Supporting Information

Vertically oriented NiCoMo sulfide nanosheet arrays on $Ti_3C_2T_x$ MXene for high performance supercapacitor

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Materials and methods

Materials: Titanium aluminium carbide (Ti_3AlC_2), Concentrated hydrochloric acid (HCl), Lithium fluoride (LiF), Nickel nitrate hexahydrate ($Ni(NO_3)_2·6H_2O$)、Cobalt nitrate hexahydrate ($Co(NO_3)_2·6H_2O$), Sodium molybdate (Na_2MoO_4), Ammonium fluoride (NH_4F), Urea ($CO(NH_2)_2$) Sodium sulphide nine hydrate ($Na_2S·9H_2O$) and Anhydrous ethanol were obtained from Sinopharm Chemical Reagent Co. and could be used without further purification. Deionized water (DI water) with a resistivity of 18.25 MΩ/cm was prepared using an ultrapure water polishing system.

Sample Preparation

(1) Preparation of less-layered $Ti_3C_2T_x$ MXene

1.6 g of LiF was added to 20 mL of hydrochloric acid solution with a concentration of 9 M. After mixing well, 1 g of Ti_3AlC_2 was added in small amounts. The product was then transferred to a water bath at 45 °C, held for 48 h and washed by centrifugation to pH≈6. The resulting product was transferred to a 50 mL centrifuge tube and sonicated in an ice-water bath for 30 min, after which the sonicated material was centrifuged in a centrifuge at 3500 rpm for 15 min. The supernatant was poured into a beaker, and the bottom of the sample was free of precipitation after several centrifugations. After several centrifugations, the bottom was free of precipitation. The obtained solution was freeze-dried to obtain MXene oligolayer.

(2) Preparation of NiCoMo-$Ti_3C_2T_x$

1 mmol $Ni(NO_3)_2·6H_2O$, 1 mmol $Co(NO_3)_2·6H_2O$, 1 mmol Na_2MoO_4, 0.3 g Urea, 0.074 g NH_4F, and 0.5 mmol $Ti_3C_2T_x$ MXene were dissolved in 50 mL Deionized water and stirred for 20 min to obtain a clarified solution. Afterwards, the mixed solution was transferred to a reactor and reacted at 150 °C for 6 h. The sample $Ni_1Co_1Mo_1$-$Ti_3C_2T_x$ was obtained by water washing, alcohol

washing and drying. control samples $Ni_2Co_1Mo_1$-$Ti_3C_2T_x$, $Ni_1Co_2Mo_1$-$Ti_3C_2T_x$ and Ni_2Co_1-$Ti_3C_2T_x$ were obtained by changing the molar mass of the added samples. in the absence of $Ti_3C_2T_x$ MXene, the sample was added to the reactor. $Ti_3C_2T_x$ MXene $Ni_2Co_1Mo_1$ LDHs were obtained.

(3) Preparation of NiCoMoS-$Ti_3C_2T_x$

20 mg of the prepared NiCoMo-$Ti_3C_2T_x$ was dissolved in 20 mL of DI water and sonicated for 10 min to obtain solution A. 40 mg of $Na_2S{\cdot}9H_2O$ was dissolved in 10 mL of DI water and stirred well to obtain solution B. Solution B was poured into solution A under stirring and mixed for 3 min and then reacted at 120 °C for 10 h to obtain the Sample NiCoMoS-$Ti_3C_2T_x$.

Characterization

In order to systematically analyze the phase composition and microstructural characteristics of the materials, this study adopts a multi-dimensional characterization method: firstly, the phase analysis is carried out by X-ray diffractometer (XRD, Cu-targeted Kα-rays, λ = 1.5406 Å) within the range of 5°-80° at a scanning rate of 8°/min, so as to accurately analyze the information on the sample's crystalline structure and phase composition; Scanning electron microscope (SEM) and transmission electron microscope (TEM) are used to implement multi-scale morphological observation, combined with energy dispersive X-ray spectroscopy (EDS) technology to achieve quantitative characterization of micro-area elemental distribution and chemical composition analysis; X-ray photoelectron spectroscopy (XPS) is used to deeply analyze the elements on the surface of the samples, revealing the chemical valence state of each element and the characteristics of the surface chemical environment; finally, the pore structure test is carried out by the fully automated specific surface and porosity analyzer (BET), systematically evaluating the specific surface area of the material, the pore size distribution and other parameters.

Electrochemical performance test

Preparation of working electrode: 0.015 g of prepared sample, 0.002 g of PVDF, 0.003 g of acetylene black and 0.2 mL of 1-methyl-2-pyrrolidone were mixed thoroughly in a mortar and then uniformly coated in a square area of 1 cm×1 cm at one end of nickel foam. It was then dried in a vacuum oven at 60 °C overnight. Finally, the dried electrodes were removed and the area covered with the samples was pressed under a tablet press at 20 MPa for 15 s and weighed by an electronic analytical balance, and then the area covered with the samples was immersed in 1 M KOH solution for 3 h and set aside.

Three-electrode and asymmetric two-electrode were used to test the electrochemical properties of the samples. In this case, the three-electrode system consists of a calomel electrode, a platinum electrode and a prepared working electrode. In the asymmetric two-electrode system, nickel foam loaded with the sample that has been prepared is used as the positive electrode, and nickel foam loaded with activated carbon is used as the negative electrode material. The mass ratio of positive and negative active materials should be in accordance with the charge balance equation:

$$\frac{m_+}{m-} = \frac{C_- \times \Delta V_-}{C_+ \times \Delta V_+} \#(1)$$

C-, C_+, ΔV- and ΔV_+ are the negative specific capacitance (F g^{-1}), positive specific capacitance (F g^{-1}), negative potential window (V) and positive potential window (V) respectively.

The electrochemical properties of the materials were systematically characterised using electrochemical testing techniques such as cyclic voltammetry (CV), constant current charge/discharge test (GCD) and electrochemical impedance spectroscopy (EIS). Specific capacitance (C), energy density (E) and power density (P) were calculated according to the following equations:

$$C = \frac{I\Delta t}{m\Delta V} \#(2)$$

$$E = \frac{C\Delta V^2}{2 \times 3.6} \#(3)$$

$$P = \frac{3600 \times E}{\Delta t} \#(4)$$

where Δt, I, ΔV and m represent the discharge time (t), current density (A g^{-1}), operating voltage window (V) and mass of active material loaded on the electrode (g), respectively.

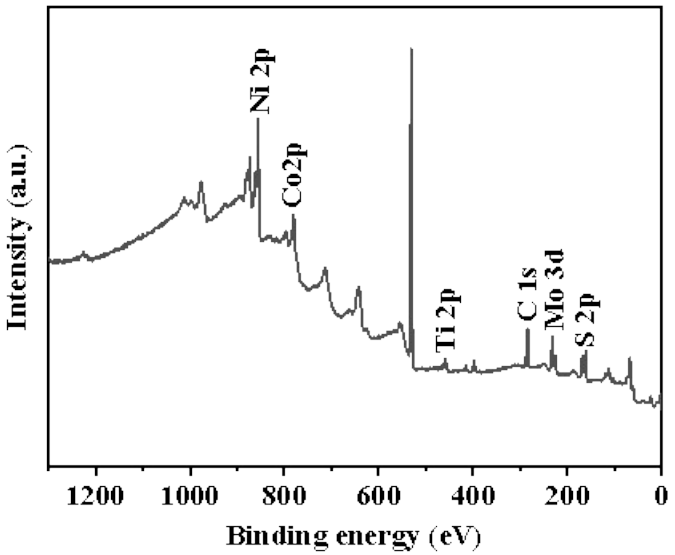
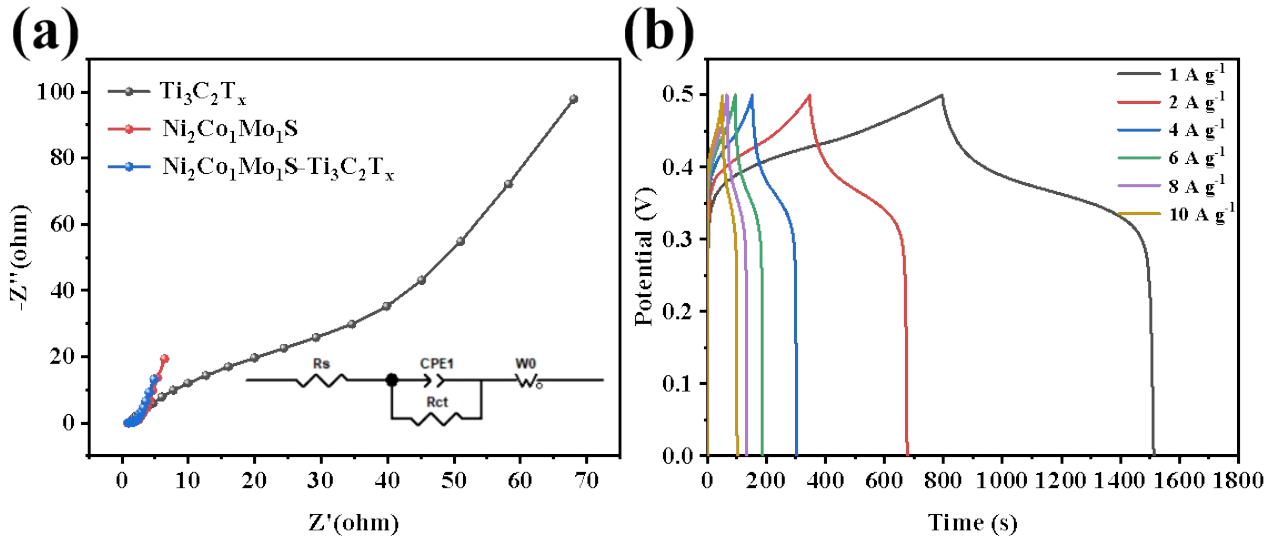
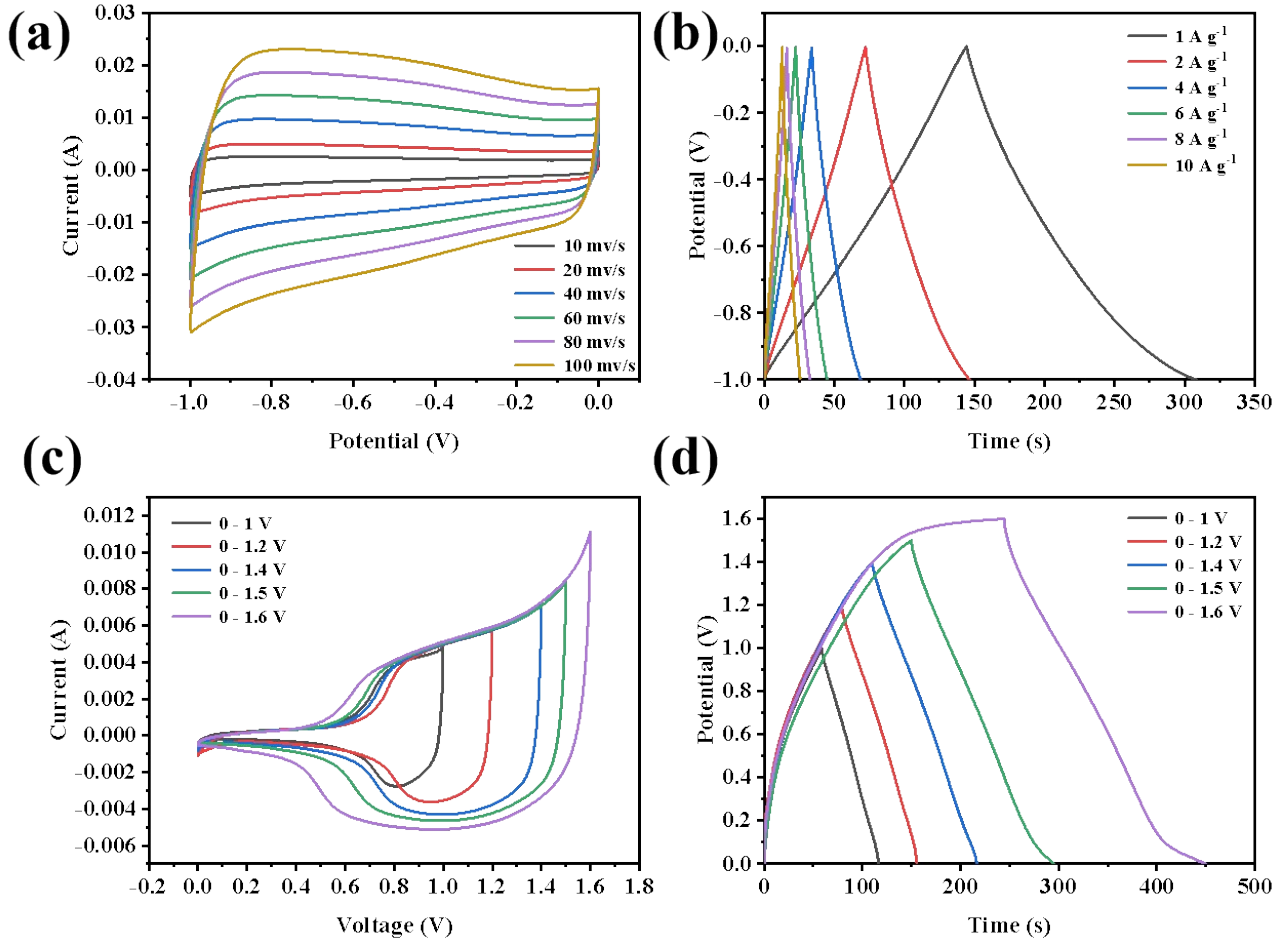


Fig.S1 Full XPS spectrum of $Ni_2Co_1Mo_1S$- $Ti_3C_2T_x$.



FigS2. (a) EIS curves of $Ti_3C_2T_x$, $Ni_2Co_1Mo_1S$ and $Ni_2Co_1Mo_1S$- $Ti_3C_2T_x$ (inset shows the fitted circuit equivalent analysis). (b) GCD curves of $Ni_2Co_1Mo_1S$- $Ti_3C_2T_x$ electrode.



FigS3. (a) CV curves as well as (b) GCD curves of activated carbon. (c) CV curves and (d) GCD curves of $Ni_2Co_1Mo_1S$-$Ti_3C_2T_x$//AC ASC at different voltage windows.

Table S1. Cycling stability of $Ni_2Co_1Mo_1S$-$Ti_3C_2T_x$//AC supercapacitor device compared to other similar devices.

Electrode	Current Density ($A\ g^{-1}$)	Number of Cycles	Retention Rate (%)	Ref.
NiCo-LDH/$Ti_3C_2T_x$//AC	10	5000	73.6	[1]
NiAl-LDH/MXene//AC	3	5000	89.1	[2]
NiGa-LDH/$Ti_3C_2T_x$ MXene// AC	10 mA cm^{-2}	5000	86.2	[3]
Ni-Co-Mo-S/NF//AC	2.5	3500	74.8	[4]
Fe_1Ni_3-LDH/$Ti_3C_2T_x$//Fe_1Ni_3-LDH/$Ti_3C_2T_x$	5	5000	80.8	[5]
NiCoFe-LDH/Ti_3C_2//rGO	5	5000	80.2	[6]
(Ni,Co)Se_2/NiCo-LDH//PC	5	3000	90	[7]
$Ni_2Co_1Mo_1S$-$Ti_3C_2T_x$//AC	10	5000	86.3	This work

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