Supporting information

Experimental Section

Synthesis of aligned carbon nanotubes (CNT). Vertically aligned CNT array was synthesized by chemical vapour deposition with ethylene as the carbon source (flow rate of 90 sccm), Fe (1.2 nm)/Al_2O_3 (3 nm) on a silicon wafer as the catalyst and a mixture of argon and hydrogen (flow rates of 400 and 30 sccm, respectively) as the carrier gas at 740 °C. The aligned CNT sheet was dry-drawn from the above array and paved onto a polytetrafluoroethylene substrate, followed by fixing on a Teflon plate. The specific surface density of the as-prepared CNT sheet was calculated as 1.4 μg cm^{-2}.

Characterization. The structure was characterized by scanning electron microscopy (SEM) (Hitachi FE-SEM S-4800 operated at 1 kV), transmission electron microscopy (TEM) (JEOL JEM-2100F operated at 200 kV), Raman spectroscopy (Renishaw in Via Reflex instrument with an excitation wavelength of 633 nm and laser power of 20 mW). X-ray photoelectron spectroscopy was recorded on an AXIS ULTRA DLD XPS System with MONO Al source (Shimadzu Corp.). Photoelectron spectrometer was recorded by using monochromatic Al KR radiation under vacuum at 5×10^{-9} Pa. All of the binding energies were referred to the C1s peak at 284.6 eV of the surface adventitious carbon. The electrochemical measurements of the fibrous supercapacitor and lithium ion battery were made by an electrochemical workstation (CHI 660D) and an Arbin electrochemical station (MSTAT-5 V/10 mA/16Ch), respectively. The bending measurements of the fibrous supercapacitor and lithium ion battery were performed at a table-top universal testing instrument (HY-0350). The photographs of the devices were taken by a camera (Nikon, J1).

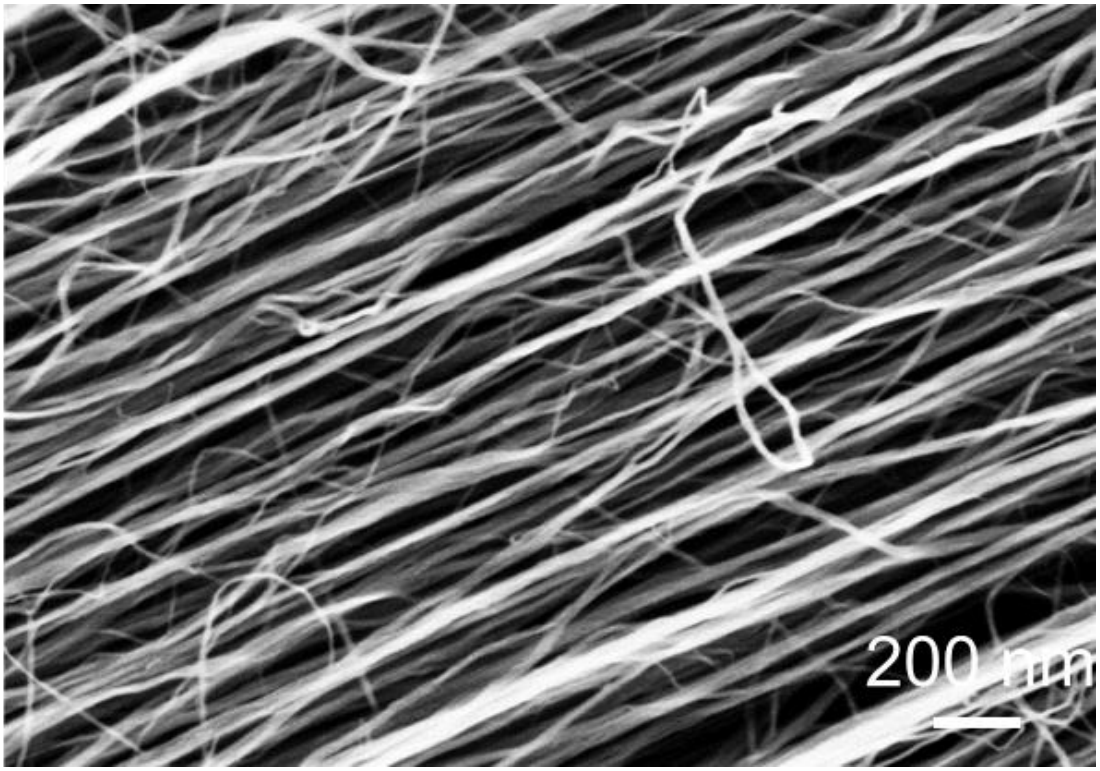

Fig. S1 Scanning electron microscopy (SEM) image of the aligned CNT sheet.

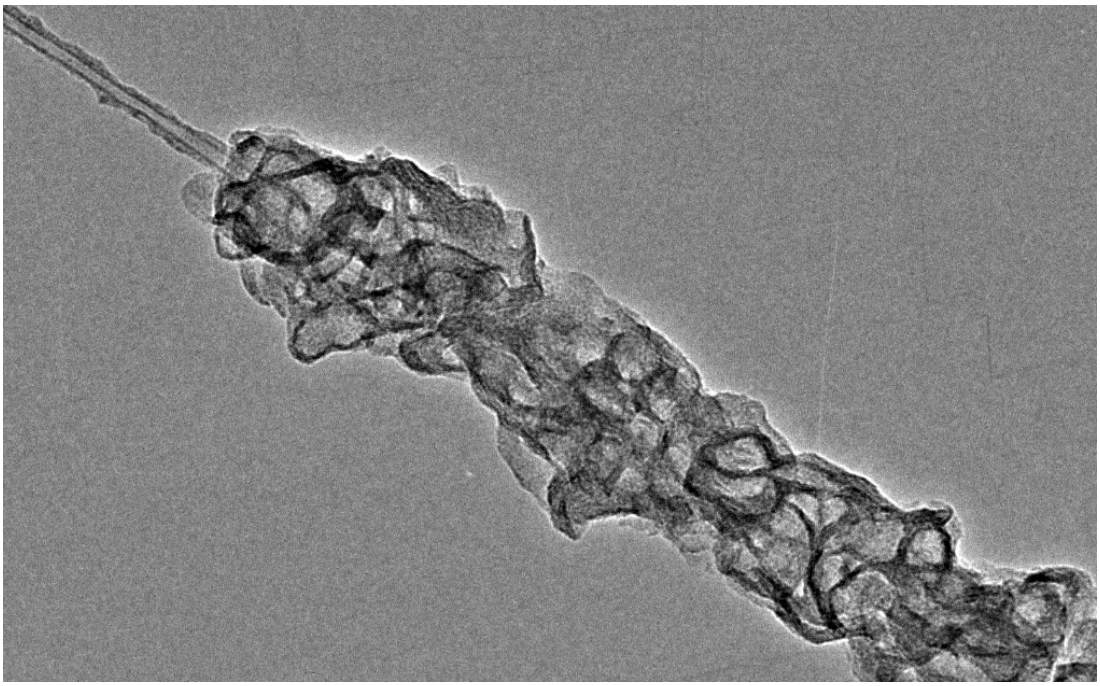

Fig. S2 Transmission electron microscopy (SEM) image of a CNT wrapped by interwoven MoS_2 nanosheets.

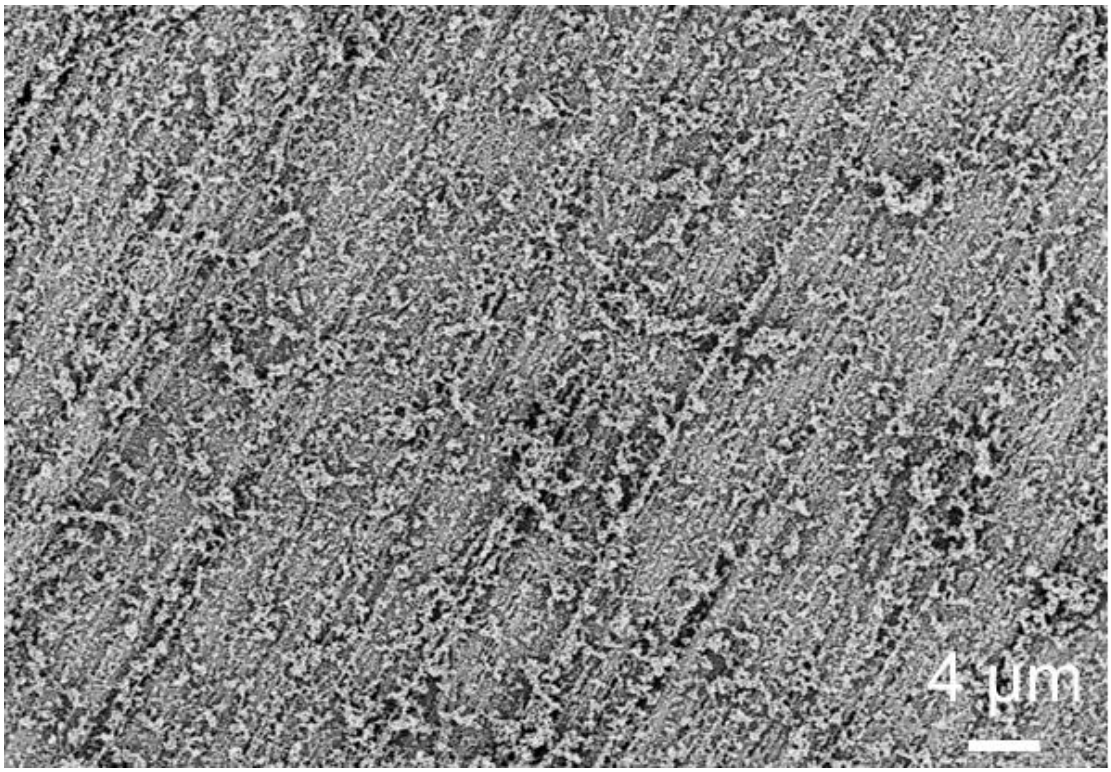

Fig. S3 SEM image of the aligned CNT/MoS_2 hybrid sheet.

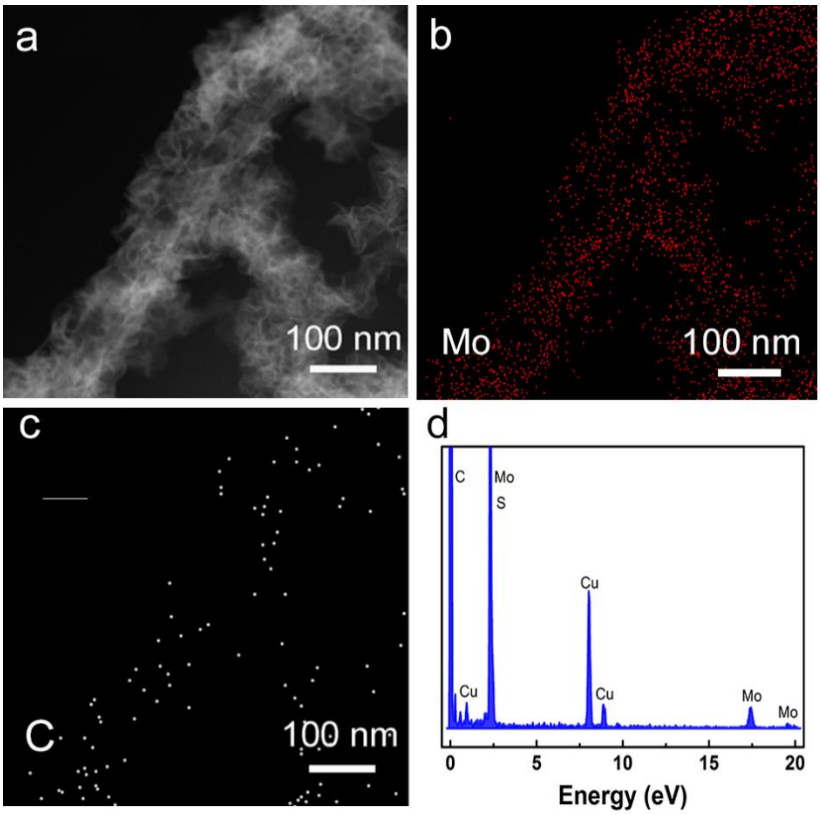

Fig. S4 (a) Scanning transmission electron microscopy image of a CNT wrapped by interwoven MoS_2 nanosheets. (b-c) Energy dispersive X-Ray mapping of Mo and C. Mo and C are marked with red and white, respectively. L-alpha peak was used for Mo. (d) Energy dispersive X-Ray spectroscopy images of CNT/MoS_2 hybrid.

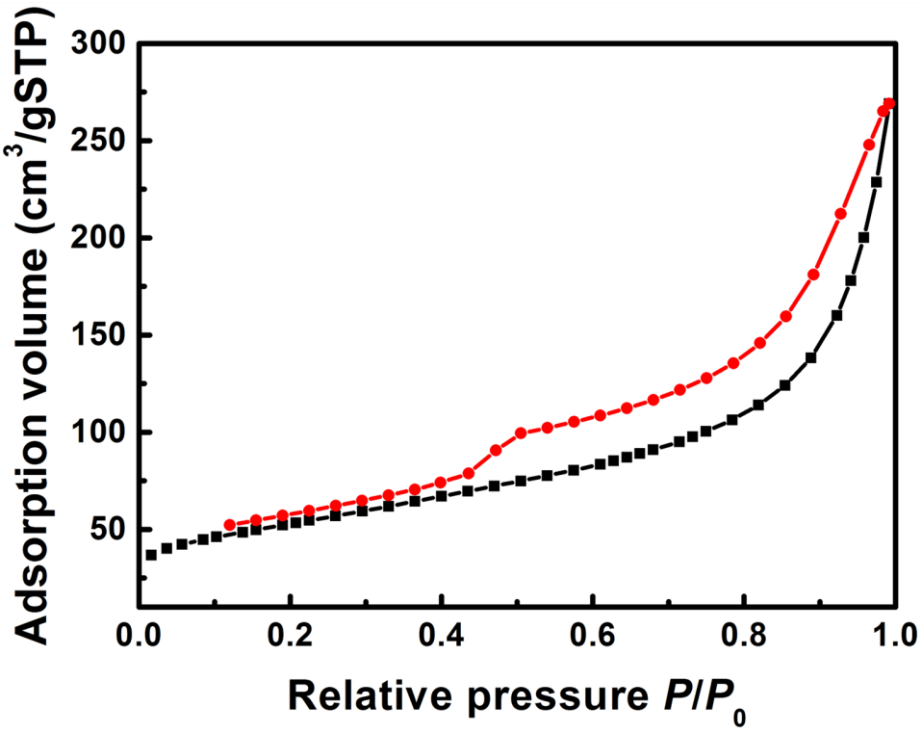

Fig. S5 Nitrogen adsorption-desorption isotherms. *P* is the measured pressure within the adsorption bulb and P_0 is the liquefaction pressure of the adsorbate.

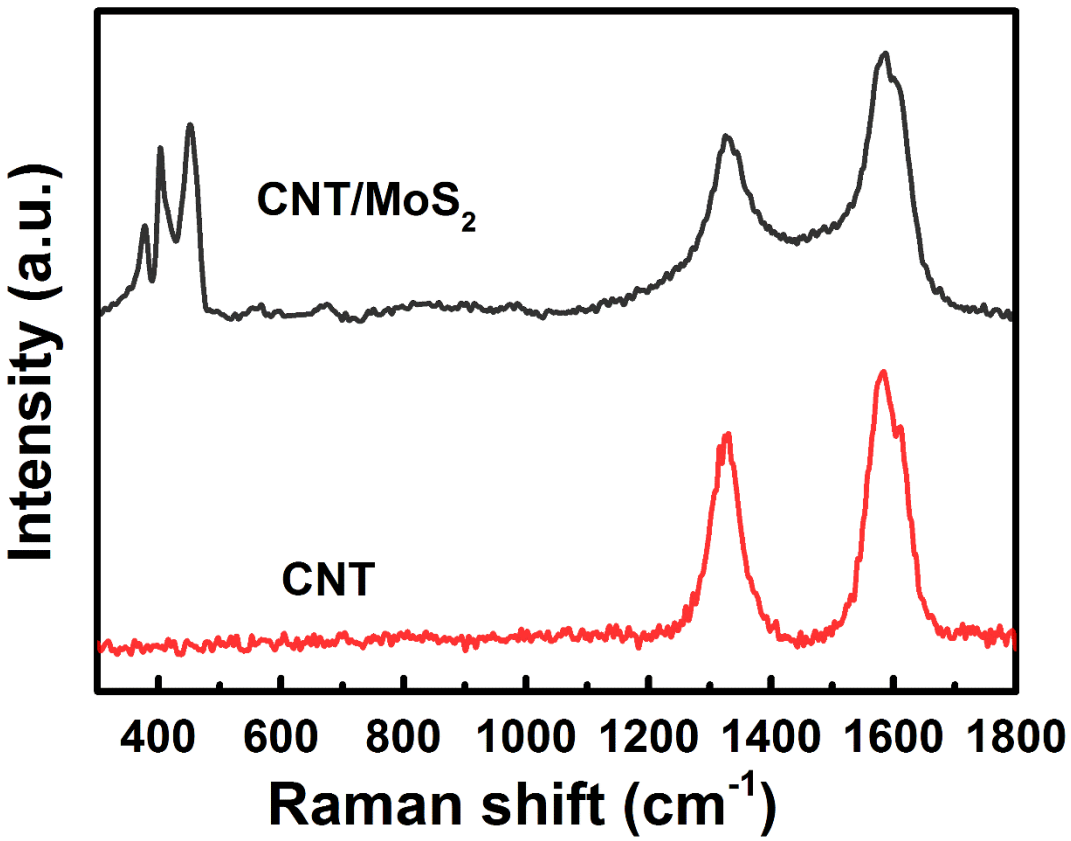

Fig. S6 Raman spectra of the aligned CNT/MoS_2 hybrid fiber and bare CNT fiber.

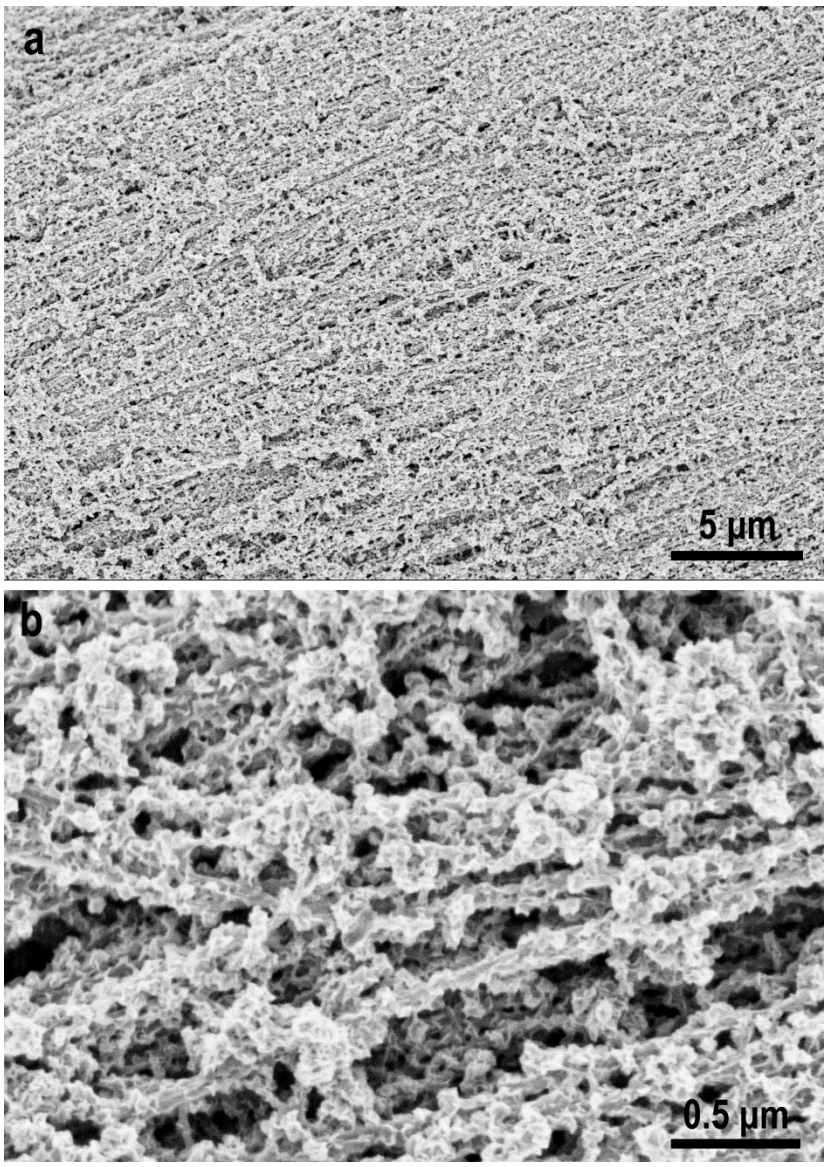

Fig. S7 SEM images of the aligned CNT/MoS_2 hybrid fiber after bending for 1000 cycles. The top and bottom images show low and high magnifications, respectively.

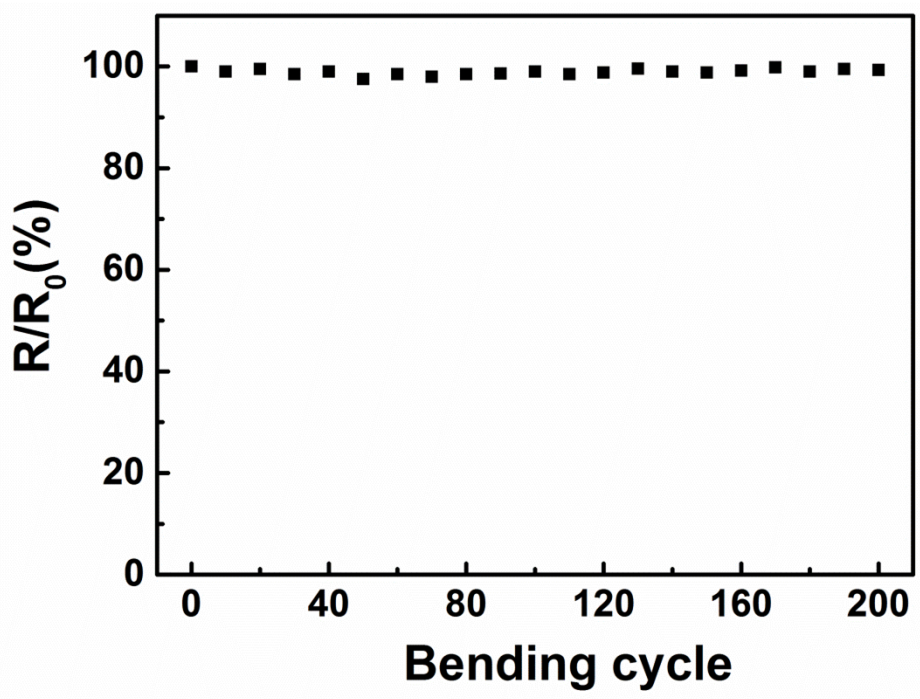

Fig. S8 Resistance variation of the aligned CNT/MoS_2 hybrid fiber after bending with increasing cycles. R_0 and R represent the resistances before and after bending for different cycles, respectively.

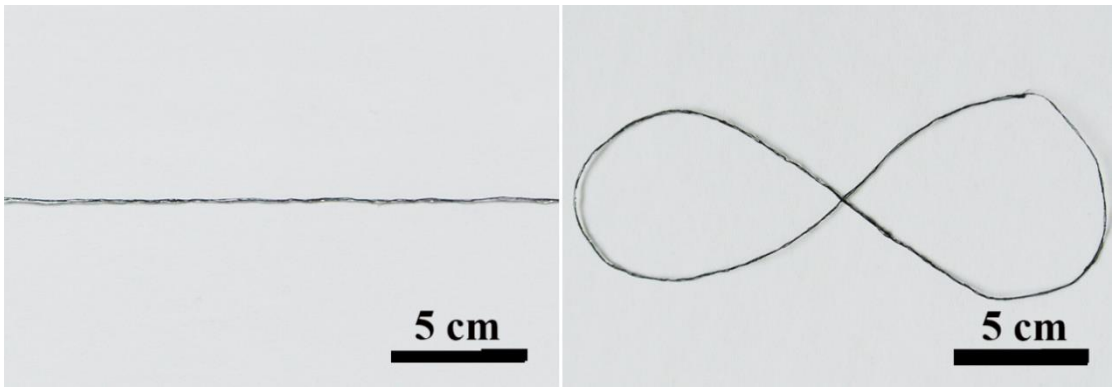

Fig. S9 Photographs of flexible fibrous supercapacitors in different formats.