Supporting Information

Ultrathin and Large-Sized Vanadium Oxide Nanosheets Mildly Prepared at Room Temperature for High Performance Fiber-Based Supercapacitor

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Experimental Section

Chemicals:

Polyvinyl alcohol (PVA, MW 85 000-124 000) was obtained from Sigma-Aldrich. Vanadium oxide powder (V_2O_5, 99.2%), hydrogen peroxide (H_2O_2, 30 wt%), and lithium chloride (LiCl, >99%) were purchased from Alfa Aesar, Sinopharm Chemical Reagent Co., Ltd, and Adamas-Beta, respectively. Ethanol (≥99.7%) was purchased from Shanghai Ling Feng Chemical Reagent Co., Ltd. All chemicals were used as received. The purity of C_2H_4 and Ar is 99.95% and 99.99%, respectively.

Preparation of ultrathin and large sized V_2O_5 nanosheets:

The commercial V_2O_5 bulk was mixed with deionized water and H_2O_2 (30 wt%) with a volume ratio of 5:1 to get a transparent red solution associated with release of O_2 bubbles. The mixture was maintained at room temperature for 20 hours and a viscous gel consisted of highly concentrated V_2O_5 ultrathin nanosheets was formed. The V_2O_5 nanosheets were then dispersed in deionized (DI) water/ethanol mixed solution (volume ration of 1:1) with a concentration of 6 mg mL^{-1} and 0.6 mg mL^{-1} for the fabrication of fiber electrode. The freeze-dried V_2O_5 nanosheets were prepared for structural and morphological characterization.

Fabrication of MWCNT/V_2O_5/MWCNT fibers:

The spinnable MWCNT array was grown by chemical vapor deposition (CVD) in a quartz tube furnace at 750 ^oC for 10 min. A 1 nm Fe film deposited on a Si/SiO_2 substrate was used as the catalyst. Ar (155 sccm) and C_2H_4 (45 sccm) were used as the carrier gas and carbon source, respectively. The well-aligned MWCNT thin film was obtained by drawing the MWCNT array using a scotch tape. The areal density of well-aligned MWCNT film was determined to be 2.12 μg cm^{-2} *via* the utilization of an ultra-accurate microbalance (METTLER TOLEDO, M×5) with a readability of 1 μg. Two-layer of the obtained MWCNT thin films with width of ~1.5 mm and length of ~8.5 cm were first stacked onto a polytetrafluoroethylene (PTFE) substrate. The V_2O_5 suspension was then drop-casted onto the MWCNT thin film. After it was dried, another two-layer of MWCNT thin films were laid on top forming a sandwiched hybrid film. Subsequently, this hybrid film was peeled off from the PTFE substrate and twisted into a fiber with a motor rotating at 200 rpm for 2 min. The obtained hybrid fibers were thermally annealed at 350 ^oC for 3.5 h to obtain MWCNT/V_2O_5/MWCNT fiber. The content of V_2O_5 nanosheets can be controlled by simply ajusting the drop-casting process.

Fabrication of the fiber-based supercapacitors:

The solid-state fiber-based supercapacitors were fabricated and tested in two-electrode configuration similar to our previous study. Typically, two MWCNT/V_2O_5/MWCNT fibers were placed in parallel on a polyethylene terephthalate (PET) substrate with a gap of ~0.5 mm, and then coated with polyvinyl alcohol (PVA)-LiCl gel electrolyte. The PVA-LiCl gel electrolyte was prepared by mixing LiCl (6 M, 10 mL) with PVA powder (1 g, MW 85 000-124 000; Sigma Aldrich) followed by heating at 90 ^oC with vigorous stirring to obtain a homogeneous gel-like suspension. Ag paste served as the conductive pads for electrochemical measurements was applied at both ends of the fibers.

Characterization:

The morphology and structure of the samples were obtained using scanning electron microscope (SEM, JEOL 6500), transmission electron microscope (TEM, JEOL 2100), X-ray diffraction (XRD, Rigaku D/Max Ultima II powder X-ray diffractometer), Raman spectroscopy (Ranishaw inVia, 633 nm), X-ray photoelectron spectroscopy (XPS, PHI Quantera X-ray photoelectron spectrometer). The mechanical properties were measured using a universal testing instrument (HY-0350) with a maximum force of 5 N. The tensile tests were performed at a strain rate of 0.6 mm s^{-1} with a gauge length of 6 mm. The electrical properties were characterized using Kaithley 2400. The electrochemical measurements were obtained using a CHI 660D electrochemical work station. The specific volumetric capacitance (C_V) of fiber-based supercapacitor was calculated from the galvanostatic charge-discharge curve according to the equation of $C_V = [i/(dV/dt)]/V_{fiber}$, where i is the discharge current, dV/dt is the slope of discharge curve, and V_{fiber} is the total volume of two fibers.

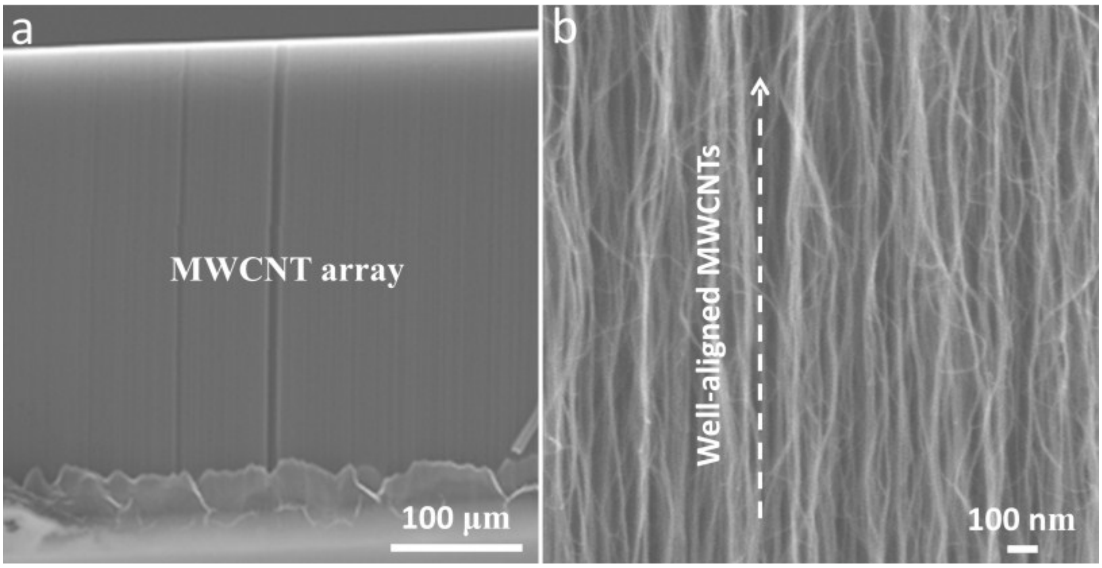

Fig. S1 (a) SEM image of vertical MWCNT array. (b) Magnified SEM image of well-aligned MWCNTs in the array. The white arrow indicates the orientation of MWCNTs.

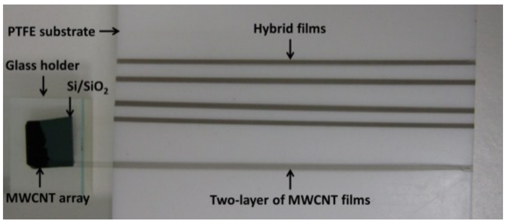

Fig. S2 Photograph of thin MWCNT film continuously drawn out from vertical MWCNT array.

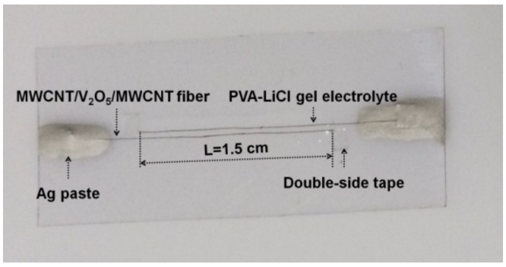

Fig. S3 Photograph of the MWCNT/V_2O_5/MWCNT fiber-based supercapacitor.

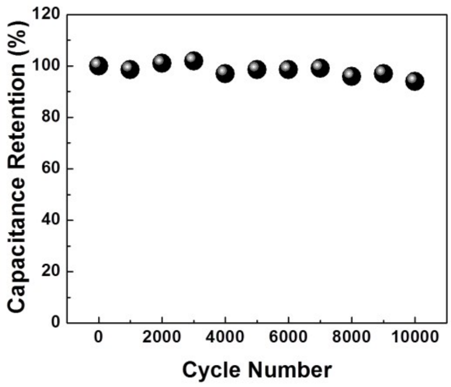

Fig. S4 Charge-discharge cycling stability of solid-state MWCNT/V_2O_5/MWCNT (90 wt% of V_2O_5) fiber-based supercapacitor at current density of 1.0 A cm^{-3}.

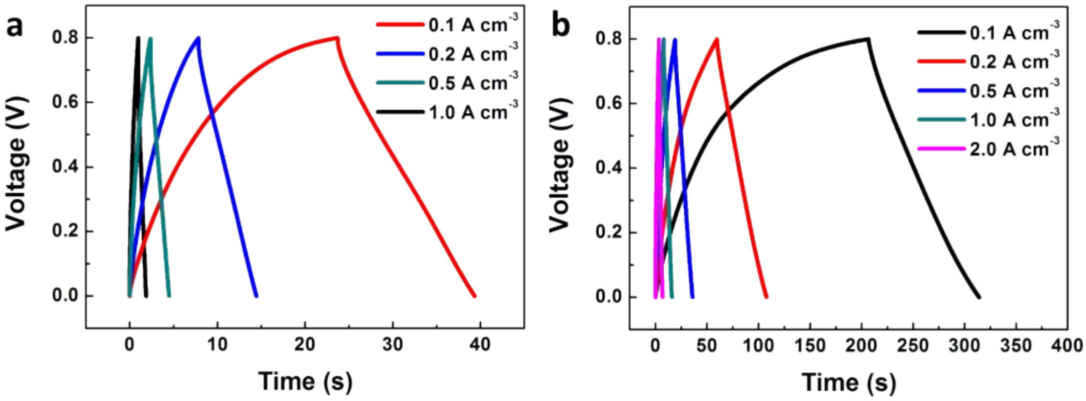

Fig. S5 Charge-discharge curves of MWCNT/V_2O_5/MWCNT fiber-based supercapacitors as a function of current density: (a) 30 wt% and (b) 60 wt% of V_2O_5.

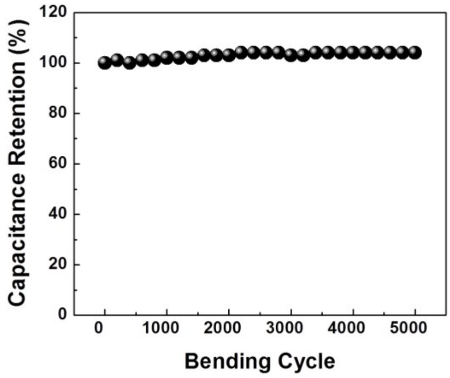

Fig. S6 Capacitance retention of the MWCNT/V_2O_5/MWCNT fiber-based supercapacitor after 5000 bending-unbending cycles.