Supplementary Electronic Information

Directionally Assembled MoS₂ with Significantly Expanded Interlayer

Spacing: A Superior Anode Material for High-Rate Lithium-Ion Battery

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Fig. S1. (a, b) TEM images of the product synthesized in the presence of MWCNTs that were not treated with oxidation. The clean surfaces of the MWCNTs indicate that the as-grown MoS_2 nanostructures cannot be grafted on the intrinsic MWCNTs due to the non-wettability between the pristine surface of CNTs and MoS_2 precursor.



Fig. S2. (top) TEM image and (bottom) XRD pattern of the sample synthesized in the absence of MWCNTs, highlighting the sphere-like assembles of MoS₂ nanosheets.



Fig. S3. Schematic illustrations of atomic structures of S-Mo-S layers: trigonal semiconducting 2H phase (left) and octahedral metallic 1T phase (right). Intercalation of lithium in MoS_2 can lead to a 2H-to-1T phase transistion. Blue and yellow balls correspond to Mo and S atoms, respectively.



Fig. S4. Initial CV graphs of pure IE-MoS₂ shown in Fig. S2. The sweep rate was 0.5 mV s⁻¹ and the measurement was performed in the voltage range of 0-3 V (versus Li/Li⁺).



Fig. S5. Initial CV graphs of pure MWCNTs at a sweep rate of 0.5 mV s⁻¹ over the voltage range of 0-3 V (versus Li/Li^+).



Fig. S6. Dicharge-charge capacities and corresponding coulombic efficiency of pure IE- MoS_2 shown in Fig. S2. The measurements over 200 cycles in the range of 0.0005-3 V are presented.



Fig. S7. (a) Rate capability of the pure MWCNTs with various current rates from C/5 to 15 C. (b) Dicharge-charge capacities and corresponding coulombic efficiency of pure MWCNTs measured at C/5 in the range of 0.0005-3 V. The long-term cycling (over 200 cycles) highlights the variation of capacities as funciton of cycle number.



Fig. S8. Replot of the 3rd discharge curve shown in Figure 3b in the main text. The red curve represents the first derivative of volatage against the specific capacity, highlighting how fast the voltage drops as discharge proceeds. The capacitance associated with the high surface area of nanostructured electrode materials may make a contribution (sometimes significant) to the overall electrochemical capacity of a battery. The capacitance-controlled capacity is usually convoluted with the capacity originated from the diffusion and the following intercalation and conversion reaction (or diffusion-controlled capacity). Although an established protocol to deconvolute the capacitance-controlled and diffusion-controlled capacity is absent, we can approximately estimate their individual contributions to the overall capacity. In general, the capacitance-controlled capacity exhibits a strong dependence on voltage while the diffusioncontrolled capacity has a weaker dependence on voltage. According to this difference, we can reasonably use the first derivative of the voltage-capacity function shown in the following figure (the 3rd cycle curve shown in Figure 3b in the manuscript) to separate their contributions. In the capacitance-controlled region, the voltage drops significantly (from 3.0 V to 1.85 V) as the discharge proceeds. The corresponding capacity is 168 mAh g^{-1} . In contrast, the variation of voltage becomes much more gradual when the discharge is in the diffusion-controlled region. The capacity of the diffusion-controlled discharge is 1487 mAh g^{-1} . Therefore, the ratio of capacitance-controlled capacity to the diffusion-controlled capacity is approximately 0.11, indicating that the electrochemical storage capacity of the IE-MoS₂/MWCNTs composite materials dominated by the diffusion-controlled capacity (i.e., ~90%).