Electronic Supplementary Information

Encapsulation of Selenium in Porous Hollow Carbon Spheres for Advanced Lithium-Selenium Batteries

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Experimental

1.1 Preparation of porous hollow carbon spheres (PHCSs) and Se/PHCS composite

The PHCSs were synthesized by a simple temple method, in which SiO2@RF spheres were synthesized using a one-step method under Stőber reaction conditions.¹ In a typical synthesis process, 1.6 mL of ammonia aqueous solution (28 wt%) was added to a mixture of 5.5 mL deionized water and 37.8 mL ethanol, then stirred for 30 minute at room temperature. Subsequently, tetraethoxysilane (TEOS) (1.5 mL), resorcinol (0.21 g) and formaldehyde solution (0.31 mL, 37 wt%) were added to the above solution at intervals of 10 minutes. After that, the mixture was vigorously stirred for 24 h at 30 °C and transfer to a 60 mL Teflon-lined autoclave under static conditions (100 °C) for another 24h. Solid SiO₂@RF products were filtered and washed with absolute ethanol and distilled water three times, after oven-drying, the solid samples were carbonized by heating at 750 °C for 2 h under argon gas flow in a tube furnace. Finally, the SiO₂ template was removed by treating the material in dilute HF solution overnight, then the final PHCSs were obtained after drying up.

The Se/PHCS composite was prepared via a facile melt-diffusion process, namely, commercial Se particle (AR, Aladdin, China) and PHCSs were mixed in a weight ratio of 7:3, the mixture were heated at 260 °C (5 K min⁻¹) for 10 h with the protection of flowing Ar gas in quartz tubes.

1.2 Material characterization

Field emission scanning electron microscopy (SEM, Nova Nano SEM 230), transmission electron microscopy (TEM, Tecnai G2 20ST) and X-ray diffraction (XRD, Rigaku3014) were applied to characterize the materials' morphology and structure respectively. Thermogravimetric analysis (TGA, SDTQ600) was conducted to determine the Se content in the composites. Nitrogen

adsorption/desorption measurements were performed by using Quantachrome instrument (Quabrasorb SI-3MP). The Raman spectra were obtained on a Jobin-Yvon LabRAM HR-800 spectrometer with excitation from an argon ion laser (514 nm).

1.3 Cell assembly and Electrochemical characterizations

In order to evaluate the electrochemical performance of the Se/PHCS composites, a CR-2025 type coin cell was fabricated, with a lithium metal sheet as counter electrode and Celgard 2400 film as separator. A slurry containing 80 wt% Se/PHCS (or mixture of pristine Se and acetylene carbon with a weight ratio of 6:4), 10 wt% acetylene black and 10 wt% sodium alginate (SA) in deionized water was spread onto aluminium foil. After dried at 60 °C overnight, the cathode material was ready. The tested cells were assembled in an argon-filled glove box (Universal 2440/750). The electrolyte used was 2 M bis(tri-fluoromethane) sulfonamide lithium salt (LiTFSI, Sigma Aldrich) in a solvent mixture of 1,3-dioxolane and 1,2-dimethoxyethane (1:1,v/v) (Acros Organics), including 0.1 M lithium nitrate as an additive. Cyclic voltammetry (CV) measurements were performed at a scan rate of 0.2 mV s⁻¹ in the voltage range of 1.0 V to 3.0 V using a PARSTAT 2273 electrochemical measurement system. A LAND CT2001A battery-testing instrument was applied to conduct galvanostatic charge/discharge tests. The cells were first discharge to 1.0 V and then the cycle number was counted. All the electrochemical tests were conducted at room temperature.

References:

1 A. B. Fuertes, P. Valle-Vigón and M. Sevilla, Chem. Commun, 2012, 48, 6124.

Raman spectra



Fig. S1 Raman spectra of the obtained PHCSs