Supporting Information for

In Situ Synthesis of MoS₂/Graphene Nanosheet Composites with Extraordinary High Electrochemical Performance for Lithium Ion Batteries

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Experimental Details

Synthesis of graphene oxide (GO): Natural graphite powder (Shanghai Colloid Chemical Plant, China) was oxidized to graphite oxide using a modified Hummers method (ref. 12 in main text). Graphite powder (0.032 g) was poured into 50 ml of concentrated H_2SO_4 under an ice bath. Then, 3 g KMnO₄ was gradually added. The mixture was stirred for 2 h and then diluted with deionized (DI) water. After that, 10 ml of 30% H_2O_2 was added to the solution until the color of the mixture changed to brilliant yellow. The as-obtained graphite oxide was re-dispersed in DI water and then exfoliated to generate graphene oxide sheets by ultrasonication. A brown homogeneous supernatant was obtained by centrifuged washing.

*Synthesis of MoS*₂/*GNS*: GO was transferred from the as-made suspension into a 200 ml beaker with the addition of 40 ml DI water. Then, 0.3 g Na₂MoO₄·2H₂O was added. After ultrasonication and stirring for 20 min, 0.1 M NaOH was added to the

solution until the pH value changed to 6.5. The mixture and 0.8 g NH_2CSNH_2 were dissolved in 80 ml DI water and then transferred into a 100 ml Teflon-lined stainless steel autoclave, sealed tightly, and heated at 240 °C for 24 h. After cooling naturally, the black precipitates were collected by centrifugation, washed with DI water and ethanol, and dried in a vacuum oven at 80 °C for 24 h.

Characterizations: The samples were characterized with X-ray diffraction (XRD; Thermo X'TRA X-ray diffractometer with Cu Kα-source), high-resolution transmission electron microscopy (HRTEM; JEOL JEM-2010, 200 kV), and field emission scanning electron microscopy (FESEM; SIRION-100), energy dispersive X-ray spectroscopy (EDX, GENENIS-4000).

Electrochemical measurements: The electrochemical tests were measured using two-electrode cells assembled in an argon-filled glove box. Li sheets served as the counter electrode and reference electrode, and a polypropylene film (Celgard-2300) was used as a separator. The electrolyte was a 1.0 M LiPF₆ solution in a mixture of EC/DMC (1:1 in volume). The working electrodes were prepared by a slurry coating procedure. The slurry consisted of 80 wt. % active material, 10 wt. % acetylene black and 10 wt. % polyvinylidene fluoride dissolved in *N*-methyl-2-pyrrolidinone. This slurry was spread on copper foil, which acted as a current collector. The coated electrodes were dried at 110 °C for 12 h in vacuum and then pressed. Galvanostatic charge/discharge cycles were carried out on a CBT-138-320 battery tester between 0.01-3.00 V at various current densities. Cyclic voltammetry measurements were

carried out on an electrochemical workstation (Zahner IM6ex) over the potential range of 0.01-3.00 V vs. Li/Li⁺ at a scan rate of 0.5 mV/s.

Supplementary Figures



Figure S1. a) SEM image of free MoS₂ obtained through the chosen method employing Na₂MoO₄·2H₂O and NH₂CSNH₂ as starting materials; b) HRTEM image of free MoS₂ indicating the layered structure with d(002)=0.62 nm; c) TEM image of graphene nanosheets obtained through the chosen method without MoS₂, and d) the corresponding XRD pattern indicating that graphene sheets can be easily restacked in the hydrothermal process, and the interlayer distance of restacked GNS is slightly larger than that of graphite (JCPDS 41-1487).



Figure S2. a) The first two charge and discharge curves of GNS obtained by a hydrothermal method employing GO and NH_2CSNH_2 as starting materials, and b) the cycling behavior at a current density of 100 mA/g.



Figure S3. The electrochemical performance of MoS_2/GNS with different molar ratios: the first two charge and discharge curves of a) MoS_2/GNS with an MoS_2 to GNS mole ratio of 1: 1 [MoS_2/GNS (1: 1)]; b) MoS_2/GNS with an MoS_2 to GNS mole ratio of 1: 4 [MoS_2/GNS (1: 4)]; c) their cycling behavior at a current of 100 mAh/g; and d) their rate capability at various current densities.