Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting Information

MXene-Derived Tetrathiafulvalene Metal-Organic Framework for Ultra-Long Supercapattery

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S1: Electrochemical calculations

The BioLogic SP-300 Modular Research Grade Potentiostat/Galvanostat/FRA electrochemical workstation was used to test the electrochemical performance of synthesized MOFs and assembled supercapattery using the cyclic voltammetry (CV), galvanostatic charge discharge (CD), and electrochemical impedance spectroscopy (EIS) techniques. The active materials V_2CT_x-TMOF/$VOSO_4$-TMOF/activated carbon were combined with PVDF binder and super-P carbon as a conducting agent in a weight ratio of 75:20:5 to create the working electrode. The *N*-Methyl-2-pyrrolidone (NMP) solvent was used to make a paste for this electrode material, which was then applied to a 2 x 4 cm^2 nickel foam substrate. The electrode materials were loaded at around 3 mg/cm^2, and the coated substrate was vacuum oven dried at 50 °C overnight. Hg/HgO and Pt foil were used as a reference electrode and a counter-electrode, respectively. For all the electrochemical analysis, 3 M KOH was used as an electrolyte. The CV and CD experiments were carried out at different scan rates and various current densities, respectively. [1,2]

The specific capacity can be calculated using the following equation:

$$Q_{sp} = \frac{I\,\Delta t}{m} \qquad (1)$$

where Q_{sp} is the specific capacity (C/g), I is the charge-discharge current (A), Δt is the total discharge time (sec), and m is the total active mass of the electrode material (g). For specific capacitance in F/g

$$Q_{sc} = \frac{I\,\Delta t}{m\Delta v} \qquad (2)$$

Total capacitance of the device is calculated using,

$$Q_t = \frac{I \times \Delta t}{M} \qquad (3)$$

$$Q_{SP} = 4 \times Q_t \qquad (4)$$

Here, Q_t is the total specific capacity of the fabricated device (C/g), Q_{sp} is the specific capacity of the electrode material (C/g), I is the discharge current (A/g), Δt is the discharge time (sec), and M is the total active mass of both electrodes (g). The specific energy and specific power of the device were calculated from the discharge profile using the following formula

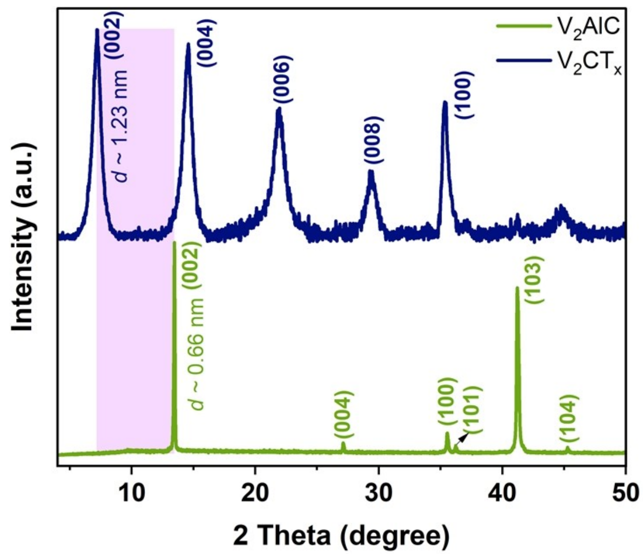
$$E = {}^{1}/_{2}\, Q_t V \tag{5}$$

$$P = \frac{E}{\Delta t} \tag{6}$$

E is the specific energy (Wh/kg), and P is the specific power (W/kg) based on the total mass of the positive and negative electrode materials. The EIS experiments were performed with 0 V bias and a sinusoidal signal of 5 mV, with a frequency range of 10 mHz to 100 kHz. EIS data were analyzed using a Nyquist plot.

S2: Material characterization

<u>Figure S1:</u> XRD profiles of commercially available V_2AlC MAX phase and etched V_2CTx MXene



<u>Figure S2:</u> SEM images of etched V_2CTx MXene.

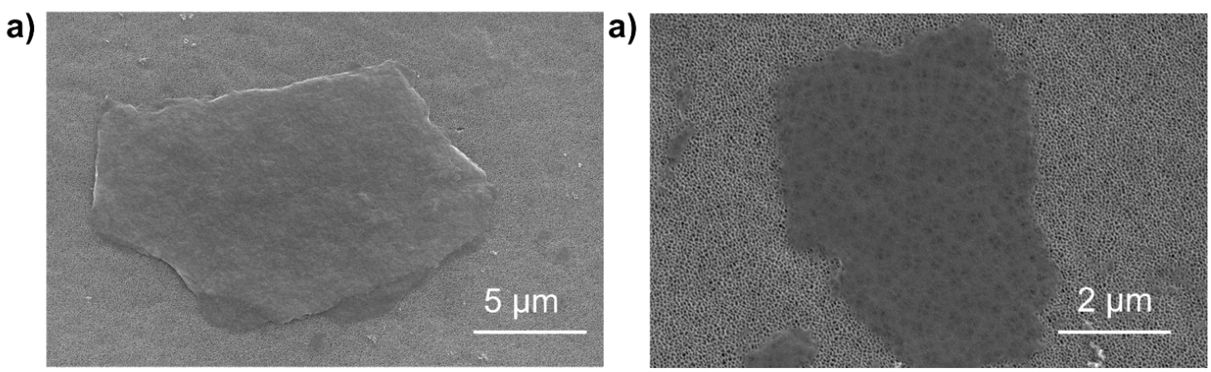


Figure S3: Pore size distribution curve of V_2CT_x-TMOF.

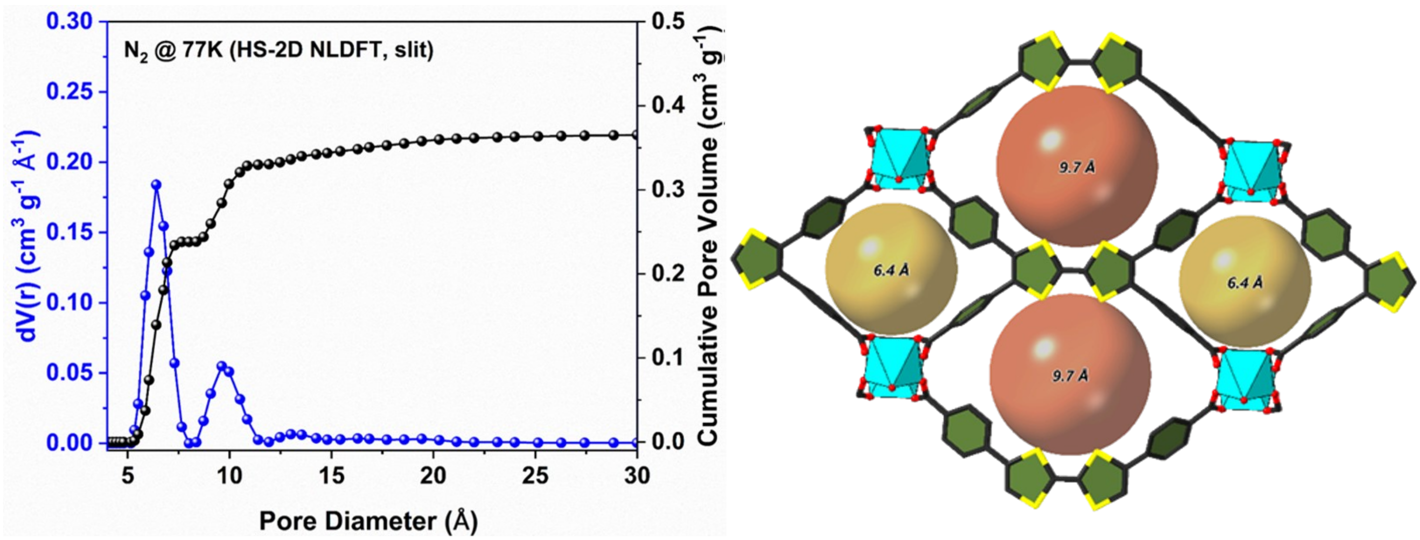


Figure S4: N_2 adsorption-desorption isotherm of $VOSO_4$-TMOF MXene.

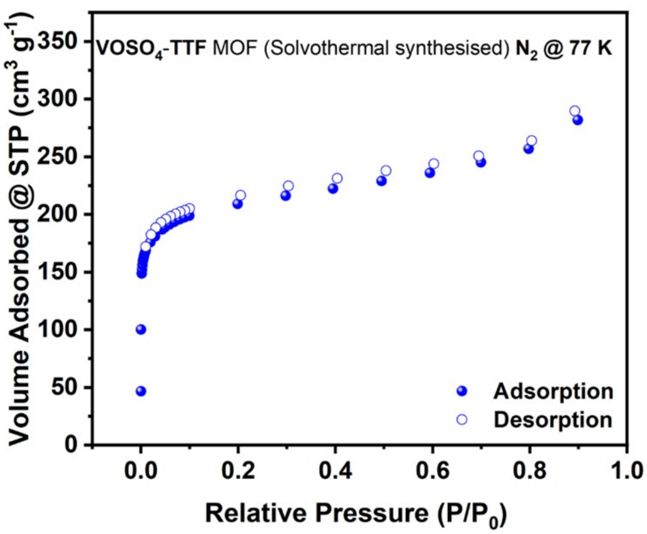


Figure S5: XRD images of V_2CTx-TMOF before and after N_2 sorption analysis

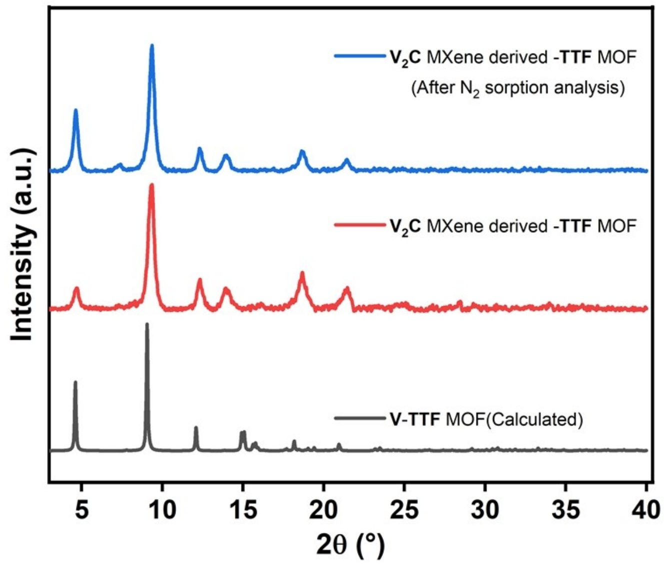


Figure S6: TGA curves of solvothermal synthesized $VOSO_4$-TMOF and MXene derived V_2CT_x-TMOF.

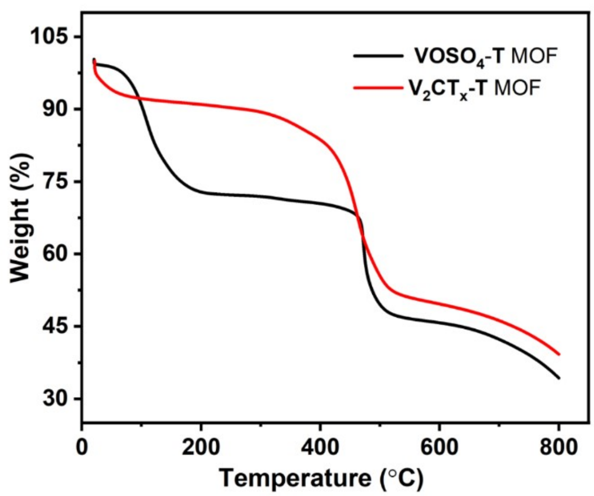
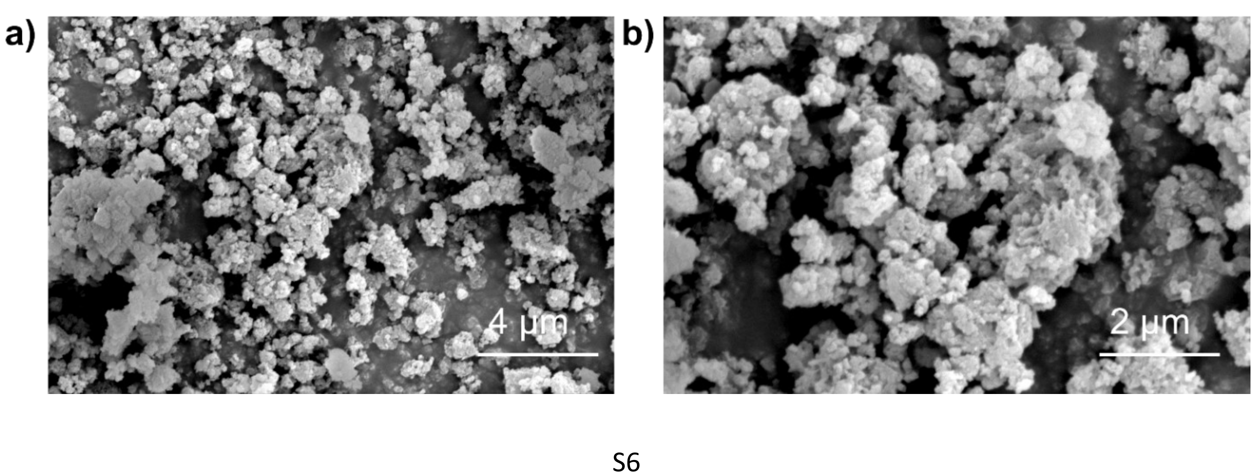


Figure S7: SEM images of solvothermal synthesized $VOSO_4$-TMOF



S3: Electrochemical characterization

Figure S8: CV, GCD curves of a, c) V_2CT_x-TMOF, b, d) Activated carbon at different scan rates and current densities

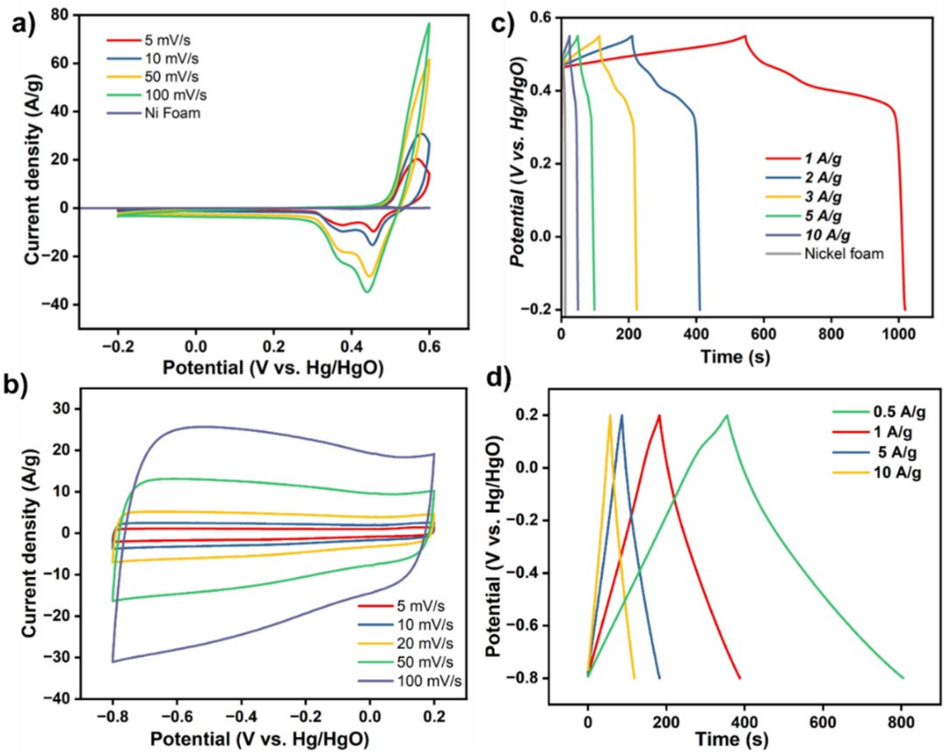


Figure S9: CV curves of V_2CT_x-TMOF//AC supercapattery deice a) device design b) device tested in different potential differences at scan rate of 5 mV/s

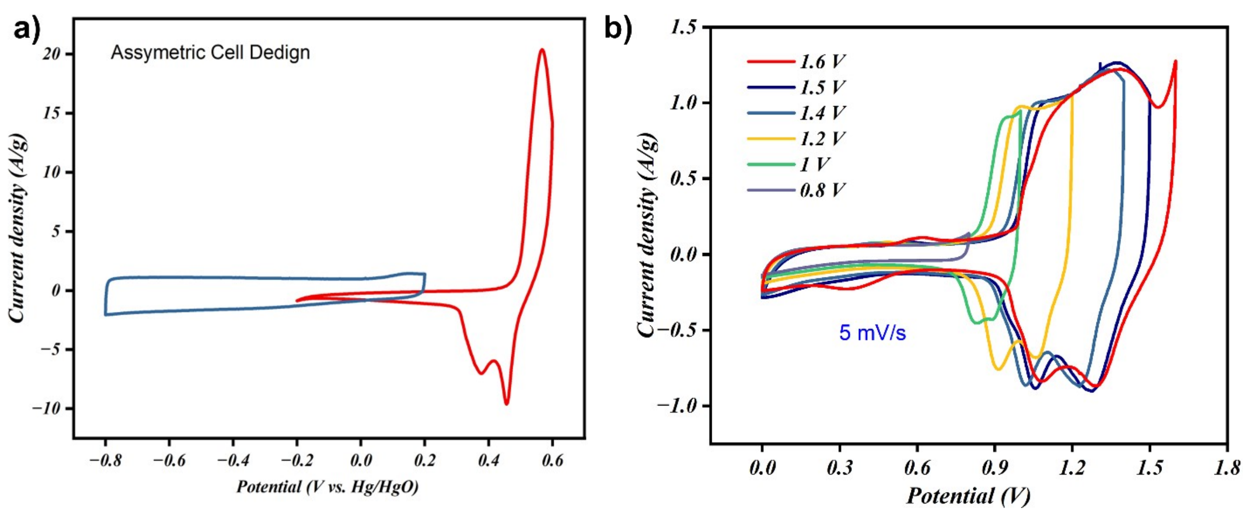
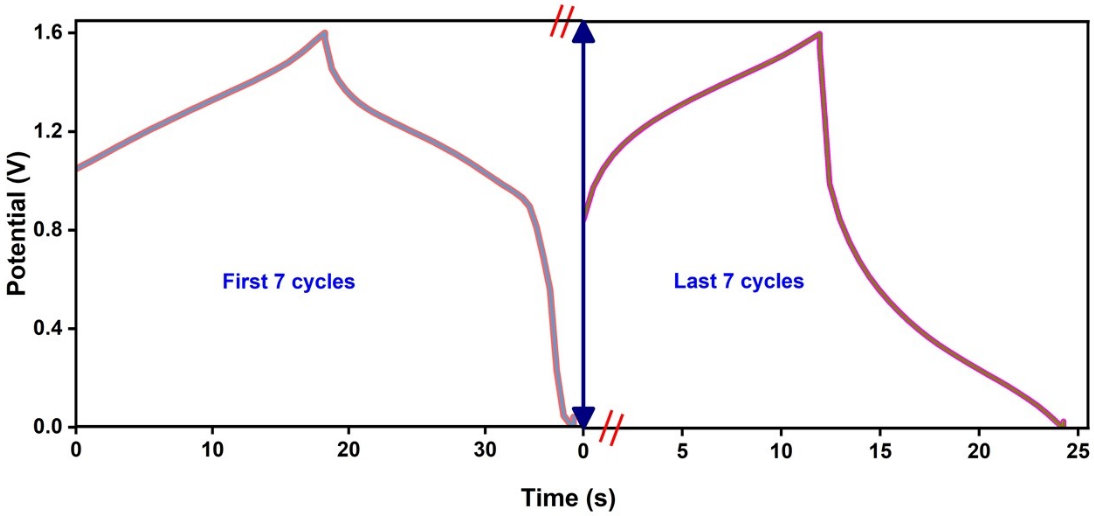


Figure S10: 7 set GCD profile of V_2CT_x-TMOF//AC supercapattery device during the initial cycles and final cycles when reaching 30,000 cycles.



S4: Electrochemical performance tables

Table S1: Three electrode capacitance (single electrode measurements) comparison for V_2CT_x-TMOF, $VOSO_4$-TMOF, and AC carbon.

V_2CT_x-TMOF			$VOSO_4$-TMOF			AC CARBON		
Current density	Specific capacity	Specific Capacitance	Current density	Specific capacity	Specific Capacitance	Current density	Specific capacity	Specific Capacitance
1 A/g	480 C/g	640 F/g	1 A/g	395 C/g	526 F/g	0.5 A/g	225 C/g	225 F/g
2 A/g	414 C/g	552 F/g	2 A/g	360 C/g	480 F/g	1 A/g	212 C/g	212 F/g
3 A/g	348 C/g	464 F/g	5 A/g	320 C/g	426 F/g	5 A/g	180 C/g	180 F/g
10 A/g	310 C/g	413 F/g	10 A/g	279 C/g	372 F/g	10 A/g	165 C/g	165 F/g

Table S2: V_2CT_x-TMOF//AC hybrid supercapattery device (Full cell device) performance, indicating specific energy and specific power with device capacitance

Current Density	Total Capacity	Total Capacitance	Specific capacitance	Specific Capacity	Specific Energy	Specific Power
1 A/g	222 C/g	138 F/g	552 F/g	888 C/g	50 Wh/kg	1264 W/kg
2 A/g	185 C/g	116 F/g	464 F/g	740 C/g	42 Wh/kg	1596 W/kg
3 A/g	170 C/g	106 F/g	424 F/g	680 C/g	38 Wh/kg	1753 W/kg
5 A/g	144 C/g	90 F/g	360 F/g	560 C/g	32 Wh/kg	2504 W/kg

S5: References

1 K. Nasrin, K. Subramani, M. Karnan and M. Sathish, *J. Colloid Interface Sci*, 2021, **600**, 264–277.

2 K. Choi, I. K. Moon and J. Oh, *J. Mater. Chem. A*, 2019, **7**, 1468–1478.