Supporting information

Dual Carbon Engineering Enabling 1T/2H MoS₂ with Ultrastable

Potassium Ion Storage Performance

Rong Hu,^a Yanqi Tong,^a Jinling Yin,^a Junxiong Wu,^{*b} Jing Zhao,^a Dianxue Cao,^a Guiling Wang,^{*a} and Kai Zhu^{*a}

^a Key Laboratory of Superlight Materials and Surface Technology, Ministry of Education, College of Materials Science and Chemical Engineering, Harbin Engineering University, Harbin 150001, China

^b Engineering Research Center of Polymer Green Recycling of Ministry of Education, Fujian Key Laboratory of Pollution Control & Resource Reuse, College ofEnvironmental and Resource Sciences and College of Carbon Neutral Modern Industry, Fujian Normal University, Fuzhou, 350117, Fujian, China

1. Experimental

1.1 Material synthesis

A modified Hummers' method was employed to synthesize graphene oxide (GO).¹ GO was dispersed in 30 mL of N, N-dimethylformamide (DMF) at a concentration of 3 mg mL⁻¹, and subsequent ultrasonication for 3 hours yielded a homogeneous GO solution. Following this, the GO solution was combined with 0.15 g of Na₂MoO₄·2H₂O, 2 g of L-cysteine, and 4 mL of deionized water. This mixture was magnetically stirred for 1 hour. Afterward, 10 mL of NH₃·H₂O was added, and the solution was stirred for an additional 30 minutes. Subsequently, the resulting solution was subjected to heating within a 50 mL Teflon-lined stainless steel autoclave for 16 hours at 180 °C. Postreaction, the autoclave's precipitate was centrifuged, then washed thrice with ethanol and deionized water, and finally dried at 80 °C for 12 hours. The resultant product is the final black powder of 1T/2H MoS₂@rGO. For comparative analysis, a pure 1T/2H MoS₂ sample was prepared under identical conditions but without the inclusion of GO.

In the synthesis of 1T/2H MoS₂@rGO@C, a systematic procedure was followed. Initially, a 30-minute stirring period involved the mixing of 50 mL of deionized water with Tris(hydroxymethyl)aminomethane (Tris) at a concentration of 10 mM and pH 8.5. Subsequently, 50 mg each of dopamine and 1T/2H MoS₂@rGO were introduced. After 24 hours of stirring, the resulting black suspension underwent filtration and was subsequently dried for 12 hours at 80 °C. A critical annealing step followed, conducted at 600 °C for 3 hours under an argon flow, targeting the resultant black powder. This procedure culminated in the successful formation of a multiphase 1T/2H MoS₂@rGO@C composite.

1.2 Materials characterization

The microstructure and morphology analysis of the specimens employed scanning electron microscopy (SEM, JEOL, JSM7500F) and transmission electron microscopy (TEM, JEOL, JEM-2100). For compositional and phase insights, Raman spectroscopy (RENISHAW, REF 2000, 514.5 nm laser) and powder X-ray diffraction (XRD, Rigaku TTR III) with Cu K α radiation ($\lambda = 1.5418$ Å) were utilized. The valence states investigation was conducted using X-ray photoelectron spectroscopy (XPS) with a Thermo ESCALAB 250 instrument. Furthermore, thermogravimetric analysis (TGA, Netzsch STA 449 F3) in an air atmosphere from 25 to 700 °C was employed to determine the carbon content of the samples.

1.3 Electrochemical measurements

To prepare the 1T/2H MoS₂@rGO@C electrode, a homogeneous slurry was achieved by blending the active material (1T/2H MoS₂@rGO@C), binder (carboxymethyl cellulose (CMC)), and conductive agent (Super P) in deionized water at a weight ratio of 7:2:1. Following this, the copper foil was uniformly coated with the mixed slurry, after which the coated copper foil was placed into a vacuum oven and dried at 60 °C for 10 hours. Subsequently, circular sections with a diameter of 12 mm were excised from these desiccated electrodes, resulting in an active material mass

loading of 1 ± 0.2 mg cm⁻².

In the assembly of the half-cell, the working electrode featured the as-prepared $1T/2H MoS_2@rGO@C$ electrode. The counter electrode employed potassium metal. Separation was facilitated by a glass fiber membrane (Whatman), while the electrolyte consisted of a mixed solution of 0.8 M KPF₆ dissolved in ethylene carbonate (EC) and diethyl carbonate (DEC) at a 1:1 (v/v) ratio. To evaluate the electrochemical performance of the $1T/2H MoS_2@rGO@C$, CR2032-type coin cells were meticulously constructed within an argon-filled glove box.

In the assembly of the full cell, the anode encompassed the prepared 1T/2H MoS₂@rGO@C electrode, while the cathode consisted of perylene tetracarboxylic dianhydride (PTCDA). The full cells employed the same electrolyte and separator as the half cells. To prepare the PTCDA cathode, initial calcination of PTCDA occurred at 450 °C under an Ar atmosphere for 5 hours at a heating rate of 10 °C min^{-1.41} A uniform slurry, achieved by blending calcined PTCDA, Super P, and CMC in deionized water at a weight ratio of 7:2:1, was uniformly coated onto an aluminum foil surface. Subsequently, vacuum-drying took place at 60 °C for 10 hours. Circular sections, 12 mm in diameter, were then excised from the prepared PTCDA electrodes, rendering them suitable for battery assembly.For the assembly of the full cell, a pre-potassization step was necessary for the 1T/2H MoS₂@rGO@C anode. Specifically, the anode electrode within the half-cell was subjected to charging and discharging for 5 cycles at 100 mA g⁻¹. The N/P ratio of the full cell is 1.24.

The Bio-Logic VMP3 workstation was employed for both cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS). Galvanostatic chargedischarge tests were conducted using a NEWARE battery tester. For the anode of 1T/2H MoS₂@rGO@C, the voltage range was maintained at 0.2-2.8 V, while the full cell operated within a voltage range of 1.0-4.2 V. Determination of the specific capacity of the full cell was based on the mass of the 1T/2H MoS₂@rGO@C anode.



Fig. S1 SEM images of (a) $MoS_2@rGO$ and (c) MoS_2 . (b) and (d) high magnification SEM image from marked area in (a) and (c), respectively.



Fig. S2 TEM image of $1T/2H MoS_2@rGO@C$.



Fig. S3 XPS analysis of GO and 1T/2H MoS₂@rGO@C: (a) XPS survey scan spectra; O 1s XPS spectra of (b) GO and (c) 1T/2H MoS₂@rGO@C. (d) Comparison of the content of oxygen-containing functional groups in the two materials.



Fig. S4 EDS mapping images of O element for 1T/2H MoS₂@rGO@C, 1T/2H MoS₂@rGO and GO.



Fig. S5 TGA profiles of 1T/2H MoS₂@rGO@C, 1T/2H MoS₂@rGO and 1T/2H MoS₂.



Fig. S6 XRD pattern of the final residue after TGA test.



Fig. S7 XPS spectra from S 2p of (a) 1T/2H MoS₂@rGO@C, (b) 1T/2H MoS₂@rGO and (c) 1T/2H MoS₂. (d) The proportion of 2H-MoS₂ and 1T-MoS₂ containing component in the three samples, respectively.



Fig. S8 XPS spectra from C 1s of (a) 1T/2H MoS₂@rGO@C and (b) 1T/2H MoS₂@rGO.



Fig. S9 Initial three CV cycles of (a) 1T/2H MoS₂@rGO@C, (b) 1T/2H MoS₂@rGO and (c) 1T/2H MoS₂ at 0.1 mV s⁻¹ 1 between 0.2 and 2.8 V. Charge-discharge profiles of (d) 1T/2H MoS₂@rGO@C, (e) 1T/2H MoS₂@rGO and (f) 1T/2H MoS₂ at a current density 100 mA g⁻¹.



Fig. S10 SEM images of (a, b) 1T/2H MoS₂@rGO@C, (c, d) 1T/2H MoS₂@rGO and (e, f) 1T/2H MoS₂ electrodes after 1000 cycles.



Fig. S11 Long-term cycling stability of 1T/2H MoS₂@rGO@C at 2000 mA g⁻¹.

References

1 H. Yu, K. Xie, J. Hu, C. Shen, J. Wanga, B. Wei, *RSC Adv.*, 2016, **6**, 17023-17028.