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A novel SnC/graphene heterostructure as an efficient host material for Li- and Na-ion batteries: computational insight

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How to cite this article: Rehman, J.; Butt, M. K.; El-marghany, A.; Li, Z.; Yang, G. A novel SnC/graphene heterostructure as an efficient host material for Li- and Na-ion batteries: computational insight. *Energy Mater.* **2025**, *5*, 500046. <https://dx.doi.org/10.20517/energymater.2024.118>

Received: 13 Aug 2024 **First Decision:** 11 Nov 2024 **Revised:** 26 Nov 2024 **Accepted:** 9 Dec 2024 **Published:** 20 Feb 2025

Academic Editor: Jiazhao Wang **Copy Editor:** Ping Zhang **Production Editor:** Ping Zhang

Abstract

The rapid growth of technologies has influenced our daily lives in building efficient energy storage systems such as lithium-ion batteries (LIBs) for various electric automobiles and portable electronic devices. Graphite, the commercial anode material for LIBs, has several limitations including low lithium storage capacity (372 mAh g⁻¹), low power rate capability, and sluggish charging for applications in grids and heavy electric vehicles. Herein, we propose a novel SnC/graphene heterostructure (SnC/G-H) as a potential anode material for LIBs and sodium-ion batteries, supported by first-principles calculations. The graphene layer in the SnC/G-H model provides high mechanical stability and electrical conductivity, enhancing device application and potentially solving the structural issues of the SnC monolayer. SnC/G-H serves as an excellent Li/Na host material, offering low average voltages (0.34-0.39 V), impressive Li/Na storage capacities of 870 mAh g⁻¹ (exceeding those of pristine SnC and graphite), and minimal activation energy barriers of 0.043/0.079 eV, which promote efficient lithiation/delithiation and sodiation/desodiation processes. These enthralling findings indicate that the SnC/G-H could serve as an efficient



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host material for rechargeable LIBs and sodium-ion batteries.

Keywords: Heterostructure, graphene, charging/discharging, first-principles calculation, Li/Na-ion batteries

INTRODUCTION

Lithium (Li)-ion batteries (LIBs) play a crucial role in energy storage systems, finding widespread use in portable electronic devices, such as smartphones, cameras, laptops, and radios, and in automotive machinery^[1-5]. For decades, LIBs have been a cornerstone of energy storage technology, and play a central role in addressing some of today's most pressing industrial and technological challenges. While much research has been focused on graphite as a key anode material, there are still significant concerns that must be addressed, particularly regarding the limitations of graphite and the broader implications for battery performance, cost, and longevity^[6,7]. Unfortunately, the low Li storage capacity of graphite (372 mA h g^{-1}) limits its suitability for high-grid applications^[8,9]. On the other hand, the scarcity of Li and its high cost pose challenges for the future of Li-based technology. As a result, researchers are exploring other energy storage systems, including sodium (Na)-ion batteries (SIBs), which benefit from abundant Na reservoirs^[10-14]. In the case of SIBs, the Na storage capacity in graphite is significantly lower (around 35 mA h g^{-1}) due to the challenges posed by Na intercalation between graphite layers, given its large ionic radii^[15-17]. Novel host materials for rechargeable batteries must be identified to address these limitations.

Addressing anode issues through electrolyte engineering in Li-battery is a dynamic and multifaceted area of research. The focus is on improving electrolyte composition, stabilizing the solid electrolyte interphase (SEI) layer, preventing dendrite formation and enabling new anode materials. Advancements in solid-state electrolytes, high-concentration electrolytes, and new solvents/additives are playing crucial roles in mitigating the challenges associated with anode performance and lifespan^[18,19]. However, there are still numerous hurdles to overcome before these technologies can be commercialized on a wide scale, especially in terms of scalability, interface stability, and cost-effectiveness. Tin (Sn) and its derivatives are promising candidates for high-density Li and Na storage. However, they suffer from significant volume variations during intercalation and deintercalation, negatively affecting anode performance by disrupting contact with the current collector and reducing cycle life^[20,21]. For instance, researchers have explored 2D graphene (G)-like SnC as a host material for alkali-ion batteries using a computational approach^[22,23]. It revealed that SnC has low average open circuit voltage (OCV), high diffusivity, and moderate binding strength with Li/K loading. In another study, Butt et al. highlighted the potential anodic applications of the SnC monolayer for SIBs^[24]. Despite these encouraging findings, challenges remain, including low electronic conductivity, poor cycling stability, and significant volume changes. Researchers have used 2D materials combined with graphene layers to overcome these obstacles. This strategy leverages the synergistic effects of the specific material and G, resulting in rapid charging/discharging rates, low average voltages, high electronic conductivity, and minimal volume variations^[25-28]. The graphene layer not only provides mechanical strength but also enhances electrical conductivity. Recent studies have highlighted metal sulfides/G and carbides/G composites as ideal energy storage materials for metal-ion batteries^[29-32]. For example, a $\text{MoS}_2/\text{G-H}$ with a molar ratio of 1:2 displayed a high reversible capacity, surpassing pure MoS_2 ^[33]. Additionally, $\text{VS}_2/\text{G-H}$ demonstrated improved performance and higher Li storage capacity than bare VS_2 and G^[34]. Similarly, $\text{SiC}/\text{G-H}$ demonstrated high Li storage capacity and fast lithiation/delithiation processes using density functional theory (DFT) predictions^[35]. Another computational study explored the V_3C_2 MXene/G-H for LIBs, revealing excellent electronic conductivity and accommodating sites for Li storage^[36]. These advances underscore the potential of heterostructures as electrode materials for enhancing energy storage performance.

This study marks the first evaluation of the SnC/graphene heterostructure (SnC/G-H) as an anode material for LIBs and SIBs employing computational insights. We address the challenges associated with SnC, such as lack of electrical conductivity and significant area expansion. While pristine graphene possesses high electrical conductivity and mechanical robustness, it is not suitable for Li/Na storage on its own. The constructed SnC/G-H, which combines the beneficial properties of both materials, could be a promising anode material for both LIBs and SIBs, offering potential improvements in battery performance.

COMPUTATIONAL METHODOLOGY

We performed DFT calculations using the Vienna Ab initio Simulation Package (VASP)^[37]. To investigate ion-electron interactions, we employed the projected augmented wave (PAW) method with the generalized gradient approximation (GGA) and the Perdew-Burke-Ernzerhof (PBE) functional^[38]. The structures were relaxed until the maximum force on each atom was reduced to 1/100 eV/Å, with a total energy convergence of 10⁻⁶ eV. Notably, the cutoff energy was chosen to be 500 eV with a k-point mesh of 9 × 9 × 1 for the optimization of the structure [Supplementary Figure 1]. A 20 Å vacuum space was introduced along the c-direction. The heterostructure of SnC/graphene consisted of a 3 × 3 × 1 supercell for SnC and a 2 × 2 × 1 supercell for graphene. We also accounted for the Van der Waals interactions using the DFT-D3 correction and spin polarization (ISPIN = 2). Before testing as a Li/Na anode, we assessed thermal stability in the temperature 300-500 K range over 5000 time steps. Additionally, we calculated phonon dispersion to evaluate dynamic stability, thermal stability, thermodynamic stability, and in-plane stiffness constants for the mechanical stability of SnC/G-H.

To determine the preferred adsorption sites of Li/Na on 2D SnC/G-H, the adsorption energy was calculated using:

$$E_{ads} = E_{(SnC/G)Li/Na} - E_{(SnC/G)} - nE_{Li/Na} \quad (1)$$

where $E_{(SnC/G)Li/Na}$ denotes the total energy of SnC/G heterostructure with Li/Na adsorption, $E_{(SnC/G)}$ shows the energy of bare SnC/G heterostructure, and $E_{Li/Na}$ describes the energy of a single bulk metallic Li/Na. Consequently, we also envisaged the charge transfer using Bader charge analysis^[39]. We determined the activation energy barrier for Li/Na on the surface and interlayer of SnC/G-H using the climbing image nudged elastic band (CI-NEB) method^[40]. More than that, the OCV for Li/Na intercalation in the heterostructure can be determined from energy differences, excluding the contributions of volume and entropy effects.

RESULTS AND DISCUSSION

Structural and electronic properties

Similar to hexagonal boron nitride (h-BN), the honeycomb hexagonal structure of SnC is composed of two atoms: carbon and tin. Also, a graphene structure comprises a hexagonal ring of a single carbon layer in a 2D atomic crystal^[41]. To maximally avoid lattice match in the SnC/G-H, we employ a 2 × 2 × 1 supercell of SnC monolayer and a 3 × 3 × 1 supercell of G to build the SnC/G-H. To eliminate the coupling effect between adjacent layers caused by periodic boundary conditions, a vacuum of 20 Å is created. The determined lattice parameters are $a = b = 7.35$ Å, Sn-C bond length of 2.12 Å, and C-C bond length of 1.415 Å. The top and side outlooks of the optimized structure of SnC/G-H are shown in Figure 1A and B. We analyzed the band structure and density of states (DOS) of the heterostructure to comprehend the effect of stacking on electronic properties. Notably, at the Fermi level, the Dirac cone of pure graphene remains intact, ensuring that this heterostructure has a high electronic conductivity. The Fermi level touches the conduction band minimum (CBM), resulting in a reduced band gap from 0.9 eV (pristine SnC)^[24] to

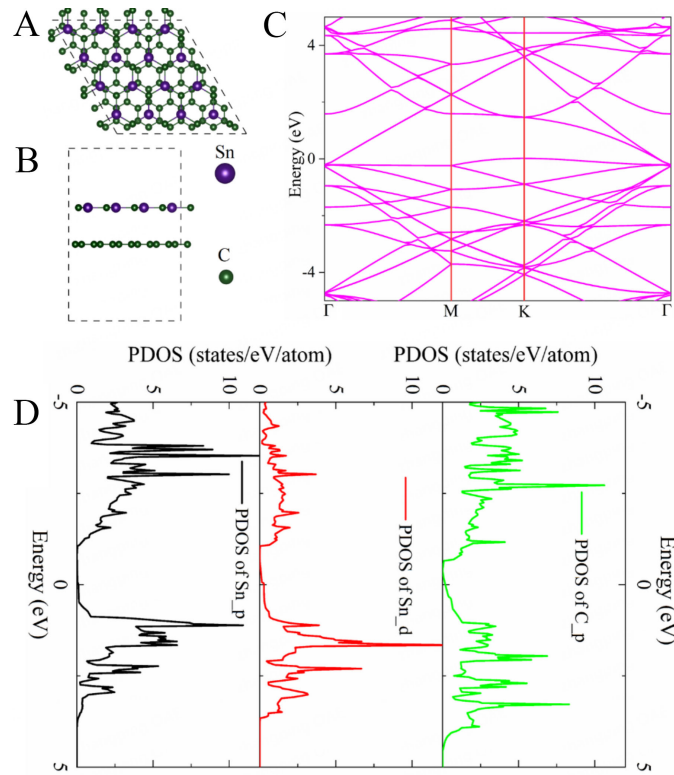


Figure 1. Structural model of SnC/G-H with (A) top and (B) side snapshots; (C) Band structure and (D) PDOS of SnC/G-H. SnC/G-H: SnC/graphene heterostructure; PDOS: Projected density of states.

zero eV in the SnC/G-H. [Figure 1C](#) depicts the band structure of SnC/G-H with the vanishing of the band gap likely attributed to the Van der Waals interaction between SnC and graphene. The projected DOS (PDOS) of the SnC/G-H is plotted in [Figure 1D](#). The p/d-orbitals of the Sn atom and the p-orbital of the C atom mainly contribute to the electronic states near the Fermi level. The whole system shows metallicity, ensuring the high electronic conductivity of the heterostructure.

Stability and experimental validity

In evaluating the experimental feasibility of the SnC/G-H, we focus on its stability for practical applications. Specifically, we investigate its thermal, mechanical, dynamic, and thermodynamic stability. To assess thermal stability, we performed ab initio molecular dynamics (AIMD) simulations at temperatures of 300 K, 400 K, and 500 K, each for a duration of 5000 fs. The results, as depicted in [Figure 2A](#), show minor fluctuations in total energy over time, confirming the thermal stability of the SnC/G structure. Furthermore, [Supplementary Figure 2](#) indicates no bond breakage in the structures post-AIMD simulation. Also, the temperature variation with AIMD time steps at 300-500 K is shown in [Supplementary Figure 3](#), further substantiating their thermal stability.

To assess the mechanical stability, we use Voigt notation to evaluate its response to applied strain. This can be estimated as^[42]:

$$U(\epsilon) = \frac{1}{2}C_{11}\epsilon_{xx}^2 + \frac{1}{2}C_{22}\epsilon_{yy}^2 + C_{12}\epsilon_{xx}\epsilon_{yy} + 2C_{66}\epsilon_{xy}^2 \quad (2)$$

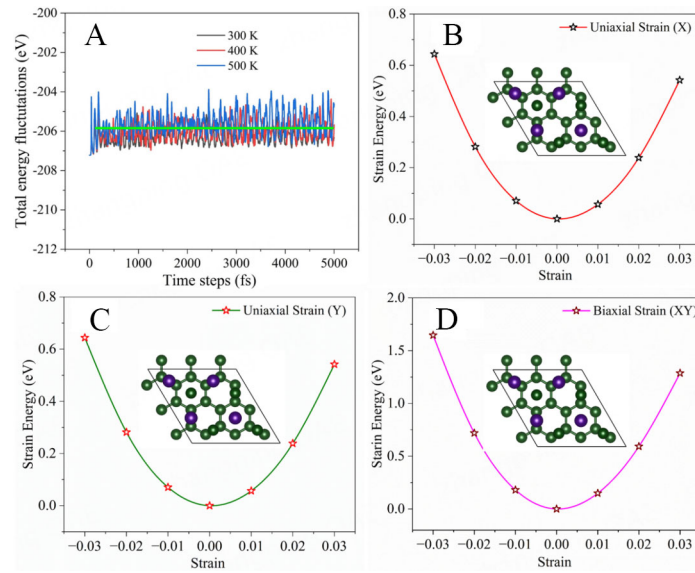


Figure 2. (A) AIMD simulation of SnC/G heterojunction at 300-500 K; (B) Variation of strain energy with applied strain along uniaxial (B) x-direction; (C) y-direction, and (D) biaxial strain along the x- and y-directions. AIMD: Ab initio molecular dynamics.

where C_{11} , C_{22} , C_{12} , and C_{66} are 2D elastic tensors; ε_{xx} is uniaxial strain along the x-direction, ε_{yy} denotes uniaxial strain along the y-direction, and ε_{xy} describes the biaxial strain along the xy-directions. The material must meet the Born-Huang criteria^[40-41] to ensure mechanical stability, given as $C_{11} > 0$, $C_{22} > 0$, $C_{11}C_{22} - C_{12}^2 > 0$, and $C_{66} > 0$. In our calculations, we applied strain from -3.0% to +3.0% with increments of 1.0%. Figure 2B-D illustrates the strain energy at different strain levels and directions. The values of C_{11} and C_{22} were determined by fitting the strain-energy data along the x-axis and y-axis. Similarly, the value of C_{12} was obtained through biaxial planar deformation in the xy-plane. C_{11} is equivalent to C_{22} for a hexagonal system, and an additional relation $C_{66} = \frac{1}{2}(C_{11} - C_{12})$ exists. The calculated values of C_{11} , C_{12} , and C_{66} for SnC/G-H are 457.45, 106.52, and 175.46 N/m, respectively, satisfying the requirements for mechanical stability. Using these relations, the in-plane Young's Modulus (Y) and Poisson's ratio (ν) are calculated from elastic constants, as determined by:

$$Y = \frac{C_{11}^2 - C_{12}^2}{C_{11}} \quad (3)$$

$$\nu = \frac{C_{12}}{C_{22}} \quad (4)$$

The calculated value of Y for SnC/G heterostructure is 432 N/m. At the same theoretical level, this value exceeds that of graphene (340.5 N/m) and the SnC monolayer (123.02 N/m), indicating that the formation of SnC/G heterostructure efficiently enhances mechanical strength. The Poisson's ratio of the SnC/G-H is 0.23, which is greater than 0, satisfying the mechanical stability criteria.

Consequently, the dynamic stability is assessed through phonon dispersion curves, specifically the phonon bands of the SnC monolayer with PHONOPY code, as shown in Supplementary Figure 4. The simulation results show no imaginary frequencies, meaning the phonon bands do not fall below zero. This means that the SnC monolayer is dynamically stable. Since graphene can be synthesized experimentally, it is possible to stack graphene with a SnC monolayer, and thus, one can realize the experimental validity of SnC/G heterostructure. Additionally, we calculate the cohesive energy of SnC/G-H to evaluate its thermodynamic stability. The cohesive energy is determined using $E_{\text{coh}} = [4E_{\text{Sn}} + 22E_{\text{C}} - E_{\text{Sn}_4\text{C}_{22}}]/26$, where $E_{\text{Sn}_4\text{C}_{22}}$, E_{C} ,

and E_{Sn} are the energies of Sn_4C_{22} , C, and Sn atoms, respectively. Notably, the calculated E_{coh} is 7.94 eV/atom, greater than the prior synthesized materials such as Si (4.6 eV/atom) and graphene (7.6 eV/atom)^[43]. Thus, it has a high possibility of being synthesized for practical applications.

Anodic applications of SnC/G heterostructure

Li/Na adsorption

To assess the diffusion and storage characteristics of metal ions within a host material, we examined the preferred adsorption sites for Li/Na on the surface and interlayer of the SnC/G-H. The three considered sites (H, B, and T) are the potential adsorption sites for the adsorption of a single Li/Na atom on the surface and interlayer of SnC/G-H. Consequently, the optimized structures of the Li and Na adsorbed sites are shown in [Supplementary Figures 5 and 6](#), respectively. We determined the adsorption energy (E_{ads}) by employing a $2 \times 2 \times 1$ supercell of SnC/G-H to minimize interactions among adjacent metal atoms and to identify the most advantageous adsorption site. As per equation (1), a more negative E_{ads} signifies an exothermic reaction between SnC/G-H and metal adatoms. The computed adsorption energies of Li/Na at various sites on the surface and interlayer of SnC/G-H are illustrated in [Figure 3A](#). The negative E_{ads} values indicate that nearly all investigated Li/Na content can be efficiently adsorbed onto SnC/G-H. The moderately negative E_{ads} ensures strong adsorption of metal ions on both the surface and interlayer of SnC/G-H. Furthermore, we analyzed the charge transfer of Li/Na on the surface and interlayer, as depicted in [Figure 3B](#). More than that, we calculated the charge density difference using^[22]:

$$\Delta\rho = \rho_{\text{Li/NaSn}_4\text{C}_{22}} - \rho_{\text{Sn}_4\text{C}_{22}} - \rho_{\text{Li/Na}} \quad (5)$$

Where $\rho_{\text{Li/NaSn}_4\text{C}_{22}}$, $\rho_{\text{Sn}_4\text{C}_{22}}$, and $\rho_{\text{Li/Na}}$ are the charge densities of Li/Na- Sn_4C_{22} , bare Sn_4C_{22} (SnC/G-H), and Li/Na, respectively. The calculated charge density difference of the most stable interface sites and the surface of the SnC/G-H with Li and Na adsorption are portrayed in [Figure 3C and D](#), and [Supplementary Figure 7](#), respectively. As shown in these images, the charge transfer of the most stable sites for Li/Na on the surface and interlayer is plotted, with the implication of the charge gathering and shortening as portrayed by the red and cyan clouds, respectively. This means an electrochemical reaction occurs between Li/Na and the host material.

Li/Na diffusion

The diffusion of alkali ions on both the surfaces and within the interlayer of material is pivotal in influencing the kinetics of electrochemical reactions, thereby influencing the charge-discharge characteristics and long-term cycling performance of rechargeable batteries. In this study, we investigate the barrier energy of Li/Na intercalation in the SnC/G heterostructure. To achieve this, we utilize the CI-NEB technique, a well-established method for analyzing the energy barrier associated with Li/Na ion diffusion in the SnC/G heterostructure. Additionally, we explore the discharging/charging process using this method, which offers insights into the minimum energy pathways (MEPs) and transitional states associated with Li/Na ions within the SnC/G structure. For that, a $2 \times 2 \times 1$ supercell of SnC/G was chosen for Li/Na-ion adsorption, with three atomic sites (H, B, and T) considered after achieving full symmetry. We investigate two pathways for Li/Na migration on both the surface and within the interlayer of SnC/G. [Figure 4A and B](#) illustrates the barrier energy of path-I and path-II for Li/Na migration, following the H-B-H and H-T-H pathways, respectively, on the surface of the SnC/G host. The calculated barrier energies for Li/Na migration along path-I are 0.11/0.14 eV and path-II are 0.32/0.23 eV. Furthermore, we examine the mobility of Li/Na within the interlayer of the SnC/G heterostructure. For this purpose, we consider two diffusion pathways for Li/Na such as H-B-H (path-i) and H-T-H (path-ii). The calculated activation barrier energies along path-i and path-ii are 0.079/0.043 eV and 0.18/0.27 eV for Li/Na [[Figure 4C and D](#)], respectively. This

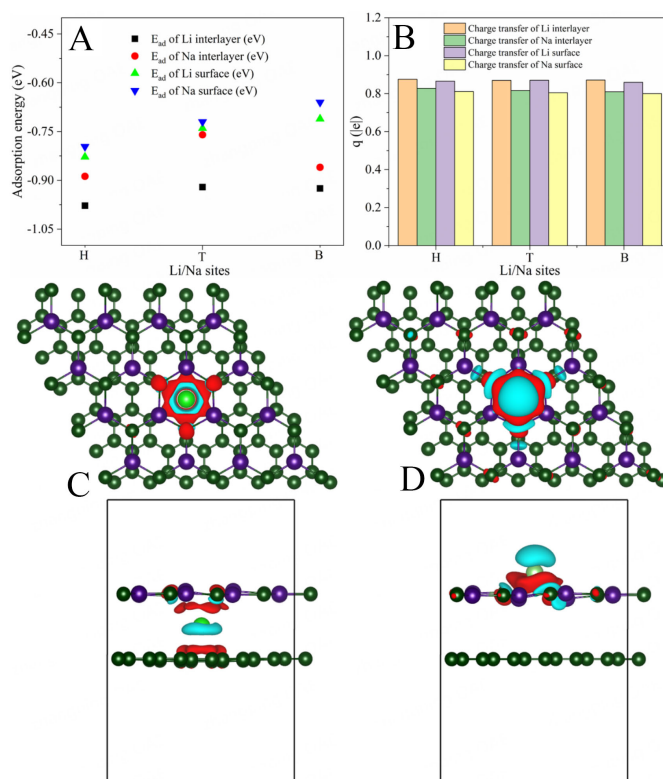


Figure 3. (A) Adsorption energy and (B) charge transfer of Li/Na adsorbed on the surface and interlayer of SnC/G; Charge density difference of the most stable site of Li/Na adsorbed on (C) interlayer and (D) surface of SnC/G.

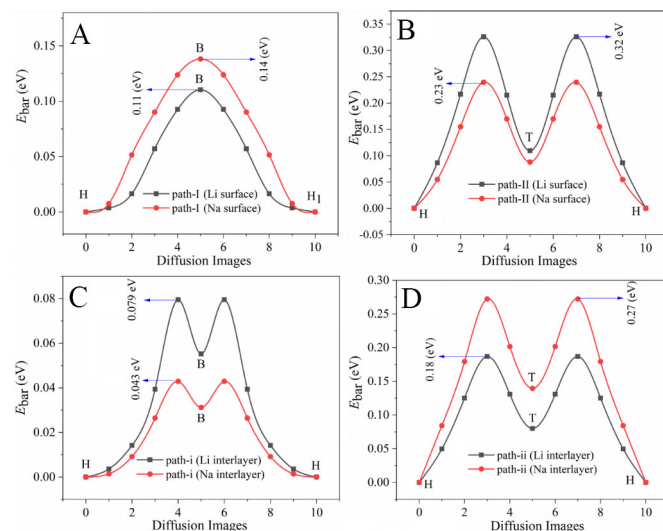


Figure 4. Diffusion energy barriers of Li/Na on the surface along (A) path-I and (B) path-II. E_{bar} of Li/Na migration on the interlayer of (C) path-i and (D) path-ii.

indicates a very low diffusion barrier of 0.079/0.043 eV for Li/Na migration due to the presence of the graphene layer. The pathways of Li/Na migration along the surface and the interlayer are depicted in [Supplementary Figure 8](#). Subsequently, the calculated barrier values are compared with other promising heterostructures reported for LIBs/SIBs [Table 1]. These findings suggest rapid lithiation/delithiation and

Table 1. Comparative analysis of Sn_4C_{22} with previously explored 2D heterojunction materials for LIBs and SIBs

Materials	E_{bar} (eV)	C (mA h g ⁻¹)	Voltage (V)	Ref.
Li@SiC/G	0.63	1229	0.29	[35]
Li@ BlueP/G	0.13	626	0.23	[54]
Li@SnC	0.33	410	0.44	[22]
Li@WS ₂ /G	0.27	588	0.18	[55]
Li@MoO ₃ /G	0.07	1411	0.35	[56]
Na@SnSe ₂	0.10	387	0.68	[10]
Na@SnS ₂ /G	0.25	586	1.0	[57]
Na@SnB	0.22	620	0.42	[58]
Na@SnC	0.17	205	0.24	[24]
Na@Si/G	0.3	487	0.84	[59]
Na@VS ₂ /G	0.08	578	0.46	[48]
Na@GeS/G	0.05	714	0.35	[60]
Li@SnC/G	0.079	870	0.39	This work
Na@SnC/G	0.043	870	0.34	This work

LIBs: Lithium (Li)-ion batteries; SIBs: Sodium (Na)-ion batteries.

sodiation/desodiation, indicating high mobility in the SnC/G heterostructure for LIBs and SIBs.

Li/Na storage capacity

Theoretically, Li/Na storage capacity stands as another crucial parameter in assessing battery performance, directly linked to the amount of Li/Na content adsorbed on host materials. Ideally, a promising anode should have extensive storage capacity and low voltage profiles. Thus, we delved into the Li/Na adsorption properties of the SnC/G structure at stable adsorption sites across varying Li/Na concentrations. We explored the high storage capacity by loading the Li/Na content to the maximum amount. The theoretical Li/Na storage capacity (C) was determined using $C = (zx_{\text{max}}F)/M_{\text{SnC/G}}$, where $M_{\text{SnC/graphene}}$ denotes the molecular weight of the SnC/graphene formula unit, x_{max} represents the maximum concentration of Li/Na adsorbed on SnC/graphene, z is the charge number of Li/Na, and F is Faraday's constant (26.801 A h mol⁻¹). To achieve maximum storage capacity, Li/Na concentrations are layered gradually onto the SnC/G-H. Even at maximum Li/Na concentration, we observed that lithiated/sodiated configurations remain stable with negative E_{ads} . All Li- or Na-adsorbed structures maintain their structural integrity well, signifying high stability. A fully lithiated and sodiated state of the SnC/G-H cell can accommodate a maximum of 6 content ($x = 6$ for Li/Na), resulting in a corresponding Li/Na storage capacity of the host material reaching 870 mA h g⁻¹, as depicted in Figure 5. This theoretical Li/Na storage capacity surpasses that of other graphene-based 2D heterostructures, as outlined in Table 1, and commercially designed anodes. Moreover, we also compared prior experimentally available 2D heterostructures with SnC/G-H [Supplementary Table 1]. Notably, the high theoretical Li/Na storage capacity designates SnC/G-H as a promising anode material for LIBs and SIBs.

Li/Na potentials

To further investigate the electrochemical performance of the SnC/G heterostructure for anodic applications, we examine the average potentials of Li/Na. The lithiation/sodiation processes of the SnC/G-H as a Li/Na anode are evaluated through the average OCV. For the proper functioning of rechargeable batteries, there should be low voltage at the anode and high voltage at the cathode side. The half-cell reactions for Li_xSnC/G-H and Na_xSnC/G-H are expressed as $\text{Sn}_4\text{C}_{22} + x\text{Li}^+ + xe^- \rightleftharpoons \text{Li}_x\text{Sn}_4\text{C}_{22}$ and $\text{Sn}_4\text{C}_{22} + x\text{Na}^+ + xe^- \rightleftharpoons \text{Na}_x\text{Sn}_4\text{C}_{22}$, respectively. In these reactions, negative charges migrate toward the external

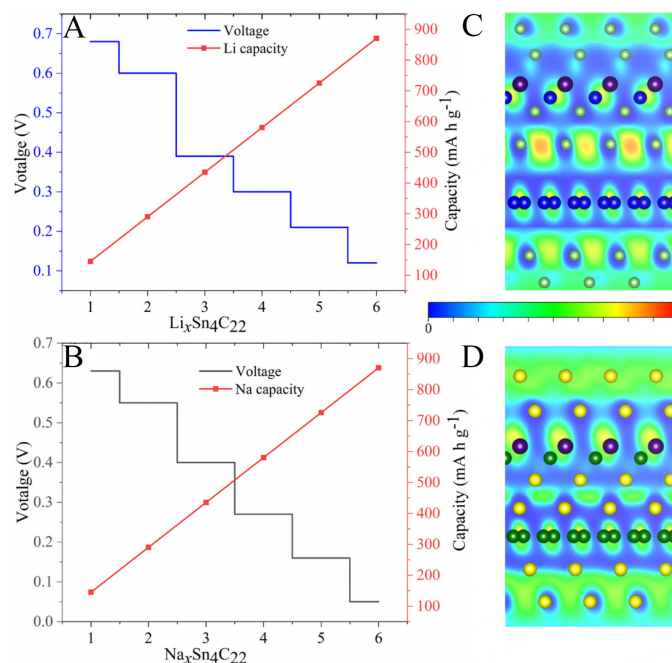


Figure 5. Open circuit voltages of (A) Li_xSnC/G and (B) Na_xSnC/G with their Li/Na storage capacities. ELF plots of fully (C) lithiated and (D) sodiated SnC/G. ELF: Electron localization function.

circuit, while positive charges move between the electrodes and electrolytes of the battery. The lithiation/sodiation processes of the SnC/G-H as Li/Na anode are estimated based on the average OCV using^[44],

$$\text{OCV} = [-E_{\text{Li/Na}_x\text{SnC/G}} + E_{\text{SnC/G}} + xE_{\text{Li/Na}}]/zxe \quad (6)$$

Where $E_{\text{Li/Na}_x\text{SnC/G}}$ is the energy of the Li/Na-adsorbed SnC/G, $E_{\text{SnC/G}}$ is the energy of the pure SnC/G, and $E_{\text{Li/Na}}$ is the energy of per bulk Li/Na. Consequently, the voltages for Na/Li are illustrated in Figure 5A and B. The results reveal a decreasing trend with increasing concentrations of Li/Na, which can be attributed to the intensification of Coulombic interactions between Li⁺-Li⁺ or Na⁺-Na⁺^[45-47]. The computed average OCVs are 0.39 V and 0.34 V for LIBs and SIBs, respectively. To assess the efficiency of our material, we compare it with previously well-documented heterostructures, such as 0.30 V for Li@SiC/graphene^[35], 0.59 V for Li@BN/VS₂^[48], 0.46 V for Na@VS₂/graphene^[49], 0.72 V for Na@BC₃N₂/graphene^[50], and commercially used materials, i.e. 0.11 V for graphite and 1.5-1.8 V for TiO₂^[51,52]. Furthermore, we have compared the results illustrated in Table 1, suggesting that 2D SnC/G heterostructure has low average voltages, and is an appealing anode material for LIBs and SIBs. We then determined the bonding characteristics by analyzing the Electron Localization Function (ELF). The results showed active interaction between the layers of Li or Na and the SnC/G-H (see Figure 5C and D). Charge transfer predominantly occurred from Li or Na ions to the carbon atoms, forming ionic bonds (Li-C or Na-C), which enhanced the stability of the adsorption layer. This illustrates that the SnC/G-H can effectively accommodate multilayer adsorption of Li/Na, leading to an improved Li/Na storage capacity over SnC MXene.

Cycling stability and comparative study of the host

Subsequently, we further examined the structural stability of the fully intercalated Sn₄C₂₂ electrode at room temperature [Figure 6A and B]. The Sn₄C₂₂ structure remains intact after the 10,000 fs simulations in all these intercalated systems. The energy fluctuation plots over time demonstrate remarkable thermal stability

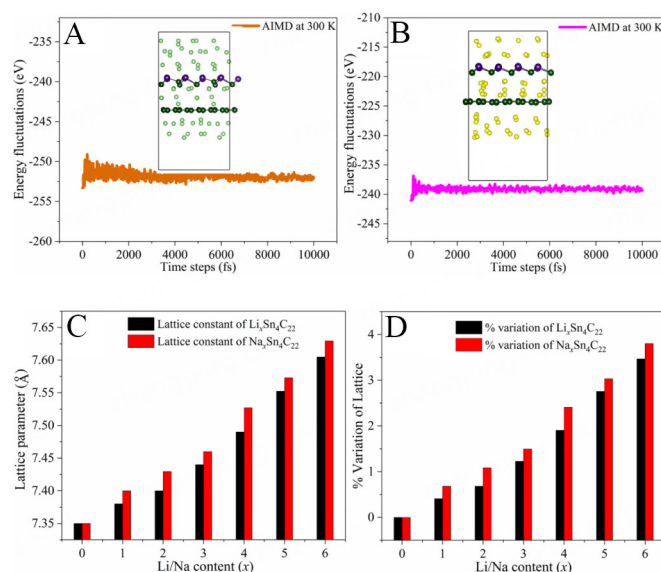


Figure 6. AIMD simulations of (A) Li₆SnC/G and (B) Na₆SnC/G with their optimized structures; (C) Calculated lattice parameters of Li_xSnC/G and Na_xSnC/G; (D) Percentage variation of Li_xSnC/G and Na_xSnC/G. AIMD: Ab initio molecular dynamics.

of the fully lithiated and sodiated SnC/G-H, as evidenced by the linear energy fluctuation trends depicted in Figure 6A and B. It is noteworthy that no significant deformations or bond breakages occur in the SnC/G during Li and Na intercalation; the adsorbed metal ions only deviate slightly from their equilibrium positions. Upon removal of all Li/Na concentrations from the intercalated SnC/G structure, AIMD simulations reveal that this slightly distorted SnC/G structure can swiftly regain its planar configuration, indicating the robust thermal stability of the SnC/G heterostructure during Li/Na-ion intercalation/extraction.

In addition, we investigate the cyclic behavior of an anode, a critical determinant in the electrochemical functionality of rechargeable batteries. Cycling stability hinges on the volume expansion resulting from intercalation/deintercalation during charging and discharging. The volume expansion was calculated by considering the lattice parameters following the full adsorption of Li/Na. In the case of the SnC/G heterostructure, the lattice parameters expanded from 7.35 to 7.605 Å for Li and from 7.35 to 7.629 Å for Na, as depicted in Figure 6C and D. This expansion amounted to 3.46% and 3.80% for fully lithiated and sodiated SnC/G, respectively, resulting in a modest volume expansion that bolsters the cycling stability of the anode. Notably, the lattice variations in Sn₄C₂₂ due to Li/Na adsorption and desorption are lesser than those observed in graphite during lithiation and delithiation, typically around 10%^[53]. This suggests that the structure of Sn₄C₂₂ remains largely unaffected after complete lithiation/sodiation, which holds practical significance. The stability of the host in aqueous environments is essential for their practical application in alkali-ion batteries. To investigate this, we conducted AIMD simulations of the SnC/G heterostructure interacting with H₂O molecules at room temperature for 5000 fs. The results, illustrated in Supplementary Figure 9, demonstrate remarkable stability in the solution, emphasizing its potential for use in rechargeable battery applications.

To assess the viability of Li@ Sn₄C₂₂ and Na@ Sn₄C₂₂ heterostructures as host materials for LIBs and SIBs, we conducted a comparative analysis of diffusion barrier energy, voltages, and Li/Na storage capacity against other commonly utilized heterostructure anode materials, as elaborated in Table 1. Our results reveal that the Sn₄C₂₂ heterojunction exhibits a notably higher Li/Na storage capacity than the materials documented in

[Table 1](#) and [Supplementary Table 1](#). Moreover, the calculated values of Energy Barrier (E_{bar}) and average OCVs are juxtaposed, showcasing enhanced mobility and swift charging/discharging rates. It is noteworthy that the Li/Na storage capacity of the Sn_4C_{22} host surpasses that of the conventional anode (graphite) by a factor of two. This examination underscores the emergence of Sn_4C_{22} heterojunction as a promising contender for Li/Na anodic applications.

CONCLUSIONS

In summary, our systematic investigation using DFT calculations focused on the SnC/G heterojunction as an anode material for LIBs and SIBs. Here are the key findings:

➤ Stability and thermodynamics

○The cohesive energy of Sn_4C_{22} is 7.94 eV per atom, indicating that the formation of the Sn_4C_{22} heterojunction is exothermic and thermodynamically stable.

○SnC/G remains stable even at elevated temperatures (300 K, 400 K, and 500 K), demonstrating robust thermal stability.

○Mechanical and dynamic stability are also confirmed, highlighting strong chemical bonds in the SnC/G heterostructure.

➤ Li/Na intercalation

○The Sn_4C_{22} heterostructure shows excellent potential for capturing Li/Na ions as an anode material.

○Li/Na intercalated structures remain stable thermally.

○Low Li/Na-ion migration barriers (0.079/0.043) suggest fast charging/discharging processes.

➤ Capacity and voltage

○ Sn_4C_{22} can store up to six Li/Na content in a single unit cell, providing a high theoretical Li/Na storage capacity of 870 mA h g⁻¹.

○Average voltages of 0.34 V (Li) and 0.39 V (Na) indicate moderate energy profiles.

Overall, the SnC/G heterostructure shows great potential as a durable electrode material for both LIBs and SIBs.

DECLARATIONS

Authors' contributions

Writing - original draft, investigation, methodology, investigation, formal analysis, data curation, and conceptualization: Rehman, J.

Writing - review and editing, methodology, investigation, formal analysis, data curation: Butt, M. K.

Writing - review and editing, conceptualization, formal analysis: El-marghany, A.

Writing - review and editing, supervision, visualization, formal analysis: Li, Z.

Writing - review and editing, supervision, formal analysis, data curation, conceptualization: Yang, G.

Availability of data and materials

The data supporting this study's findings are available from the corresponding author upon reasonable request.

Financial support and sponsorship

This work was supported by the National Natural Science Foundation of China (No. 52350410474 and No. 22372142), Yanshan University, China, and the Researchers Supporting Project Number (RSPD2024R667), King Saud University, Riyadh, Saudi Arabia.

Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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