

Binder-free layered $\text{Ti}_3\text{C}_2/\text{CNTs}$ nanocomposite anodes with enhanced capacity and long-cycle life for lithium-ion batteries

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1. Experimental Section

Synthesis of Ti_3C_2 nanosheets: Ti_3C_2 nanosheets were synthesized as described by Gogotsi's group previously.¹ Briefly, 2.0 g Ti_3AlC_2 powders (Beijing Jinhezhi Materials Co., Ltd. >98 wt.% purity) were dispersed in 20 mL 25% hydrofluoric acid (Sinopharm Chemical Reagent Co., Ltd.) under stirring for 10 hours. Then the suspensions were washed several times using ultrapure water until the pH of the liquid reached 5. The obtained sample was collected and dried in air. Next, 0.5 g obtained Ti_3C_2 powder was dispersed in 25 mL of dimethyl sulfoxide (DMSO) under stirring at room temperature for 20 h. Then the intercalated powder was collected by centrifugation from the colloidal suspension. The purified powder was redispersed in 250 mL ultrapure water under the ultrasonication for 1 hour and centrifuged with 3500 rpm. At last, the upper solution has a concentration of around 0.6 mg/mL was obtained.

Synthesis of layered $\text{Ti}_3\text{C}_2/\text{CNTs}$ nanocomposite: Oxidized water-soluble CNTs with a concentration of 0.2 mg/mL were prepared as presented previously.² A certain amount of Ti_3C_2 suspension and 15 mL CNTs dispersion was mixed together and sonicated for 1 h. Subsequently, $\text{Ti}_3\text{C}_2/\text{CNTs}$ lamellar films were prepared by filtering the mixed suspension on a polycarbonate (PC) membrane with 200 nm pores (Millipore). The route of preparation of $\text{Ti}_3\text{C}_2/\text{CNTs}$ paper is demonstrated in Scheme 1. Three samples with mass ratios of Ti_3C_2 to CNTs of 1:1 (6 mg), 2:1 (9 mg), 1:2 (4.5 mg), and pure Ti_3C_2 paper were synthesized for comparison.

Electrode preparation: Pure Ti_3C_2 and $\text{Ti}_3\text{C}_2/\text{CNTs}$ hybrid lamellar paper were directly used as electrodes, respectively, and incorporated into a coin-type cell with lithium foils as both counter and reference. The electrolyte employed in the cell was 1M LiPF₆ in a 50:50 (w/w) mixture of ethylene carbonate and diethyl carbonate. The galvanostatic (GV) charge-discharge performances of the cell were tested in the voltage range of 0.05-3.0 V under a constant current density of 0.5 C by EQ-BST8-WA battery-test system, cyclic voltammetry (CV) was recorded by a CHI 660D electrochemical workstation in the voltage range 0.05-3.0 V with a scan rate of 0.1 mV s⁻¹. Electrochemical impedance spectroscopy (EIS) results were obtained from a frequency range of 100 kHz to 1 Hz.

Characterization: The phase of the composite is obtained by X-ray diffraction (XRD) at room temperature using an X'Pert PRO (PANalytical, Netherlands) instrument with Cu $K\alpha$ radiation. The morphologies were characterized using scanning electronic microscopy (SEM) (Hitachi S-4800), transmission electron

microscopy (TEM, Philips CM200).

2. Cross-section SEM images of the $\text{Ti}_3\text{C}_2/\text{CNTs}$ paper with corresponding energy-dispersive X-ray spectroscopy (EDS) maps for C and Ti.

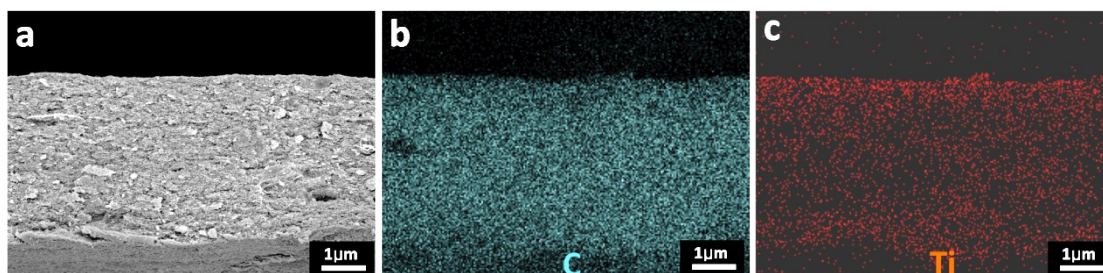


Fig. S1 (a) Cross-section SEM images of the $\text{Ti}_3\text{C}_2/\text{CNTs}$ paper with corresponding energy-dispersive X-ray spectroscopy (EDS) maps for C (b) and Ti (c).

3. Morphology of delaminated Ti_3C_2 nanosheets

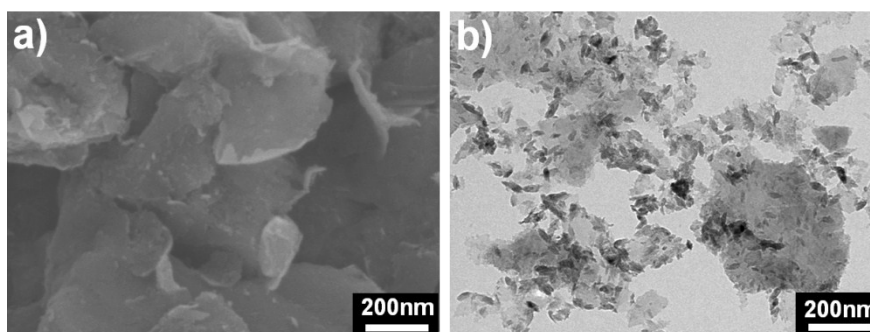


Fig. S2 SEM and TEM images of delaminated Ti_3C_2 nanosheets

4. Cycling performance of $\text{Ti}_3\text{C}_2/\text{CNTs}$ composites with different mass ratios of Ti_3C_2 to CNTs at 1 C

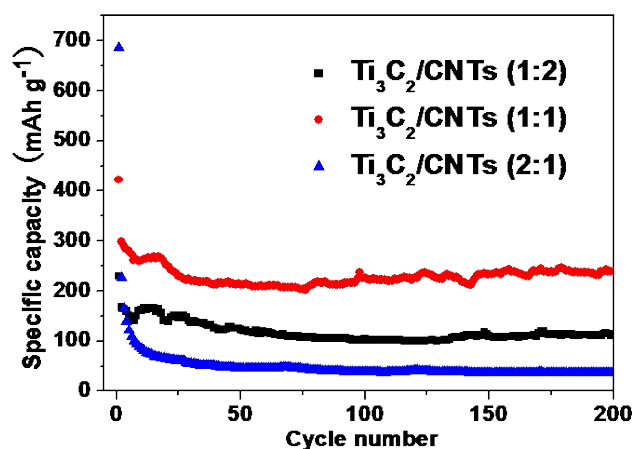


Fig. S3 Cycling performance of $\text{Ti}_3\text{C}_2/\text{CNTs}$ composites with different mass ratios of Ti_3C_2 to CNTs at 1 C.

5. Surface area of Ti_3C_2 , $\text{Ti}_3\text{C}_2/\text{CNTs}$ composites and CNTs.

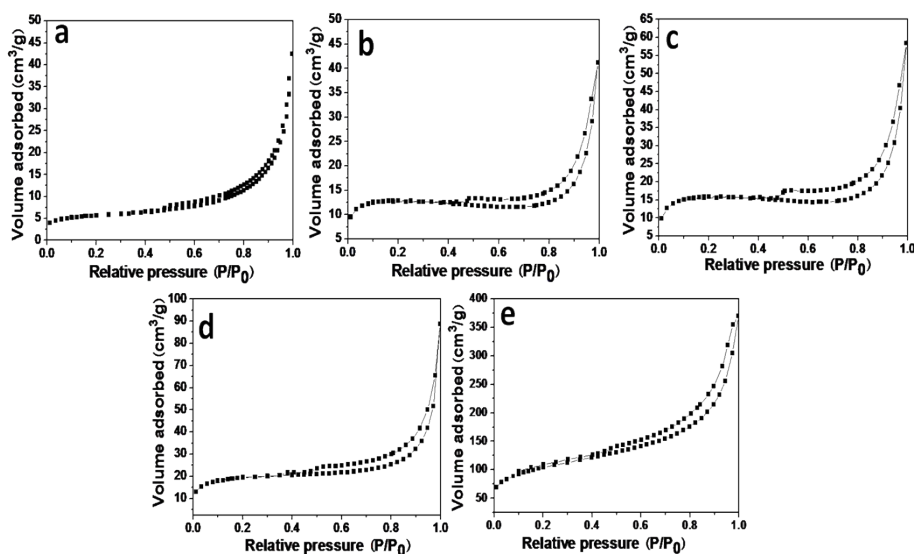


Fig. S4 (a) Nitrogen adsorption and desorption isotherms of Ti_3C_2 , $\text{Ti}_3\text{C}_2/\text{CNTs}$ (2:1), $\text{Ti}_3\text{C}_2/\text{CNTs}$ (1:1), $\text{Ti}_3\text{C}_2/\text{CNTs}$ (1:2) and CNTs.

The surface area of Ti_3C_2 , $\text{Ti}_3\text{C}_2/\text{CNTs}$ (2:1), $\text{Ti}_3\text{C}_2/\text{CNTs}$ (1:1), $\text{Ti}_3\text{C}_2/\text{CNTs}$ (1:2) and CNTs were 20, 46, 74, 97 and $366 \text{ m}^2 \text{ g}^{-1}$, respectively.

References

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