# Supporting Information

## Molten-Salt Etching Synthesis of Delaminatable MXene

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#### **Experimental Section**

#### Synthesis of MXene by molten-salt etching

The Ti<sub>3</sub>AlC<sub>2</sub> MAX phase (1.0 g) was uniformly mixed with anhydrous CuCl<sub>2</sub> (2.2g), KCl (1.0 g), NaCl (0.6 g) and LiF (1.0 g) under dry conditions. This mixture was annealed at 750 °C for 5 h in Ar flow. The obtained products were repeatedly rinsed with deionized water and an aqueous solution of ammonium persulfate (APS, 0.5 M) to remove the salt and Cu by-product, yielding accordion-like MXene.

The accordion-like MXene was immersed into an aqueous solution of TBAOH solution (25 %) for 36 h. After the removal of the TBAOH by centrifugation, the TBAOH intercalated accordion-like MXene was exfoliated to highly dispersed MXene nanosheets (denoted as LiF/CuCl<sub>2</sub>-MXene) under ultrasonic for several hours in an ice bath. The dark green supernatant was collected by centrifugation at 2,000 rpm and stored at 4 °C in the refrigerator before use

As a comparison, the accordion-like MXene was also prepared similarly in the absence of LiF, which is hard to be exfoliated even with the assistance of TBAOH intercalation agent.

#### Synthesis of MXene by solution etching.

The MXene (denoted as LiF/HCl-MXene) was synthesized by etching the  $Ti_3AlC_2$  MAX phase (1.0 g) with a mixture solution of LiF (1.32g) and HCl (6 M, 20 mL) at 35 °C for 24 h. The obtained product was harvested by several centrifugation-rinsing cycles with deionized water, followed by exfoliation under ultrasonic for 1 h in an ice bath. The dark green supernatant was collected by centrifugation at 2,000 rpm and stored at 4 °C in the refrigerator before use

### Material Characterization.

The SEM and TEM images were taken with scanning electron microscopy (SU8220) and transmission electron microscopy (HT7700 EXALENS). The X-ray diffraction (XRD) analysis was done on a Bruker D8 Advance X-ray spectrometer (Cu K $\alpha$ ,  $\lambda$  = 1.5406 Å). The mass change of the electrode was tested by electrochemical quartz

crystal microbalance (QCM 200). The XPS measurements were performed using Thermo ESCALAB MK II X-ray photoelectron spectrometer with C 1s (284.6 eV) calibration. The Raman analysis was conducted on a Thermo Fisher Scientific DXR Raman microscopy using laser excitation ( $\lambda = 532$  nm). The FT-IR spectra were measured by a Thermofisher Nicolet-IS50 Fourier transform infrared spectrometer.

#### **Supercapacitor tests**

The supercapacitor tests were performed on an IVIUM vertex. C. EIS electrochemical workstation with a standard three-electrode system. A graphite rod and an Ag/AgCl electrode with saturated KCl solution were employed as counter and the reference electrode, respectively. The electrolyte was 3 M H<sub>2</sub>SO<sub>4</sub>. The working electrode was made by casting a slurry consisting of Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene (10 mg), deionized water (0.49 mL), ethanol (0.49 mL) and 30 wt% Nafion solution (0.02 mL) on a carbon paper. The electrode has an average mass loading of ca. 0.2 mg cm<sup>-2</sup>. The cyclic voltammetry (CV) was tested in a voltage window between –0.6 and 0.3 V (*vs.* Ag/AgCl) at different scan rates. The galvanostatic charge-discharge tests were conducted between –0.5 and 0.3 V (*vs.* Ag/AgCl) at desired current densities. The gravimetric capacitance was calculated from constant charge/discharge curves by the following formula:

$$C = \frac{I_{\rm cons}t}{m \Delta V} \tag{1}$$

where  $I_{cons}$  is the constant discharge current (A), t is the discharge time (s), m is the active mass in the electrode (g), and  $\Delta V$  is the voltage range.

The Electrochemical impedance spectroscopy (EIS) measurements were performed at open-circuit voltage over the frequency range of 100 kHz to 0.01 Hz and an amplitude of 10 mV.



Fig. S1 SEM image of LiF/CuCl<sub>2</sub>-MXene.



Fig. S2 Tyndall effect of LiF/CuCl<sub>2</sub>-MXene colloids.



**Fig. S3** Element mapping showing the uniform distribution of Ti, O and F element on LiF/HCl-MXene.



Fig. S4 XPS spectra of a) LiF/CuCl<sub>2</sub>-MXene and b) LiF/HCl-MXene.



**Fig. S5** (a) Zeta potential of LiF/CuCl<sub>2</sub>-MXene and accordion-like MXene without -F group. (b) XRD patterns showing the peak from the (002) plane of LiF/CuCl<sub>2</sub>-MXene before and after TBAOH intercalation. A shift in the peak position to the lower angle indicates the expansion of interlayer spacing.



**Fig. S6** Galvanostatic charge-discharge curves of a) LiF/HCl-MXene and b) accordion like-MXene at various current densities from  $0.5 \text{ A g}^{-1}$  to  $4.0 \text{ A g}^{-1}$ .



**Fig. S7** (a) CVs at a scan rate of 100 mV s<sup>-1</sup> and (b) galvanostatic charge-discharge curves a current density of 0.5 A g<sup>-1</sup> for LiF/CuCl<sub>2</sub>-MXene, LiF/HCl-MXene and accordion like-MXene with a mass loading of *ca.* 2.0 mg cm<sup>-2</sup>.



**Fig. S8** Normalized real and imaginary capacitances of a) LiF/CuCl<sub>2</sub>-MXene and b) LiF/HCl-MXene.



**Fig. S9** CVs of a) LiF/CuCl<sub>2</sub>-MXene, b) LiF/HCl-MXene and c) accordion like-MXene at various scant rates in 3 M H<sub>2</sub>SO<sub>4</sub>.

Element ratio / At. %	Ti	С	0	F	Cl
CuCl <sub>2</sub> -MXene	12.79	69.23	15.88	0.82	0.2
LiF/HCl-MXene	10.32	74.85	13.43	1.15	trace

 Table S1. The element composition of LiF/CuCl<sub>2</sub>-MXene and LiF/HCl-MXene.