Supplementary Information for

Flexible MXene/Holey Graphene Films as Multifunctional Electrodes for High-Performance Energy Storage and Pressure Sensing

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Methods

Preparation of few-layer Ti_3CNT_x suspension

The few-layer Ti_3CNT_x suspension is prepared by etching Ti_3AlCN powder with a LiF/HCl mixed solution. Specifically, 4.8 g of LiF was added to a reaction kettle containing 40 mL of 9 M HCl and stirred at room temperature for a few minutes to ensure the full reaction between LiF and HCl. Subsequently, 3 g of Ti_3AlCN powder was slowly added to the reaction kettle, and the mixture was reacted at 30 °C for 24 h. After the reaction, the product was washed by centrifugation multiple times with pure water at a rotation speed of 3500 rpm until the supernatant was neutral. After each centrifugation, water was added to the precipitate and manually shaken to promote the gradual delamination of Ti_3CNT_x. Finally, the upper layer liquid of few-layer Ti_3CNT_x was extracted.

Preparation of porous graphene oxide

Weigh 0.4 g of graphene oxide (GO) and add it to 100 mL of deionized water. Then, perform ultrasonic treatment for 1h to ensure that the graphene oxide can be uniformly dispersed in water, forming a homogeneous dispersion. Next, measure 50 mL from the dispersion prepared above (the concentration of GO at this time is 4 mg mL^{-1}), and slowly add 10 mL of a 30wt% H_2O_2 aqueous solution to it. Place the mixed system in an environment at 100 °C and continuously stir for 5 h to allow H_2O_2 to etch the graphene oxide. After the etching process is completed, let the resulting porous graphene oxide (HGO) aqueous solution cool naturally to room temperature. Then, use high-speed centrifugation to remove the excess H_2O_2 in the solution.

Electrochemical tests

Three-electrode assembly and testing: All electrochemical tests, including cyclic voltammetry (CV), galvanostatic charge - discharge (GCD), and electrochemical impedance spectroscopy (EIS), were conducted using an electrochemical workstation (CHI 660e). To measure the electrochemical performance of the composite film, a three - electrode Swagelok cell was employed. Specifically, the MXene electrode was used as the working electrode, activated carbon as the counter electrode, the Hg/Hg_2SO_4 electrode as the reference electrode, and a 2M H_2SO_4 solution as the electrolyte. In the cyclic voltammetry tests, the voltage window was set from -0.3 V to -0.7 V, and different scan rates (2 - 2000 mV s^{-1}) were selected for the measurements. For the galvanostatic charge - discharge tests, the experiments were carried out within the same voltage window of -0.3 V to -0.7 V at different current densities (0.5 - 20 A g^{-1}). The electrochemical impedance spectroscopy tests were performed under open - circuit potential conditions, with the frequency range set from 0.01 Hz to 100 kHz.

Two-electrode assembly and testing: All electrochemical tests, namely cyclic voltammetry (CV), galvanostatic charge - discharge (GCD), and electrochemical impedance spectroscopy (EIS), were carried out using an electrochemical workstation (CHI 660e). In the experiments, the MXene material was used as both the working electrode and the counter electrode. A 2 M H_2SO_4 aqueous solution and a PVA-H_2SO_4 gel electrolyte were selected as the electrolytes respectively for the tests. In the cyclic voltammetry tests, the voltage window was set from 0 V to 0.7 V, and measurements were taken at different scan rates (5 - 200 mV s^{-1}). The galvanostatic charge - discharge tests were also conducted within the voltage window of 0 V to 0.7 V at different current densities (0.5 - 20 A g^{-1}). The electrochemical impedance spectroscopy tests were performed under open - circuit potential conditions, with the frequency range set from 0.01 Hz to 100 kHz

Pressure sensing test

A glass substrate of appropriate size was washed multiple times with ethanol and acetone solutions. After drying, a copper tape was pasted on the glass substrate. Another identical glass substrate was treated in the same way. The prepared 95-M/rHG film was sandwiched between the two glass substrates with copper tapes, and the composite film was connected to the copper tapes. The copper tapes were used as electrodes and connected to an electrochemical workstation to capture the change in current. Different pressures between 0 - 30 N were applied to the assembled pressure sensor device using a pressure testing machine. The change in the current signal was observed through the electrochemical workstation to evaluate its piezoresistive performance.

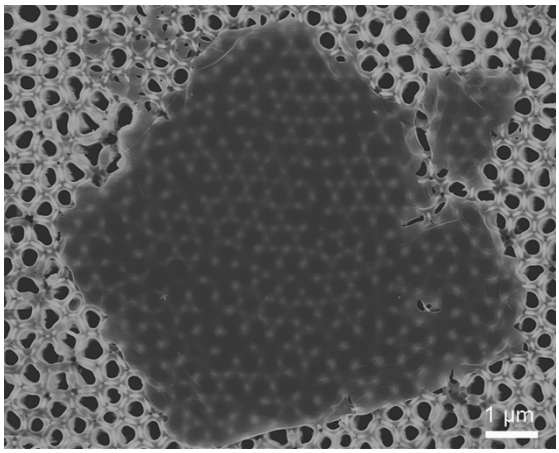

Figure S1. SEM image of few-layer Ti_3CNT_x nanosheets.

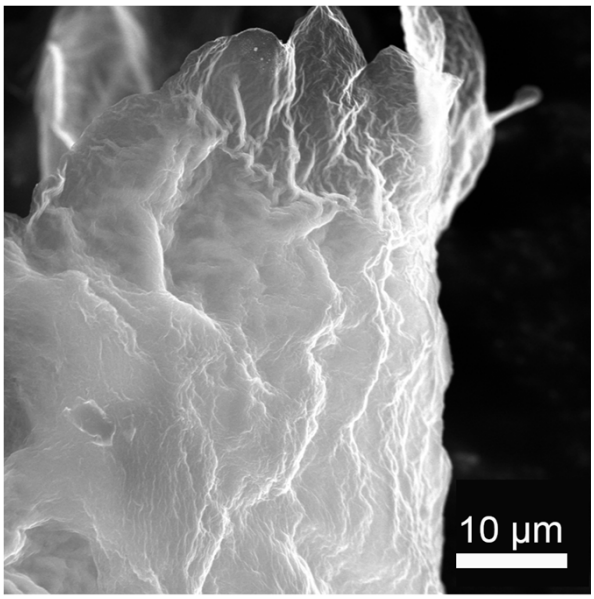

Figure S2. SEM image of GO.

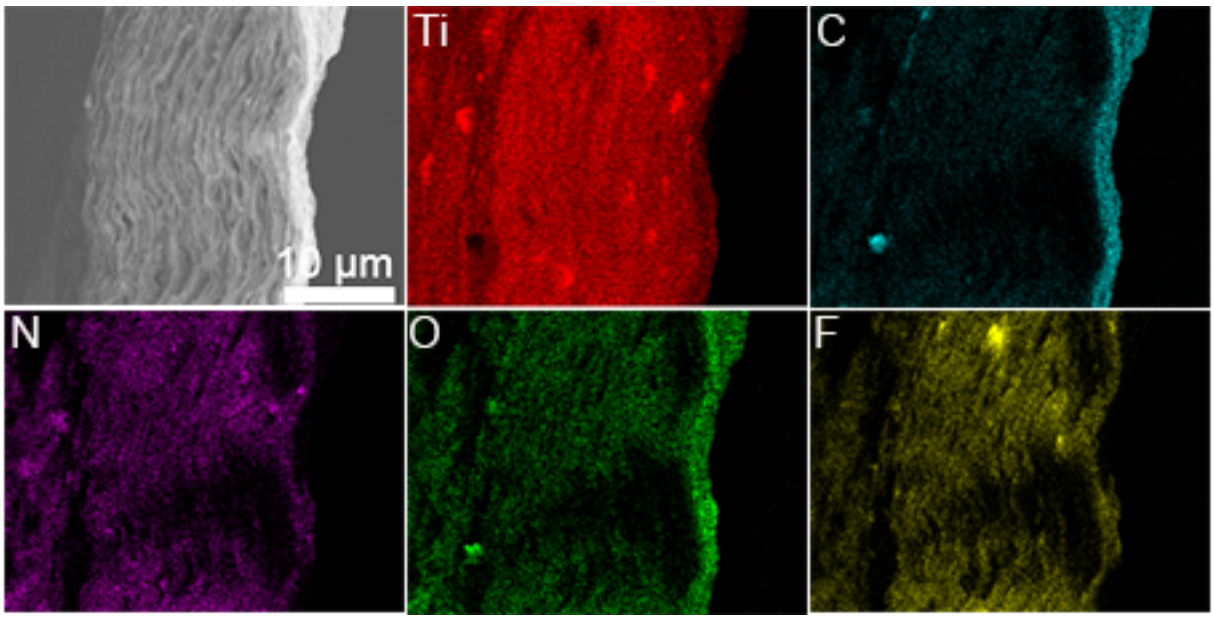

Figure S3. The cross-sectional EDS mappings of 95-M/rHG.

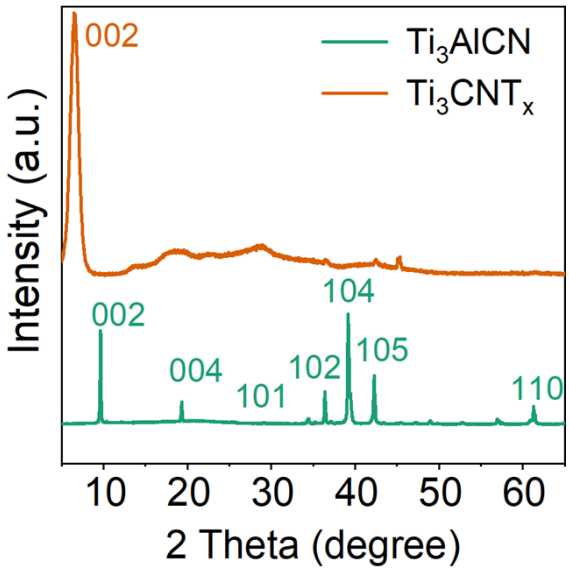

Figure S4. XRD patterns of Ti_3AlCN and Ti_3CNT_x.

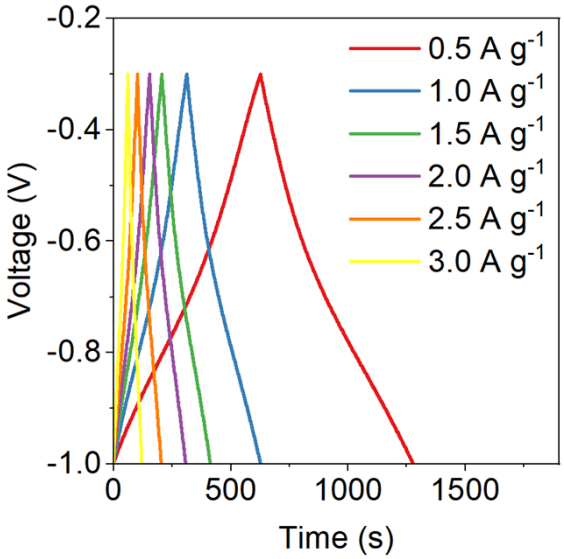

Figure S5. GCD of 95-M/rHG.

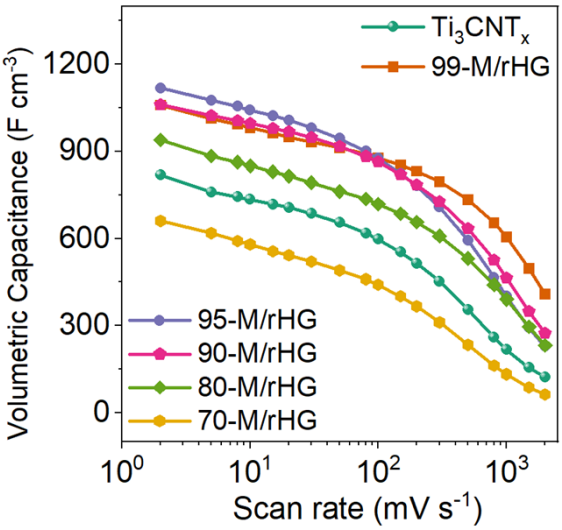

Figure S6. Rate performance of Ti_3CNT_x and x-M/rHG.

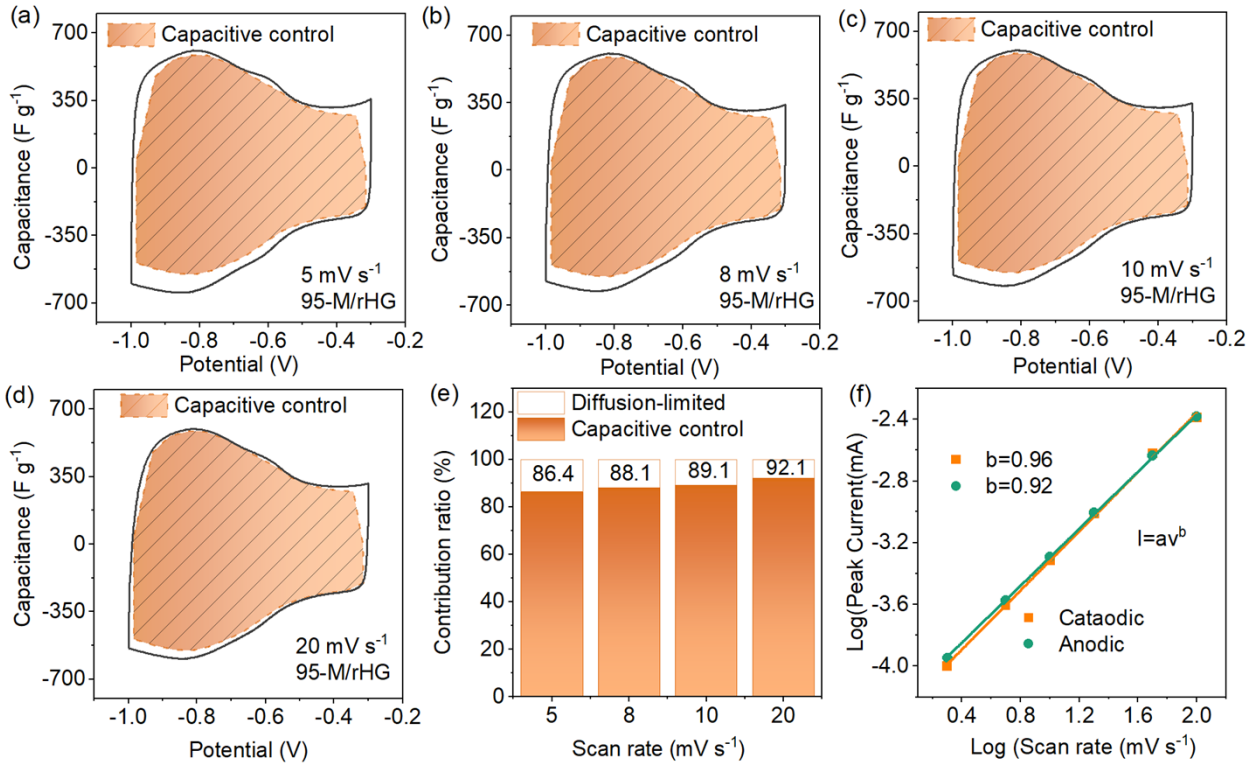

Figure S7. CVs with hatched portions of the capacitive contribution at (a) 5 mV s^{-1}, (b) 8 mV s^{-1}, (c) 10 mV s^{-1}, (d) 20 mV s^{-1}, respectively. (e) Normalized contribution ratio of the capacitive at various scan rates. (f) The power law dependence of current on scan rate for 95-M/rHG.

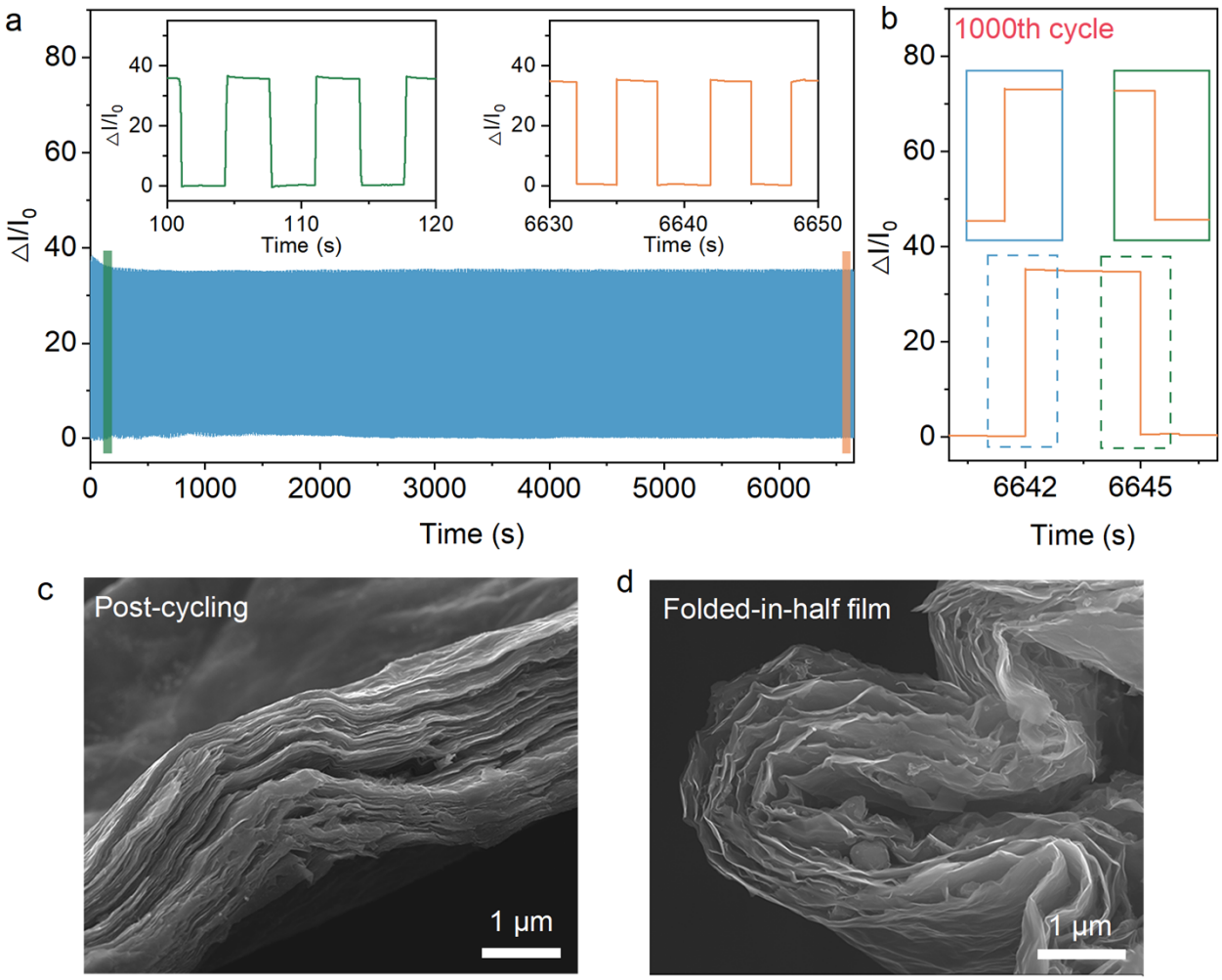

Figure S8. Long-term piezoresistive sensing stability and mechanical durability of the 95-M/rHG. (a) Piezoresistive response of the sensor over 1,000 continuous pressure cycles. (b) Magnified view of the piezoresistive response around the 1,000th cycle. (c) Cross-sectional SEM image of the electrode film after 1,000 pressure cycles. (d) SEM image of the electrode film completely folded in half.

To evaluate the long-term piezoresistive stability, the sensor was subjected to 1,000 continuous pressure cycles, exhibiting no obvious degradation in baseline or peak signals (Fig. S8a, b). Post-cycling cross-sectional SEM (Fig. S8c) confirms the structural resilience of the lamellar framework without plastic deformation. Furthermore, the composite film can be completely folded in half without fracturing (Fig. S8d), verifying its exceptional flexibility and mechanical durability.