{1-x}\text{Ba}_x\text{LiH}_3$: Inverse perovskites for high-volumetric-capacity hydrogen storage

S. Saadoun^{a,b}, T. Ghellab^{a,b,*}, H. Baaziz^{a,b,*}, T. Saadi^{a,b}, 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

ARTICLE INFO

Keywords:

Inverse perovskite hydrides
 $\text{Sr}_{1-x}\text{Ba}_x\text{LiH}_3$ alloys
 Hydrogen binding
 Cation substitution
 Desorption temperature

ABSTRACT

This computational study investigates the structure-property relationships in $\text{Sr}_{1-x}\text{Ba}_x\text{LiH}_3$ ($0 \leq x \leq 1$) inverse perovskite hydrides using DFT. We demonstrate how controlled Ba substitution systematically tunes formation energies (-60.4 to -58.8 kJ/mol-H₂) and hydrogen binding energies (-1.19 to -1.24 eV), enabling precise optimization of hydrogen storage performance. The alloys exhibit an unusual structural evolution from cubic ($Pm\bar{3}m$) to tetragonal ($P4/mmm$) and back to cubic ($Pm\bar{3}m$) symmetry, with the re-entrant cubic phase at $x = 0.875$ showing exceptional stability (cohesive energy -17.1 eV). Electronic structure calculations reveal composition-dependent bandgap transitions (3.45 – 4.13 eV, mBJ-GGA) that alternate between direct and indirect character. For hydrogen storage, the materials' key advantage lies in their outstanding volumetric capacity of 188 g H₂/L, which significantly exceeds the capacities of many benchmark storage systems, while they maintain a moderate gravimetric capacity of 2.1 – 3.2 wt%. Desorption temperatures (449 – 462 K) are directly correlated with the hydrogen binding energy landscape. The study identifies an optimal composition window ($x = 0.25$ – 0.5) that balances favorable formation energy (-59.5 kJ/mol-H₂), moderate hydrogen binding (-1.22 eV), and robust thermodynamic stability. These findings establish a quantitative framework for designing perovskite hydrides with tailored hydrogen storage properties through controlled cation substitution, specifically targeting applications where high volumetric density is a critical requirement.

1. Introduction

Projections indicate that the world's petroleum reserves could be exhausted within 41 years, while fossil fuel consumption is expected to increase by 56% by 2040, intensifying concerns over energy security and the long-term viability of environmental systems [1]. These challenges are compounded by predictions of a 2.5°C rise in global temperatures by the century's end [2,3]. The convergence of an escalating global energy crisis and rising pollution levels significantly hampers efforts to achieve sustainable development. This underscores the urgent need for cleaner energy sources and technological advancements to mitigate carbon emissions and reduce air pollution [4,5]. In response to these growing demands, the quest for renewable energy alternatives has brought silicon-based solar cells to the forefront as a promising and viable solution [6]. Research into alternative photovoltaic materials is gaining momentum due to the challenges associated with the production complexity and relatively low conversion efficiency of silicon-based solar cells [7,8]. As the global drive for green energy accelerates,

hydrogen is emerging as a highly promising alternative due to its abundant supply, superior energy density, and potential for producing carbon-free energy [6,9]. With current annual hydrogen consumption at 70 million tons, projections suggest a 70% increase in global demand by 2050 [10]. Despite its promise as a clean energy carrier, hydrogen's widespread adoption is hindered by the challenges related to establishing safe, efficient, and economically viable storage and distribution systems [11]. Traditional hydrogen storage methods, such as high-pressure compression and cryogenic liquefaction, are plagued by high costs and significant safety risks. In contrast, metal hydrides are garnering increasing interest as a viable alternative due to their high hydrogen storage capacity, reversibility, and favorable reactivity properties [12].

Among the various methods, solid-state hydrogen storage is gaining recognition as a highly viable, efficient, and secure approach for long-term energy storage solutions [13]. Storage systems for hydrogen typically rely on a range of substances, including alloys and hydrides of metals [14,15], layered two-dimensional materials [16], and highly

* corresponding authors at: Department of Physics, Faculty of Science, University of M'sila, 28000 M'sila, Algeria.

E-mail addresses: torkia.ghellab@univ-msila.dz (T. Ghellab), hakim.baaziz@univ-msila.dz (H. Baaziz).

<https://doi.org/10.1016/j.comptc.2026.115663>

Received 10 November 2025; Received in revised form 20 December 2025; Accepted 7 January 2026

Available online 11 January 2026

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