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Full Length Article

Theoretical predication of a two-dimensional structure of non-layered ScC as an excellent electrode material for rechargeable Na-ion battery

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ABSTRACT

Non-layered two-dimensional structures with the atomic thickness recently attract much attention. In this work, we predicated a two-dimensional structure of non-layered ScC, denoted as ScC sheet, and evaluated its potential as the electrode material of Na-ion battery. The stability of ScC sheet is determined by the cohesive energy, phonon spectrum and potential energy curve calculations. Interestingly, the proposed ScC sheet can exhibit many unexpected electrochemical properties. It is found that ScC sheet can possess strong Na adsorption capability. Both pristine and Na-interacted ScC sheets represent good electronic conductivity due to the metallic nature. The structural degradation of the ScC during the sodiation and desodiation processes will not be anticipated. Besides, our results reveal that the ScC sheet has a minimal diffusion barrier of 9 meV and the calculated theoretical capacity of ScC sheet can reach to 940 $mAh{\cdot}g^{-1}$ on the basic of the second layer adsorption. Thus, benefiting from these features, ScC sheet is expectable to be served as a promising electrode material for the Na-ion battery.

1. Introduction

In order cope with the stress of the gradual depletion of fossil energy and ever-increasing global population, renewable and sustainable energy storage systems move to the center of current research. Among all kinds of energy storage systems, Li-ion battery (LIB) attracts immense interest [1,2]. Until now, LIB with the unique features, such as high energy density, long cycle life and desirable power performance, has been used in different electronic devices [3,4]. For example, the mobile phone and the laptop are closely associated with the LIB. As a comparison, Na-ion battery (NIB) can own not only the low cost due to the abundant natural sodium deposits but also the operational safety and environmental friendship [5,6]. Thus, the development of NIB is essential and indispensable that the NIB is expectable to become an ideal candidate for the application of energy storage in the future.

Similar to the LIB, a critical problem for the development of advanced rechargeable NIB is to search for suitable electrode materials. To tackle this issue, two-dimensional (2D) nanomaterials begin to attract more and more attention [7,8]. Comparing with the bulk structure, the reduction of the dimensionality can effectively limit the expansion of the volume during the charge and discharge cycling processes [9]. Furthermore, the flat surface and large surface area of 2D structure can establish them as high energy densities and fast ion motilities that cannot occur in the bulk analogue [10–13]. Currently, 2D structures, such as graphene derivatives [14,15], transition-metal dichalcogenides [16–18], phosphorene [19], MXenes [20–24] and borophene [25–28], have been received considerable interest for the NIB due to the experimentally effective syntheses. Meanwhile, other 2D materials, such as β_0-PC [29], Mo_2C sheet [30,31], Y_2C sheet [32], Ca_2N sheet [33] and Mo_2N sheet [34], also have been predicated and confirmed as excellent Na-electrode materials. Nevertheless, the footstep seeking for suitable electrode material that can deliver all desirable electrochemical properties including strong adsorption capability, high electronic conductivity, stable cycling performance, superior rate capability and high capacity will never be stopped.

Recently, non-layered 2D structures with the atomic thickness, such as SnO_2, α-Fe_2O_3 and TiO_2 sheets, entered our awareness [35–37]. In this work, by combining cohesive energy, phonon spectrum and potential energy curve calculations, we predicated a new non-layered 2D structure, ScC sheet. ScC sheet can be viewed as the 2D counterpart of the rocksalt ScC structure, as shown in Fig. 1S in Supporting Information. Via the calculations of electrochemical properties including adsorption behavior, electronic conductivity capability, structural stability, diffusion capability and theoretical capacity, we systematically evaluated the potential of ScC sheet as the electrode material for NIB. Our results revealed that Na atom can bind strongly to the ScC sheet,

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and both pristine and Na-interacted ScC sheets represent excellent electronic conductivity due to the intrinsic metal property. Besides, the structural degradation of the proposed ScC during the sodiation and desodiation processes will not be anticipated. More importantly, we found ScC sheet possesses a very low Na ion diffusion barrier of 9 meV and a high theoretical capacity of 940 $mAhg^{-1}$. All of these interesting features suggest that the non-layered ScC sheet is expectable to be applied for the electrode material of NIB.

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.apsusc.2018.08.138.

2. Computational methods

Our calculations have been carried out in the framework of density functional theory (DFT) as implemented in the Vienna *ab initio* simulation package (VASP). The exchange correlation energy is treated by the generalized gradient approximation (GGA) in the scheme proposed by Perdew-Burke-Ernzerhof (PBE). The electron wave functions were expanded by a plane wave cutoff of 550 eV. For the simulations of adsorption and diffusion of single Na atom, the ScC sheet was modeled by using a $2\sqrt{2} \times 2\sqrt{2}$ supercell, and for the simulation of Na adsorption at high concentration, the ScC sheet was modeled by using a $2\sqrt{2} \times \sqrt{2}$ supercell. For both supercells, a vacuum space of 25 Å was applied to avoid the interaction between neighboring images. On the basic of Monkhorst and Pack scheme, a $8 \times 8 \times 1$ grid for k-point sampling was used for the calculation of $2\sqrt{2} \times 2\sqrt{2}$ supercell and a $8 \times 16 \times 1$ grid for k-point sampling was used for the calculation of $2\sqrt{2} \times \sqrt{2}$ supercell. To investigate the diffusion behavior of Na on ScC sheet, we have performed a series of nudge elastic band calculations. The relaxation was stopped until the change of the force on each atom was smaller than 0.02 eV/Å.

3. Results and discussion

To the beginning, we firstly investigated the geometric structure and stability of ScC sheet. The physical model of ScC sheet obtained from the rocksalt ScC structure was used as the initial configuration for the optimization. As shown in Fig. 1a, we can see that the ScC sheet exhibits a rectangular configuration and each ScC sheet consists of two atomic layers. In ScC sheet, per Sc atom binds to five neighboring C atoms and per C atom binds to five neighboring Sc atoms. For 1×1 unit cell, the optimized lattice constant, bond length and thickness of ScC sheet are about 3.22, 2.28 and 2.34 Å, respectively. To access the feasibility of the predicated ScC sheet, we calculated its cohesive energy defined as:

$$E_{coh} = (nE_C + nE_{Sc} - E_{ScC})/2n$$

where E_C, E_{Sc} and E_{ScC} are the energies of a single C atom, a single Sc atom and the ScC sheet, respectively. According to our computations, ScC sheet has a cohesive energy of 5.85 eV/atom, which is comparable with that of previous reported 2D structures, such as Cu_2Si sheet (3.46 eV/atom) [38], Mn_2C sheet (3.35 eV/atom) [39], Be_2C sheet (4.86 eV/atom) [40], h-BeS sheet (4.86 eV/atom) [41], FeB_2 sheet (4.87 eV/atom) [42] and MoS_2 sheet (5.05 eV/atom) [43]. Then, we investigated its phonon spectrums. Since a 1×1 unit cell that contains four atoms (2 Sc atom and 2 C atoms) was used during the calculation, 12 phonon branches including 4 acoustic branches and 8 optical branches were clearly observed in Fig. 1b. Note that, all vibrational modes are found to be real in the whole Brillouin Zone. The highest frequency of the ScC sheet located at the symmetric X and Y points reaches up to 500 cm^{-1}. Although this value is smaller than that of Mn_2C sheet (700 cm^{-1}) [39], Be_2C sheet (1020 cm^{-1}) [40], h-BeS sheet (760 cm^{-1}) [41] and FeB_2 sheet (870 cm^{-1}) [42], it is still larger than the values of experimentally achieved MoS_2 sheet (473 cm^{-1}) [44] and phosphorene (450 cm^{-1}) [45]. Thus, ScC sheet is kinetically stable. In addition, *ab initio* molecular dynamics (AIMD) simulation was also performed to further evaluate its thermal stability. The simulations were carried out for 5000 steps with a time step of 1 fs. As shown in Fig. 1c, we found that the average value of the temperature is about 300 K and the average value of the potential energy remains nearly a constant during the entire simulation. Besides, after optimization, we further found that the framework of ScC sheet was kept in its original structure very well, as given in Fig. 1S in Supporting Information. Thus, ScC sheet can be thermally stable at the 300 K. These results indicate that it is possible to realize a freestanding 2D ScC structure.

A fundamental requirement of a material as the Na-electrode is that the material must have strong adsorption capability to the Na atom. According to this, we investigated the adsorption energy of Na atom on ScC sheet. Herein, a $2\sqrt{2} \times 2\sqrt{2}$ supercell was employed to avoid the interaction between the adjacent Na atoms. Four high-symmetry adsorption sites, denoted as S_1, S_2, S_3, S_4 sites, were considered. As depicted in Fig. 2, the S_1 site is directly above a C atom, the S_2 site is above the center of a tetragonum composed by 2C and 2 Sc atoms, the S_3 site is directly above a Sc atom, and the S_4 site is above the bridge of C-Sc binding. The stabilities of the adsorption structures were determined by calculating their adsorption energies, which are defined as:

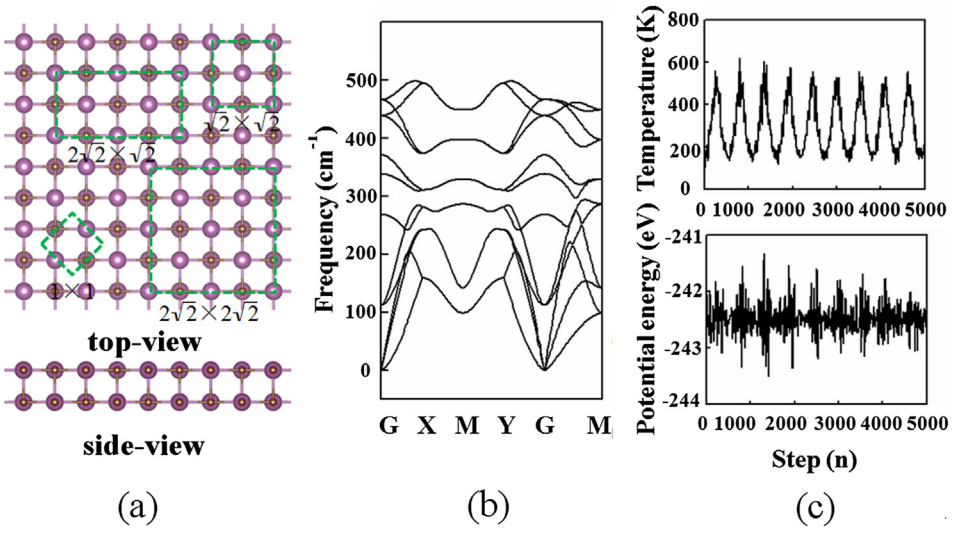

Fig. 1. (a) Top and side views of ScC sheet. The purple and brown balls represent Sc and C atoms, respectively. (b) The phonon dispersion curves of ScC sheet. (c) Variation of temperature and potential energy of ScC sheet with respect to the simulation step.