Three-dimensional holey MXene hydrogel framework for high rate performance supercapacitor

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INTRODUCTION: Two-dimensional (2D) MXenes show great potential for energy storage due to their exceptional electrical conductivity, large surface area, intrinsic surface reactivity, and easy solution processability.[1] Nonetheless, nanoscale designing of MXene electrode architecture is essential to fully exploit their distinctive attributes and enabling their practical energy storage applications. Herein, we develop a 3D freestanding MXene hydrogel framework (MXH) by controlled electrostatic self-assembly process. The subsequent, mild oxidative etching of the hydrogel leads to the formation of holey MXene hydrogel (HMXH). This HMXH exhibits superior charge-storage and rate performance than MXH.

METHODS: The self-assembly of MXene nanosheets were initiated by divalent cations $[Zn(NH3)4]^{2+}$ at room-temperature, which gives rise to MXH. The subsequent formation of HMXH was achieved by a mild acid-etching of MXH at low temperature.

RESULTS: The as-developed MXH and HMXH have a 3D porous architecture as evident from the morphological characterizations. More importantly, after acid-etching, nanopores can be found on the surface of the MXene sheets, as observed in transmission electron microscopy (TEM) image (Figure 1a). Both MXH and hierarchically porous HMXH were used as electrodes for supercapacitor.

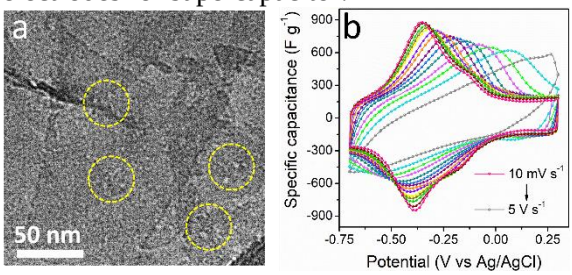

Fig. 1: (a) TEM image and (b) cyclic voltammetry (CV) profiles of HMXH. The circular regions in (a) indicates the region of pores on MXene.

From cyclic voltammetry (CV) data (Figure 1b), we found that HMXH showed improved specific capacitance (359 F g^{-1} at 10 mV s^{-1}) in comparison to pristine MXH (252 F g^{-1}). Moreover, HMHX maintains up to 79% of this capacitance at an ultrafast scan rate of 5000 mV s^{-1}. Even with the commercial mass loading (10.1 mg cm^{-2}) the electrode retains over 52% of its initial capacitance at 1000 mV s^{-1}.

DISCUSSION & CONCLUSIONS: The improved capacitance and charge storage properties of HMXH is attributed to the following factors:

1. The holey MXene framework without extra insulating/low-conducting binders, maintains the high electrical conductivity of the hydrogel.
2. The 3D porous framework also restricts the restacking of MXene nanosheets which helps to maintain high surface area as well.
3. The hierarchically porous structure of the hydrogel electrode with nanopores on the surface of MXene enable 3D transportation of the electrolyte ions, increase the electrochemically active surface area and thereby result in the outstanding capacitive performance.

This study provides a new strategy toward the design principle of MXene based electrodes for practical energy storage systems.

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