

## Supporting Information

### Fast assembly of MXene hydrogel by interfacial electrostatic interaction for supercapacitors

Mengke Peng,<sup>a,#</sup> Weizu Yang,<sup>a,#</sup> Longbin Li,<sup>a</sup> Kaiyang Zhang,<sup>a</sup> Li Wang,<sup>a</sup> Ting Hu,<sup>b</sup> Kai Yuan,<sup>a,\*</sup> and Yiwang Chen<sup>a,c,\*</sup>

<sup>a</sup>Institute of Polymers and Energy Chemistry (IPEC), College of Chemistry, Nanchang University, Nanchang 330031, China. Email: kai.yuan@ncu.edu.cn (K. Yuan); ywchen@ncu.edu.cn (Y. Chen)

<sup>b</sup>School of Materials Science and Engineering, Nanchang University, Nanchang 330031, China

<sup>c</sup>Institute of Advanced Scientific Research (iASR), Key Laboratory of Functional Small Molecule Ministry of Education, Jiangxi Normal University, 99 Ziyang Avenue, Nanchang 330022, China

<sup>#</sup> These authors contributed equally to this work.

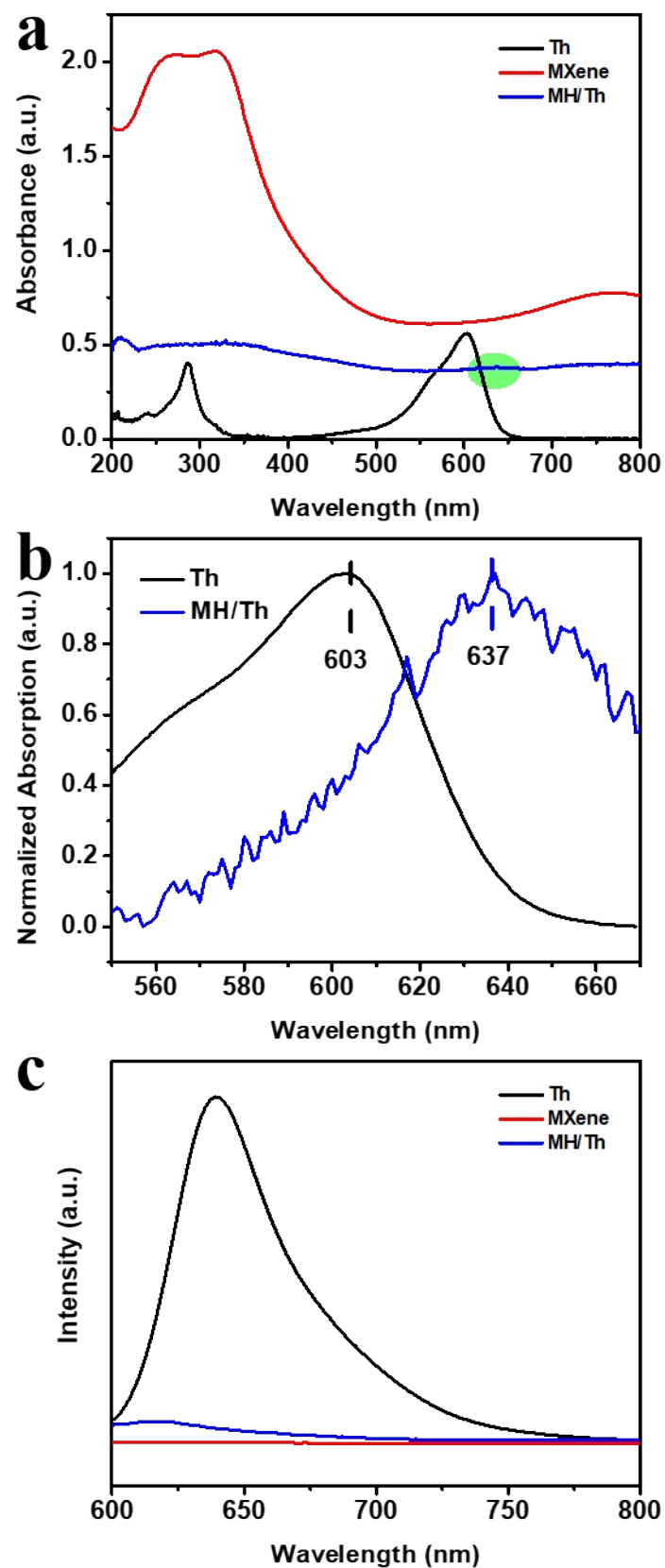
**Characterizations:** UV-vis absorption spectra were obtained by Agilent Cary100. Fluorescence spectrophotometer was performed on a F-7000, Hitachi High-Technologies Corporation. Fourier transform infrared (FTIR) spectroscopy studies were conducted on a SHIMADZU FT-IR Prestige-21. XRD patterns studies were conducted on a Bruker D8 Advance X-ray diffractometer. X-ray photoelectron spectroscopy (XPS) was performed by ESCALAB 250Xi (THERMO SCIENTIFIC Co.), conducting Al K $\alpha$  source as an internal standard. The SEM and TEM were carried out at a Quanta 200F environmental scanning electron microscopy and transmission electron microscopy (TEM, JEOL, JEM-2100F), respectively. The nitrogen adsorption-desorption measurements were performed on a Brunauer-Emmett-Teller (Quantachrome, Autosorb-iQ). The performance of samples was

evaluated by Autolab electrochemical workstation (AutoLab, PGSTAT302N1).

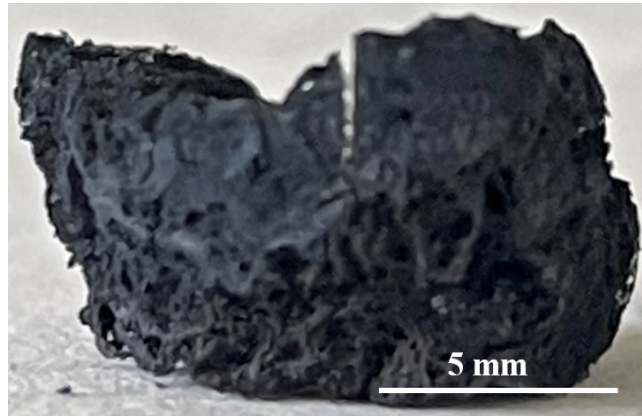
**Preparation of  $\text{Ti}_3\text{C}_2\text{T}_x$  flakes.** MAX phase ( $\text{Ti}_3\text{AlC}_2$ ) was purchased from 11 Technology Co., Ltd. Generally, 2 g LiF and 40 mL of 9 M HCl was mixed in a Teflon beaker to form HF solution. Then, 2 g  $\text{Ti}_3\text{AlC}_2$  power was slowly and carefully added into above solution and constantly stirred at 35°C for 24 hours. After the etching was completed, the resulting suspension was repeatedly washed by centrifugation at 3500 rpm with deionized water until the pH of suspension turned to about 6. The single- or fewer-layer MXene flakes was collected by sonicate the above mixture with a certain amount of water under  $\text{N}_2$  atmosphere for 1 hour and followed by centrifuged at 3500 rpm for 1 hour.

**Preparation of MXene hydrogel.** Typically, MXene hydrogels were assembled by add 1 mL of MXene suspension into 200  $\mu\text{L}$  protonated thionine solutions (2 mg  $\text{mL}^{-1}$ , 3 mg  $\text{mL}^{-1}$ , 5 mg  $\text{mL}^{-1}$ ) with 10  $\mu\text{L}$  hydrochloric acid. The hydrogel was shaped within a few seconds. Then, the MXene hydrogels were immersed into deionized water for several hours to remove excess thionine molecules. The obtained MXene hydrogels were labeled as MH/Th-1, MH/Th-2, MH/Th-3 with 2 mg  $\text{mL}^{-1}$ , 3 mg  $\text{mL}^{-1}$ , 5 mg  $\text{mL}^{-1}$  protonated thionine crosslinking, respectively. MH/Th-2 was used as a representative in UV-vis absorption spectra, Fluorescence spectra, FTIR, XRD, XPS, BET and TEM studies and recorded as MH/Th.

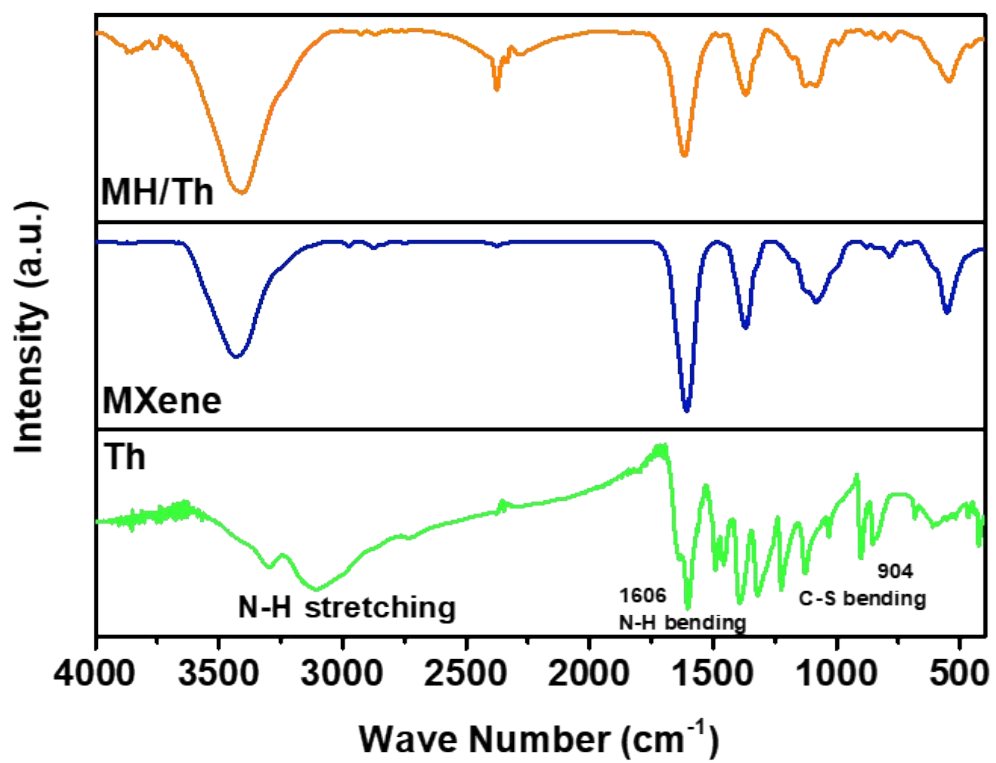
**Electrochemical measurements.** A certain volume of washed MH/Th hydrogel was gently clamped on the working electrode without further processing. The counter electrode was platinum plate and the reference electrode was Ag/AgCl. The potential range was set from -0.2 to 0.4 V and 1 M  $\text{H}_2\text{SO}_4$  solution was used as electrolyte. The electrochemical impedance spectroscopy (EIS) was measured from 0.01 Hz to 100 kHz. The specific capacitance of the materials was evaluated from GCD profiles according to the formula:  $C = (\int i dt) / (m \Delta V)$ . Where  $C$  ( $\text{F g}^{-1}$ ) is the specific capacitance,  $i$  (A) is the discharge current,  $t$  (s) is the discharge time,  $m$  (g) is the mass of the active materials and  $\Delta V$  (V) is potential window.



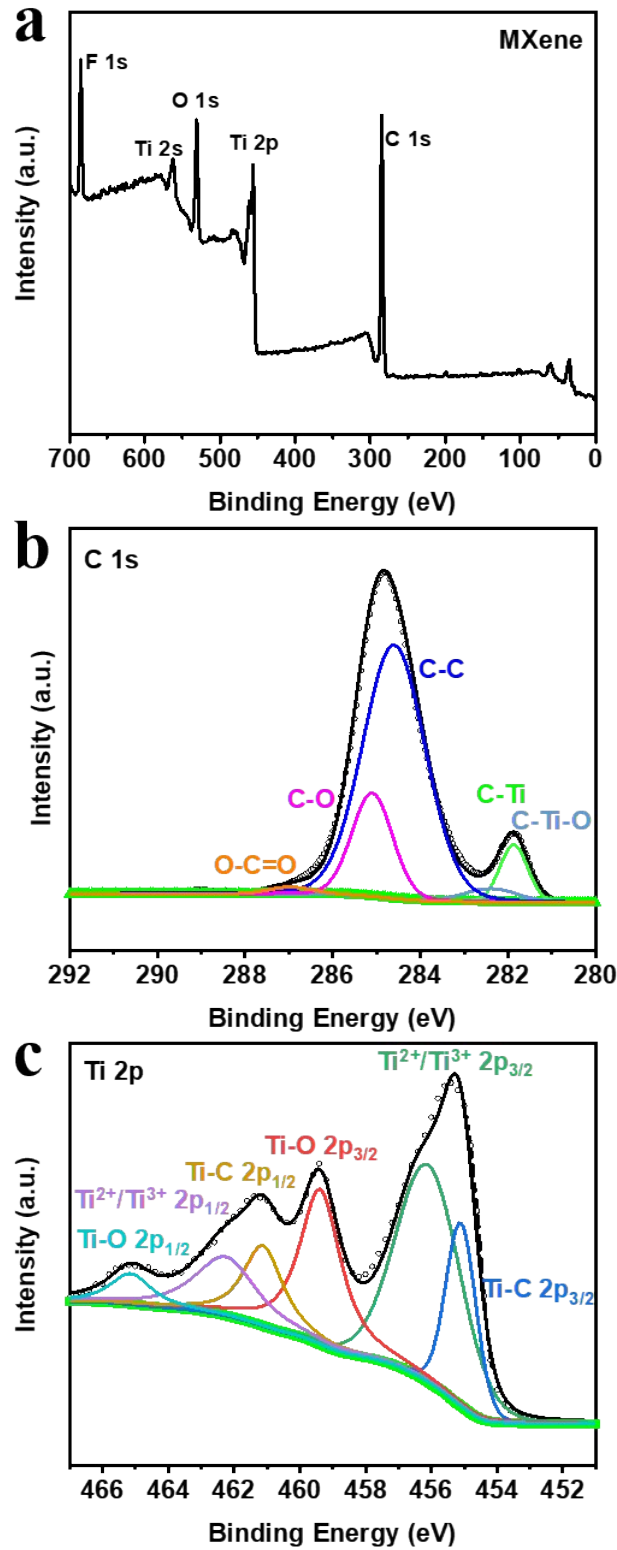
**Figure S1.** a) UV-vis absorption spectra and b) the corresponding enlarged UV-vis spectra of Th, MXene and MH/Th aqueous solutions. c) Fluorescence spectra of Th, MXene and MH/Th aqueous solutions.



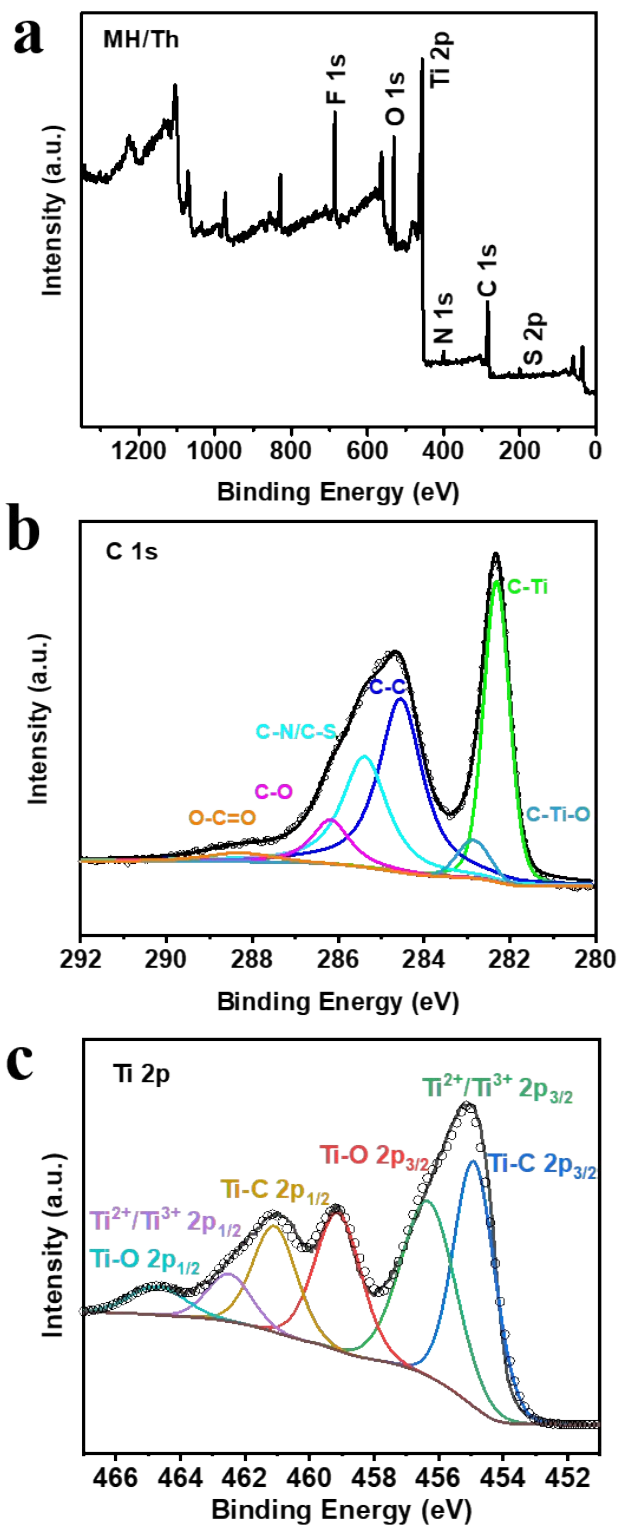
**Figure S2.** Optical image of freeze-drying MH/Th.



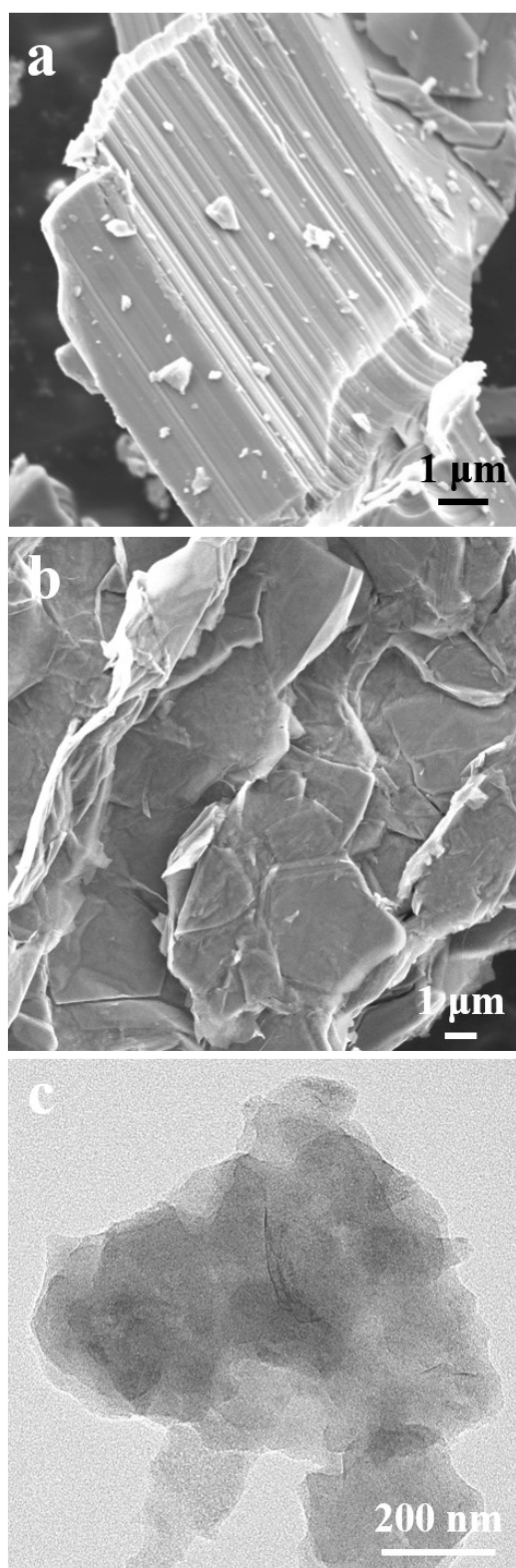
**Figure S3.** FTIR spectra of thionine, MXene, and the MH/Th nanocomposite.



**Figure S4.** a) XPS spectra of MXene and corresponding high-resolution XPS spectra of b) C 1s and c) Ti 2p.

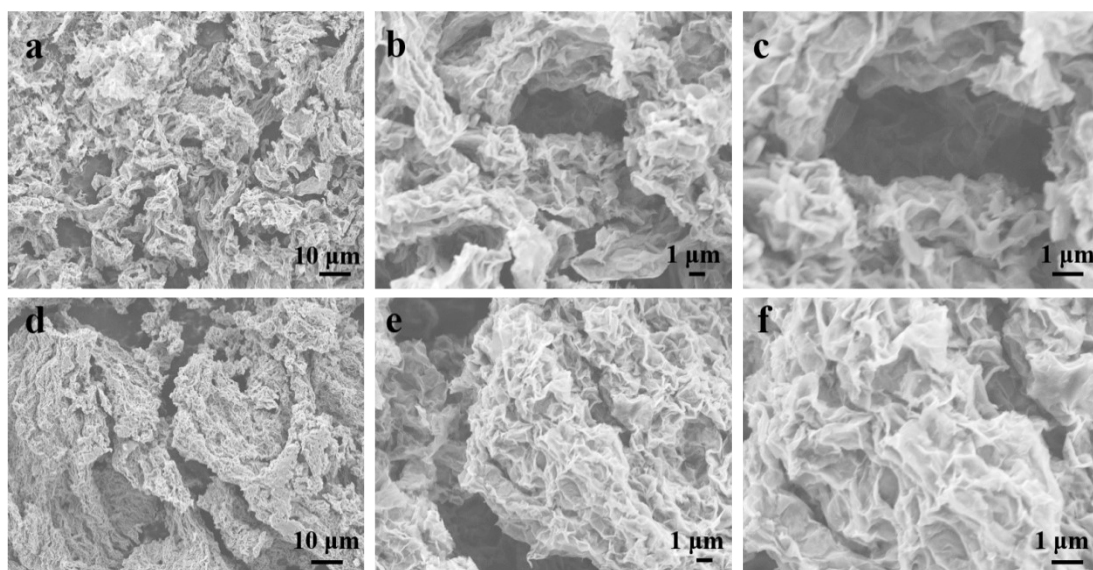


**Figure S5.** a) XPS spectra of MH/Th and corresponding high-resolution XPS spectra of b) C 1s, c) Ti 2p.

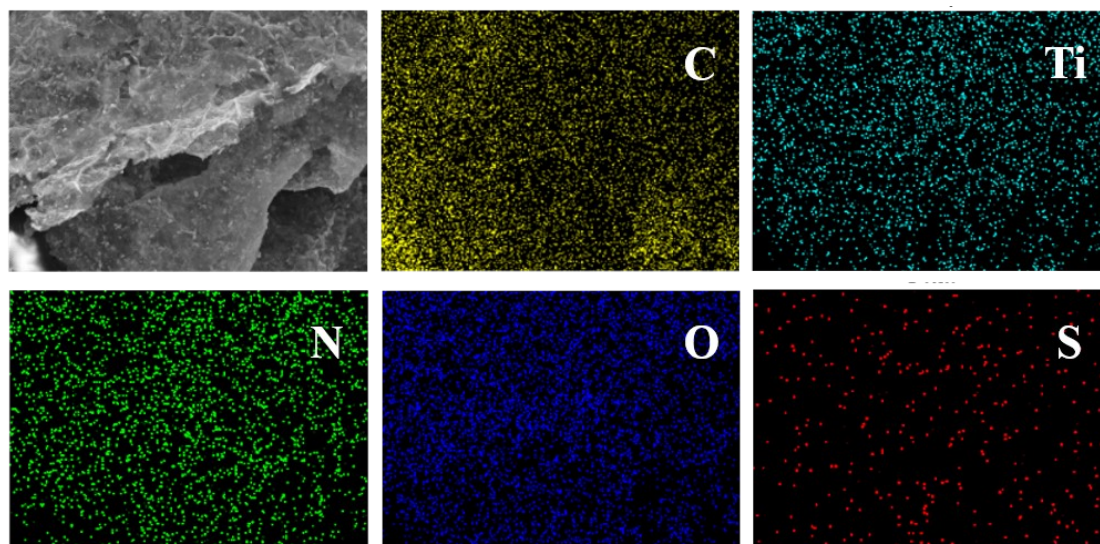


**Figure S6.** SEM images of a)  $\text{Ti}_3\text{AlC}_2$  and b) MXene. c) TEM image of MXene.

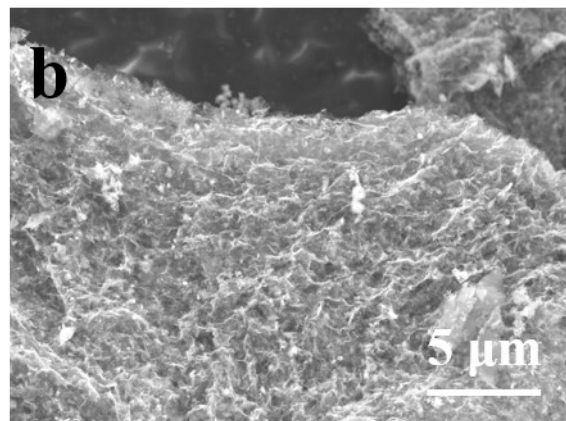
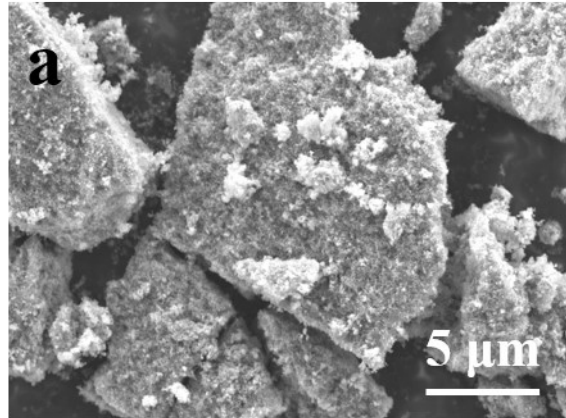




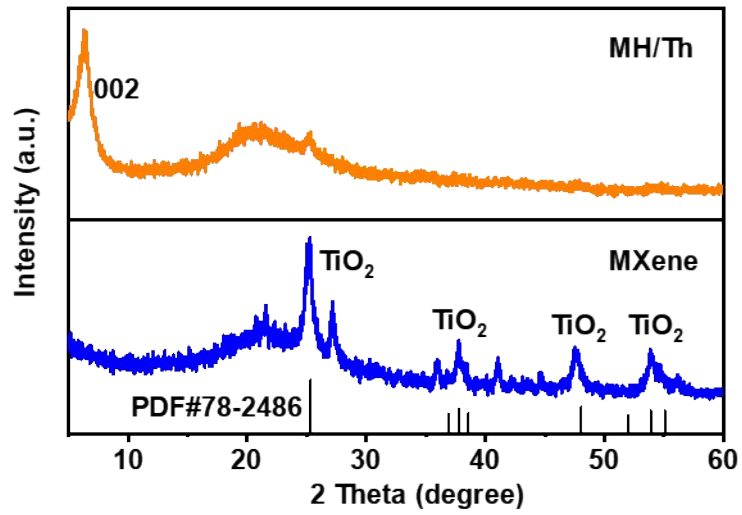
**Figure S7.** a-c) SEM images of MH/Th-1 (prepared by thionine solution of  $2 \text{ mg mL}^{-1}$  with MXene) in different resolutions. d-f) SEM images of MH/Th-3 (prepared by thionine solution of  $5 \text{ mg mL}^{-1}$  with MXene) in different resolutions.



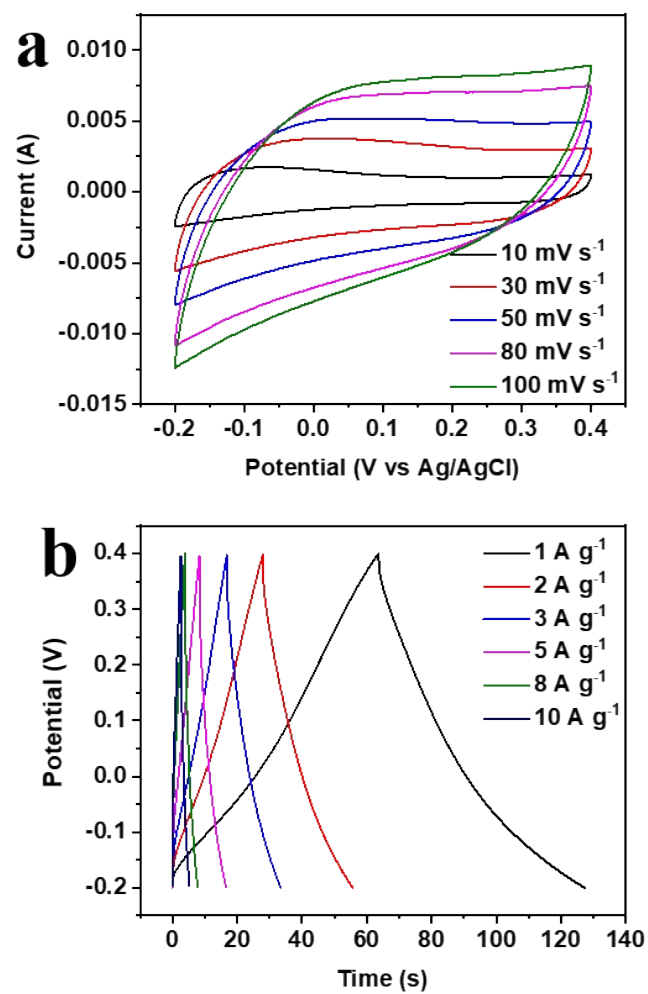
**Figure S8.** Element distribution mapping of MH/Th.



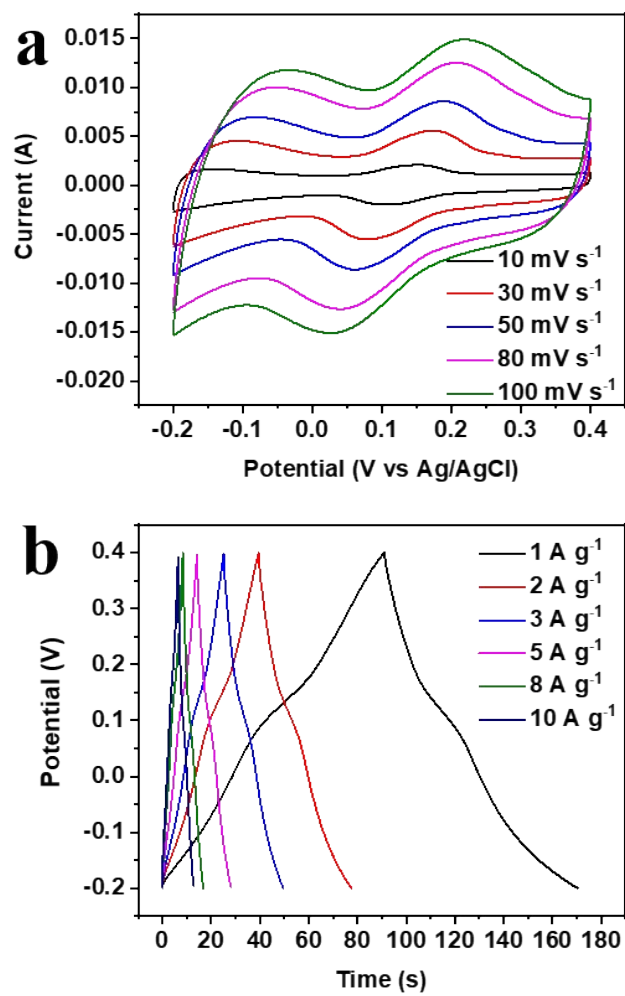
**Figure S9.** SEM images of a) MXene and b) MH/Th after 14 days under ambient conditions.



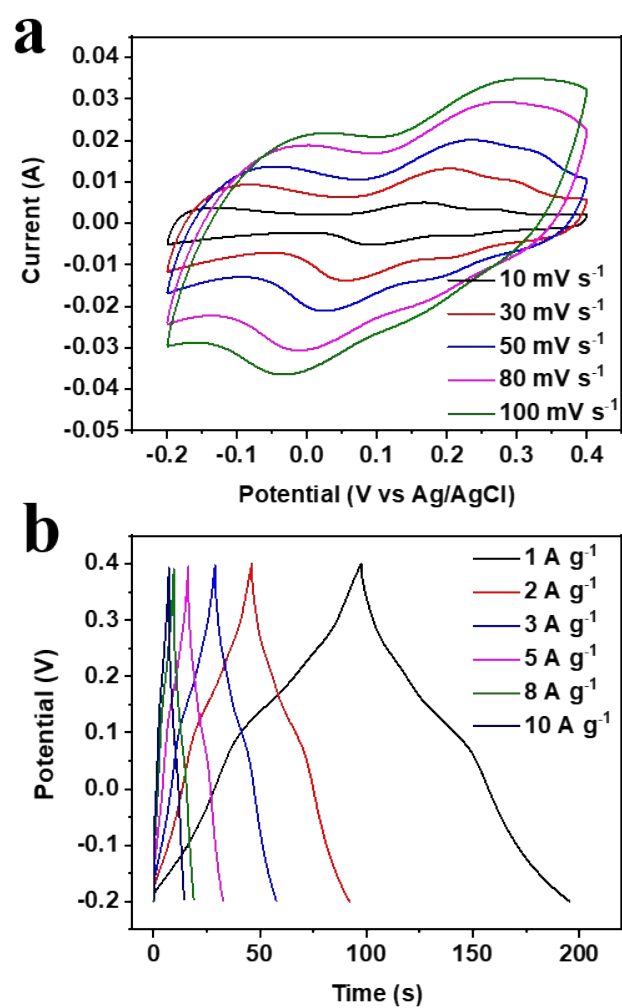
**Figure S10.** XRD patterns of MXene and MH/Th after 14 days under ambient conditions.



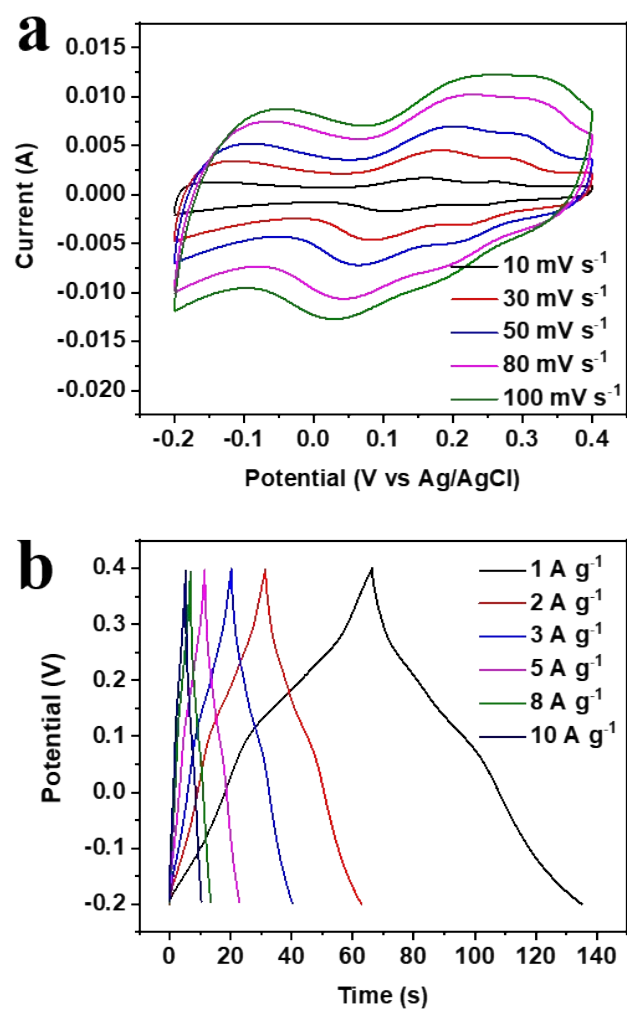
**Figure S11.** a) CV curves at various scan rates, b) GCD curves at different current densities of MXene.



**Figure S12.** a) CV curves at various scan rates, b) GCD curves at different current densities of MH/Th-1 (prepared by thionine solution of  $2 \text{ mg mL}^{-1}$  with MXene).

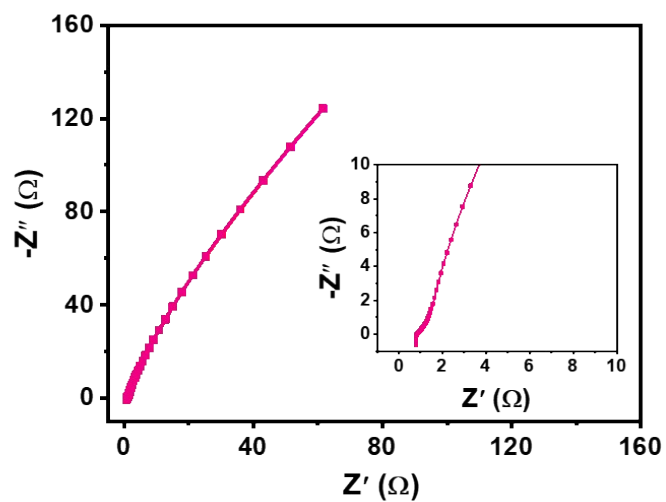


**Figure S13.** a) CV curves at various scan rates, b) GCD curves at different current densities of MH/Th-2 (prepared by thionine solution of 3 mg mL<sup>-1</sup> with MXene).



**Figure S14.** a) CV curves at various scan rates, b) GCD curves at different current densities of MH/Th-3 (prepared by thionine solution of 5 mg mL<sup>-1</sup> with MXene).





**Figure S15.** Nyquist plots of the symmetric supercapacitor fabricated by coupling two identical MH/Th-2.