Supporting Information

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

Xiaojie Shen¹,²,³, Liqing Li⁴, Yuecheng Xiong³,⁵,⁶, Fei Yu¹*, Jie Ma³,⁴*

¹ College of Marine Ecology and Environment, Shanghai Ocean University, Shanghai 201306, P.R. China, E-mail: fyu@vip.163.com
² Department of Environmental Science & Engineering, College of Chemical Engineering, Beijing University of Chemical Technology, 15 North Third Ring Road, Beijing 100029, P. R. China
³ Research Center for Environmental Functional Materials, College of Environmental Science and Engineering, Tongji University, 1239 Siping Road, Shanghai 200092, P.R. China
⁴ Faculty of Materials Metallurgy and Chemistry, Jiangxi University of Science and Technology, Ganzhou, 341000, P.R. China, E-mail:liliqing79@126.com
⁵ Department of Chemistry, City University of Hong Kong, Hong Kong, 999077 China
⁶ Hong Kong Branch of National Precious Metals Material Engineering Research Center (NPMM), City University of Hong Kong, Hong Kong, 999077 China
Experimental methods

1. Preparation of 2D delaminated Ti$_3$C$_2$Tx MXene

One gram of LiF (>98%) was dissolved in 20 mL of 9 M HCl. Then, 1 g of sieved Ti$_3$AlC$_2$ 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$_3$C$_2$Tx 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$_3$C$_2$Tx 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:

\[ C = \int i dV / \Delta V m v \]  

(1)

where \( i \) is the current (A), \( m \) is the mass of the active material (g), \( \Delta V \) 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 \( \Gamma \), removal rates \( v \) and energy consumption (kWh/kg-NaCl) are defined as follows:

\[ \Gamma = (C_0 - C_e) \times V / m_t \]  

(2)

\[ v = \frac{\Gamma}{t} \]  

(3)

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

(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.

<table>
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<tr>
<th>Electrode materials</th>
<th>C_0 (mg/L)</th>
<th>Applied voltage/current density</th>
<th>SAC (mg/g)</th>
<th>Long term-stability</th>
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<td>MnO_2</td>
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<td>Ag coated carbon composite</td>
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<td>M-NTO/rGO</td>
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References


