Supporting Information

Rolled-up island-bridge (RIB): A new and general electrode configuration design for wire-shaped stretchable micro-supercapacitor array

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Experimental section

Preparation of few-layered Ti_3C_2Tx flakes toward flexible and free-standing MXene film electrode. Firstly, an acidic solution consisting of 1.0 g dissolved LiF and 20 ml 6 M HCl was used to etch the Ti_3AlC_2 MAX raw powder (1.0 g), which was kept at 40 °C for 48 hours. Next, the reaction mixture was centrifuged, and the precipitate was further washed until the pH value of the washing liquor achieving 6. After drying, the multi-layered Ti_3C_2Tx product was acquired, which was further dispersed into 100 mL DI water (0.2 g). Followed intense ultrasound for 60 minutes under ice bath and then centrifugal removal of unexploited multi-layered Ti_3C_2Tx powders (4000 rpm/5 min), the colloidal solution of few-layered Ti_3C_2Tx flakes (~0.5 mg mL^{-1}) was obtained. Finally, the colloidal solution of few-layered Ti_3C_2Tx flakes was filtrated into flexible and free-standing MXene film electrode for further use.

Preparation of flexible and free-standing CNTs@PPy film electrode. Flexible and free-standing CNTs@PPy film electrode (2 cm×2 cm) was prepared via conformal electrodeposition of PPy on the surface of CNT fibers constituting the CNTs paper with a three-electrode system, in which CNT paper serves as working electrode, platinum plate serves as counter electrode, and Ag/AgCl serves as reference electrode. Aqueous solution (100 ml) containing 0.01 mol L^{-1} $LiClO_4$ (AR grade, Aladdin) and 0.2 mol L^{-1} pyrrole monomer (AR grade, Aladdin) was used as electrolyte. Electrodeposition was carried out at 2 mA cm^{-2} for 80 to 95 minutes with an electrochemical workstation (CHI 660E).

Preparation of GaIn-Ni semi liquid metal. Firstly, 8 g commercial GaIn liquid metal consisting of 74.5 wt% gallium and 24.5 wt% indium (Northeast Non-ferrous Metals Market Co., Ltd.) with a low melting point (16 °C) was agitated with a glass bar at 90 ℃. Then, 0.8 g Nickel (Ni) powders (Aladdin, diameter in the range from 20 to 100 nm, 99.9%) are slowly added until the Ni powders mixed in GaIn totally. After natural cooling, the semi liquid metal defined as GaIn-Ni was obtained for directly screen printing.

Fabrication of 1D Wire-shaped stretchable micro-supercapacitor array. Firstly, interdigital MXene negative electrode and CNTs@PPy positive electrode were prepared via laser-cutting above-obtained flexible and free-standing MXene film electrode and CNTs@PPy film electrode (Figure 4a). Then, the MG tissue paper (15 mm×9 mm) with a thickness of 20 μm and a Young's modulus of 3.5 MPa is chosen as the stiff substrate to fix the interdigital MXene negative pole and CNTs@PPy positive pole to assemble the hybrid MSC active units. Square grooves (9 mm×9 mm) are pre-prepared via reverse mould method on the elastomer of Ecoflex film (Ecoflex 00-30, ~400 μm in thickness) to fill the gel electrolyte films (PVA/1 M H_2SO_4 gel electrolyte). Finally, the hybrid MSCs were deployed in the "islands" region buried with PVA/1 M H_2SO_4 gel electrolyte films and interconnected by screen-printed GaIn-Ni based circuits "bridge" on bottom elastic silicone film. After coating a top layer of uncured Ecoflex on the bottom silicone film and complete curing at room temperature, the 1D Wire-shaped stretchable micro-supercapacitor array can be obtained through further rolling up the elastic silicone film matrix embracing the asymmetric MSC active islands connected by conductive/stretchable bridges and edge-sealing with semi-cured silicone as shown in Figure 1.

Electrochemical measurements. Electrochemical performance was assessed with CV, EIS, and GCD measurements via an electrochemical workstation (CHI 660E, Chenhua, Shanghai). Impedance spectroscopy measurement was performed at open circuit voltage with ±10 mV amplitude.

The linear/areal capacitance (C_l/C_s, mF cm^{-1}/mF cm^{-2}) and energy density (W_l/W_s, mWh cm^{-1}/mWh cm^{-2}) of the devices were calculated according to the equations:

$$C = Q/\Delta E = I\Delta t/\Delta E \quad (1)$$

$$C_l = C/l = I\Delta t/l\Delta E \quad (2)$$

$$C_s = C/S = I\Delta t/S\Delta E \quad (3)$$

$$W_l = 0.5C(\Delta E)^2/3600l \quad (4)$$

$$W_s = 0.5C(\Delta E)^2/3600s \quad (5)$$

Here, C represents the total capacitance, Q represents the total charge, I represents the discharge current, Δt represents the discharge time, ΔE represents the potential window during the discharge process after IR drop, and l and S represents the total length and surface area of the device.

3D full-field strain measurement and analysis. The real-time measurement and analysis of strain field of the object surface during deformation were performed via the combination of non-contact digital image correlation (DIC) technology with binocular stereo vision technology by tracking the speckle image of the object surface (XTOP 3D Technology Co., LTD., XTDIC-CONST). A thin layer of black paint is sprayed on the surface of sample, then the sprayed white paint to prepare the random speckle white pattern on the samples before test. The sample stretching is realized by using a home-made motorized test stand at a constant speed during the working of 3D full-field strain measurement and analysis system. With the built-in analysis software, the grayscale distribution from the speckle patterns among images before and after the deformation were analyzed to generate the corresponding 3D strain mapping of the devices.

Material Characterization. The micromorphology of the samples was revealed by Field-emission scanning electron microscopy and transmission electron microscopy. The phase composition characterizations of the samples were carried out via an X-ray diffractometer (XRD Bruker D8-ADVANCE) with Cu Ka radiation (λ=1.54056 Å), X-ray photoelectron spectroscopy, Fourier transform infrared spectroscopy. Sheet resistances of the films were measured via a four-

point probe method. The resistance of Ni-EGaIn lines was measured with a multimeter (VICTOR 86E). Contact angle (CA) values of the samples were obtained via a contact angle measuring instrument (Shanghai innuo Precision Instrument Co., Ltd., CA100D). The sampling volume is 10 μl.

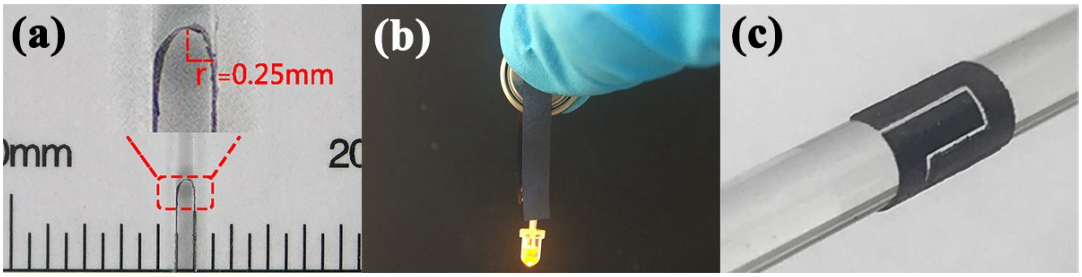

Figure S1. Photos demonstrate the flexible and free-standing few-layered MXene self-assembled film a) undergoing bend at a small curvature radius of 0.25 mm, b) functioning as conducting wires to light an LED, and c) conforming well to a curved surface;

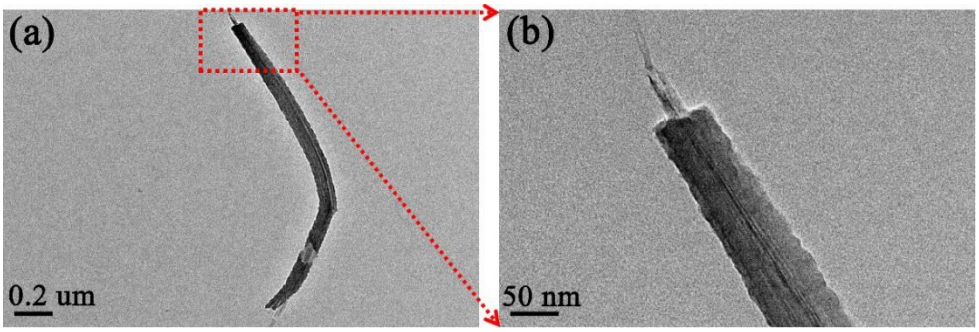

Figure S2. Typical TEM images of single CNTs@PPy fiber scraped off from the fabricated CNTs@PPy film electrode.

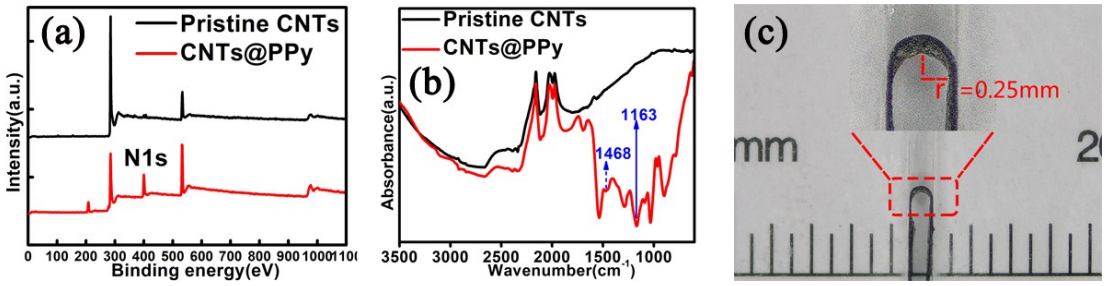

Figure S3. a) XPS survey spectrum and b) FTIR spectra of the pristine CNTs film and CNTs/PPy film; c) Optical image of the flexible and free-standing CNTs/PPy film undergoing bend at a small curvature radius of 0.25 mm;

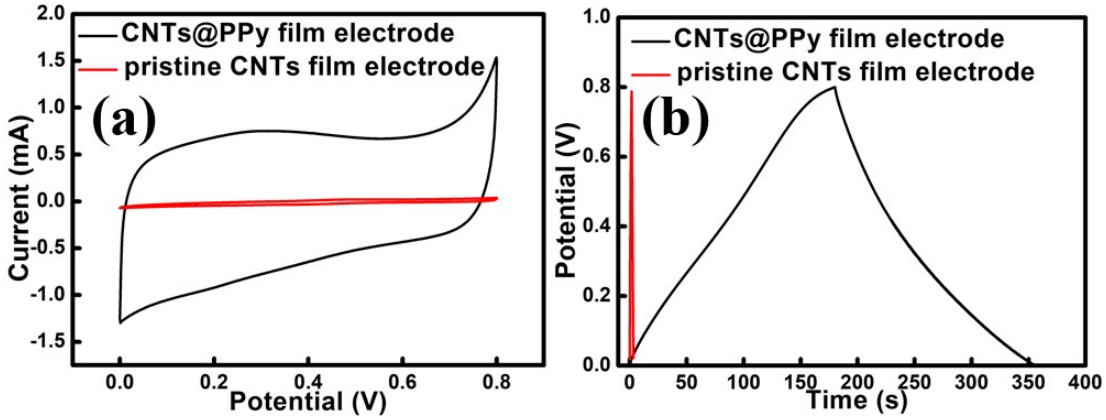

Figure S4. a) CV and b) GCD curves of pristine CNTs film electrode and the fabricated CNTs@PPy film electrode.

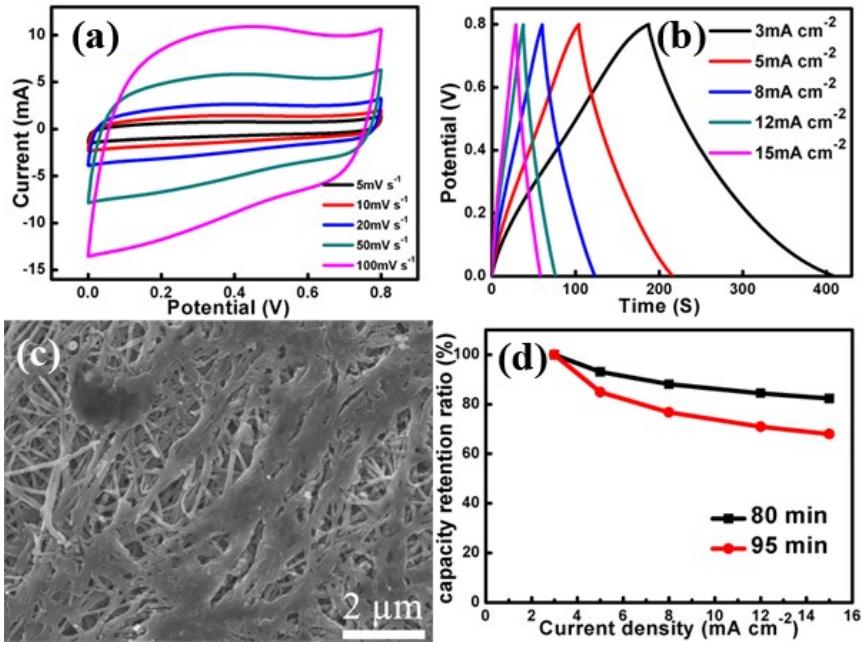

Figure S5. a) CV and b) GCD curves of the fabricated CNTs@PPy film electrode with an electrodeposition time of 95 min; c) The corresponding SEM image; d) Capacitance retention of the CNTs@PPy film electrodes based on different electrodeposition times as a function of current densities.

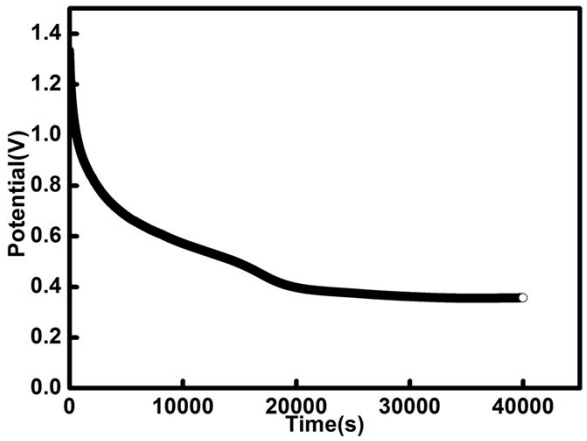

Figure S6. The time courses of the open-circuit voltage of the hybrid MSC assembled from interdigital MXene negative electrode and CNTs@PPy positive electrode

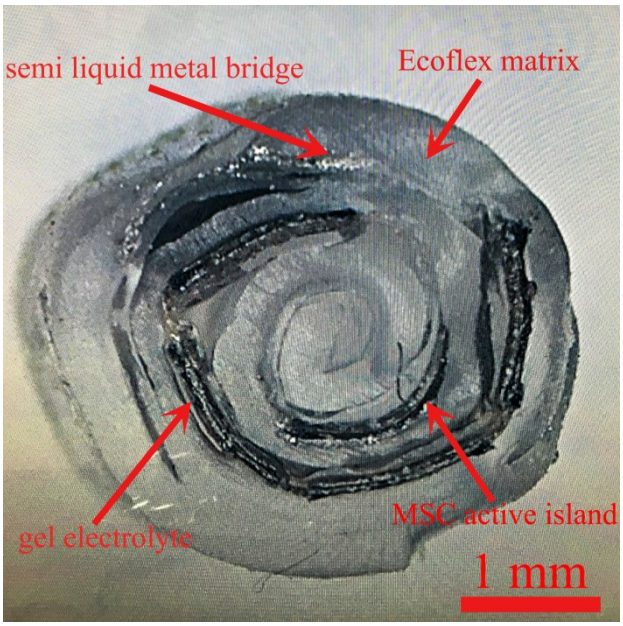

Figure S7. A cross-sectional optical microscope image of the WSS-MSCA.

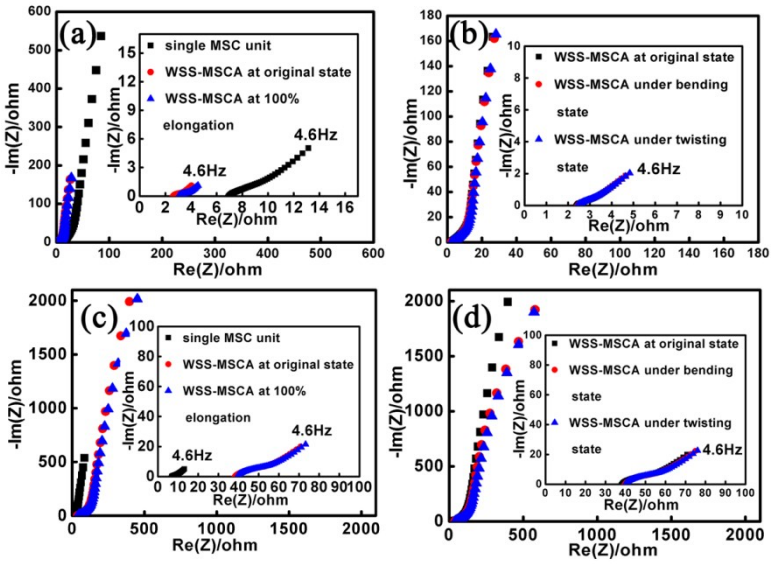

Figure S8. Real-time recorded Nyquist impedance curves (inset is a magnification of the high-frequency band) of the WSS-MSCA consisting of four hybrid MSC units, connected in (a, b) series and (c, d) parallel, stretched from 0% to 100% elongation, as well as bent to 180° and twisted by 360°.

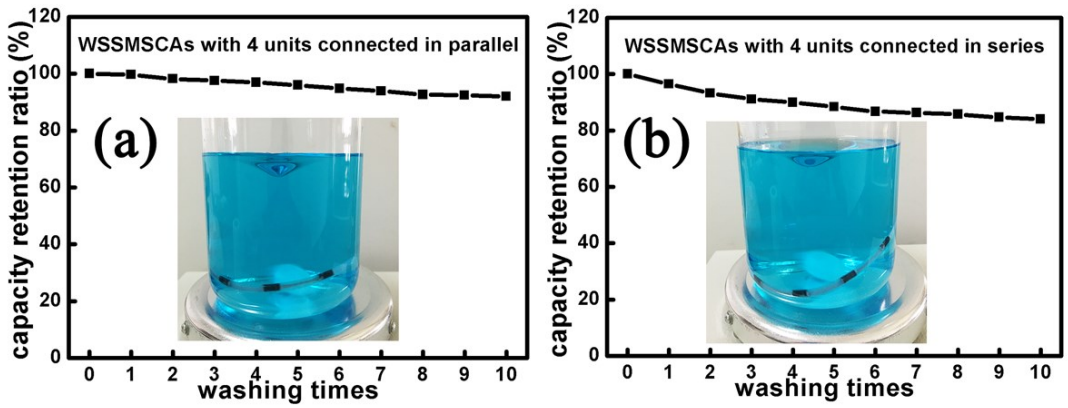

Figure S9. Electrochemical performance of the WSS-MSCA in wash tests.