Creating a synergistic interplay between tubular MoS₂ and particulate Fe₃O₄ for improved lithium storage

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S1. Experimental details

S1.1 Hydrothermal synthesis of tubular MoS₂

The process for the hydrothermal synthesis of tubular MoS₂ was adopted from a previous report.^[1] Briefly, sulphur powder (2.4 mmol) was dissolved in octylamine (28 mL) and absolute ethanol (24 mL), to which ammonium molybdate (0.15 mmol) was added. The mixture was stirred for 30 min, transferred to a Teflon-lined autoclave, sealed, and heated at 200 °C for 24 h. After being cooled naturally to ambient temperature, this mixture was sequentially washed by absolute ethanol and deionised water, and vacuum-dried at 60 °C overnight. The resulting black powder was collected, dissolved in *N*-methyl-2-pyrrolidinone (NMP), centrifuged, and oven-dried.

[1] P. Wang, H. Sun, Y. Ji, W. Li and X. Wang, Adv. Mater., 2014, 26, 964.

S1.2 Hydrothermal synthesis of particulate Fe_3O_4

The hydrothermal synthesis of particulate Fe_3O_4 was reported by us previously.^[2] In a typical experiment, iron(III) acetylacetonate (9 mmol) was dissolved in oleylamine (90 mL) and octadecene (90 mL) under nitrogen protection, heated at 280 °C for 1 h, and cooled naturally to ambient temperature. Absolute ethanol (50 mL) and ethyl acetate (100 mL) were added, and the resulting black powder was separated out by a magnet, washed by absolute ethanol, and vacuum-dried at 60 °C overnight.

[2] L. Pan, X.-D. Zhu, X.-M. Xie and Y.-T. Liu, Adv. Funct. Mater., 2015, DOI:10.1002/adfm.201404348.

S1.3 Hierarchical assembly of particulate Fe_3O_4 on tubular MoS_2

In a typical experiment, particulate Fe₃O₄ and tubular MoS₂ were separately dissolved in THF to obtain

0.1 mg mL⁻¹ solutions, which were mixed at different Fe_3O_4/MoS_2 weight ratios (1/1 and 1/2). The mixtures were subjected to 70 W sonication at ambient temperature for 12 h, during which particulate Fe_3O_4 was spontaneously attracted to the naked surfaces of tubular MoS_2 through van der Waals interactions. The resulting 3D MoS_2 @ Fe_3O_4 nanohybrids were separated out by a magnet, rinsed by THF, and vacuum-dried at 60 °C overnight.

S1.4 Electrochemical evaluation

The anodes for the electrochemical evaluation were prepared by coating copper foils with a slurry containing 70 wt% active material, 10 wt% acetylene black and 20 wt% poly(vinylidene fluoride) dissolved in NMP. The anodes were vacuum-dried at 120 °C for 12 h, and equipped in half cells according to the configuration of (–) Li | liquid electrolyte | anode (+) in a vacuum glove box. The liquid electrolyte was 1 M solution of LiPF₆ in ethylene carbonate/dimethyl carbonate at a volume ratio of 1/1. The separators were microporous polypropylene membranes.

S2. Supplementary figures



Fig. S1 – SEM images of as-synthesised tubular MoS_2 .



Fig. S2 – XRD pattern of tubular MoS_2 (JCPDS card No. 37–1492).



Fig. S3 – Nitrogen adsorption/desorption isotherms of tubular MoS_2 revealing a BET surface area of 102.83 m² g⁻¹, and pore size distribution (insert) of tubular MoS_2 revealing an average pore size of 5.66 nm.



 $\label{eq:Fig.S4} \textbf{Fig. S4} - \text{TEM} \text{ images of 3D } MoS_2 @Fe_3O_4 \text{ nanohybrid at an } Fe_3O_4 / MoS_2 \text{ wt ratio of } 1/1.$



Fig. S5 – XRD patterns of tubular MoS₂, particulate Fe_3O_4 and 3D MoS₂@Fe₃O₄ nanohybrid at an Fe_3O_4/MoS_2 wt ratio of 1/2.



Fig. S6 – CV curves of tubular MoS_2 .