Supporting Informaiton

Graphene-Assisted Ti₃C₂ MXene-Derived Ultrathin Sodium Titanate for Capacitive deionization with Excellent Rate Performance and Long Cycling Stability

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Experimental methods

1. Preparation of 2D delaminated $Ti_3C_2T_x$ MXene

One gram of LiF (>98%) was dissolved in 20 mL of 9 M HCl. Then, 1 g of sieved Ti_3AlC_2 powders (400 mesh) was added. The mixture was kept at 40°C for 24 h under stirring with a magnetic stirrer. The resulting solid residue was washed several times with deionized water and centrifuged at a speed of 3500 rpm until the pH of the supernatant was approximately 6. The powder was mixed with deionized water and sonicated for 1 h under an ice-bath, through which Ar gas was bubbled. The resulting solution was centrifuged for 40 min at a speed of 4000 rpm. Finally, the 2D delaminated $Ti_3C_2T_x$ colloidal was obtained. To determine the volumetric density, a syringe was used to pull out 3 ml of the colloidal suspension to filter a film. The remaining suspension was stored in sealed bottles under Ar. After drying in air, it was weighed. For all the work conducted herein, the 2D delaminated $Ti_3C_2T_x$ MXene solution concentration was approximately 3 mg/mL.

2. Material characterization

Scanning electron microscopy (SEM, Hitachi 54800, Japan) and transmission electron microscopy (TEM, JEOL-2010F, Japan) were used to analyze the morphology and microstructure. The crystal structures were analyzed using X-ray diffraction (XRD, D8 Advance, Bruker, Germany) operated at 40 mA and 45 kV with Cu Kα radiation (λ=0.15418 nm, 5°/min, 3-65°). The Brunauer-Emmett-Teller (BET) isotherms and specific surface area (BET surface area) were obtained using a Belsorp Mini-II instrument (Japan) at 77 K. The pore size distribution profile was analyzed using the Barrett-Joyner-Halenda (BJH) model. X-ray photoelectron spectroscopy (XPS Kratos Axis UltraDLD SHIMADZU, Japan) was performed using monochromated Al Ka X-rays at a base pressure of 1×10^{-9} Torr.

3. Electrochemical measurement

The AC EDL electrode consisted of 80% active material, 10% acetylene black and a 10% polyvinylidene difluoride (PVDF) binder in N-methyl-2-pyrrolidone (NMP) on a graphite sheet (mass ratio). Constant voltage (CV) and galvanostatic charging-discharging (GCD) tests were performed on a three-electrode electrochemical workstation (CHI660D, Chenhua Instruments Co., China), using Pt as the counter electrode, Ag/AgCl as the reference electrode, and 1 M NaCl as the electrolyte. The specific capacitance (C, F/g) can be obtained from the CV curves using the following

equation:

$$C = \int idV/\Delta Vmv \qquad (1)$$

where i is the current (A), m is the mass of the active material (g), ΔV is the voltage window (V), and v is the scan rate (V/s).

Electrochemical impedance spectroscopy (EIS) was applied via a CHI660D instrument with a calomel reference electrode, and the data were obtained using a 5mV amplitude in the frequency range from 10^5 Hz to 0.1 Hz.

4. Desalination experiments

The electrosorption experiments were conducted in a batch mode system with an HCDI unit cell, which included an activated carbon (AC) anode, an MXene cathode, an anion exchange membrane (AEM) and a cation exchange membrane (CEM). All the experiments were performed by applying a 30 mA/g electric current density with a flow rate of 50 ml/min, and the feed water was pumped through plastic tubes via a peristaltic pump. The conductivity of the solution was monitored by a conductivity meter (METTLER TOLEDO S230, Switzerland). The volume and temperature of the solution were maintained at 45 mL and 25°C, respectively. The relationship between the conductivity and the concentration was calibrated prior to the deionization experiments. The desalination capacity (Γ), removal rates (v) and energy consumption (kWh/kg-NaCl) are defined as follows:

$$\Gamma = (C_0 - C_e) \times V/m_t \qquad (2)$$

$$v = \frac{\Gamma}{t} \qquad (3)$$

$$\text{Energy consumption} = \frac{i \times \int v\, dt}{3.6 \times (C_e - C_0) \times V} \qquad (4)$$

where C_0 and C_e (mg/L) are the initial and final NaCl concentrations, respectively, m_t (g) is the mass of the MXene electrode, i is the current (A), and V (L) is the volume of the NaCl solution.
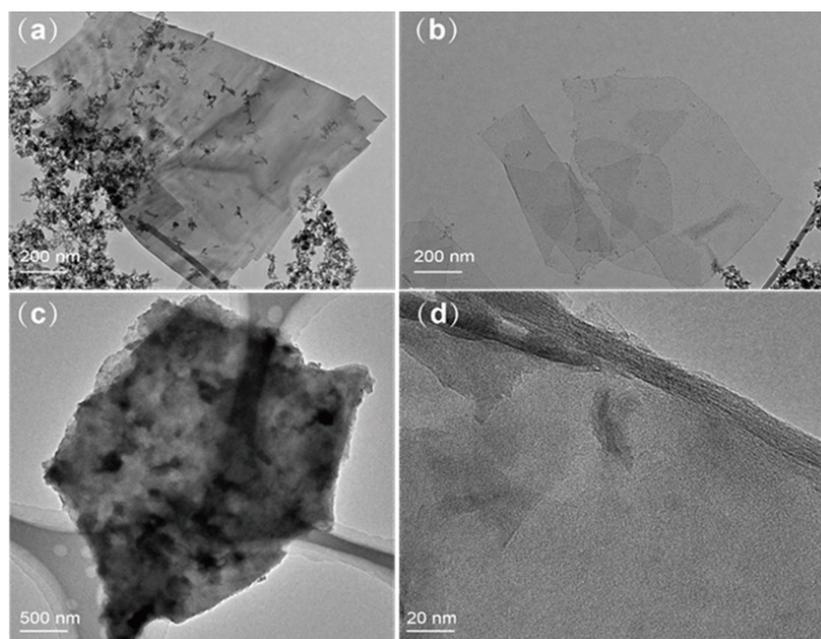
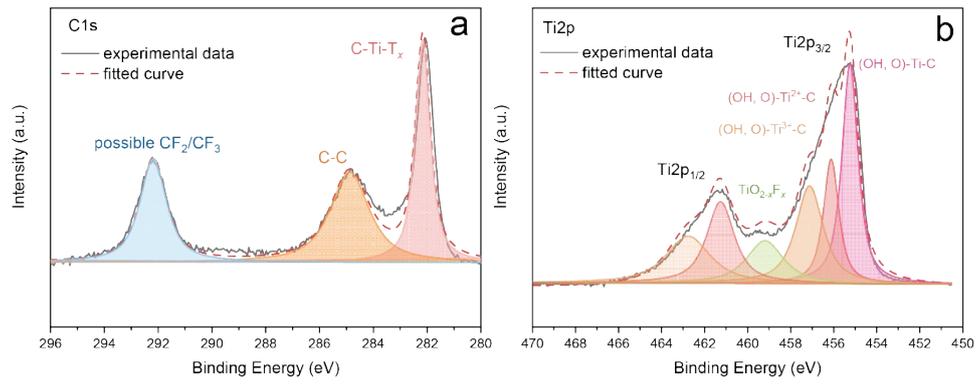Figure S1. TEM images of Ti3C2Tx (a, b) and M-NTO/rGO (c, d).

Figure S2 typical high-resolution XPS of C1s (a) and Ti 2p (b) of MXene.

Table.S1 Comparison of various reported electrodes applied for CDI.

Electrode materials (Cathode)	C_0（mg/L）	Applied voltage/current density	SAC (mg/g)	Long term-stabillity	Ref.
MnO$_2$	500	1.4	14.9	350	[1]
Hybrid-MnO$_2$	850	1.2	27.3	-	[2]
Na$_4$Mn$_9$O$_{18}$	580	1.2	31.2	-	[3]
aNa$_2$FeP$_2$O$_7$	580	1.2	30.2	-	[4]
AC-Ti-S	500	1.2	10	-	[5]
Ag coated carbon composite	580	0.7	15.6	-	[6]
Grapheme@Na$_4$Ti$_9$O$_{20}$	250	1.4	41.8	-	[7]
M-NTO/rGO	1000	30mA/g/1.4V	57.57	100	This work

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