## **Supporting Information**

## Interconnected N-doped MXene Spherical Shells for Highly Efficient Capacitive

## Deionization

Gujia Zhang <sup>a</sup>, Luhua Wang <sup>a</sup>, Rongjian Sa <sup>b</sup>, Chao Xu <sup>a</sup>, Zhaohui Li <sup>a</sup> and Lianzhou Wang <sup>c</sup>

 <sup>a</sup> State Key Laboratory of Photocatalysis on Energy and Environment, College of Chemistry, Fuzhou University, Fuzhou, No. 2 Wulongjiang North Avenue, 350108, P.
R. China.

<sup>b</sup> Fujian Key Laboratory of Functional Marine Sensing Materials, Institute of Oceanography, Minjiang University, Fuzhou, 350108, P. R. China.

<sup>c</sup> School of Chemical Engineering and Australian Institute for Bioengineering and Nanotechnology, The University of Queensland, Brisbane, QLD. 4072, Australia.

The capacitive behaviors of electrodes can be studied in detial according to the followed equations. Generally, a power-law relationship between the peak CV current  $(i_p)$  and scan rate (v) is adopted to explain the kinetics of charge storage in the electrodes,

$$i_{\rm p} = av^b \tag{1}$$

where *a* and *b* are variables, and a plot of log *i* versus log *v* should result in a straight line with a slope equal to *b*. The *b*-value provides important information on the charge

storage kinetics: b=1 indicates capacitive storage, while b=0.5 is characteristic for diffusion-limited processes.

The contribution of capacitive and diffusion (intercalation pseudocapacitive) effects was calculated according to the equation

$$i(V) = k_1 v + k_2 v^{1/2}$$
(2)

where i(V),  $k_1v$ ,  $k_2v^{1/2}$  and v are the current at a fixed potential, capacitive and diffusion-controlled currents, and scan rate, respectively.  $k_1$  and  $k_2$  are variable parameters, which change with the variation of the applied voltage. The contribution of capacitive (diffusion) process at a certain scan rate can be determined by calculating the value of  $k_1$  ( $k_2$ ). By plotting  $v^{1/2}$  versus  $i/v^{1/2}$ ,  $k_1$ can be determined from the slope of a straight line. At a specific voltage,  $k_1v$  is the contribution of the pseudocapacitance to the i(V). A series of  $k_1v$  can be calculated under different voltages, which can be utilized to draw the curves of pseudocapacitance. By comparing the integral areas, the contribution of capacitive and diffusion (intercalation pseudocapacitive) can be calculated.



Figure S1. (a) Zeta-potential values of samples. (b) The contents of  $Ti_3C_2T_x$  in core-shell composites.



Fig. S2. FESEM images of N-doped samples with (a) single layer and (b) three layer of  $Ti_3C_2T_x$ 



Fig. S3. (a) XRD patterns of 3DM-S and N-3DM-L samples, (b) Deconvolution of high resolution XPS spectra for Ti, and (c) the XPS spectrum of control N-3DM-L sample



Fig. S4. Nitrogen adsorption-desorption isotherms of (a) N-3DM-S and (b) N-3DM-L.

Mercury penetration curves of (c) N-3DM-S and (d) N-3DM-L.



Fig. S5. Galvanic charging curves of (a) N-3DM-L and (b) 3DM-S electrodes



Fig. S6. Contact angle tests of (a) N-3DM-S and (b) 3DM-S.