Supporting Information for

Sandwich structured graphene-like MoS₂/C microspheres for rechargeable Mg batteries

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Experimental details

Materials Synthesis

In a typical synthesis, 0.30 g Na₂MoO₄·2H₂O and 0.40 g NH₂CSNH₂ were dissolved in 60 mL deionized water under vigorous stirring, and then different amounts of glucose (0.50, 1.0, 2.0 g) were added into the above solution with ultrasonication for 30 min, the obtained clear solution was sealed in a 100 mL Teflon-lined stainless steel autoclave, and maintained at 210 °C for 24 h. After cooled to room temperature naturally, the resulting black precipitates were collected by centrifugation with deionized 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. In order to investigate the effects of glucose on crystal structures and morphologies of the samples, the MoS₂ was prepared by the same procedure without the addition of glucose and denoted as S1.

The electrolyte solution (0.4 M $Mg_2Cl_3^+ AlPh_2Cl_2^-/THF$) 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 solution was added dropwise to a predetermined quantity of 2 M 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. The 2θ angular regions between 3° and 80° were investigated at a scan rate of 4° min⁻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 (EDS). The pore size distributions were measured using the Brunauer-Emmett-Teller (BET) method (NOVA 2200e).

Electrochemical Measurements

Electrochemical performances 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 80 °C for 12 h in vacuum oven and then pressed. The active materials of working cathode were about 1.5 mg/cm² without wiping off the mass of carbon. The counter and reference electrode was AZ31 alloy which was cut into wafers (diameter 14mm, thickness 0.2mm), 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 a current density of 50mA/g, and the voltage was kept below 2.4 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 1mV 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. It is necessary to point out that the electrochemical tests were carried out using active materials with the same weight.

Supplementary Figures



Fig. S1 Cyclic voltammograms of S1 and S3 electrodes at a scan rate of 1mV/s in a typical steady state (after 5-6 preliminary cycles).