Supporting Information for

Synthesis of rGO-supported layered MoS₂ for

high-performance rechargeable Mg batteries

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Experimental details

Synthesis of Graphene Oxide (GO). Natural graphite powder (Sigma) was oxidized to graphene oxide using a modified Hummer's method.¹ 2.0 g graphite powder and 1.0 g NaNO₃ were poured into 46 ml concentrated H₂SO₄ (98 %) under an ice bath, after stirring for 10 min, 6 g KMnO₄ was gradually added. The mixture was stirred at 35 °C for 1 h and then diluted with 92 ml deionized (DI) water. After stirring at 98 °C for another 30 min, 20 ml H₂O₂ and 280 ml DI water were added to the solution until the color of the mixture changed to brilliant yellow. After washing the as-made colloid with DI water several times until pH value changed to 4, the as-obtained graphene oxide (GO) was collected by centrifuging and freeze drying.

Synthesis of MoS₂/graphene (MoS₂/rGO) hybrids. In a typical synthesis, 0.30 g $Na_2MoO_4 \cdot 2H_2O$ and different amounts of GO (14, 28, 56 mg) were dissolved in 40 ml DI water with the help of vigorous stirring and ultrasonication. Then 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 100 ml Teflon-lined stainless steel autoclave, sealed tightly, and maintained at 210 °C for 24 h. After cooled to room temperature naturally, the resulting black precipitates were collected by centrifugation with DI water and ethanol for several times, and then dried in a vacuum over at 60 °C for 12 h followed by further thermal treatment at 800 °C in an Ar environment for 2 h. The products were named as S2, S3 and S4 for the sake of simplicity. In order to investigate the effects of GO on crystal structures and

morphologies of the samples, the pristine MoS_2 was prepared by the same procedure without the addition of GO and denoted as S1.

Synthesis of Electrolyte. The electrolyte solution (0.4 M Mg₂Cl₃⁺·AlPh₂Cl₂⁻/THF) was synthesized in a glove box filled with high purity argon gas. First, AlCl₃ (J&K, anhydrous, 99.999%) was added slowly to the vigorously stirred THF (high purity after redistilled) solvent to obtain the desired concentration AlCl₃ solution. Then, this added dropwise to predetermined solution was a quantity of 2 Μ Phenylmagnesiumchloride (PhMgCl) solution in THF (J&K). Both reactions are very exothermic. The resulting solution was stirred for additional 16 h or more (at room temperature).

Materials Characterizations. X-Ray Diffraction (XRD) analysis was performed on a Rigaku D/Max-2500 with Cu-K α radiation (λ = 1.54178 Å). The 2 θ angular regions between 3° and 80° were investigated at a scan rate of 4° min⁻¹. Fourier transform infrared (FT-IR) spectra of the samples were collected at room temperature by using a FTIR-650 spectrometer (Tianjin Gangdong) at a resolution of 4 cm⁻¹. The morphology and microstructure were observed by scanning electron microscopy (SEM, HITACHI S-4800) and transmission electron microscopy (Tecnai G2 F20 TEM). The element content of the samples was analyzed by GENENIS-4000 energy dispersive X-Ray spectroscopy (EDX). X-ray photoelectron spectrometer (XPS, PHI 5000 Versaprobe, ULVAC PHI) and Raman spectrum (Renishaw inVia, excitation 514.5 nm) were also used to characterize the synthesized materials.

Electrochemical Measurements. Electrochemical tests were carried out using a two-electrode cell assembled in an argon-filled glove box with water and oxygen contents below 5 ppm. The working electrodes were fabricated by mixing active materials, acetylene black and polyvinylidene fluoride (PVDF) binder with a weight ratio of 80:15:5 in N-methyl-pyrrolidinone (NMP) solvent to form homogeneous slurry, and then the mixtures were coated on a Cu foil. The coated electrodes were dried at 60 °C for 12 h in vacuum oven and then pressed. The active materials in per working cathode were about 1.2 mg without wiping off the mass of graphene. The counter and reference electrode was AZ31 Mg alloy which was cut into wafers

(diameter 14 mm, thickness 0.2 mm) and polished with metallographic sandpaper to remove the oxides and impurities on the surface. The separator was Celgard 2320 microporous film. Galvanostatic charge/discharge tests were performed on a LAND battery-test instrument (CT2001A) at different current densities, and the voltage was kept below 2.3 V (*vs.* Mg/Mg²⁺) to avoid corrosion of the copper current collector. There are 2 mins rest between both charge and discharge voltages. Cyclic voltammograms (CV) were conducted by a CHI660B electrochemical workstation at a scan rate of 1 mV s⁻¹. Electrochemical impedance spectras (CHI660B) were obtained by applying a sine wave with amplitude of 5.0 mV over the frequency range from 100 KHz to 500 mHz. All the tests were performed at room temperature.

Supplementary Figures



Fig. S1 FT-IR spectra of as-prepared GO and MoS₂/rGO hybrids (S2-S4).



Fig. S2 EDX spectra of of MoS₂ (S1) and MoS₂/rGO hybrids (S2-S4).



Fig.3 XPS full spectrum of S3 after discharged (Mg-intercalation). It is clear that Mg has intercalated into MoS₂ after discharged.



Fig. S4 XPS spectra of S3: (a,b) Mo 3d spectra before and after discharged (Mg-intercalation);(c,d) S 2p spectra before and after discharged. Obviously, the valence states of Mo and S have changed and the redox reactions occur with the introduction of Mg.

References

1. W. S. Hummers, R. E. Offeman, J. Am. Chem. Soc., 1958, 80, 1339.