

# Ultrahigh Energy and Power Densities of d-MXene-Based Symmetric Supercapacitors

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## Electrochemical Experiment and measurement:

All electrochemical tests were conducted by using an Electrochemical workstation CHI660E by using two types of setup, a three-electrode setup, and two electrodes as shown in Fig. S1a and S1b. In the three-electrode setup, a platinum electrode was used as the counter electrode, both MXene and MX/AuNPs free-standing films are used as a working electrode and Ag/AgCl was the reference electrode. Aqueous 1M and 3 M H<sub>2</sub>SO<sub>4</sub> electrolyte and a cellulose membrane was used as the separators. All the electrodes were pre-cycled for 20 cycles using Cyclic Voltammetry (CV) at 20 mV s<sup>-1</sup> before the actual electrochemical tests were performed. Moreover, the electrodes were dipped in the aqueous solution H<sub>2</sub>SO<sub>4</sub> for 24hrs before testing. The CV tests for different MXene electrodes were performed at scan rates of 5-100 mV s<sup>-1</sup>. Galvanostatic charge-discharge (GCD) cycling was performed at different current densities. To check the cyclic stability and capacitance retention CV cycle was performed at 50 and 100 mVs<sup>-1</sup> for 5000cycles. The specific capacitance was calculated from the CV curves in the three-electrode setup can be calculated using the following formula.

$$C_s = \frac{\int I dV}{mv\Delta V} \quad (S1)$$

Where,  $\Delta V$  (V) is the potential window,  $m$  (g) is the mass of the working electrode,  $V$  (V) is the potential versus reference electrode,  $I$  (A) is the response current,  $v$  (V s<sup>-1</sup>) is the scan rate,  $C_s$  (F g<sup>-1</sup>) is specific capacitance.

In the two-electrode cell,  $C_s$  values of the electrodes in the symmetric supercapacitor can be obtained by the following equation using the CV curves.

$$C_s = \frac{\int I dV}{Mv\Delta V} \quad (S2)$$

where  $M$  (g) is the mass of both electrodes and  $v$  (V s<sup>-1</sup>) is the scan rate. The volumetric capacitance of film electrodes can be calculated according to Eqs. 3 and 4:

$$C_v = \rho C_s \quad (S3)$$

$$\rho = \frac{m}{sd} \quad (S4)$$

where  $S$  (cm<sup>2</sup>) and  $d$  (cm) are the surface area and thickness of film electrode, respectively.

The Energy density (Wh kg<sup>-1</sup>) and power density (W Kg<sup>-1</sup>) can be calculated by using following equations.

$$E = \frac{0.5 \times C_s \times (\Delta V)^2}{3.6} \quad (S5)$$

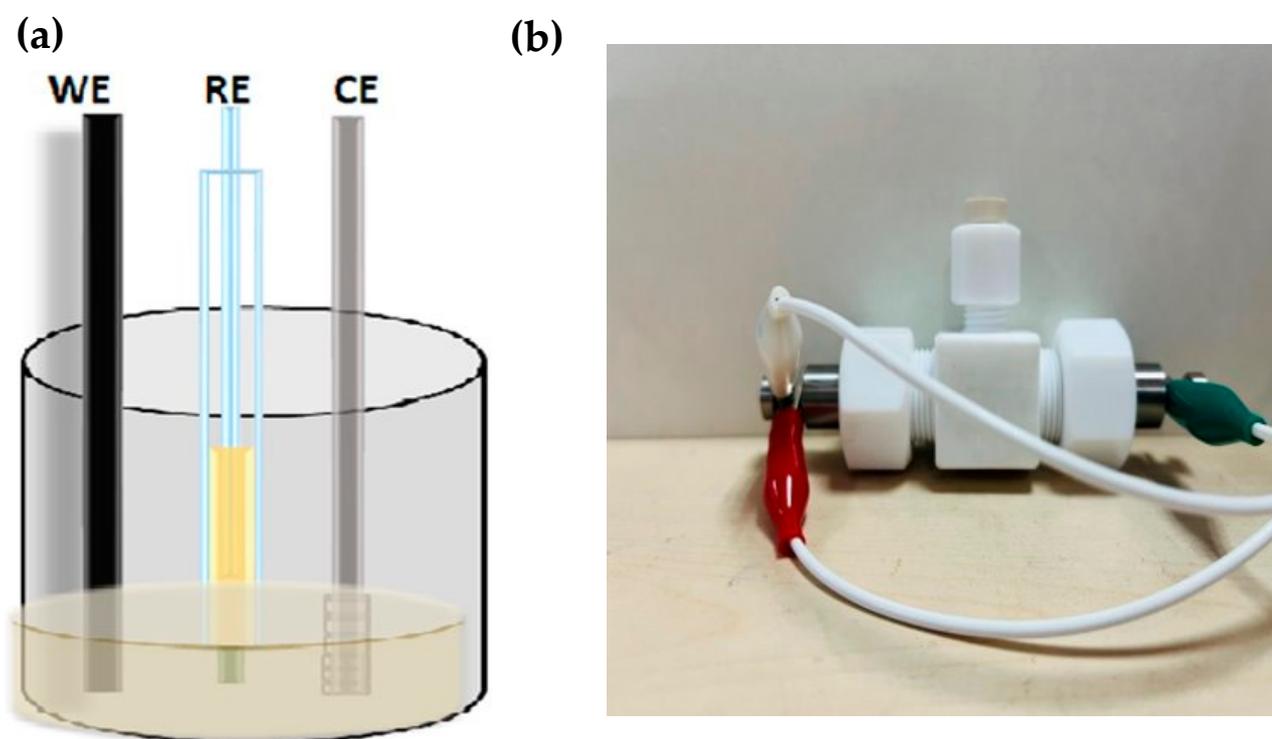
$$p = \frac{E \times 3600}{\Delta t} \quad (S6)$$

where  $\Delta t$  is the discharge time according to the CV curves ( $\Delta t = \frac{\Delta V}{v}$ )

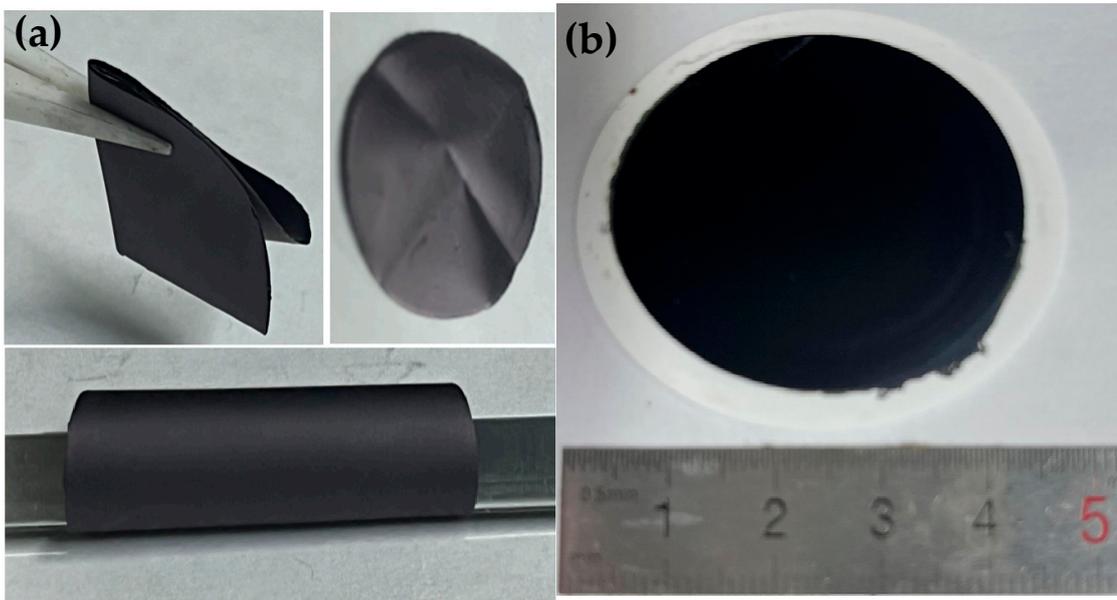
The coulombic efficiency was calculated from GCD curves according to the following equation:

$$C_E = \frac{\Delta t_d}{\Delta t_c} \quad (S7)$$

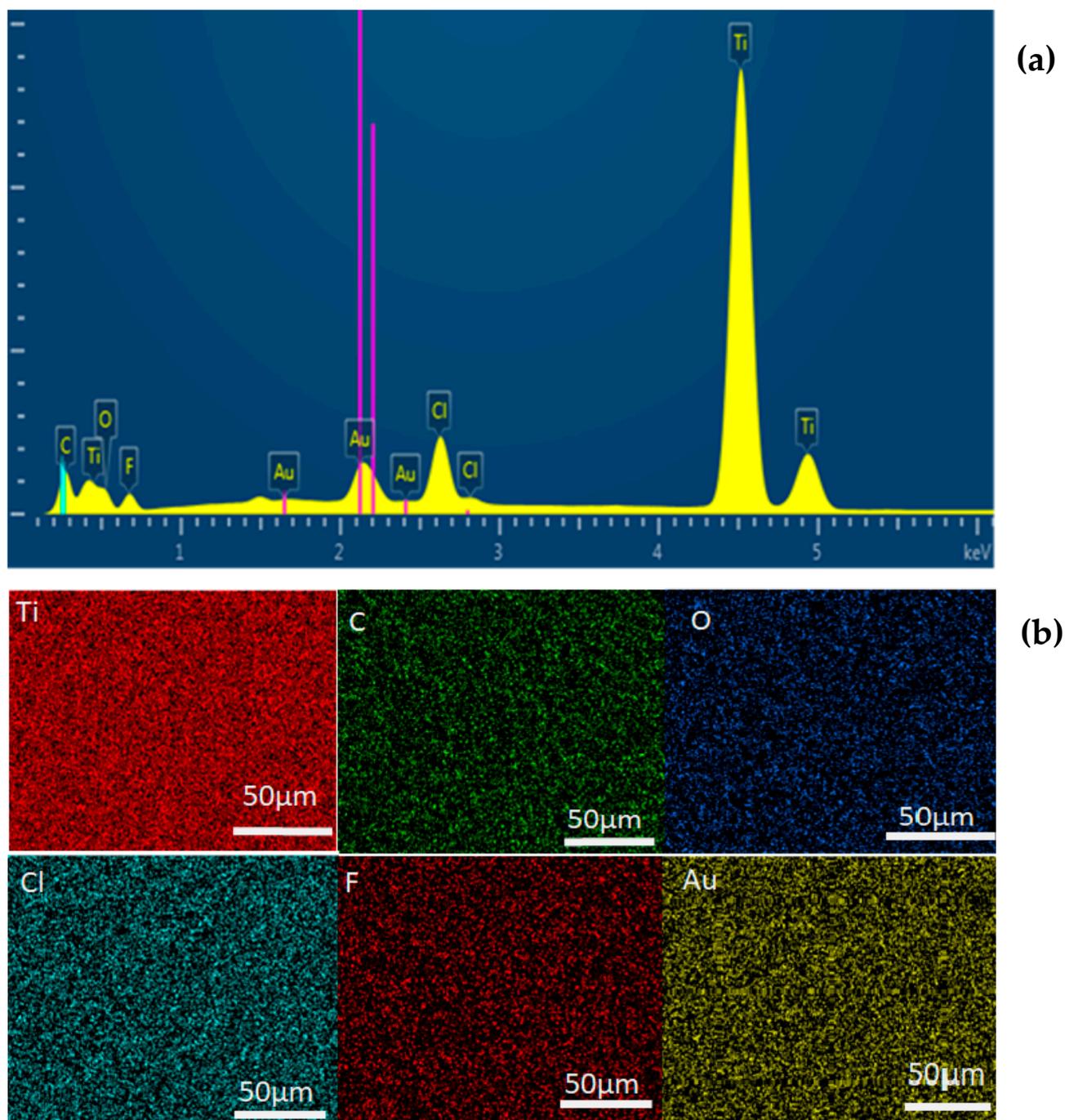
Where,  $\Delta t_d$  and  $\Delta t_c$  are the time of discharging and charging, respectively, and CE is coulombic efficiency.



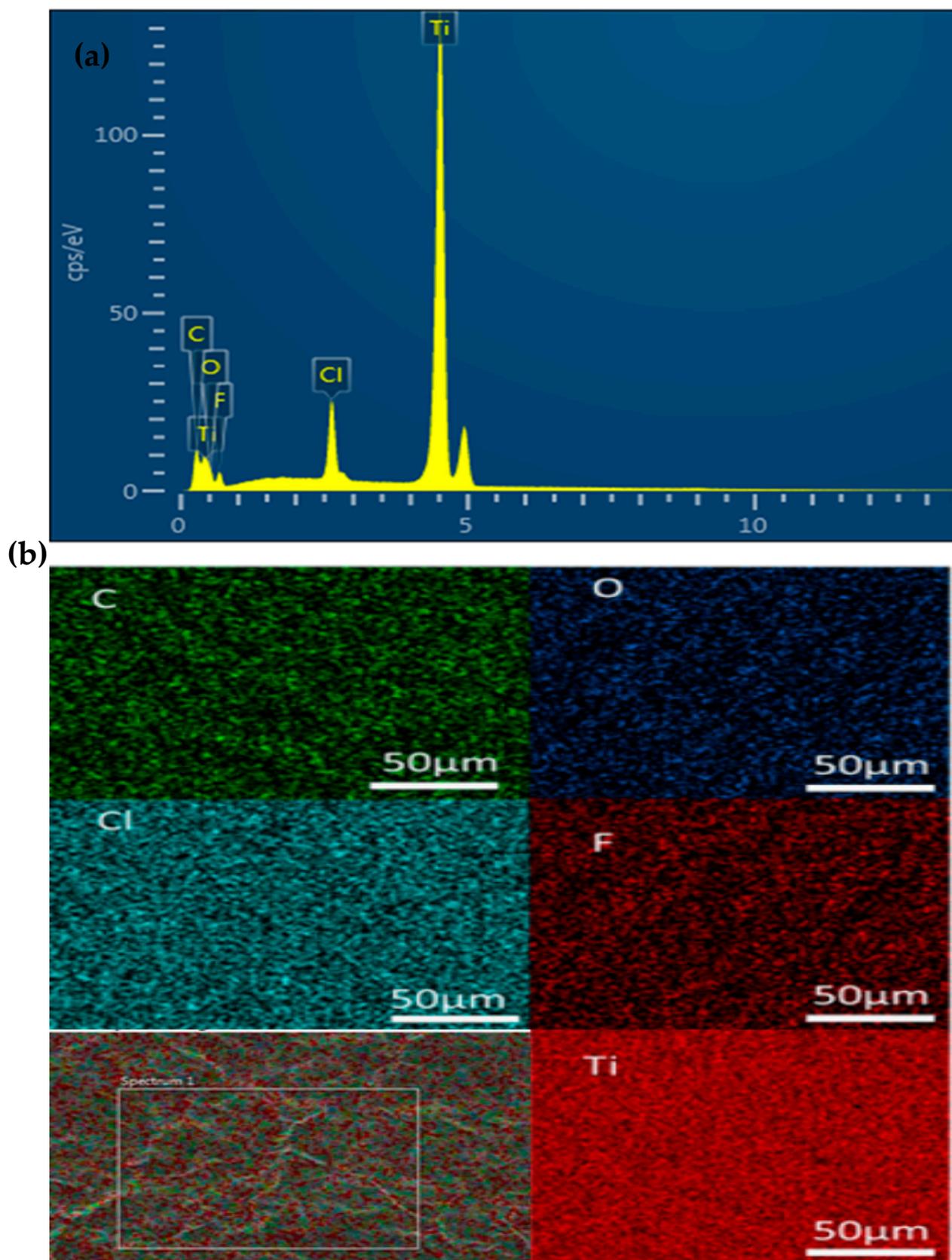
**Figure S1.** (a) Illustration of three-electrode setup; (b) Digital photo of two setup using Swagelok cell.



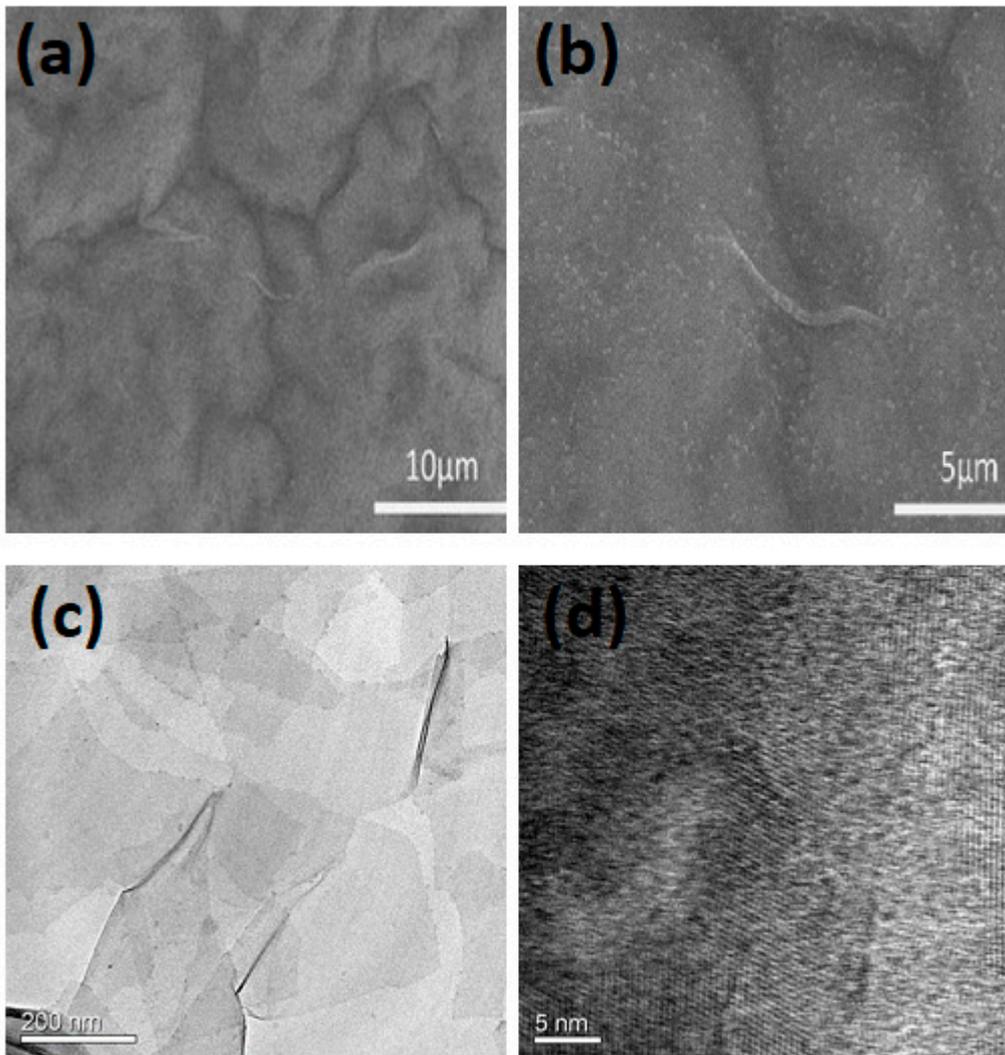
**Figure S2.** (a) the digital photograph of the free-standing film, showing excellent flexibility at different deformation statuses, which can be bent, rolled, and even folded; (b) digital photo along the scale.



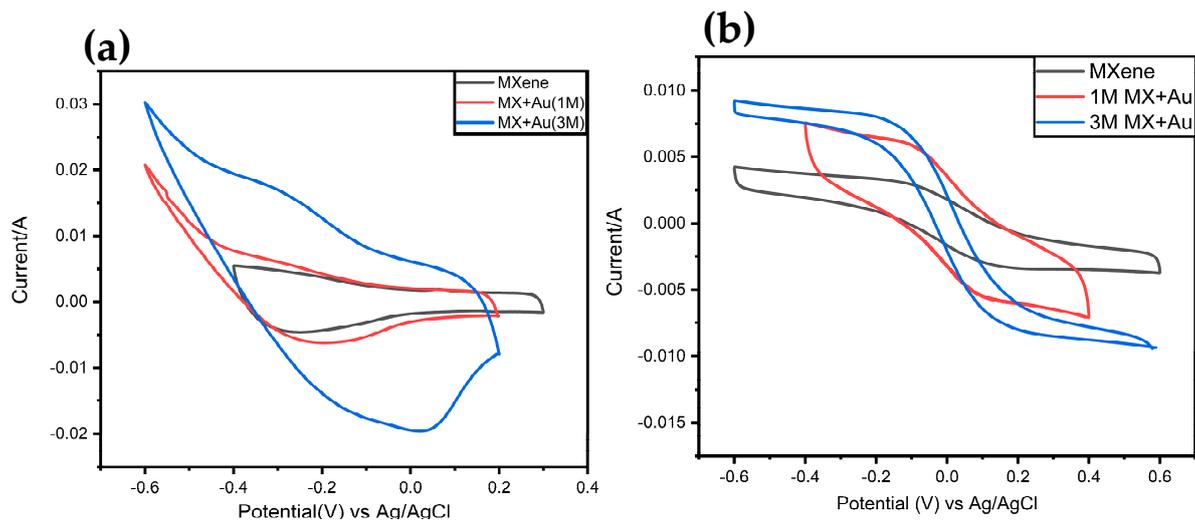
**Figure S3.** (a) EDS spectrum of MX/AuNP film; (b) The elemental mapping of MX/Au free-standing film.



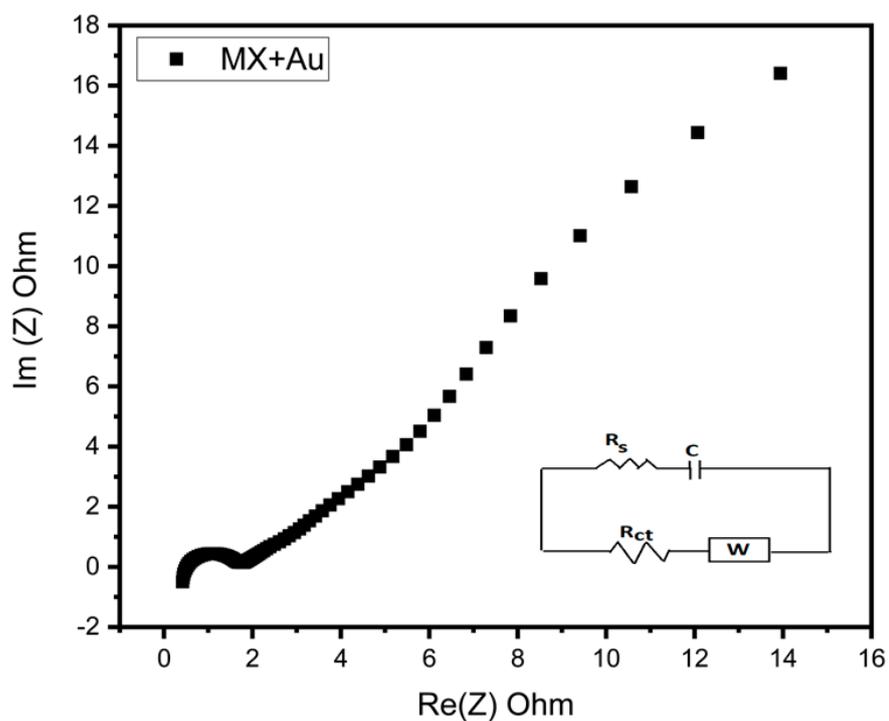
**Figure S4.** (a) EDS spectrum of MXene film; (b) The elemental mapping of MXene free-standing film.



**Figure S5.** (a, b) SEM images of Mxene films; (c)TEM image of Mxene film (d) HRTEM image of Mxene film.



**Figure S6.** (a) CV curves of MX and MX/AuNPs in three electrodes setup at 5mVs<sup>-1</sup>; (b) CV curves Symmetric SCs of MXene and MX/AuNPs at 5mVs<sup>-1</sup>.



**Figure S7.** Nyquist plots of symmetric SC of MX/AuNPs, Inset shows the equivalent circuit model.

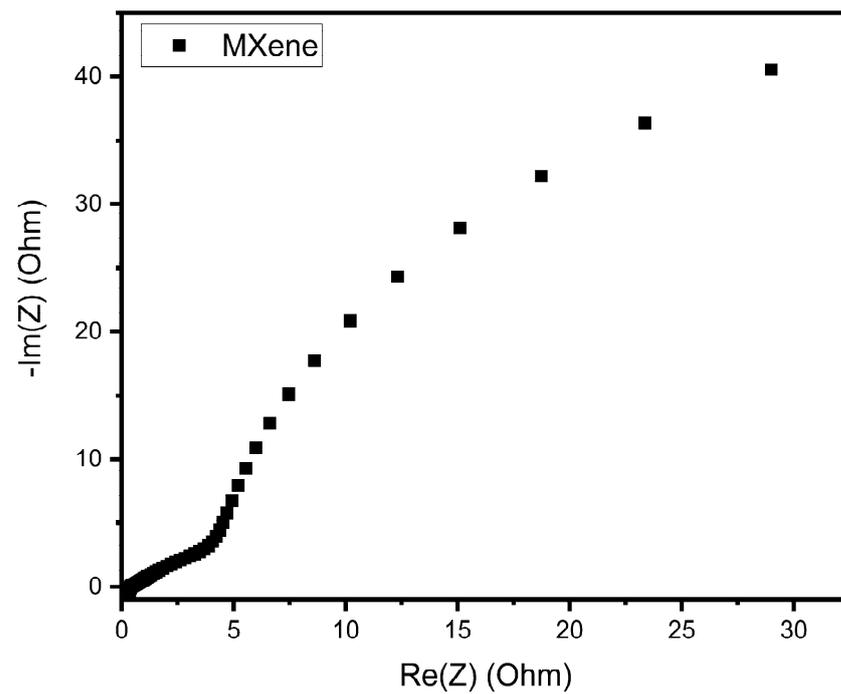


Figure S8. Nyquist plots of symmetric SC of Mxene.

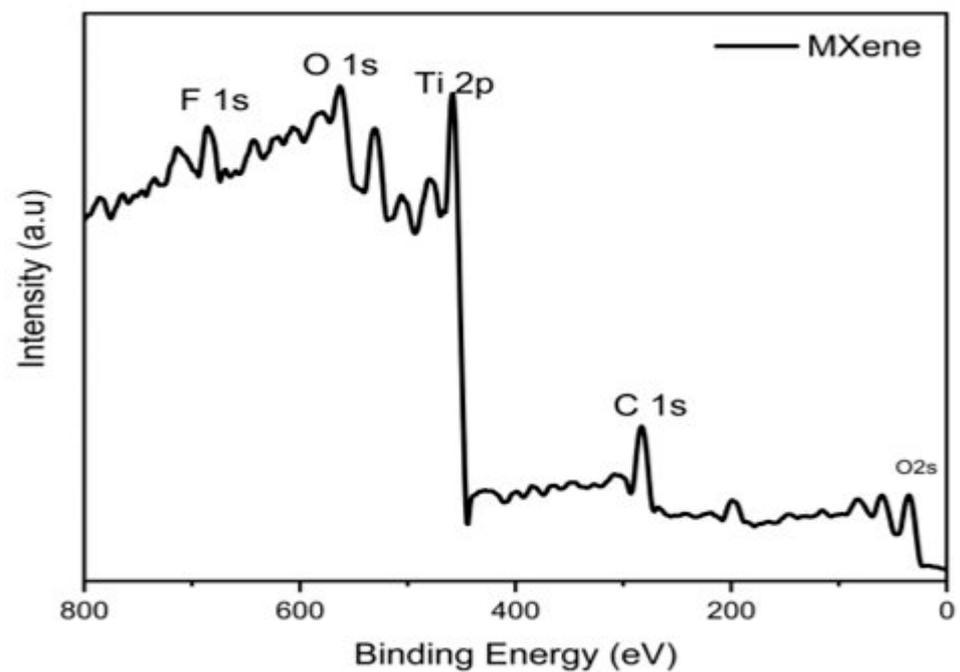
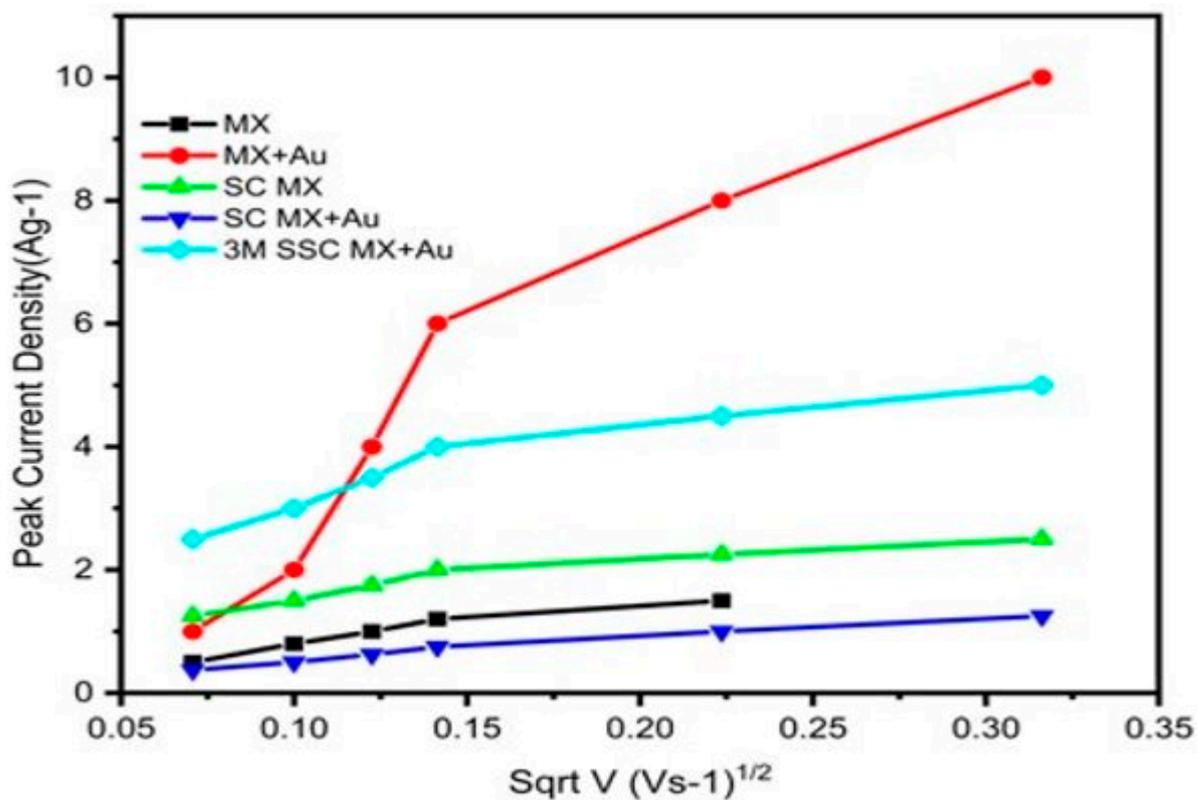
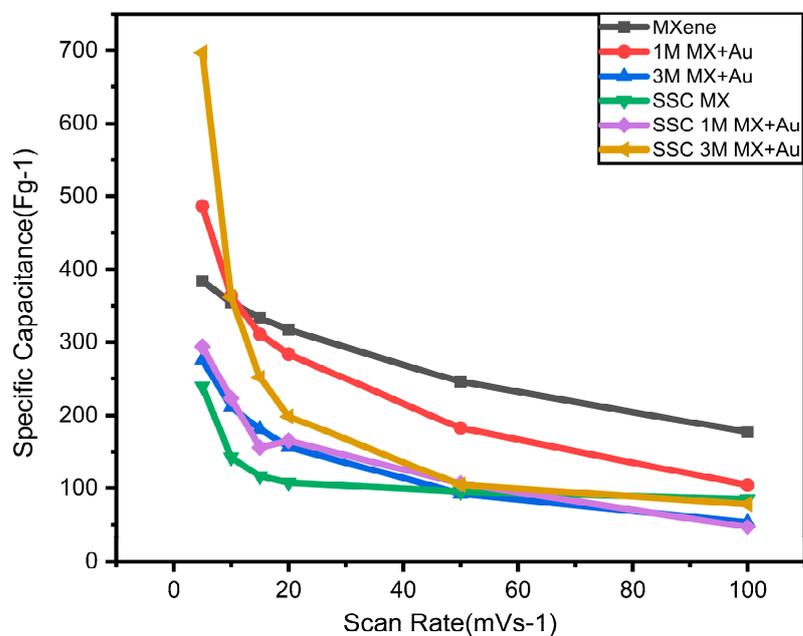


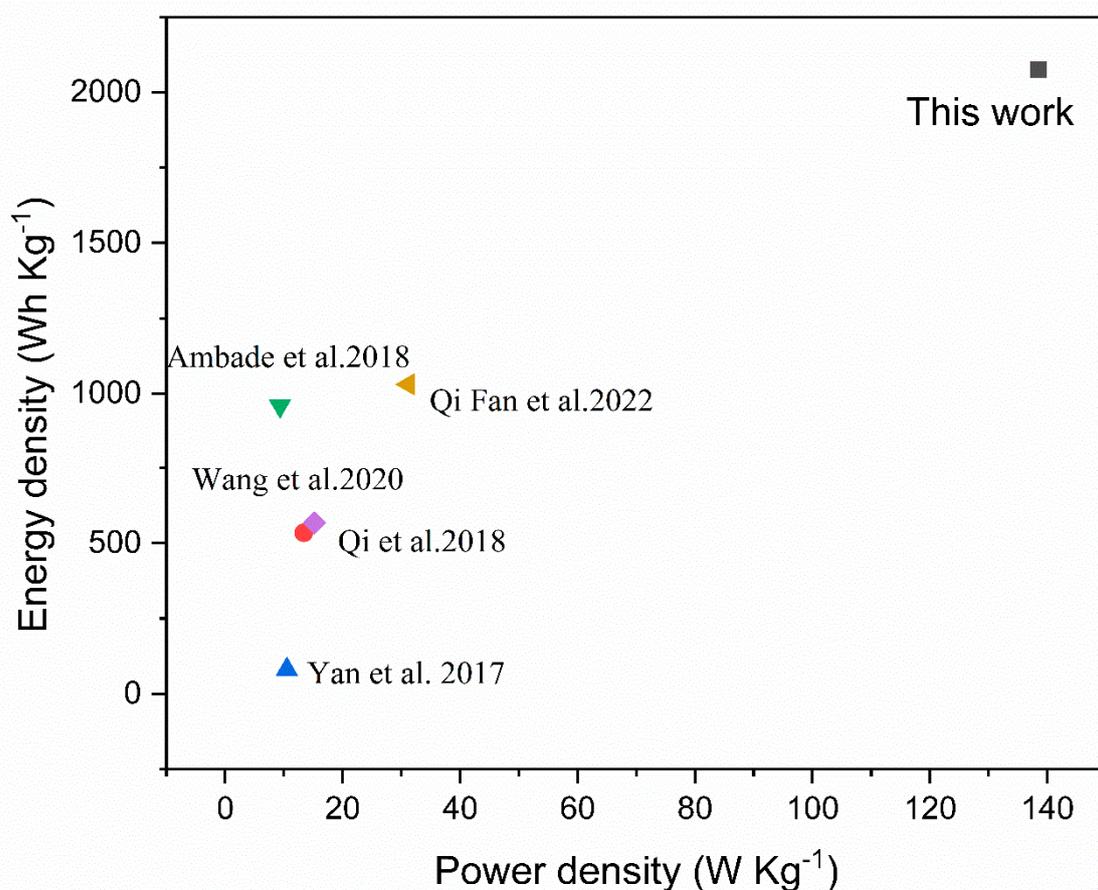
Figure S9. The survey spectra of Mxene film.



**Figure S10.** Graph between the peak current densities of all the tested electrodes Vs square root of Scan Rate.



**Figure S11.** Specific capacitance of MXene and MX/AuNPs electrode and Symmetric SCs vs. the scan rate.



**Figure S12.** Ragone plots displaying energy and power densities of MX/AuNPs symmetric supercapacitor in comparison with other MXene-based supercapacitors.

**Table S1.** EDS results of the Mxene and MX/AuNPs.

Element	MXene		MX/AuNps	
	Atomic %	Weight %	Atomic %	Weight %
Ti	36.18	62.61	32.92	32.92
C	26.04	11.30	28.42	28.42
O	19.77	11.43	18.36	18.36
F	14.16	9.72	15.84	15.84
Cl	3.86	4.94	3.48	3.48
Au	-	-	0.98	6.80

**Table S2.** Comparison of electrochemical performance of different MXene-based materials as a symmetric supercapacitor.

Electrode material	Electrolyte	Specific Capacitance (Fg <sup>-1</sup> )	Energy Density (Wh Kg <sup>-1</sup> )	Power Density (W kg <sup>-1</sup> )	Ref.
MX/AuNP	3M H <sub>2</sub> SO <sub>4</sub>	696.67	138.44	2076	This work
M/ MoO <sub>3</sub>	1M H <sub>2</sub> SO <sub>4</sub>	545	13.4	534.6	1
M/G-5%	3M H <sub>2</sub> SO <sub>4</sub>	335	10.5	80.3	2

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MXene/WO <sub>3</sub>	0.5M H <sub>2</sub> SO <sub>4</sub>	566	9.32	960	3
TCBOC	1 M KOH	247.8	15.2	567.4	4
PPy-MXene	Ionic Liquid	51.85	31.2	1030.4	5

## References

1. Wang, Y.; Wang, X.; Li, X.; Liu, R.; Bai, Y.; Xiao, H.; Liu, Y.; Yuan, G., Intercalating ultrathin moo<sub>3</sub> nanobelts into mxene film with ultrahigh volumetric capacitance and excellent deformation for high-energy-density devices. *Nano-micro letters* **2020**, *12*, 1-14.
2. Yan, J.; Ren, C.E.; Maleski, K.; Hatter, C.B.; Anasori, B.; Urbankowski, P.; Sarycheva, A.; Gogotsi, Y., Flexible mxene/graphene films for ultrafast supercapacitors with outstanding volumetric capacitance. *Advanced Functional Materials* **2017**, *27*, 1701264.
3. Ambade, S.B.; Ambade, R.B.; Eom, W.; Noh, S.H.; Kim, S.H.; Han, T.H., 2d ti<sub>3</sub>c<sub>2</sub> mxene/wo<sub>3</sub> hybrid architectures for high-rate supercapacitors. *Advanced Materials Interfaces* **2018**, *5*, 1801361.
4. Xia, Q.X.; Shinde, N.M.; Yun, J.M.; Zhang, T.; Mane, R.S.; Mathur, S.; Kim, K.H., Bismuth oxychloride/mxene symmetric supercapacitor with high volumetric energy density. *Electrochimica Acta* **2018**, *271*, 351-360.
5. Fan, Q.; Zhao, R.; Yi, M.; Qi, P.; Chai, C.; Ying, H.; Hao, J., Ti<sub>3</sub>c<sub>2</sub>-mxene composite films functionalized with polypyrrole and ionic liquid-based microemulsion particles for supercapacitor applications. *Chemical Engineering Journal* **2022**, *428*, 131107.