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Full Length Article

A flexible three-dimensional MoS_2/carbon architecture derived from melamine foam as free-standing anode for high performance lithium-ion batteries

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ABSTRACT

MoS_2 has been investigated extensively as novel energy storage materials because of its unique physical and chemical properties. Herein, a flexible Three-Dimensional (3D) free-standing composite is rationally designed and synthesized by growing MoS_2 nanosheets onto carbonized melamine foam. Afterwards, nitrogen-doped carbon (NC) coating is created on the bare MoS_2 nanosheets depends on dopamine self-polymerization. The composite can be used directly as a free-standing anode for lithium-ion batteries. It exhibits high reversible lithium storage capacity with good cyclic performance (573.8 mAh g^{-1} at current density of 500 mA g^{-1} after 200 cycles) and rate capability (536.9 mAh g^{-1} at current density of 2000 mA g^{-1}). The outstanding electrochemistry properties are mainly attributed to the well-designed architecture. The interconnected carbon skeleton provides 3D fast transport paths for electron. NC coating on MoS_2 nanosheets and the carbon filled in the interspaces between MoS_2 nanosheets serves as conductive matrix which enhances the electrical conductivity, expands the path of electron transport and accelerates electron transfer.

1. Introduction

Lithium-ion batteries (LIBs) have been widely used in portable electronics, power tools, and has been extensively concerned as power sources of electrical vehicles. Exploring new electrode materials with high capacity and excellent cyclic stability is the key to meet the requirements of high-performance LIBs [1–3]. Currently, transition metal dichalcogenides (TMDs) MX_2 (M = Mo, W, V, Ti; X = S, Se) have gained considerable research interests because of their unique physical and chemical properties. These materials have layered structures formed by van der Waals interaction between nanosheets. Transition metal dichalcogenides has been investigated extensively as an anode material for LIBs [4–6].

As a typical layered transition metal dichalcogenide, molybdenum disulfide (MoS_2) delivers a similar structure to graphene, which consists of three atom layers (S-Mo-S) superimposed together. The layered structure and weak van der Waals interaction make it easier for lithium ions intercalation and extraction without significant MoS_2 volume change [7,8]. As an anode materials for LIBs, MoS_2 delivers a higher theoretical specific capacity of 670 mAh g^{-1} than that of 372 mAh g^{-1} for commercial graphite [9,10]. However, it suffers from capacity decay rapidly and poor rate performance for its low conductivity, which are the main obstacles that limit its practical applications. To address these problems, researchers devised various strategies to enhance the electrochemical performance of MoS_2 by introducing conductive carbon-based materials, including graphene, carbon nanotube, conducting polymers and other carbon matrix composite [11–16].

Generally, MoS_2 composites in powder form have to be evenly mixed with conductive additives (such as acetylene black) and polymer binders, and lastly coated on current collectors (Cu foil). The additional conductive additives, binders and metallic current collectors reduce the energy and power density of batteries greatly, and they also have a negative impact on the rate and cycle performance of LIBs [17]. Recently, a new kind of electrode design strategy has been applied to fabricate free-standing LIBs electrodes by direct coating and growing active materials on self-supported 3D materials without conductive additives and binders [18,19]. These materials provide highly efficient electronic transport due to their 3D conductive paths. And they can greatly accelerate the exchange of lithium-ions during the charging/discharging. The intercalation and extraction of lithium-ions are much faster than that of bulk materials. In addition, 3D architecture can effectively avoid the aggregation of active materials and furnish more

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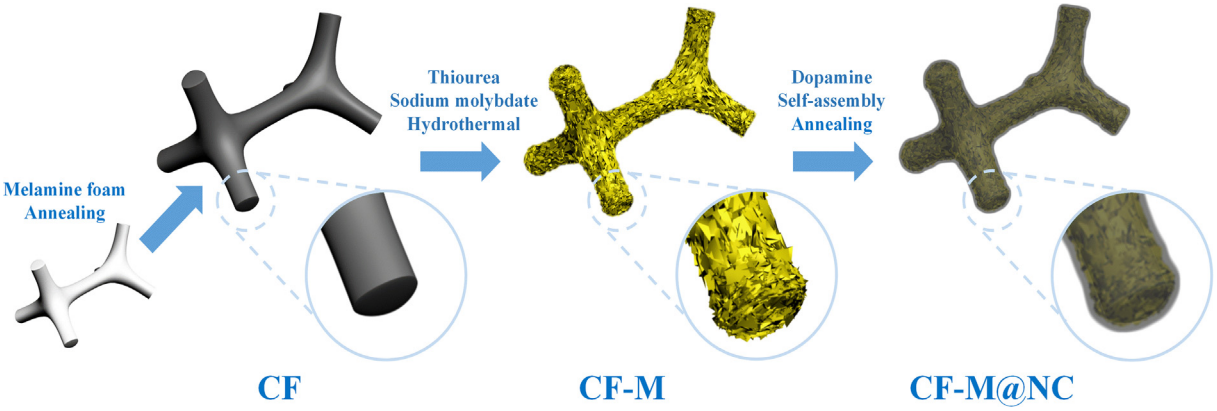

Fig. 1. Schematic illustration of the synthesis process of CF-M@NC.

active sites for lithium-ions, which can enhance the capacity and cycle performance of active materials in batteries [20–23].

In this work, we fabricate a flexible 3D free-standing composite material by growing MoS_2 nanosheets on the surface of a carbonized melamine foam (CF), and create a polydopamine (PDA) coating on the MoS_2 nanosheets surface. In practice, both CF and PDA can introduce nitrogen element, creating N-doped carbon structure during annealing progress [24,25]. As an anode material for LIBs, the interconnected carbon acts as the framework and provide fast transport paths for electrons and lithium-ions. The composite material as a free-standing electrode exhibit high reversible capacity, long cycle ability and excellent rate capability.

2. Experimental

2.1. Materials

All chemicals are analytical grade without any further purification. Commercial melamine foam (MF) was purchased from SINOYQX Co. Ltd., Sichuan. Thiourea, Sodium molybdate and Dopamine hydrochloride (DOPA-HCl) were purchased from Aladdin-reagent Ltd.

2.2. Synthesis of CF-M and CF-M@NC

MF was first washed with distilled water and dried at 80 °C. Then, the clean MF was calcined in tube furnace at 800 °C for 1 h with a heating rate of 5 °C min^{-1} under flowing argon gas. After cooling to ambient temperature, carbonized melamine foam (CF) was obtained. Next, CF was punched into circular disks with a diameter of 12 mm and the weight is about 1.8 mg per disk.

CF-M was synthesized by using a simple one-step hydrothermal method. A typical procedure was as follows. 1 mmol of Sodium molybdate and 4 mmol of Thiourea were dissolved in deionized water (DI)/ethanol mixture (4:1 in volume) of 40 mL under the continuously stirring for 30 min. The obtained CF disks were immersed in above solution. Next, the mixture was transferred into a 50 mL Teflon-lined stainless steel autoclave and heated at 200 °C for 12 h. The obtained disks were washed with DI water, and dried at 60 °C. Then, 3D carbon foam (CF) hybrids coated with MoS_2 were obtained and designated as CF-M. The weight is about 4.0 mg per CF-M hybrid.

Appropriate amount of dopamine hydrochloride was dispersed in Tris-buffer aqueous solution (30 mL, 10 mM, and pH = 8.5). Subsequently, CF-M disks were immersed in the solution for 24 h at ambient temperature. Afterwards, the CF-M@PDA disks were washed with DI water and ethanol, and dried overnight at 80 °C under vacuum. The obtained sample was heated in a furnace tube to 800 °C for 2 h with a heating rate of 5 °C min^{-1} under flowing argon atmosphere. Since the PDA coating converted to N-doped carbon coating, the obtained sample was denoted as CF-M@NC. Each slice weighs about 5.0 mg.

2.3. Structural and morphological characterization

The morphology characterization was studied by a field-emission scanning electron microscope (FESEM) of Hitachi S4800 (accelerating voltage, 5 kV) equipped with an energy-dispersive X-ray spectroscopy (EDX; Oxford XMax) system. The high-resolution transmission electron microscopy (HRTEM) was performed using a FEI Tecnai G2 F30 microscope at 300 kV. X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Focus diffractometer using Cu Kα radiation generator at a scan rate of 5° min^{-1} in the range of 10–70°. The thermogravimetric analysis (TGA) was studied using a STA449 thermal analyzer in air with a heating rate of 10 °C min^{-1} from room temperature to 800 °C. X-ray photoelectron spectroscopy (XPS) analysis was observed by a Thermo Scientific Escalab 250 XI X-ray photoelectron spectrometer.

2.4. Electrochemical characterization

For the electrochemical performance tests, CR2032 coin-type cells were assembled in a dry glove box filled with protective argon gas, directly using the resultant CF-M@NC composites as cathode, lithium foil as anode, and Celgard 2400 membrane as the separator. 1 M $LiPF_6$ in a mixture of ethylene carbonate (EC) and dimethyl carbonate (DMC) (1:1 by volume) was used as the electrolyte. Galvanostatic charge-discharge measurements were performed on Neware battery tester (BTS-5 V 10 mA) at various current rates in the voltage range of 0.01–3 V at room temperature. Cyclic voltammetry (CV) and Electrochemical impedance spectroscopy (EIS) measurements was tested on an electrochemistry workstation of Princeton Applied Research while the CV test was between 0.01 and 3.0 V at a scanning rate of 0.1 mV s^{-1} and the EIS test was in a frequency range of 10^{-2}–10^5 Hz with an AC voltage amplitude of 5 mV.

3. Results and discussion

The fabrication process for CF-M@NC is illustrated in Fig. 1. Firstly, CF is prepared from melamine foam through a one-step pyrolysis process. After carbonized, CF also maintains a good flexible 3D structure. After hydrothermal progress, the grown MoS_2 nanosheets are anchored onto the skeleton of CF by self-assembly. Dopamine can easily coat on various material surfaces by self-polymerization in Tris-buffer solution [26–28]. After adding dopamine, CF-M@PDA is obtained by a homogeneous and ultrathin polydopamine membrane created on the surface of MoS_2 nanosheets. The PDA also fills the interspaces between MoS_2 nanostructures in the CF-M. Finally, the CF-M@NC with an N-doped carbon layer is obtained through a carbonization process of the obtained CF-M@PDA. NC layers filling in the interspaces are considered to

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