Supporting Information

Dual design of electrode and electrolyte ensures flexible symmetric micro-supercapacitors with high energy density

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Experimental Section

The synthesis of MXene nanosheets

Typically, the etching solution was prepared by adding 2.0 g LiF to 20 mL HCl (6 mol/L). Next, 1.0 g Ti_3AlC_2 MAX powder was added to the above etching solution. The mixed solution was stirred at 40 °C for 48 hours. The centrifuged product was then washed with deionised (DI) water to pH=7. After then, a multilayer $Ti_3C_2T_x$ powder was obtained after drying. 0.3 g multilayer $Ti_3C_2T_x$ powder was added into 100 mL DI water. After ultrasonic treatment for 0.5 h at 4 °C, the unstripped multilayer $Ti_3C_2T_x$ were removed via centrifugation (4000 rpm/5 minutes). Finally, a colloidal solution containing 2D MXene nanosheets (2 mg/mL) was obtained.

The preparation of ZV-AgNP-MXene film electrodes

The ZV-AgNP-MXene film was prepared using an efficient self-reducing process of silver nitrate ($AgNO_3$) in MXene colloidal solution. Firstly, $AgNO_3$ (15 mg) was dissolved into 10 mL DI water. Then, 20 mL of MXene colloidal solution (2 mg/mL) was added to the above solution and stirred for 30 minutes. The ZV-AgNP-MXene film was then obtained by filtering the above mixed solution, which was further peeled off from the filter membrane and dried for 24 hours. The Ag content of ZV-Ag-NP-MXene film is about 19% (MXene: Ag nanoparticle=40 mg: 9.5 mg). In addition, pure MXene films and highly loaded ZV-AgNP-MXene (HD-Ag-NP-MXene: ~28% Ag content) films were also prepared using the same procedure.

The preparation of PAM/1M Na_2SO_4 hydrogel electrolytes

3.0 g acrylamide (AM) was first dissolved in 20 mL Na_2SO_4 (1 mol/L) solution under stirring. After being deoxygenated for 30 minutes with nitrogen gas, 0.01 g initiator ammonium persulfate was added and stirred rapidly for 10 s. Finally, after heating at 80°C for 2 h, polyacrylamide (PAM)/Na_2SO_4 gel electrolyte was obtained.

The assembly of flexible AS-MSCs devices

Firstly, the interdigital electrodes were fabricated via laser-engraving the above obtained MXene and ZV-AgNP-MXene film electrodes. Then, the obtained MXene and ZV-AgNP-MXene interdigital electrodes were taped with double faced adhesive onto a silicone elastomer substrate (Ecoflex 00-30). After coating the obtained PAM/Na_2SO_4 hydrogel on the electrodes as the top solid electrolyte for both the ionic conductor, the AS-MSCs can be obtained. After that, the AS-MSC device units are interconnected by copper foils and further encapsulated with semi-cured silicone resin (Ecoflex 00-30, two-component mixture at the ratio of 1:1 by weight). After further curing at 45 °C for 1 h, the packaged flexible AS-MSCs devices are finally fabricated.

Electrochemical measurements.

Cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) measurements were conducted using an electrochemical workstation (CHI 660E, Chenhua). The areal capacitance (Cs, mF cm^{-2}) and energy density (Ws, μWh cm^{-2}) were calculated based on the GCD curves according to the following equations:

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

$$Cs = C/S = I\Delta t/S\Delta E \quad (2)$$

$$W\mathrm{s} = 0.5C(\Delta E)^2/3600s \quad (3)$$

Where C, Q, I, Δt represents the total capacitance, total charge, discharge current, and discharge time, respectively. ΔE is the potential window during the discharge process after IR drop, and S is the total area of the film electrodes.

Material Characterizations.

The microstructure and phase composition of the film samples were characterized by field-emission scanning electron microscopy (FE-SEM, S-8200, Hitachi, Japan), transmission electron microscopy (TEM, JEM-2100, JEOL, Japan), X-ray diffractometer (XRD, Bruker D8-ADVANCE), in-situ Raman spectroscopy (RXNI-785, RAMAN RXN SYSTEMS, Kaiser), and X-ray photoelectron spectroscopy (XPS, Mg K_{α} achromatic X-ray source).

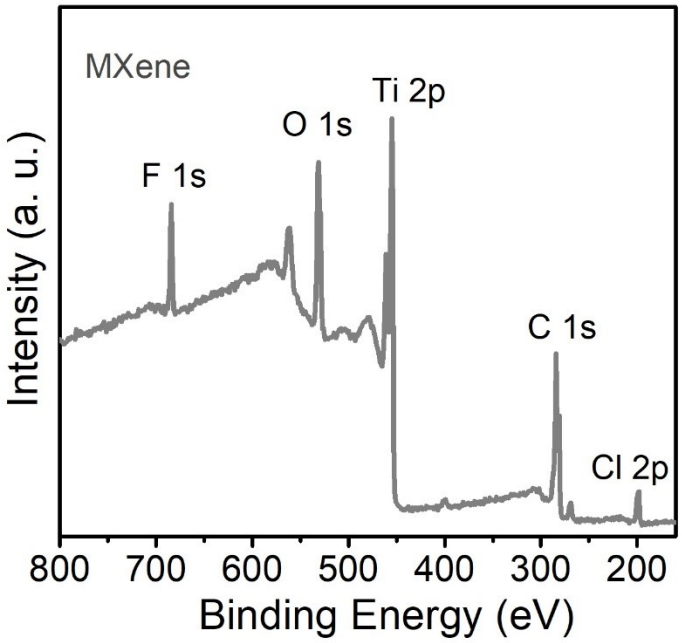

Fig. S1 XPS patterns of MXene film.

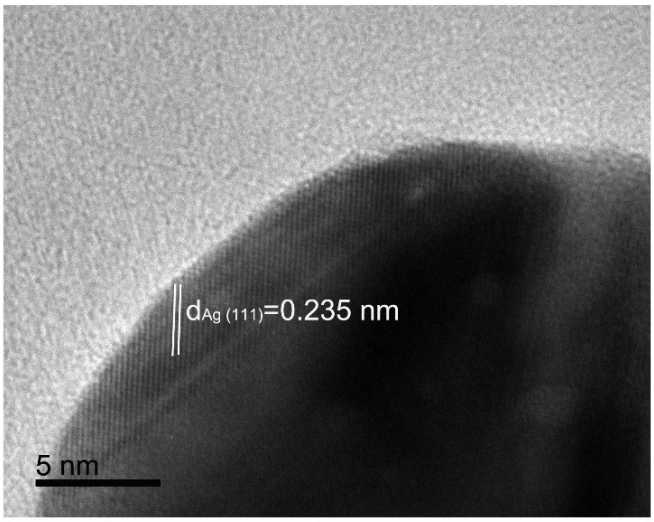

Fig. S2 HRTEM images of ZV-AgNP-MXene nanosheets.

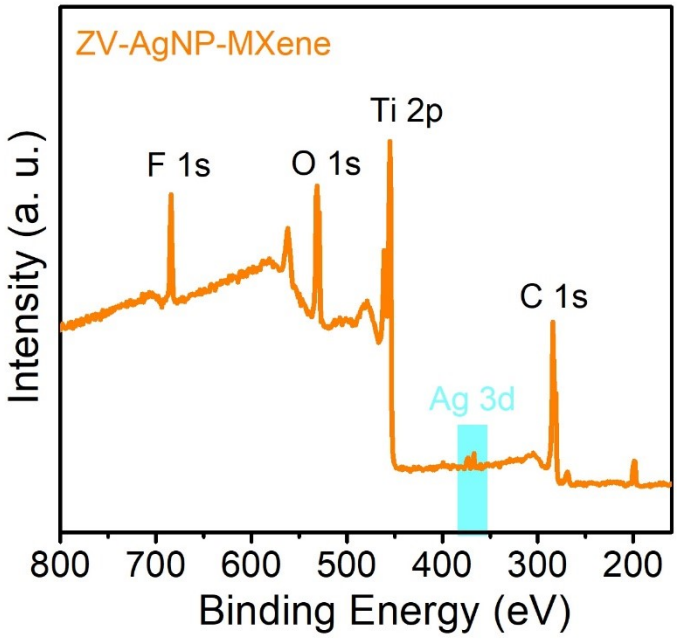

Fig. S3 XPS patterns of ZV-AgNP-MXene film.

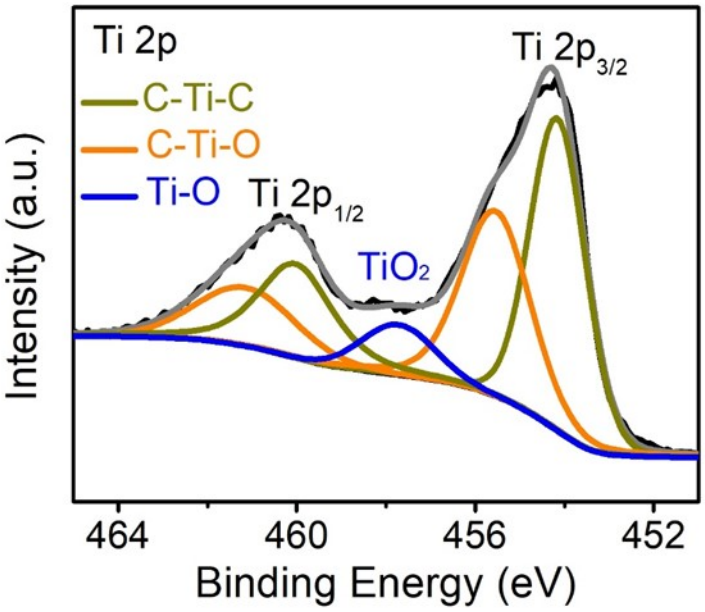

Fig. S4 High-resolution XPS spectra of Ti 2p of ZV-AgNP-MXene film.

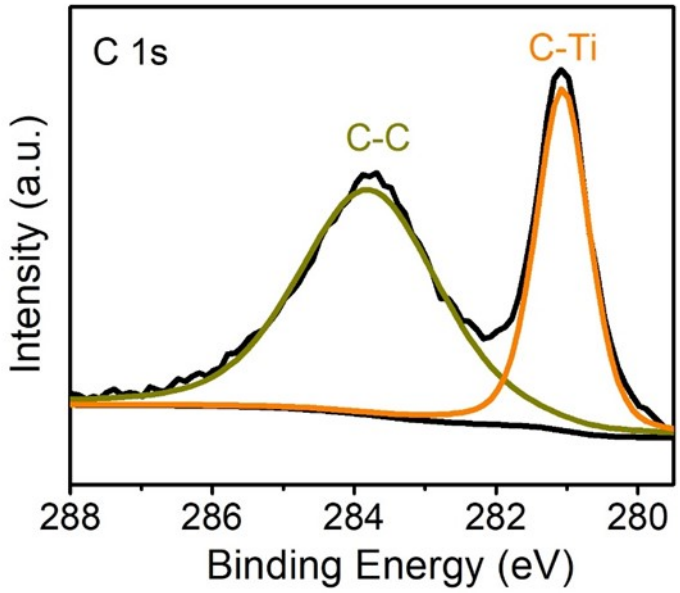

Fig. S5 High-resolution XPS spectra of C 1s of ZV-AgNP-MXene film.

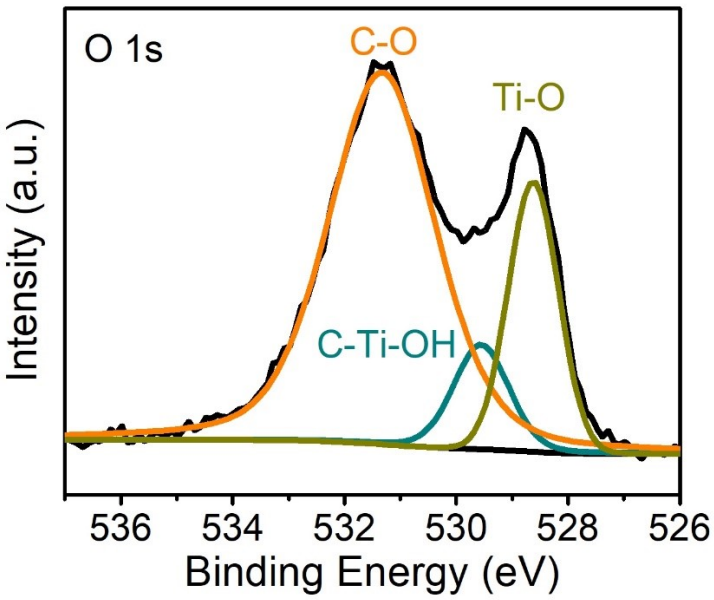

Fig. S6 High-resolution XPS spectra of O 1s of ZV-AgNP-MXene film.

Fig. S7 Photograph of the colloidal solution containing ZV-AgNP-MXene nanoflakes.

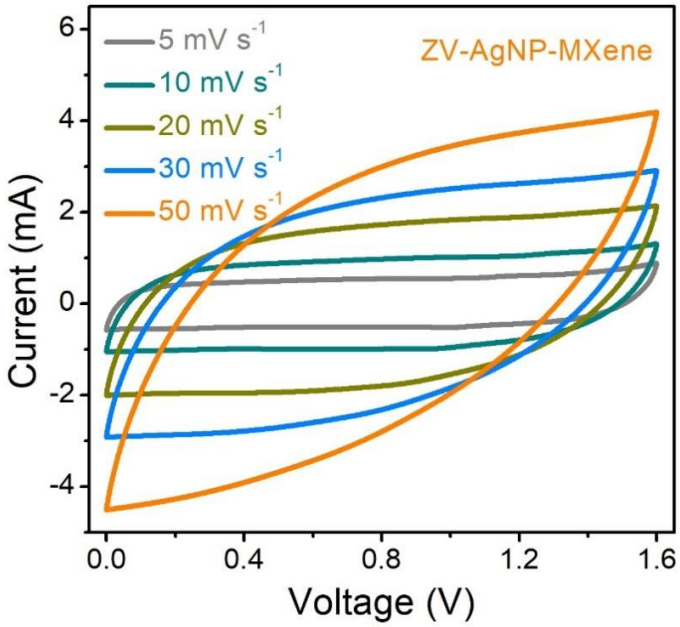

Fig. S8 CV curves of the AS-MSCs based on ZV-AgNP-MXene film electrodes at different scanning rate.

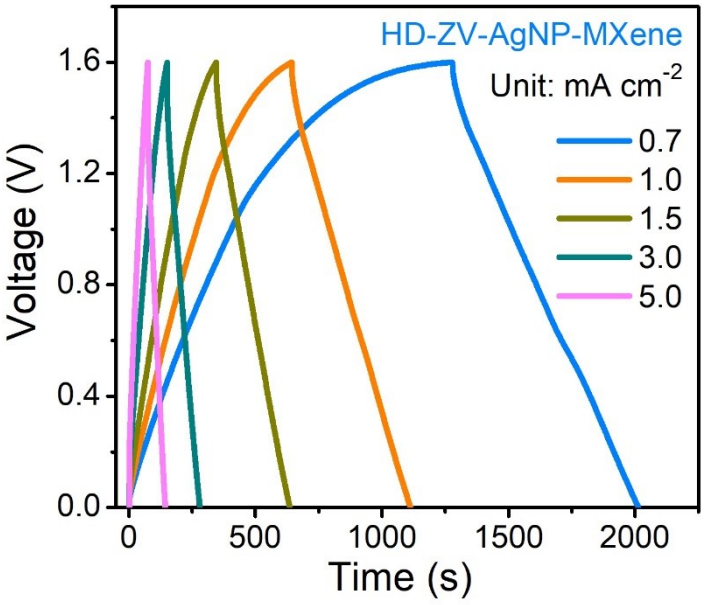

Fig. S9 GCD profiles of the AS-MSCs based on HD-ZV-AgNP-MXene film electrodes at different current densities.

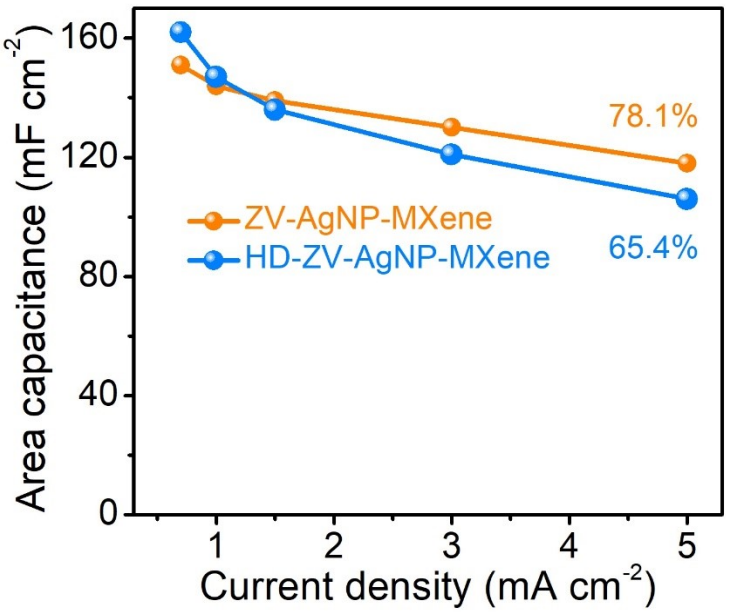

Fig. S10 The areal capacitance of AS-MSCs based on HD-ZV-AgNP-MXene and ZV-AgNP-MXene film electrodes.

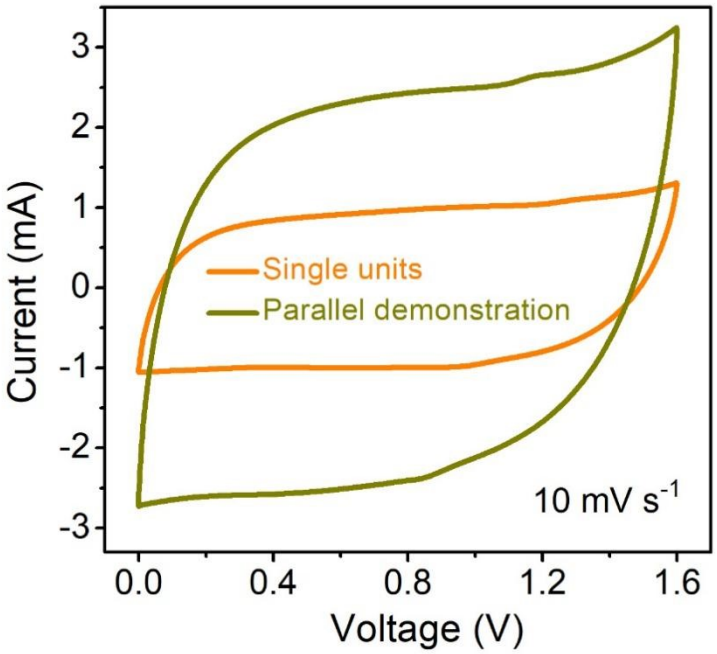

Fig. S11 CV curves of single AS-MSCs unit and parallel-connected AS-MSCs devices.

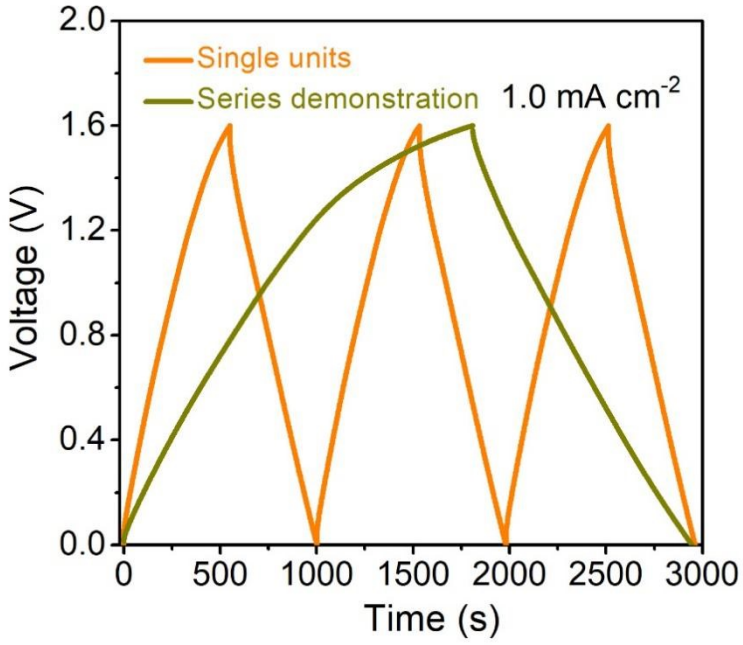

Fig. S12 GCD profiles of single AS-MSCs unit and parallel-connected AS-MSCs devices.

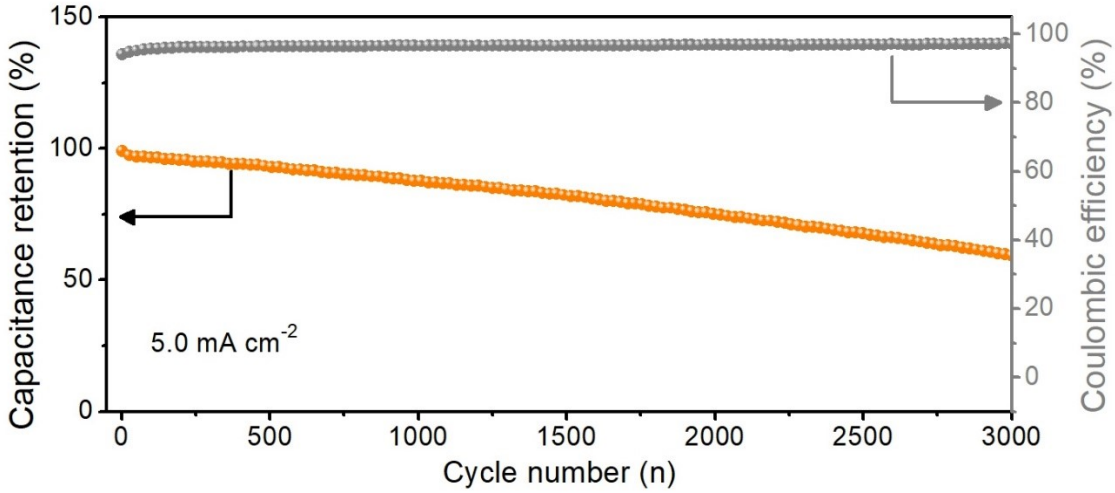

Fig. S13 Cycling stability of series-connected AS-MSCs devices.