

Supporting information

For

**Low-cost Wearable Yarn Supercapacitor Constructed by Highly Bended
Polyester Fiber Electrode and Flexible Film**

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Experimental Section

Synthesis of NiCo₂O₄/polyester fiber nanosheet arrays

All the chemicals were of analytical grade and were used without further purification. The samples were synthesized by a facile hydrothermal method. First, a polyester fiber was immersed into abundant acetone and ethyl alcohol, ultrasonically cleaned for 30 min and rinsed with deionized water. Subsequently, it was sensitized by aqueous solution with 40 mL L⁻¹ 38% HCl, Then 0.29g Ni(NO₃)₂·6H₂O, 0.582g Co(NO₃)₂·6H₂O, 0.186g NH₄F and 0.6g urea were dissolved in 50 mL of deionized water under magnetically stirring for 30 min in air. The as-obtained solution was transferred into a 100mL Teflon-lined stainless steel autoclave and a polyester fiber was immersed into the reaction solution. The autoclave was sealed and kept at 120°C for 2.5 h. After that, the solution was cooled down to room temperature. The product

on the polyester fiber was washed with distilled water repeatedly, and then dried at 60°C for 12h. The weight NPNs is about 2.5 mg.

Preparation of MoO₃ nanobelts

The preparation MoO₃ nanobelts is through our previous method.^[1] In a typical synthesis, 40 mL of H₂O₂(30%) was added dropwise into 4 g of molybdenum powders in the ice-water bath under magnetic stirring for 4 h to remove the redundant H₂O₂, forming a clear orange solution. The orange solution was transferred and sealed in a 100 mL Teflon-lined stainless autoclave. The autoclave was heated at 180°C for 24h, then cooled to room temperature.

Preparation of CNT/MoO₃ films electrode

CNT/MoO₃ films electrodes were obtained by vacuum filtration (The CNTs were purchased by Time@nano). First, 1g dispersant and 60 mg CNT were dissolved in 100 mL deionized water under magnetically ultrasonic for 1 h in air. Then, 0.1g MoO₃ nanobelts were added in above solution and ultrasonic 15 minutes again. Finally, The as-obtained solution was filtered on the PTFE membrane filter and the film was then dried and can be easily peeled off from the filtration paper.

Fabrication of fiber-shaped supercapacitor(FSC)

The FSC was assembled by using CNT/MoO₃ film as the negative electrode and NiCo₂O₄/ polyester fiber nanosheet arrays as the positive electrode with a separator (NKKTF40, 40μm thickness, low ESR type, purchased from SCM industrial Chemical CO.,LTD) and PVA/Na₂SO₄/KOH gel as a solid electrolyte. PVA/Na₂SO₄/KOH gel was prepared as follows: in a typical process, 4g PVA was

dissolved in 40mL deionized water with stirring at 85°C for 1h. Then, 1mol L⁻¹ Na₂SO₄ and 1mol L⁻¹KOH were slowly dropped into the above solution at 80 °C under stirring until the solution became clear, respectively. The CNT/MoO₃ film, NiCo₂O₄/polyester fiber electrode were soaked in the gel for about 5min and then were assembled together. The device was kept at 60 °C for 12 h to remove excess water in the electrolyte. The volume of this device is 0.39 cm³

Characterization

The obtained product was characterized by scanning electron microscopy (SEM, Hitachi-4800), and X-ray powder diffraction (XRD, Rigaku Dmax-rB, Cu K α radiation, $\lambda=0.1542\text{nm}$, 40kV, 100mA). X-ray photoelectron spectroscopy(XPS) was carried out by using the Physical Electronics PHI model 5700 instrument and a field emission transmission electron microscopy (FETEM) (JEM-2100). The optical properties of the as-synthesized samples were investigated by Raman spectroscopy (HR800). The specific surface area of the as-prepared material was estimated using the Brunauer-Emmett-Teller (BET) equation based on the nitrogen adsorption isotherm obtained with a Belsorp-max. The stress-strain test was performed on the universal testing machine (Instron 3344). The strain rate during stress-strain tests was set at 5 mm/min. Electrochemical characteristics of the as-obtained products were studied on an CHI660 electrochemical work station (Chenhua, Shanghai) using cyclic voltammetry, chronopotentiometry and electrochemical impedance test by configuring the sample in to a three-electrode cell, where the sample was used as the working electrode, Pt foil as the counter electrode and a saturated calomel electrode

(SCE) as the reference electrode.

Calculation Methods

The specific volumetric capacitance (C_v) of the electrode were calculated based on its charge/ discharge curve with the following equation:

$$C_v = I\Delta t / V\Delta U \quad (1)$$

where I , Δt , V , and ΔU are the discharge current, discharge time, effective active material volume in the electrode and voltage window, respectively.

The volume of the active material was calculated by assuming it as a cylinder. So the volume can be obtained using the following equation:

$$V = A \times L = \pi \times R^2 \times L \quad (2)$$

where A is the cross-sectional area of the polyester fiber, R is the radius of the polyester fiber, and L is the length of the polyester fiber immersed into the electrolyte.

For the FSC, the specific volumetric capacitance of the whole device ($C_{\text{cell},v}$) was calculated using the following equation:¹

$$C_{\text{cell},v} = I\Delta t / V_{\text{cell}}\Delta U \quad (3)$$

where V_{cell} refers to the whole device volume, including the volume of a polyester fiber, a film and the surrounding electrolyte. The other symbols are the same as above.

Energy density(E) and average power density (P) could be calculated as:²

$$E_v = C_{\text{cell},v} \times U^2 / 2 \times 3600 \quad (4)$$

$$P_v = E_v \times 3600 / \Delta t \quad (5)$$

It is necessary to maintain charge balance between the positive and the negative electrodes in a NPCM device. That is

$$q^+ = q^- \quad (6)$$

Where q^+ and q^- are electrical charges stored in the positive and negative electrodes, respectively. The optimal volumetric ratio between electrochemically active species in the positive and negative electrodes should satisfy the following equation:

$$R = \frac{V_+}{V_-} = \frac{C_- \Delta V_-}{C_+ \Delta V_+} \quad (7)$$

Where V_+ and V_- are the effective active material volume, C (F cm^{-3}) is the specific volumetric capacitance (C_v) of electrode and ΔV is the voltage change (V).

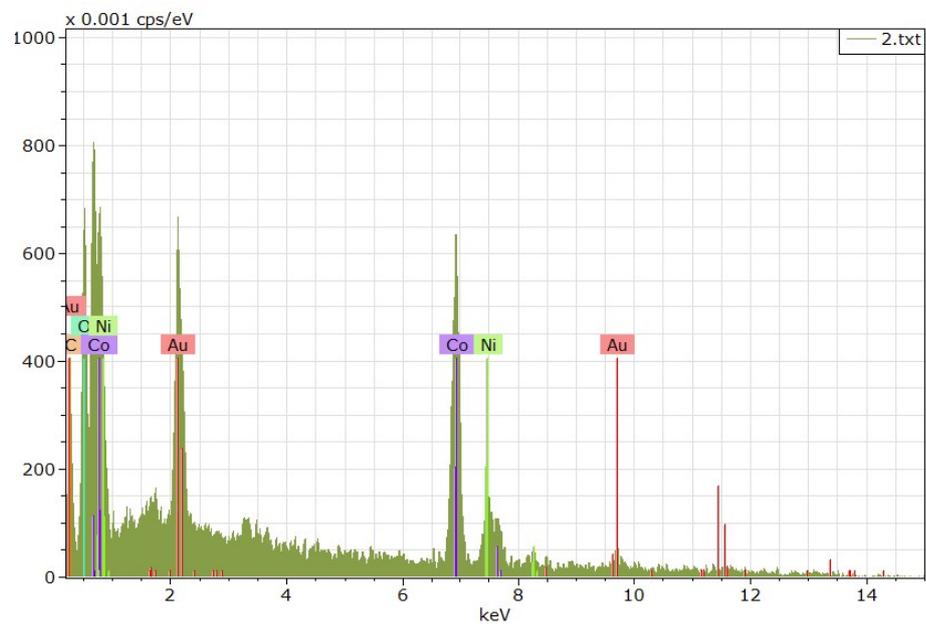


Figure S1 EDS image of NPNs

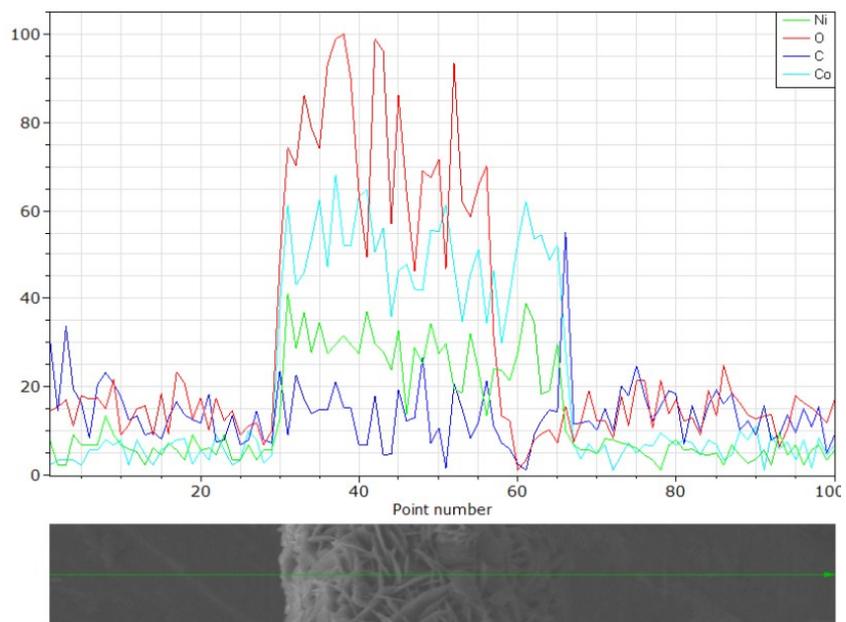


Figure S2 Line scan by EDAX of C, Ni, Co and O across the fiber

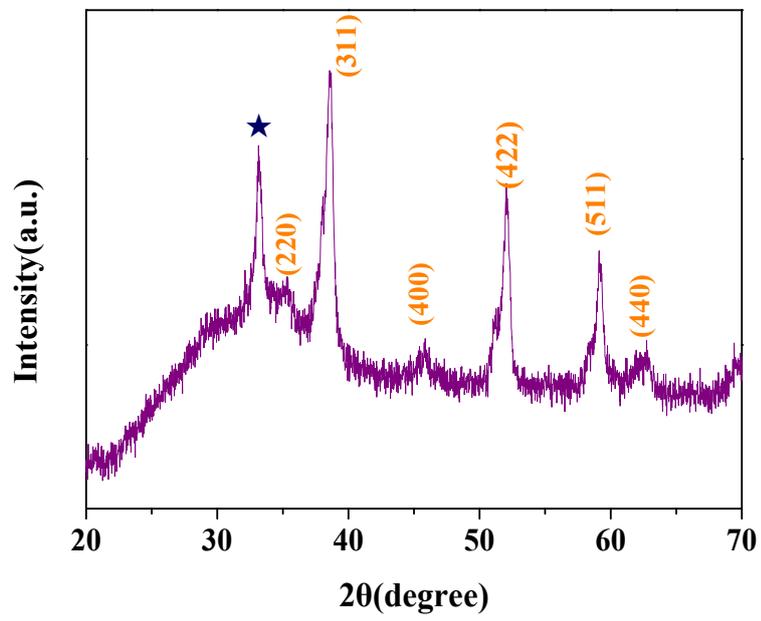


Figure S3 The XRD pattern of NPNs

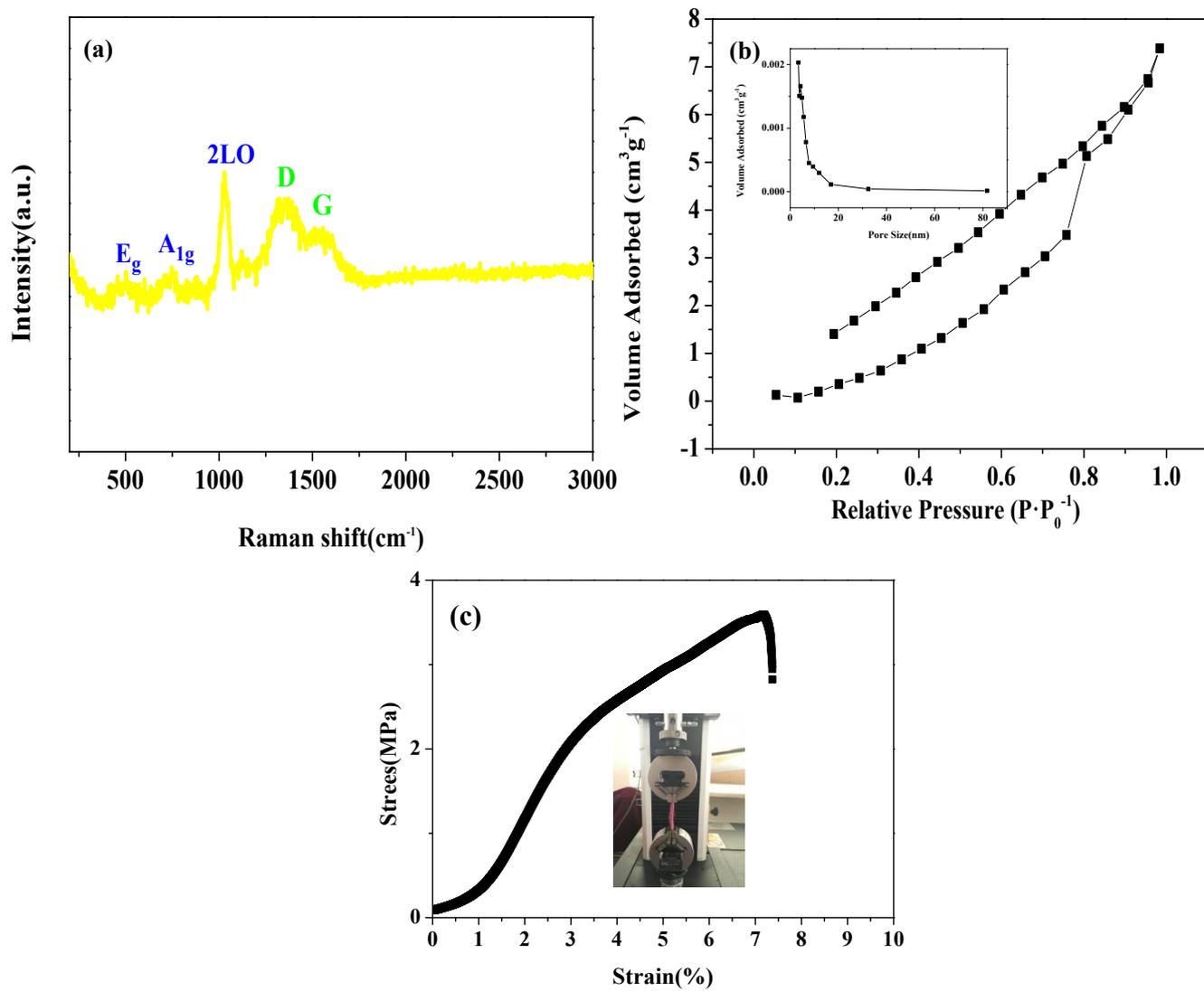


Figure S4 (a) Raman image of the NPNs composites.(b) Nitrogen adsorption/desorption isotherm and BJH pore size distribution plot of the as-synthesized product.(c)Stress-strain curves of the NPNs.

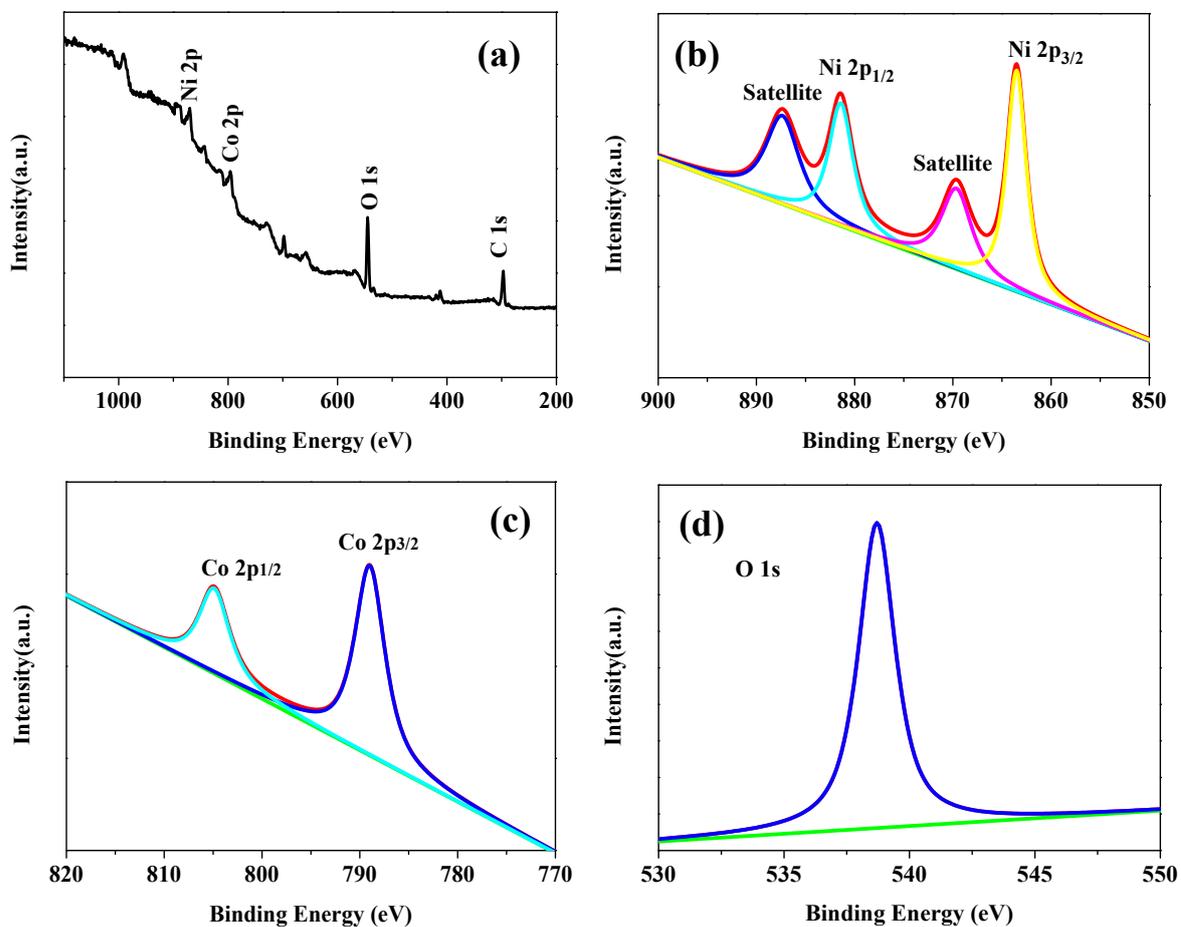


Figure S5 XPS spectra of (a) survey spectrum, (b) Ni 2p, (c) Co 2p, and (d) O 1s for NPNs

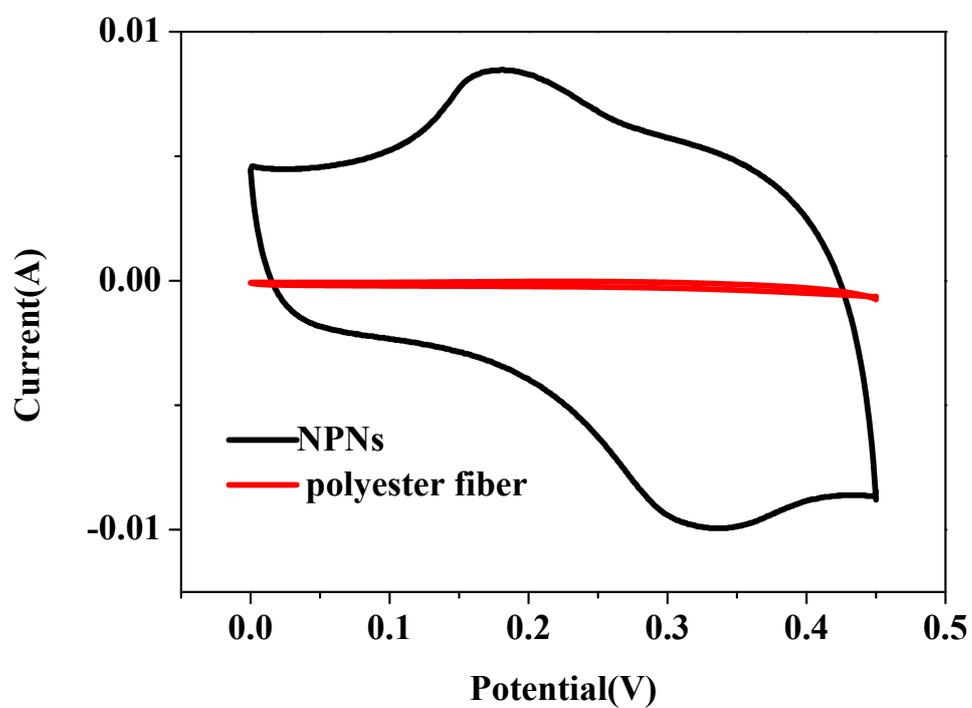


Figure S6 The CV curves of NPNs and the pure polyester fiber at a scanning rate of 100 mVs^{-1}

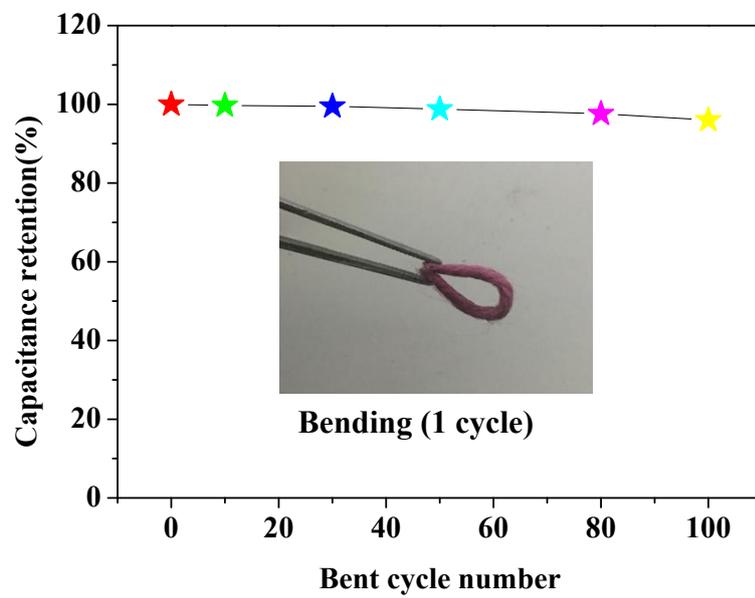


Figure S7 Capacitance retention after bending for various cycles

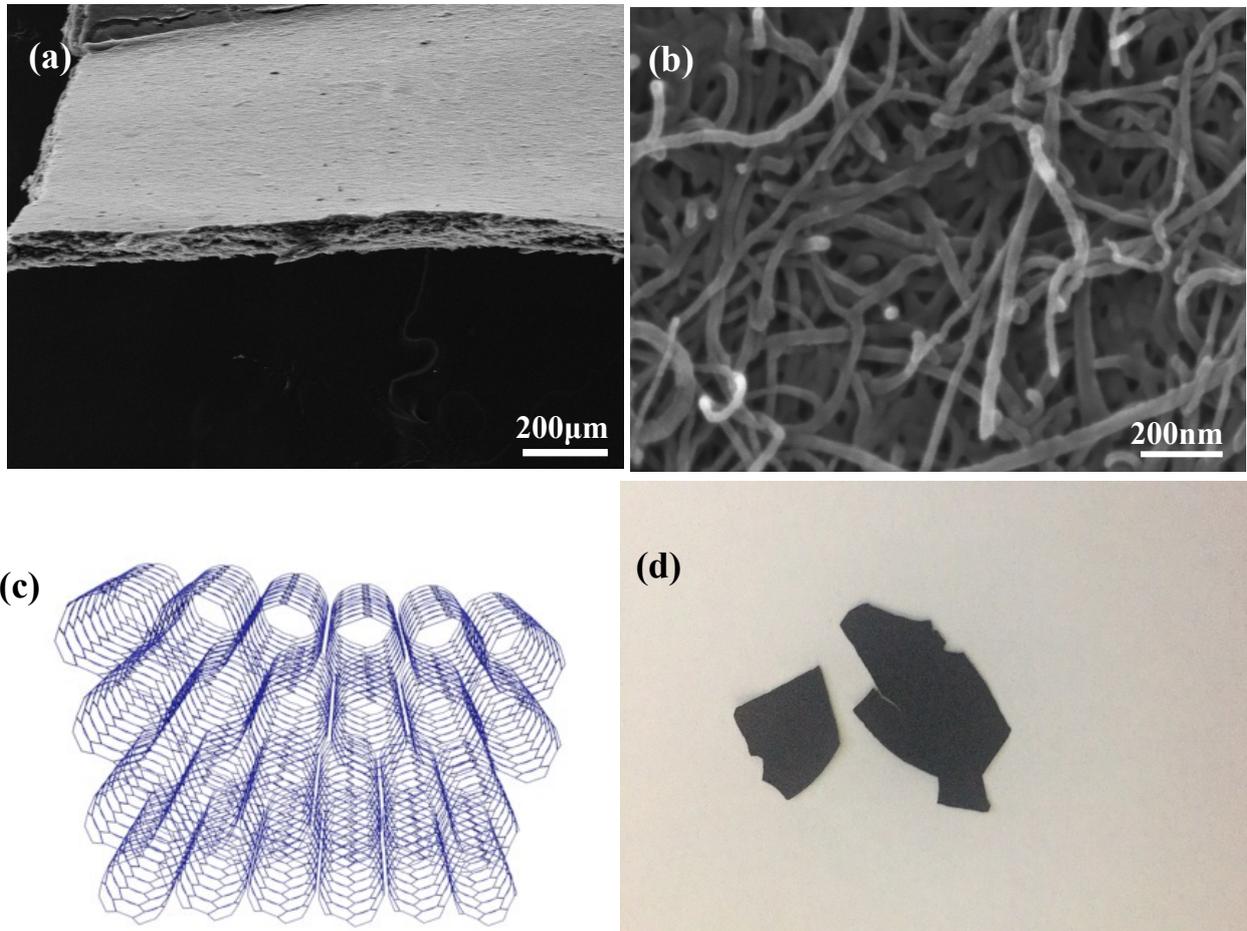


Figure S8 (a,b) SEM images of the CNT at low and high magnification, respectively.

(c) Schematic illustration showing the crystal structure of the CNT. (d) The photographs when the CNTs content is low.

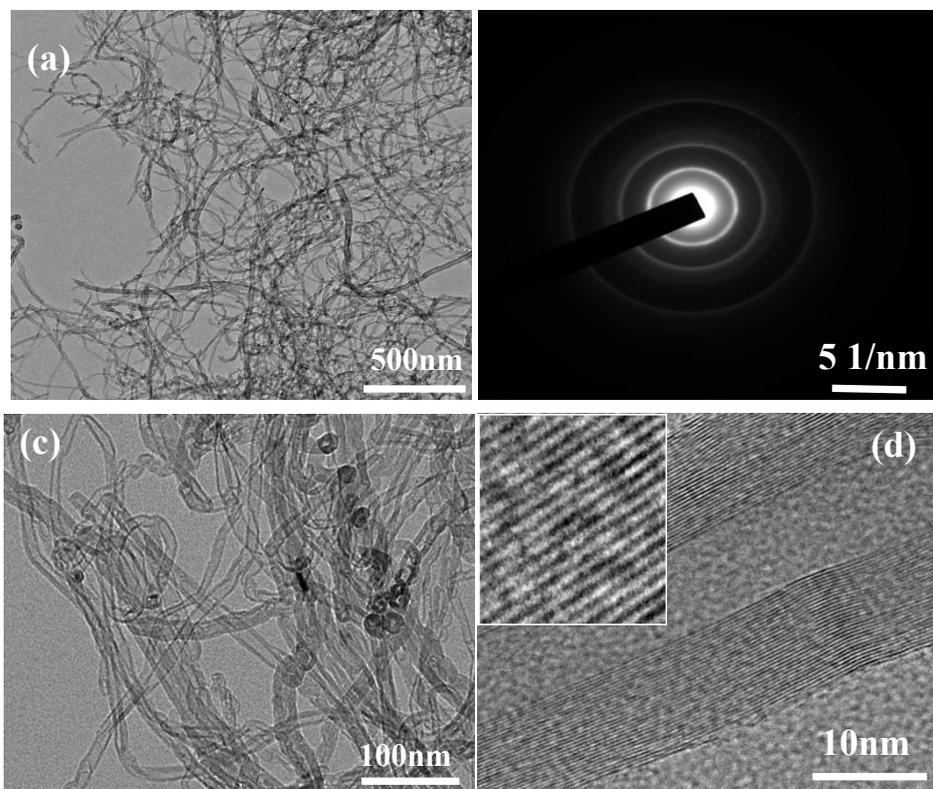


Figure S9 (a,c) TEM of CNT.(b) SAED pattern of CNT.(d) HRTEM image of CNT

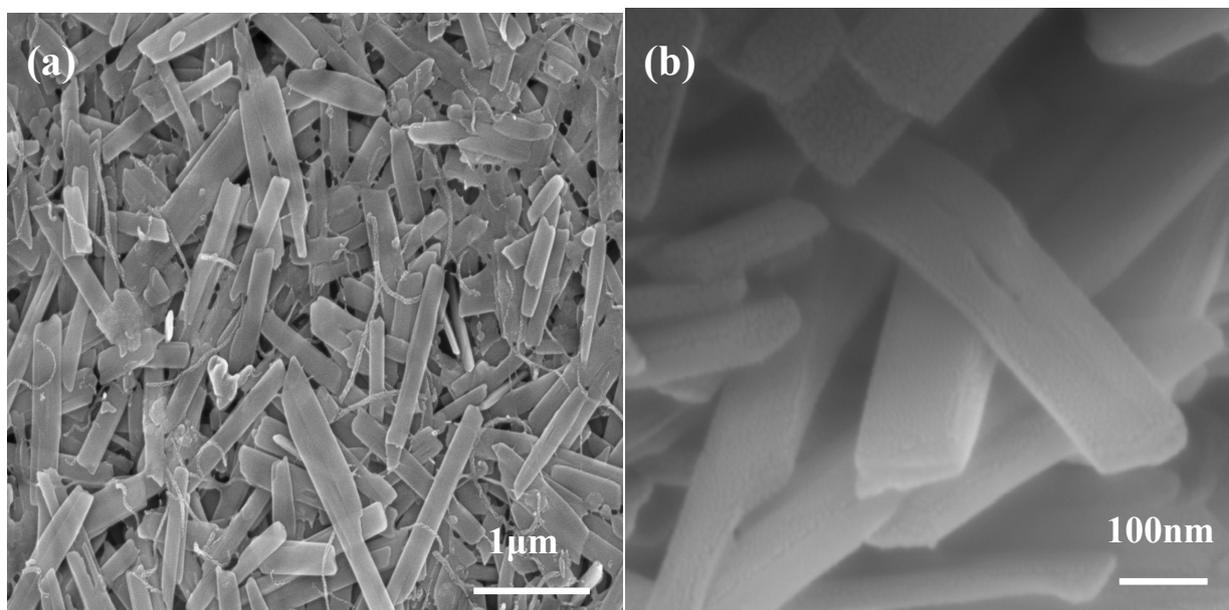


Figure S10 (a-b)SEM images of the MNBs at low and high magnification, respectively.

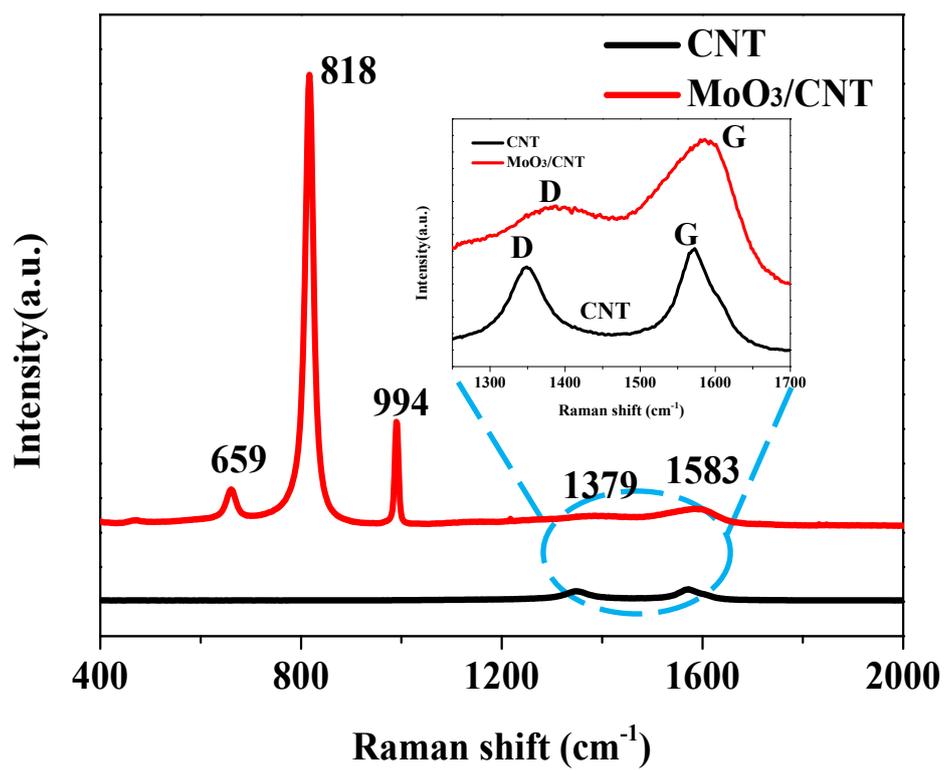


Figure S11 Raman spectra of the pure CNT and MoO₃/CNT nanocomposites. Inset shows carbon scattering bands of the pure CNT and MoO₃/CNT

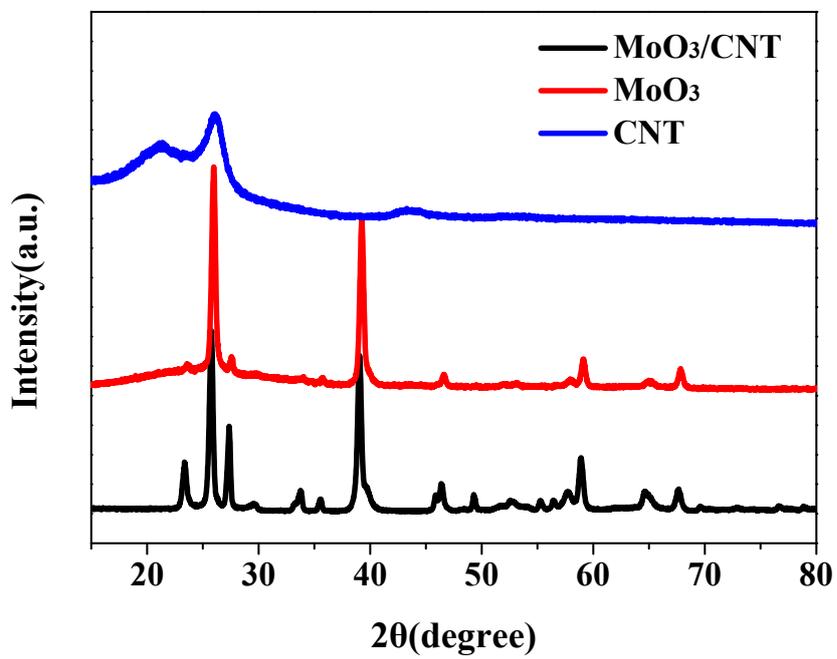


Figure S12 XRD patterns of the pristine MoO₃ nanobelts , MoO₃/CNT, and CNTs

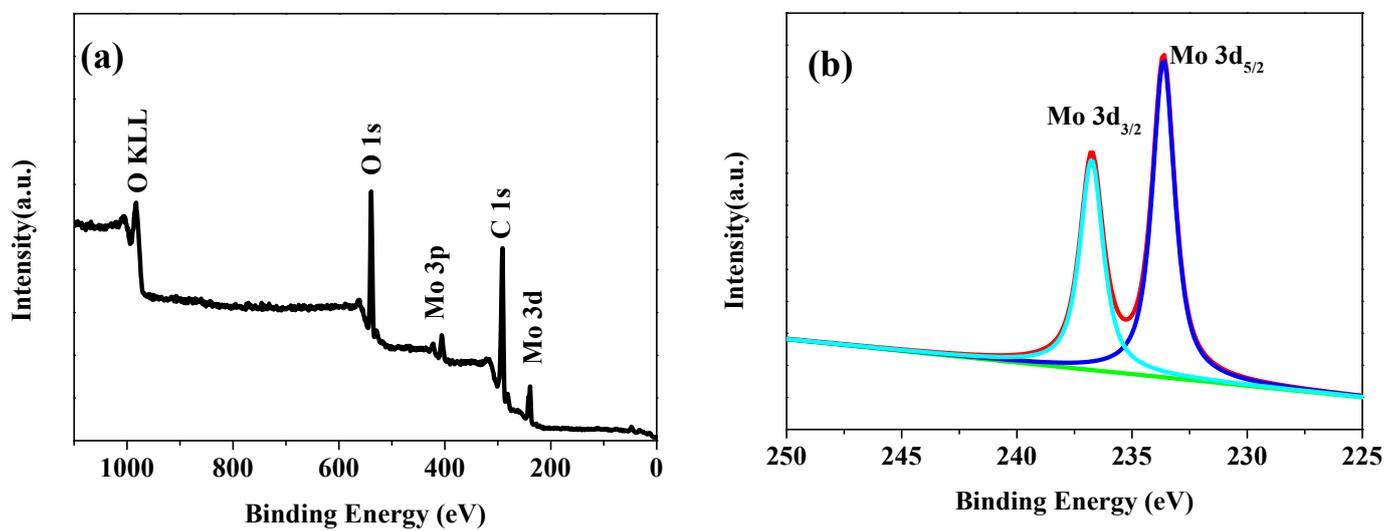


Figure S13 XPS spectra of (a) survey spectrum, (b) Mo 3d,

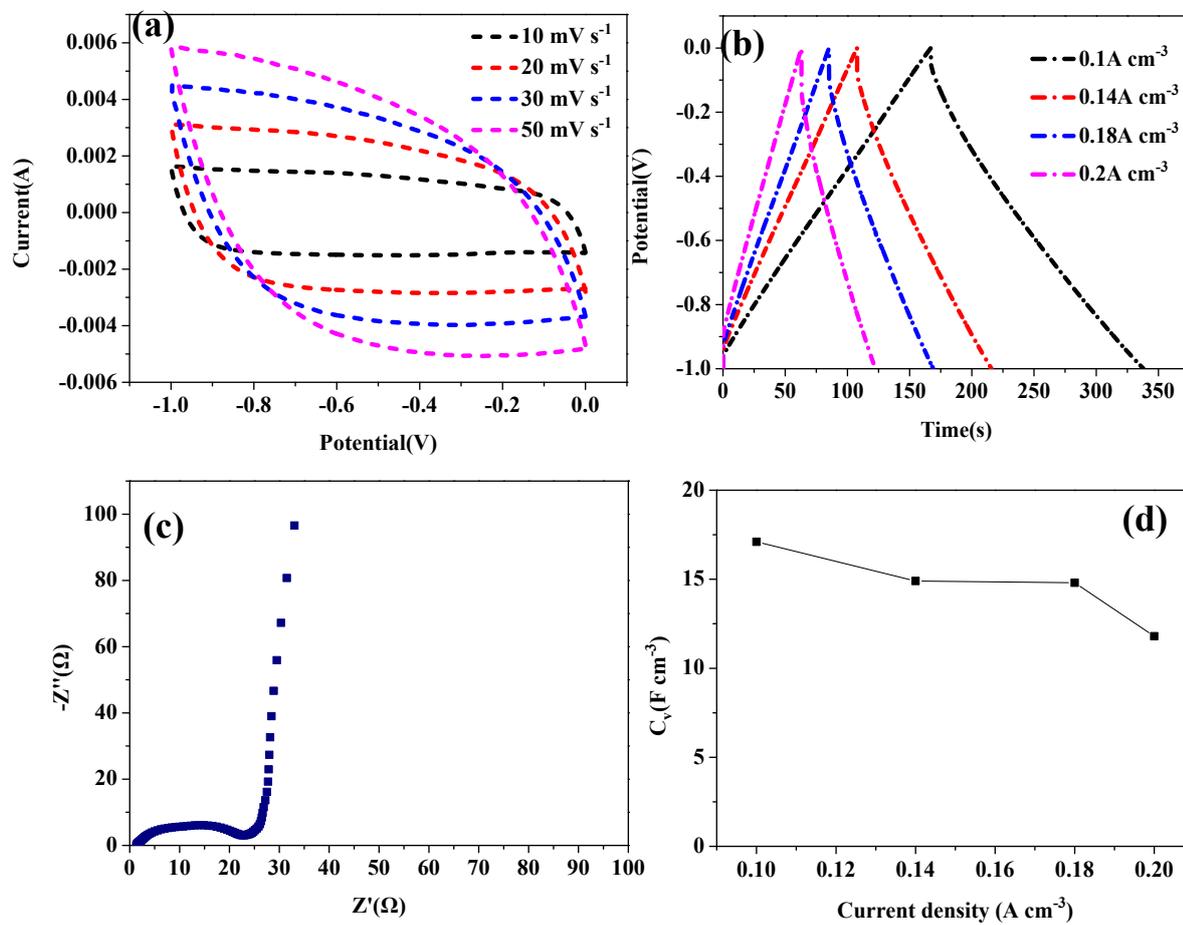


Figure S14 (a,b) GCD and CV curves of CNTs.(c) EIS of CNTs (d) Specific volumetric capacitances of CNTs

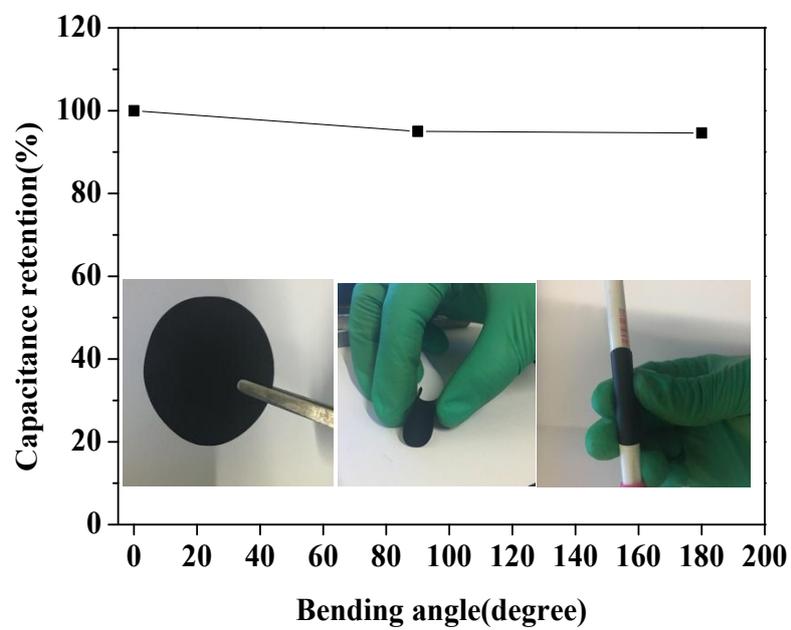


Figure S15 capacitance retention under different bending states

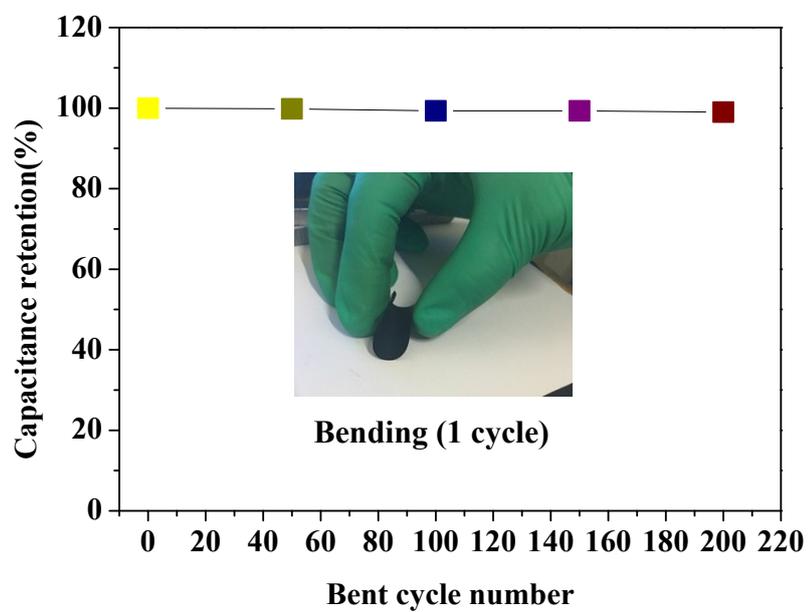


Figure S16 Capacitance retention after bending for various cycles

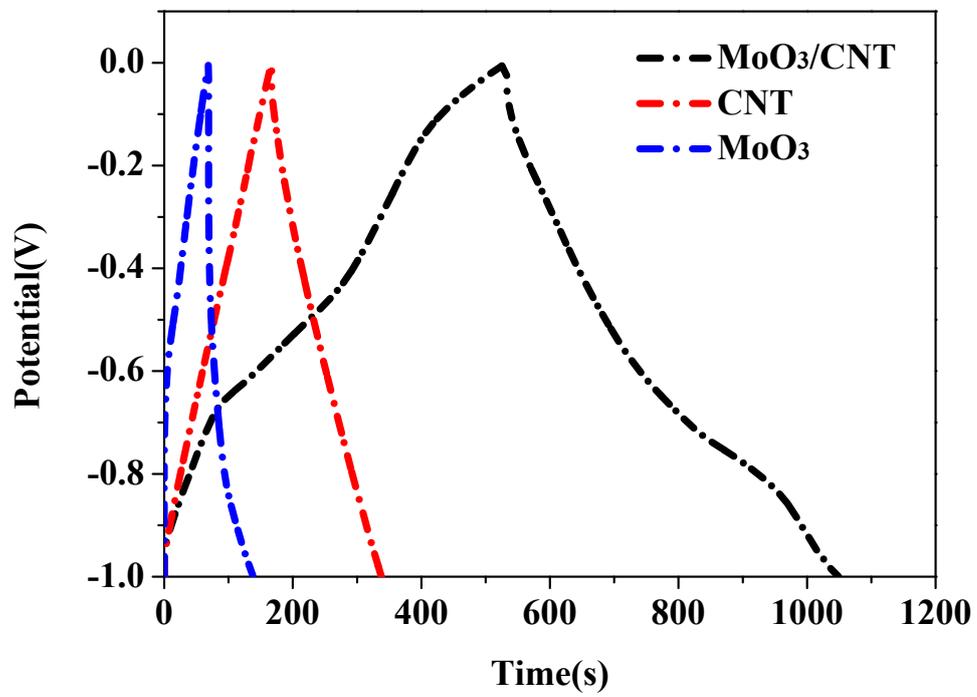


Figure S17 GCD curves of MoO₃ nanobelts, MoO₃/CNT, and CNTs

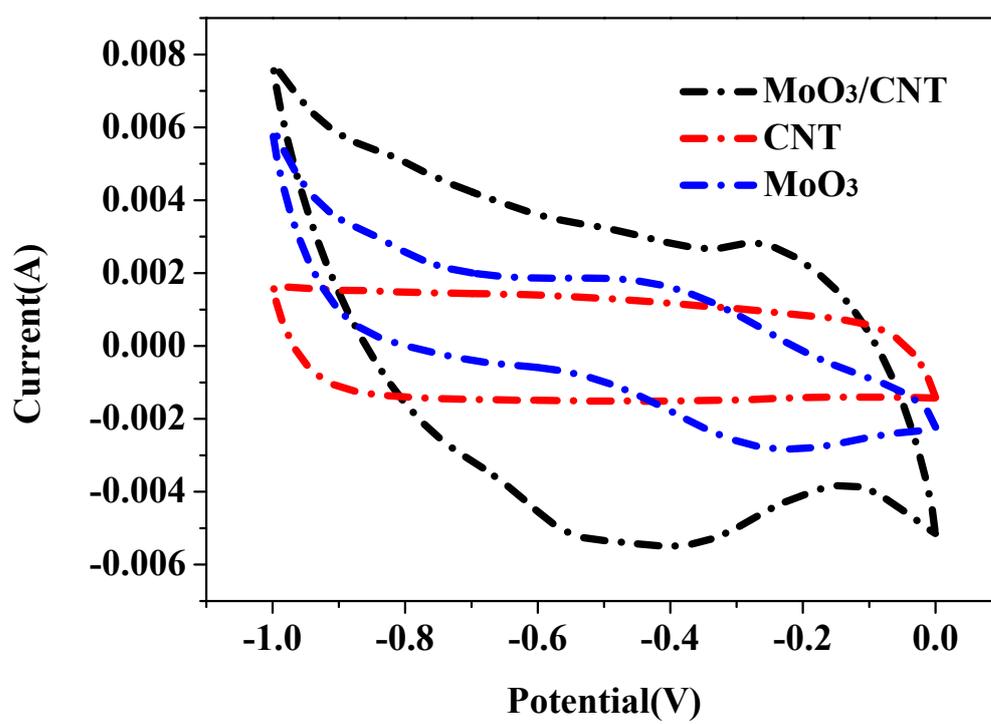


Figure S18 CV curves of MoO₃ nanobelts, MoO₃/CNT, and CNTs

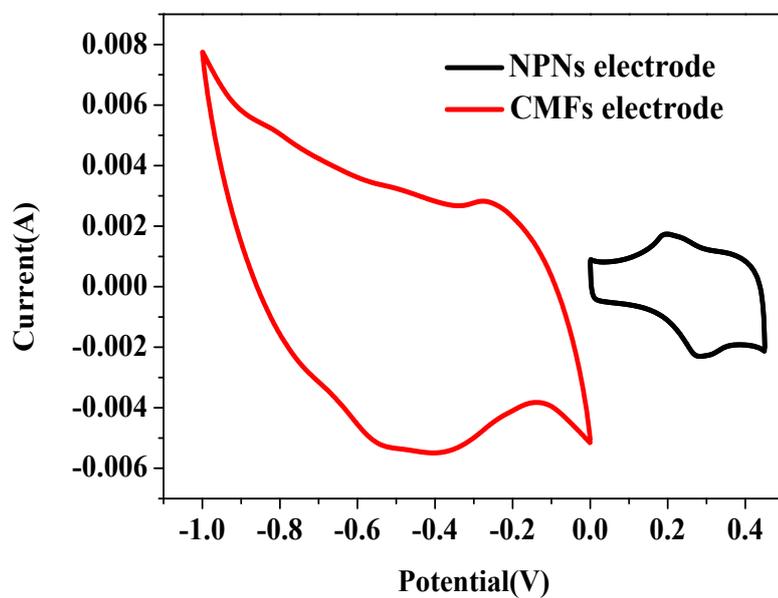


Figure S19 Comparative CV curves of NPNs electrode and CMFs electrode obtained

at a scan rate of 10 mV s⁻¹



Type	Electrolyte	Device	Energy Density (mWh cm ⁻³)	Ref.
NPCM-FSC	PVA/Na ₂ SO ₄ /KOH	Micro-Asymmetric	7.9	The work
rGO/CNT//rGO/ MnO ₂ FAS	PVA/LiCl	Micro-Asymmetric	5.5	3
CNT/MnO ₂ yarn (CMY) Supercapacitor	PVA/H ₃ PO ₄	Micro-Asymmetric	3.52	4
N-rGO/CNT@ MnO ₂ /rGO/CNT fiber	PVA/H ₃ PO ₄	Micro-Asymmetric	5	5
CNT-graphene FMS	PVA/H ₃ PO ₄	Micro-Asymmetric	6.3	6
graphene-based in-plane IMS	PVA/H ₃ PO ₄	Micro-Asymmetric	2.5	7
CNT/MnO ₂ micro- supercapacitor	H ₃ PO ₄ /poly	Micro-Asymmetric	1.73	8

Figure S20 The future tendency of the NPCM-FSC.

Table 1

PEDOT/CNT supercapacitor	PVA/H ₃ PO ₄	Micro-Asymmetric	1.4	9
Carbon/MnO ₂ supercapacitor	PVA/H ₃ PO ₄	Micro-Asymmetric	0.22	10

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