Supporting Information

Three-Dimensional Crumpled Reduced Graphene Oxide/MoS₂ Nanoflowers: A Stable Anode for Lithium-Ion Batteries

Fangyu Xiong, ‡ Zhengyang Cai, ‡ Longbing Qu, Pengfei Zhang, Zefang Yuan, Owusu Kwadwo Asare, Wangwang Xu, Chao Lin, Liqiang Mai*

State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070.

*E-mail: mlq518@whut.edu.cn

EXPERIMENTAL SECTION

All the reagents used in the experiment are analytical grade without further purification. Commercial carbon fiber cloths were cleaned in 0.1 M HCl solution, deionized water and ethanol with ultrasonic treatment for 30 minutes, respectively.

Preparation of MS and MS-CG. Nanosheet-assembled MoS₂ nanoflowers were prepared through a facile hydrothermal method. In a typical procedure, Na₂MoO₄ (1 mmol) and CSN₂H₄ (9 mmol) were dissolved in deionized water (65 mL) under stirring to get a transparent solution. Then the solution was transferred into a 100 mL Teflon-lined stainless steel autoclave with a piece of cleaned carbon fiber cloth (3*4 cm²) immersed into the reaction solution. The autoclave was sealed and maintained at 180 °C for 24 hours, and cooled to room temperature. The carbon fiber cloths were taken out, ultrasonically cleaned for 5 minutes in deionized water and rinsed with ethanol for several times, dried at 70 °C for 12 hours and finally annealed at 750 °C for 2 hours in a H₂/Ar (5%/95%) atmosphere. The MS-CG were obtained via replacing the deionized water (65 mL) by the deionized water (60 mL) and graphene oxide suspension (5 mL, synthesized through a modified Hummer's method).

Preparation of plain RGO/MoS₂ nanoflowers (MS-PG). The procedure was carried out in a three-electrode system, using the MS electrode as the working electrode, Ag/AgCl as the reference electrode and Pt foil as the counter electrode. Electrolyte for electrochemical deposition was obtained by dissolving amount of graphene oxide suspension (synthesized through a modified Hummer's method). into Na₂HPO₄ solution (100 mL, 0.1 M). The graphene oxide was deposited and partial

reduced through a chronopotentionmetry method at a constant current of 2 mA/cm^2 in a potential range of (-2)-0 V for 300 seconds. And then the electrode was rinsed with deionized water several times and dried at 70 °C for 6 hours.

Characterizations. X-ray diffraction (XRD) measurements were performed by using a Bruker D8 Advance X-ray diffractometer with a non-monochromated Cu Kα X-ray source. Raman spectra were acquired using a Renishaw RM-1000 laser Raman microscopy system. X-ray photoelectron spectroscopy (XPS) analysis was done on VG Multilab 2000. Field emission scanning electron microscopy (FESEM) images were collected using a JEOL JSM-7100F at an acceleration voltage of 20 kV. Transmission electron microscopy (TEM) and high resolution TEM (HRTEM) images were recorded using a JEOL JEM-2100F STEM/EDS microscope. The sample of TEM and HRTEM was peeled off from MS-CG by ultrasonication. Energy dispersive spectroscopy (EDS) were recorded by using Oxford EDS IE250.

Electrochemical Measurements. The electrochemical properties were characterized by means of 2016 coin cells using lithium metal foil as the anode, and as-prepared materials without any further treatment as cathode. The electrolyte was 1 M LiPF₆/EC (ethylene carbonate) + DMC (dimethyl carbonate) (1:1 vol/vol). The cells were assembled in an argon filled glove-box. Galvanostatic charge–discharge measurements were performed using a multichannel battery testing system (LAND CT2001A). Cyclic voltammetry (CV) and electrochemical impedance spectra (EIS) were performed using an Autolab PGSTAT 302N. All the measurements were carried out at room temperature.



Figure S1. (a) XRD pattern and (b) FESEM image of the MS precursor without heat treatment under H_2/Ar (5%/95%).



Figure S2. Stick and ball lattice structure of the hexagonal 2H phase MoS_2 . Blue and yellow spheres represent molybdenum (Mo) and surplus (S) atoms, respectively. In detail, the S atoms in the lattice adopt a hexagonal close-packing structure. The Mo atoms are sandwiched between two atomic layers of S, and each Mo is coordinated with six S atoms.



Figure S3. The high resolution C 1s spectrum of MS-CG.



Figure S4. CV curves of the first three cycles of the MS-CG (a) and the MS-PG (b).



Figure S5. FESEM images of MS-CG materials after 250 cycles.



Figure S6. (a, b) FESEM images of bare carbon fiber cloth. (c) Cycling performance and (d) charge/discharge profiles of bare carbon fiber cloth at a current density of 100 mA/g.