

Supporting Information

Experimental Section

Material Synthesis

Synthesis of SPNTs: The bamboo-like PNTs were prepared through a modified method according to a literature method.⁴⁶ 3 g of PNTs were dispersed into 90 g of concentrated sulfuric acid and followed by a sonication for 10 min. The mixture was then stirred at 40 °C for 24 h, and then the resulting pink precipitate was collected by centrifugation, washed with deionized water and ethanol for several times.

Synthesis of SiO₂@SPNTs: 0.5 g of SPNTs and 0.5 g of tetraethoxysilane (TEOS) were added to a mixture of 10 g of water and 40 g of ethanol under stirring at room temperature for 12 h, then the yellow products were collected by centrifuged and washed with ethanol.

Synthesis of SiO₂@ACNTs and ACNTs: SiO₂@SPNTs was treated in the atmosphere of N₂ at 800 °C for 2 h with a heating rate of 1 °C min⁻¹. The SiO₂ shell was subsequently removed by HF etching (2 wt%). The brown product was harvested by centrifugation. After been washing with ethanol and deionized water for several times, the as-obtained ACNTs was drying at 60 °C overnight.

Synthesis of MoS₂@ACNTs: 50 mg of as-prepared ACNTs were dispersed into glucose solution (35 mL, 0.05 M) under a ultrasonication for 5 seconds. Then, 0.6 g of thiourea (CN₂H₄S) and 0.3 g of sodium molybdate (Na₂MoO₄·2H₂O) were added in the above dispersion. After stirring for 5 min, the reaction solution was transferred to a 50 mL polytetrafluoroethylene (Teflon)-lined stainless-steel autoclave and kept in an electric oven at 200 °C for 24 h. The autoclave was then taken out from the oven and left to cool down to room temperature. The black precipitate was

then harvested by centrifugation, washed thoroughly with ethanol for several times, and dried at 60 °C. The as-prepared MoS₂@ACNTs sample was further treated in the atmosphere of 10% H₂ balanced by N₂ at 500 °C for 4 h with a heating rate of 1 °C min⁻¹ to obtain highly crystalline MoS₂ nanosheets. MoS₂ flakes were prepared through a similar route but without adding ACNTs.

Materials Characterization

Field-emission scanning electron microscopy (FESEM; HITACHI, su-8010) and transmission electron microscopy (TEM; JEOL, JEM-2100) were employed to investigate the morphology of products. The crystal structure of the sample was acquired by powder X-ray diffraction (XRD; SHIMADZU, Lab X XRD-6000). Specific surface area and pore size distribution of the sample were measured by a Brunauer–Emmett–Teller instrument (BET; ASAP 2020M) at 77 K. Thermogravimetric measurements (METTLER-TOLEDO TGA 1) were carried out with a flow of air at a heating rate of 10 °C min⁻¹ from room temperature to 700 °C.

Electrochemical measurements

Two-electrode coin-type cells (CR2016) were assembled for the electrochemical tests. The working electrode was prepared by spreading a mixture of MoS₂@ACNTs composites, conductive agent (carbon black, C-ENERGY™ Super C65) and polymer binder (poly(vinylidenedifluoride), PVDF, Aldrich) in a weight ratio of 70: 20: 10 on a copper foil. The loading mass of the active materials is around 0.5 mg cm⁻². The electrolyte used in the cells was 1 M NaClO₄ dissolved in ethylene carbonate/diethyl carbonate/fluoroethylene carbonate mixture (1:1:0.1 v/v/v). Both of the counter and reference electrode are served by sodium only. Every cell was assembled in argon-filled dry glovebox. The galvanostatic charge-discharge tests were performed on a NEWARE battery tester. Cyclic voltammetry (CV) was carried out using an electrochemical workstation (CHI 660D).

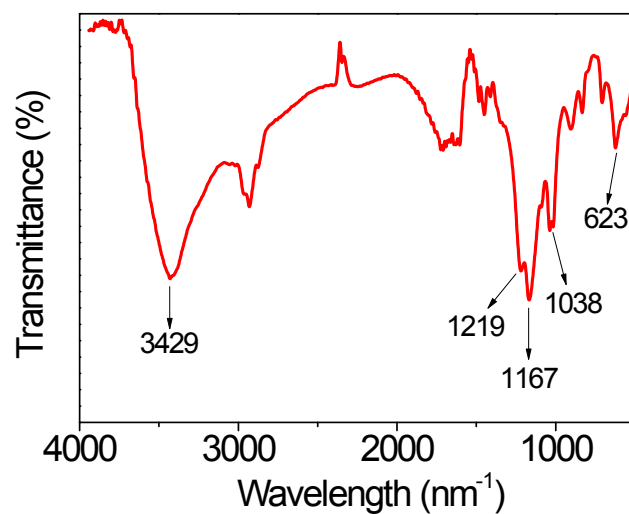


Fig. S1 FT-IR spectra of SPNTs

FT-IR spectra of SPNTs is shown in Fig. S1. As can be seen, a strong peak located at 3429 cm^{-1} can be assigned to vibration of hydroxyl group (-OH). The characteristic peaks at 1219, 1167 and 623 cm^{-1} can be assigned to the sulfonic acid group ($-\text{SO}_3\text{H}$). The band at 1038 cm^{-1} is related to the sulfone group ($-\text{SO}_2-$).

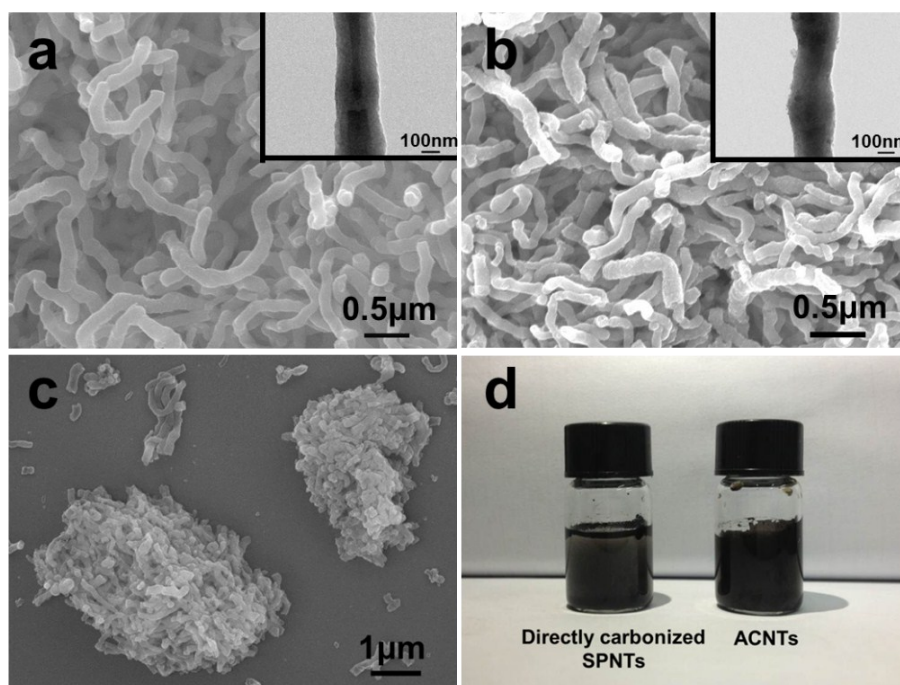


Fig. S2 SEM and TEM (inset) images of SiO₂@SPNTs (a) and SiO₂@ACNTs (b). (c) SEM image of the directly carbonized SPNTs. (d) Digital photo shows the directly carbonized SPNTs and the as-prepared ACNTs dispersed in water under sonication for 5 minutes and 10 seconds, respectively.

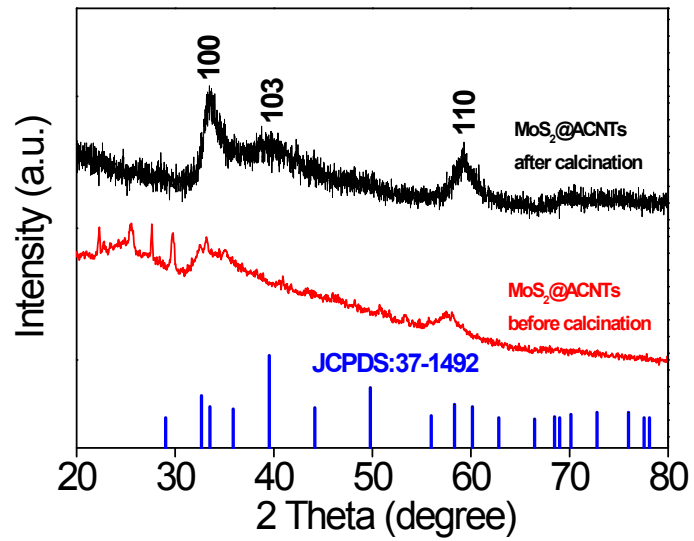


Fig. S3 XRD patterns of MoS₂@ACNTs before and after a heat treatment at 500 °C for 4 h.

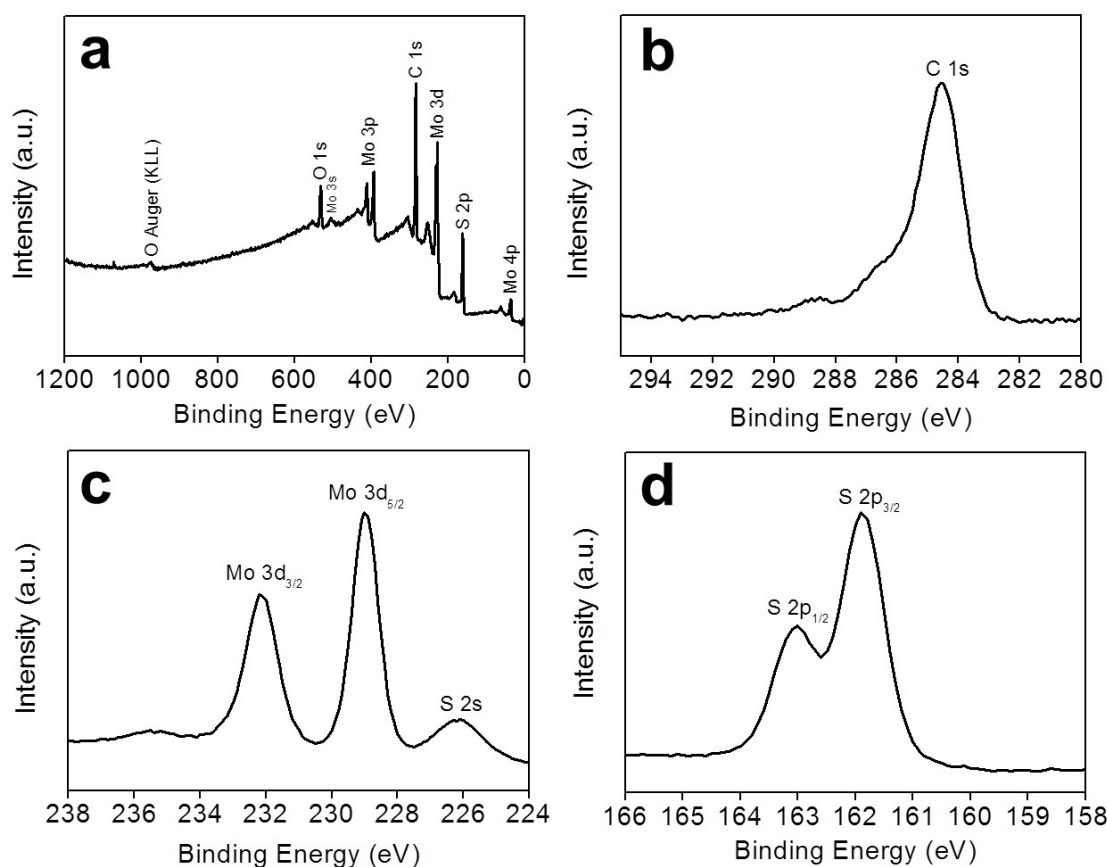


Fig. S4 XPS spectra of MoS₂@ACNTs: (A) Full spectrum; (B) C 1s; (C) Mo 3d; (D) S 2p.

To gain further information about the structure and composition of MoS₂@ACNTs, we resort to X-ray photoelectron spectroscopy (XPS) measurements with the result shown in Fig. S4. Specifically, the peak at 284.6 eV arises from the graphite-like carbon atoms of the ACNTs (Fig. S4b). [*Adv. Mater.*, 2006, **18**, 181.] In Fig. S4c, the peaks at 229.0 and 232.2 eV are characteristic of Mo⁴⁺ in MoS₂, which are very close to the reported literature. [*J. Phys. Chem.*, 1993, **97**, 11020.] In addition, a small S 2s peak is located at a slightly lower binding energy (226.1 eV) than the Mo 3d_{5/2} peak. In Fig. S4d, there are two strong peaks at 161.9 eV and 163.0 eV, which can be attributed to S 2p_{3/2} and S 2p_{1/2} binding energies, respectively, which are also in good agreement with the values reported in the literature. [*J. Phys. Chem.*, 1993, **97**, 11020.]

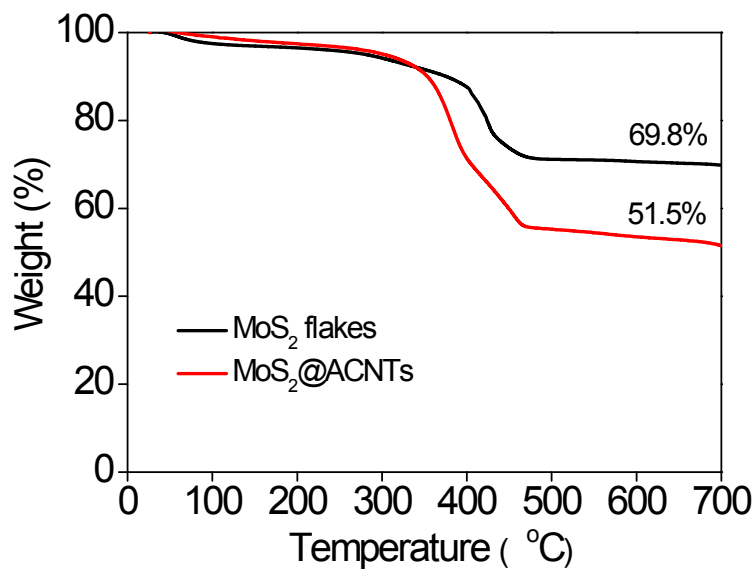


Fig. S5 TGA curves of MoS₂@ACNTs and MoS₂ flakes at a temperature ramp of 10 °C min⁻¹ in air. Let the weight percentage of MoS₂ in the MoS₂@ACNTs to be x . Assuming ACNTs are completely removed after combustion, one has $0.698x = 0.515$. Therefore $x = 0.738$.

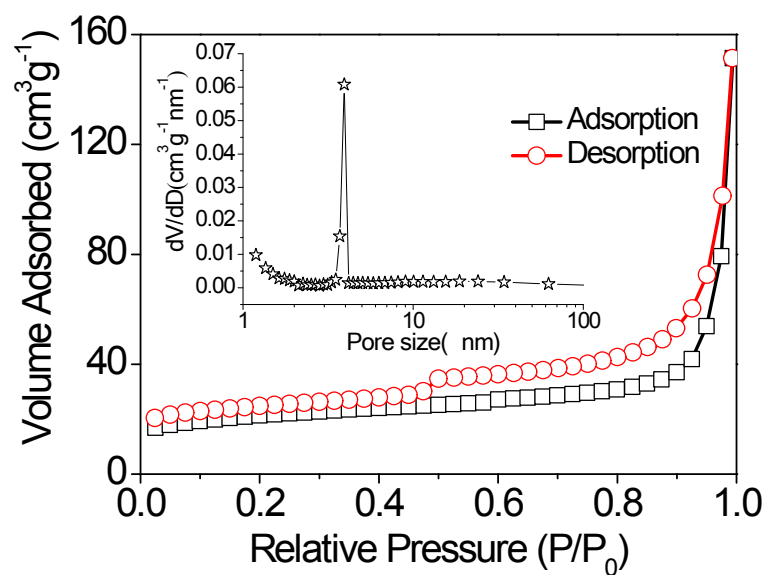


Fig. S6 N₂ adsorption–desorption isotherms of the MoS₂@ACNTs composite, and the inset shows the pore-size distribution calculated from desorption branch.

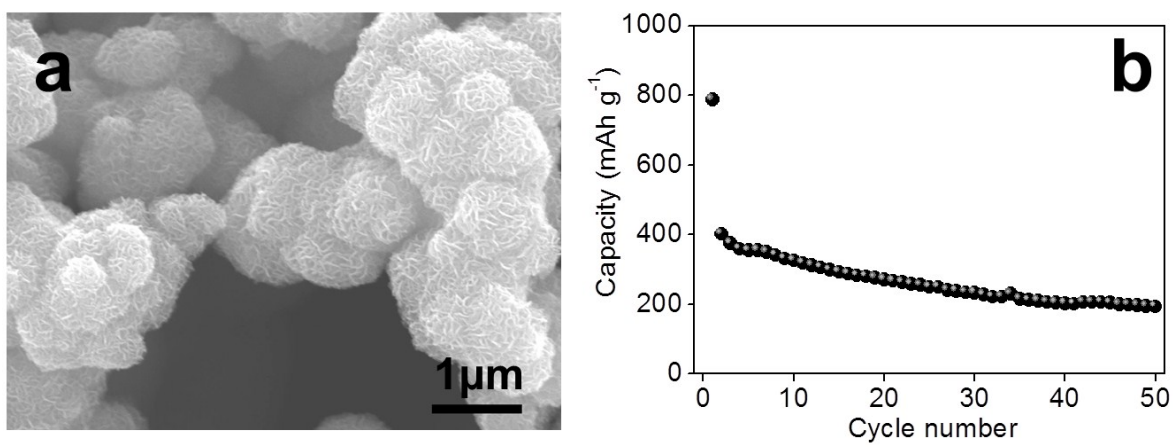


Fig. S7 The morphology of as-prepared MoS₂ flakes (a) and their cycling performance at a current rate of 500 mA g⁻¹ (b).

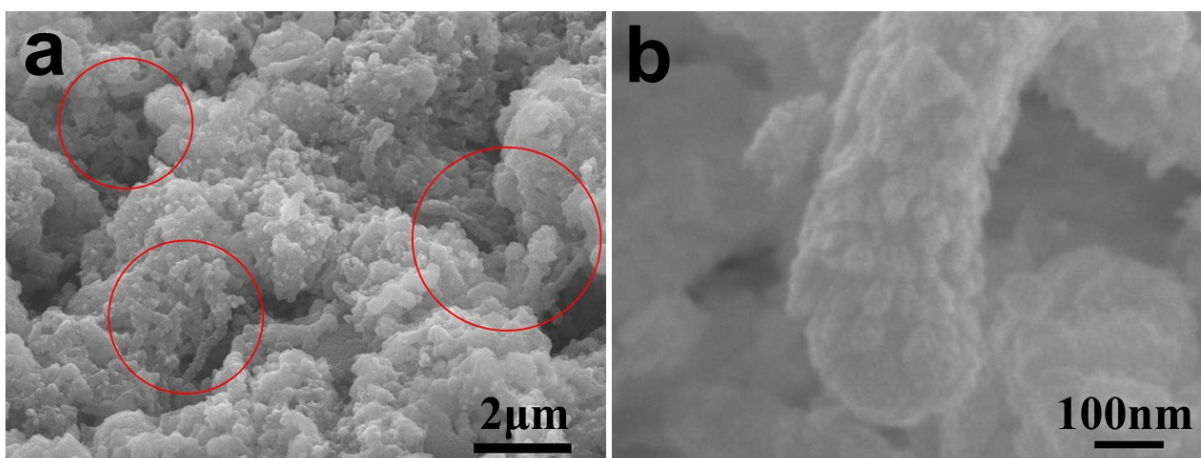


Fig. S8 SEM images of MoS₂@ACNTs after cycling for 160 cycles.

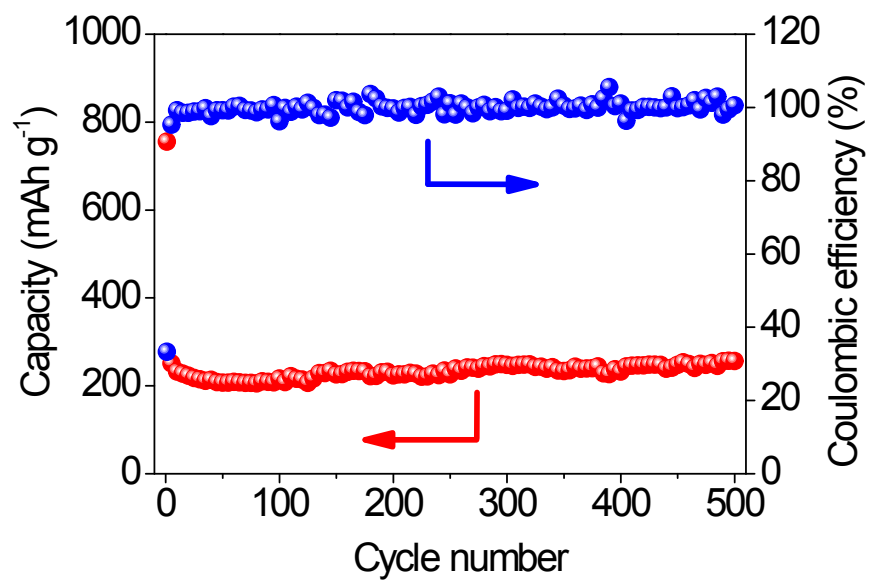


Fig. S9 Cycling performance and corresponding Coulombic efficiency of ACNTs at a current rate of 500 mA g⁻¹.

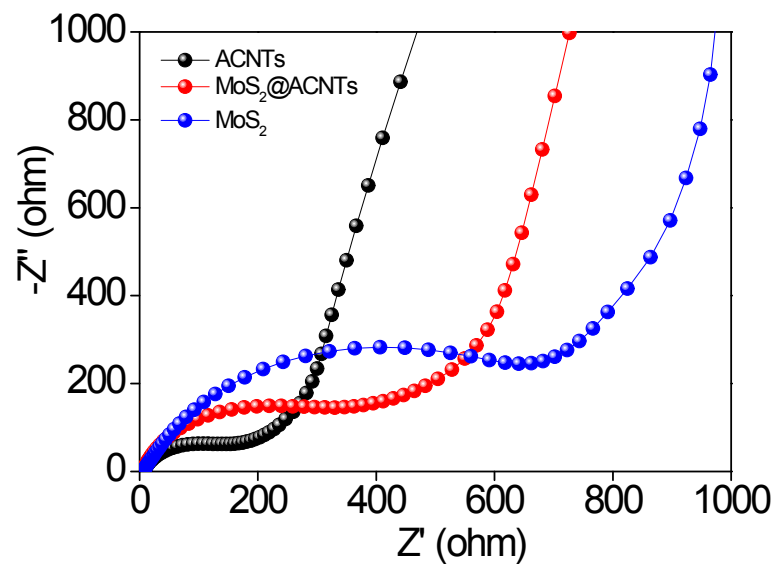


Fig. S10 Nyquist plots of MoS₂@ACNTs, pure MoS₂ flakes and ACNTs.

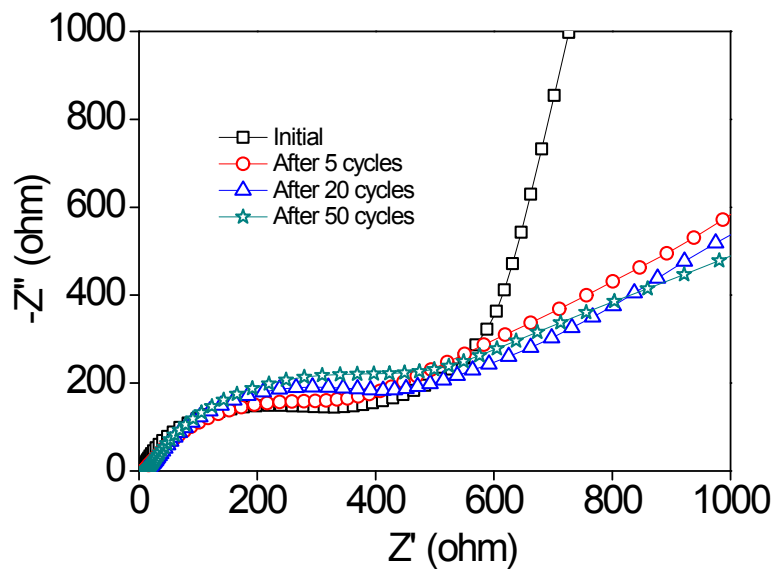


Fig. S11 Nyquist plots of MoS_2 @ACNTs electrode after different cycles.