Electronic Supplementary Information

Selective Lithiation-Expansion-Microexplosion Synthesis of Two-Dimensional Fluoride-Free MXene

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EXPERIMENTAL METHODS

Electrochemical intercalation

The lithium intercalation of MAX phase Ti₃AlC₂ was performed in a test cell using the Li foil as anode and 1M LiTFSI as electrolyte, which was dissolved in TEGDME solution. Ti₃AlC₂ was prepared as cathode by mixing with poly(vinylidene fluoride) (PVDF) binder dispersed in N-methylpyrrolidone (NMP) solutions. In the mixed slurry, the mass ratio of the Ti₃AlC₂ and PVDF was 95:5. The resulting slurry was then uniformly coated on a copper foil and dried under vacuum overnight. The assembly of the test cells was conducted in an Ar-filled glove box. The electrochemical intercalation in the test cell was accomplished in a Neware battery test system at room temperature. The electrochemical intercalation was performed using galvanostatic discharge at 0.10 mA. After the discharge process, the Li-intercalated sample was washed with acetone with ultrasound to remove electrolyte and PVDF binder for several times, followed by exfoliation and ultrasonication in distilled water. After the suspension was centrifuged and washed six times with water, it was collected for further characterizations.

Materials characterization

The morphologies were tested through Hitachi field emission scanning electronic microscope. Micromorphology and fine structure were measured *via* FEI Tecnai F20 field emission transmission electron microscope. The crystal structure was confirmed by Phillips X'pert ProMPD diffractometer. An ESCALAB 250Xi spectrometer (Thermo Fisher) was employed to measure the X-Ray

photoelectron spectra (XPS). The loading of CoHPO was measured using inductively coupled plasma mass spectroscopy (Thermo XSeries II) by dissolving 1 cm \times 1 cm electrode in 100 mL aquaregia.

Electrochemical measurements

The working electrode was prepared by mixing the as-prepared $Ti_3C_2T_x$ MXene and polyvinylidene fluoride (mass ratio=95 : 5) in ethanol to form a slurry. Then the slurry was coated onto the nickel foam current collector with coating area of 1 cm² and dried at 100 °C for overnight in a vacuum oven. The electrochemical tests of the individual electrode were carried out using a standard three-electrode cell with 6 M KOH aqueous solution as the electrolyte. Ni foam coated with electroactive materials was used as the working electrode, platinum foil and Hg/HgO electrode were used as the counter and reference electrodes, respectively. All tests were performed using a CHI660D electrochemical workstation. The gravimetric specific capacitance (C) from the galvanostatic charge/discharge curves was calculated using the formula:

$$C = \frac{\mathrm{I}\Delta t}{m\Delta V} \qquad (1)$$

 $C = (\int I dV) / \mu mV$ (2)

here I is the current density, m is the total mass from MXene electrodes, ΔV is the potential change within the discharge time Δt . V is potential, v is the potential scan rate, and m is the mass of the electroactive materials.



Figure S1 The discharge curve of Ti_3AlC_2 in Li-ion battery with galvanostatic discharge at 0.10 mA.



Figure S2 X-ray diffraction patterns of Ti_3AlC_2 and lithiation Ti_3AlC_2 without reaction with

water.



Figure S3 The $Ti_3C_2T_x$ solution with Tyndall effect



Figure S4 the XPS survey spectra of $Ti_3C_2T_x$



Figure S5 a) AFM image and e) the relevant height profiles of $Ti_3C_2T_x$ MXene from Ti_3AlC_2MAX .



Figure S6 a) AFM image and e) the relevant height profiles of Ti_3AlC_2MAX .



Figure S7 X-ray diffraction patterns of Ti_3SiC_2 and $Ti_3C_2T_x$.



Figure S8 a) AFM image and e) the relevant height profiles of MXene phase $Ti_3C_2T_x$ from MAX phase Ti_3SiC_2MAX .



Figure S9 The dependence of the current sensitivity (the CV curves at -0.7 V (vs. Hg/HgO) for the charge and discharge) for $Ti_3C_2T_x$ electrode.

(-)//(+)	pseudocapacitive	rate	Specific capacitance	Ref.
MXene film	6 M KOH	0.5 A g ⁻¹	338 F g ⁻¹	This work
rGO/Mxene	6 M KOH	$0.2 A g^{-1}$	148.5 F g ⁻¹	J. Mater. Chem. A 2017, 5, 17442
Ti ₃ C ₂ T _x -NOMC	3 М КОН	1 A g ⁻¹	329 Fg ⁻¹	Nano Energy 2019, 65, 103991
Ti ₃ C ₂ T _x	6 M KOH	1 A g ⁻¹	$52 \ F \ g^{-1}$	Nano Energy 2018,50, 479–488
Ti ₃ C ₂ T _x Mxene	1 М КОН	2 mV s ⁻¹	135 F g ⁻¹	Energy Storage Materials, 2019, 18, 456-461
MXene/rGO	1 М КОН	1 A g ⁻¹	80.3 F g ⁻¹	Adv. Funct. Mater. 2017, 27, 1701264
Clay-like Ti ₃ C ₂ T _x	1 M H ₂ SO ₄	2 mV s ⁻¹	245 F g ⁻¹	Nature, 2014, 516, 78-81.
ϵ -MnO ₂ /Ti ₃ C ₂ T _x	6 M KOH	1 A g ⁻¹	210 F g ⁻¹	ACS Appl. Mater. Interfaces, 2016, 8, 18806
Ti ₃ C ₂ T _x	1 M H ₂ SO ₄	0.2mAcm ⁻²	356.8 F cm ⁻³	Energy Environ. Sci. 2016, 9, 2847
Ti ₃ C ₂ T _x hydrogel	1 M H ₂ SO ₄	2 mV s ⁻¹	380 F g ⁻¹	Nat. Energy, 2017, 17105, 1–7

Table S1. The table of Ti₃C₂T_x-based pseudocapacitive supercapacitors

Ti ₃ C ₂ T _x clay	1 M H ₂ SO ₄	2 mV s ⁻¹	325 F g ⁻¹	Electrochem. commun., 2014, 48, 118-122
Ti ₃ C ₂ T	6 М КОН	1 A g ⁻¹	51 F g ⁻¹	Chem. Mater. 2015, 27, 5314-5323
Ti ₃ C ₂ T _x /CNT	1 M MgSO ₄	2 mV s ⁻¹	150 F g ⁻¹	Adv. Mater., 2015, 27, 339-345
Ti ₃ C ₂ Tx	6 M KOH	$0.2 \mathrm{A g}^{-1}$	118 F g ⁻¹	J. Electrochem. Soc., 2015, 162, B16–B21
PANI@TiO ₂ /Ti ₃ C ₂ T _x	6 M KOH	0.5 A g ⁻¹	108.6 F g ⁻¹	Electrochimica Acta, 2017, 228, 282–289