

## Supporting Information

### **Converting mesoporous polydopamine coated MIL-125 (Ti) to core-shell heterostructure for efficient water desalination**

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**Chemicals.** Tetra-n-butyl titanate  $\text{Ti}(\text{OC}_4\text{H}_9)_4$  was purchased from Sinopharm Chemical Reagent Co., Ltd. Terephthalic acid ( $\text{H}_2\text{BDC}$ ), dopamine hydrochloride, and 1, 3, 5-trimethylbenzene (TMB) were purchased from Aladdin Industrial Corporation. Pluronic F127 and were purchased from Sigma-Aldrich. Carbon black was purchased from Alfa Aesar. Tris-buffer was purchased from Bio-Rad Laboratories, Inc. Polytetrafluoroethylene (PTFE), anhydrous ethanol, methanol, sodium chloride (NaCl), and N, N-dimethylformamide (DMF) were obtained from Nanjing Chemical Reagent Co., Ltd. All chemicals were used as received without further purification.

**Materials Characterization.** The morphology and structure of the samples were conducted by scanning electron microscopy (SEM, JEOL 7800 system) and transmission electron microscopy (TEM, FEI Tecnai G2 F30 S-Twin). The composition was investigated by X-ray diffraction (XRD, BRUKER D8,  $\text{Cu K}\alpha$ ) at 40 kV and 40 mA ( $\lambda = 1.5418 \text{ \AA}$ ). The  $\text{N}_2$  adsorption-desorption isotherms were collected using the Micromeritics ASAP-2020 instrument. X-ray photoelectron spectroscopy (XPS) spectra were obtained by PHI Quantera II ESCA System with Al  $\text{K}\alpha$  radiation at 1486.8 V. Thermogravimetric analysis (TGA) measurements were conducted by the SDT Q600 thermogravimetry/differential thermal analyzer. Raman spectroscopy was conducted using the Renishaw in Via reflex spectrometer system. Fourier transform infrared (FTIR) spectra of the samples were obtained using FT-IR spectrometer (Bruker HYPERION, Germany).

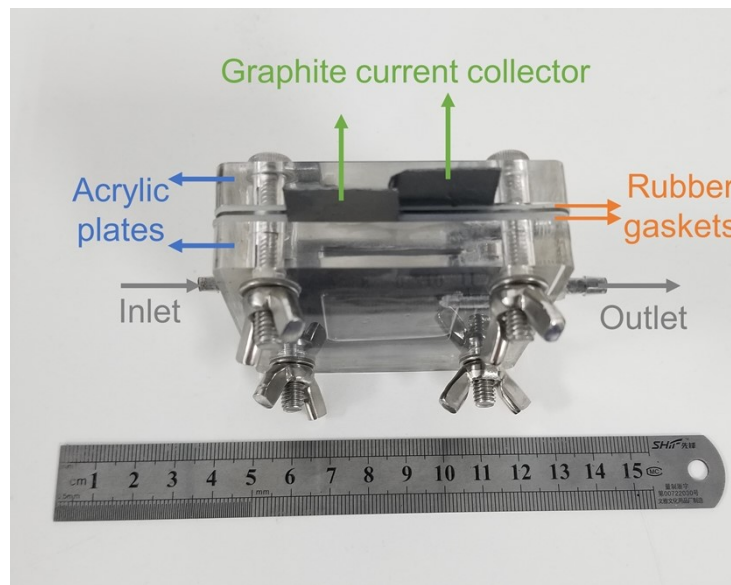


Fig. S1. The digital photograph of HCDI device.

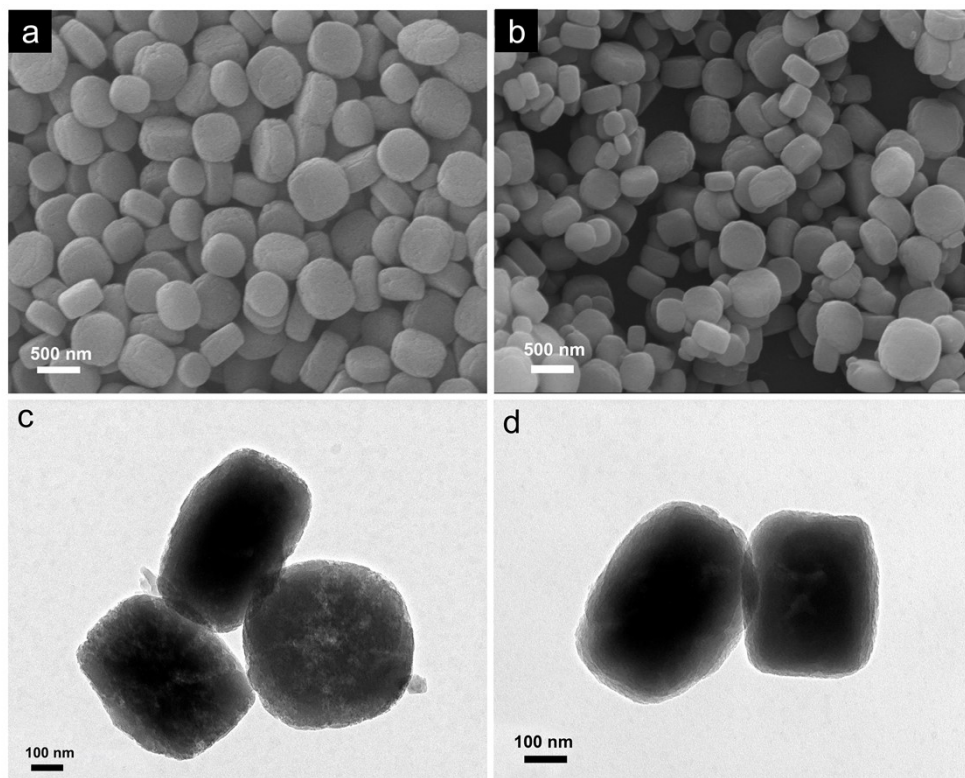


Fig. S2. (a) SEM image and (c) TEM image of MIL-125. (b) SEM image and (d) TEM image of MIL-125@PDA.

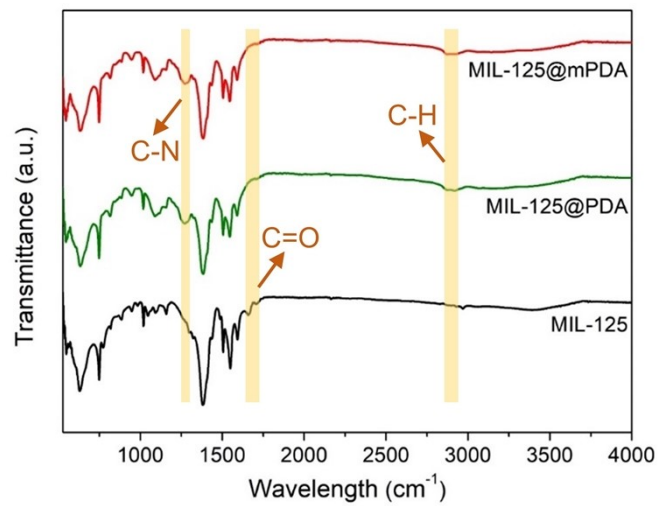


Fig. S3. Fourier transform infrared (FTIR) spectra of MIL-125, MIL-125@PDA, and MIL-125@mPDA.

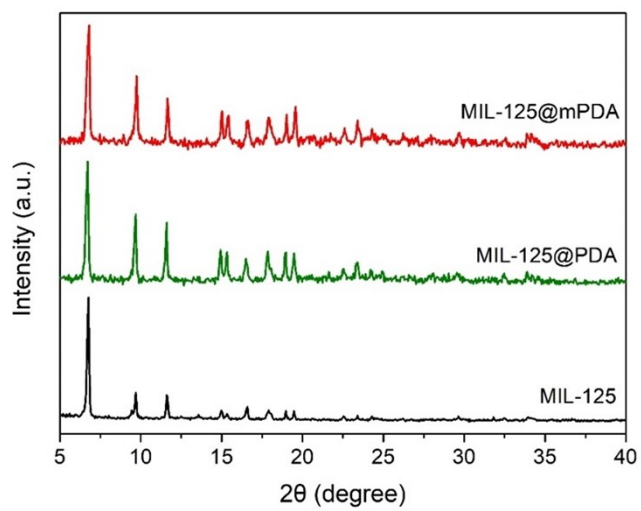


Fig. S4. X-ray diffraction (XRD) patterns of MIL-125, MIL-125@PDA, and MIL-125@mPDA.

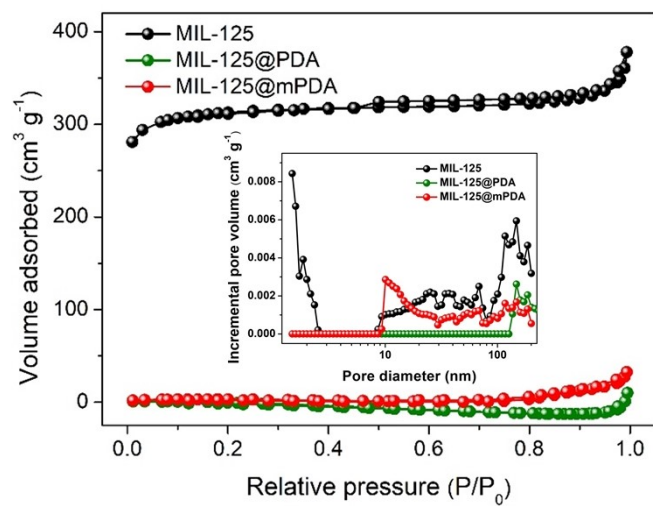


Fig. S5. N<sub>2</sub> adsorption/desorption isotherms and pore size distributions (insert) of MIL-125, MIL-125@PDA, and MIL-125@mPDA.

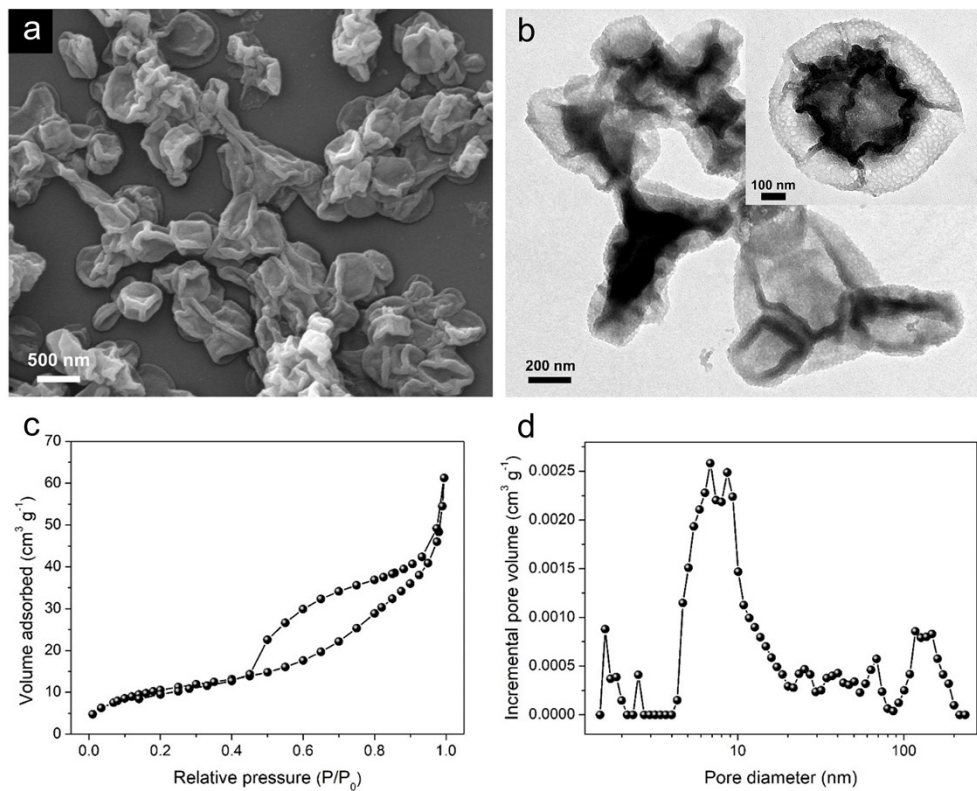


Fig. S6. (a) SEM image, (b) TEM image, (c) N<sub>2</sub> adsorption/desorption isotherm, and (d) pore size distribution of the mesostructured PDA shell after the alkali etching of MIL-125 core.



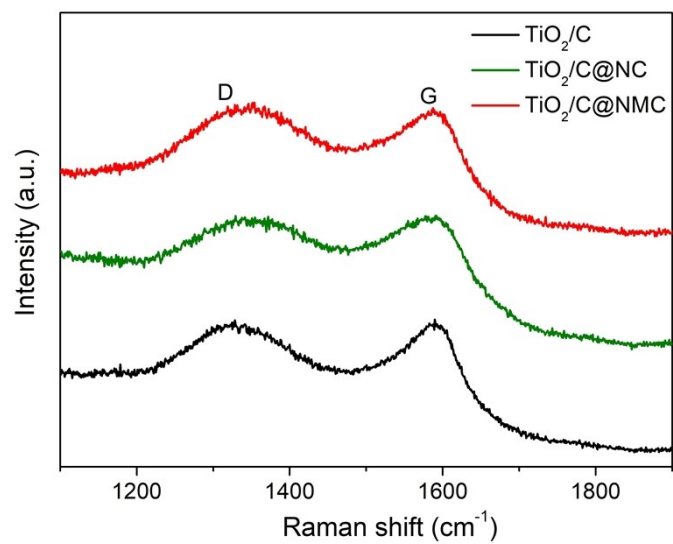


Fig. S7. Raman spectra ranging from 1100 cm<sup>-1</sup> and 1900 cm<sup>-1</sup> of TiO<sub>2</sub>/C, TiO<sub>2</sub>/C@NC, and TiO<sub>2</sub>/C@NMC.

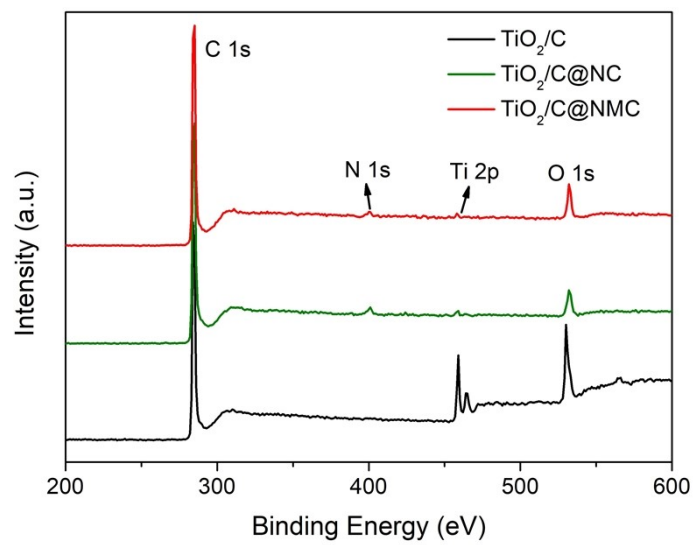


Fig. S8. XPS full surveys of  $\text{TiO}_2/\text{C}$ ,  $\text{TiO}_2/\text{C@NC}$ , and  $\text{TiO}_2/\text{C@NMC}$ .

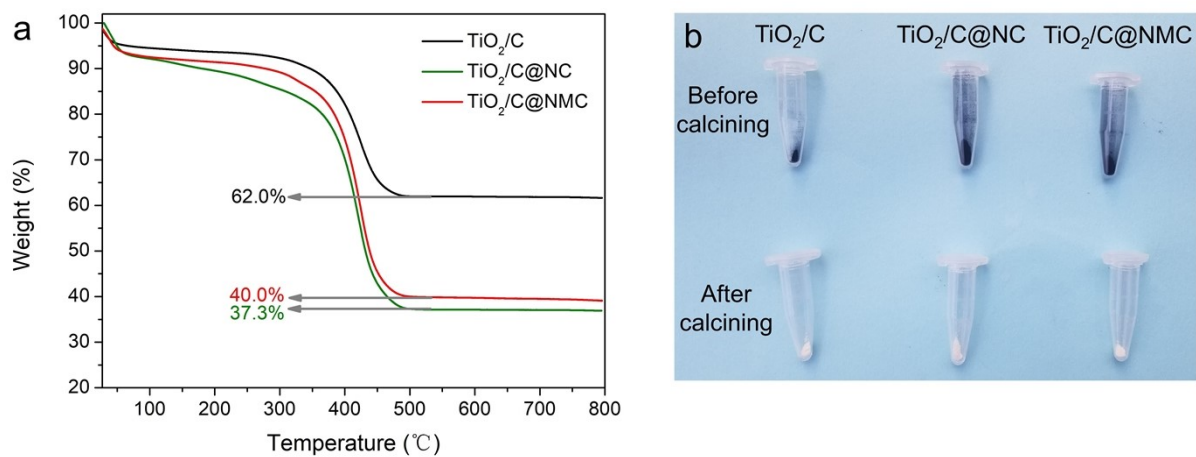


Fig. S9. (a) TGA curves of TiO<sub>2</sub>/C, TiO<sub>2</sub>/C@NC, and TiO<sub>2</sub>/C@NMC tested in air atmosphere with a heating rate of 10 °C min<sup>-1</sup> and (b) the digital photograph of TiO<sub>2</sub>/C, TiO<sub>2</sub>/C@NC, and TiO<sub>2</sub>/C@NMC before and after calcining.

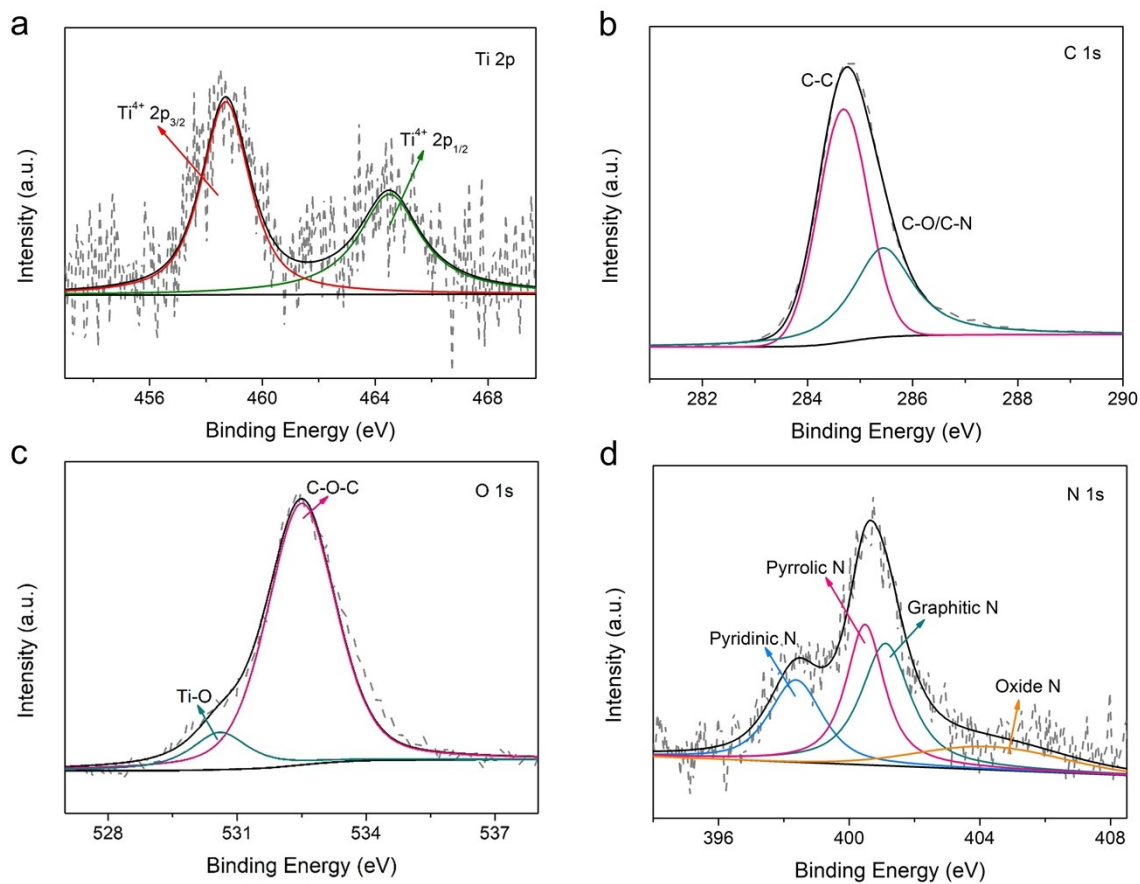


Fig. S10. (a) Ti 2p spectra, (b) C 1s spectra, (c) O 1s, and (d) N 1s of  $\text{TiO}_2/\text{C}@/\text{NMC}$ .

Table S1. Structural parameters and elemental compositions of TiO<sub>2</sub>/C, TiO<sub>2</sub>/C@NC, and TiO<sub>2</sub>/C@NMC.

Sample	$S_{\text{BET}}$ (m <sup>2</sup> g <sup>-1</sup> )	$V_{\text{pore}}$ (cm <sup>3</sup> g <sup>-1</sup> )	Surface elemental composition (%)				Ti content calculated by TGA (%)
			C	O	Ti	N	
TiO <sub>2</sub> /C	330.6	0.22	79.9	14.6	5.5	/	37.1
TiO <sub>2</sub> /C@NC	189.7	0.09	89.4	6.3	0.8	3.5	22.3
TiO <sub>2</sub> /C@NMC	202.3	0.12	89.7	7.3	0.7	2.3	24.0

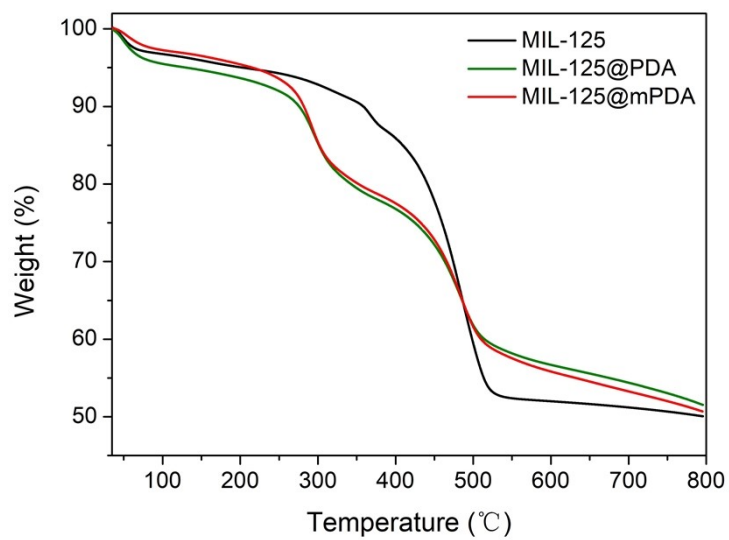


Fig. S11. TGA curves of MIL-125, MIL-125@PDA, and MIL-125@mPDA tested in N<sub>2</sub> atmosphere with a heating rate of 5 °C min<sup>-1</sup>.

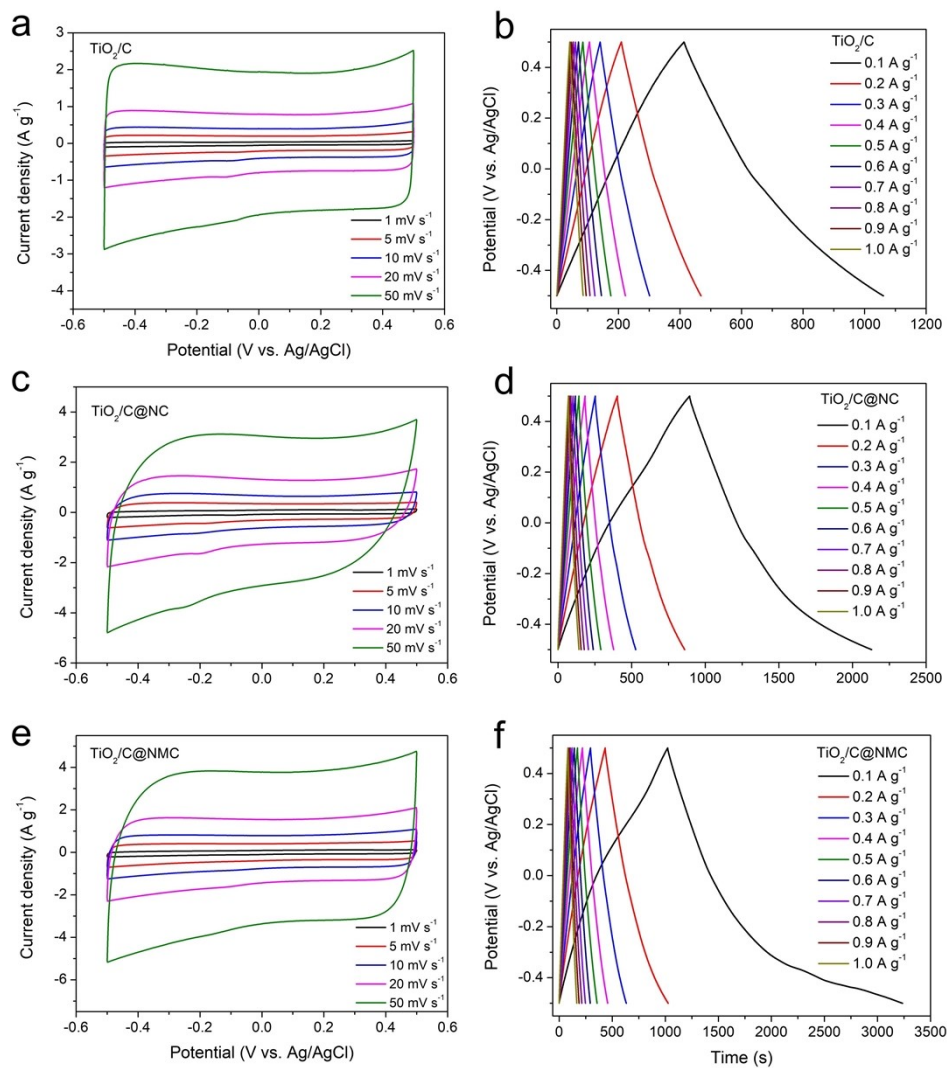


Fig. S12. CV curves and GCD plots of (a, b)  $\text{TiO}_2/\text{C}$ , (c, d)  $\text{TiO}_2/\text{C}@\text{NC}$ , and (e, f)  $\text{TiO}_2/\text{C}@\text{NMC}$  at different scan rates and current densities in 1 M NaCl solution.

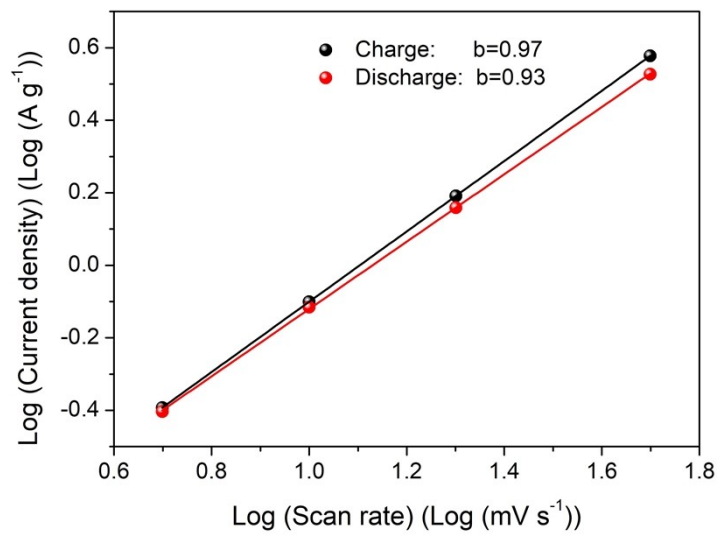


Fig. S13. The  $b$  values determined by using the relationship between the current density and the scan rate.



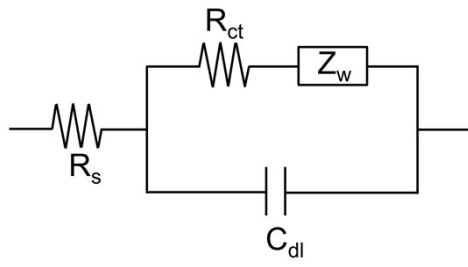


Fig. S14. The equivalent circuit diagram of EIS.

Table S2.  $R_s$  and  $R_{ct}$  of  $\text{TiO}_2/\text{C}$ ,  $\text{TiO}_2/\text{C}@\text{NC}$ , and  $\text{TiO}_2/\text{C}@\text{NMC}$  electrodes.

Sample	$R_s$ ( $\Omega$ )	$R_{ct}$ ( $\Omega$ )
$\text{TiO}_2/\text{C}$	2.81	3.56
$\text{TiO}_2/\text{C}@\text{NC}$	2.84	3.33
$\text{TiO}_2/\text{C}@\text{NMC}$	2.71	2.72

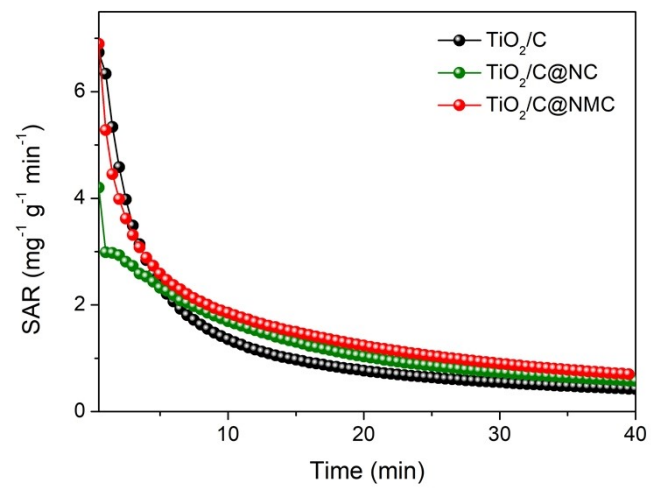


Fig. S15. The SAR variations versus time f TiO<sub>2</sub>/C, TiO<sub>2</sub>/C@NC, and TiO<sub>2</sub>/C@NMC.

Table S3. The charge efficiency ( $\mathcal{A}$ ) and energy consumption ( $E$ ) of  $\text{TiO}_2/\text{C}$ ,  $\text{TiO}_2/\text{C}@\text{NC}$ , and  $\text{TiO}_2/\text{C}@\text{NMC}$ .

Sample	Charge efficiency (%)	Energy consumption ( $\text{kWh kg}_{\text{NaCl}}^{-1}$ )
$\text{TiO}_2/\text{C}$	73.1	0.75
$\text{TiO}_2/\text{C}@\text{NC}$	79.4	0.69
$\text{TiO}_2/\text{C}@\text{NMC}$	87.3	0.61

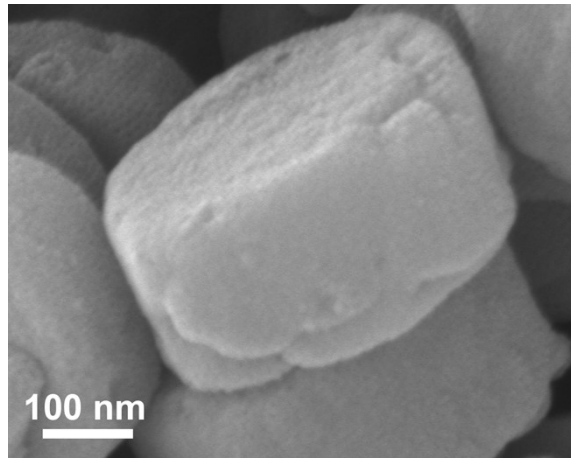


Fig. S16. SEM image of TiO<sub>2</sub>/C@NMC after regeneration tests.

Table S4. Comparison of TiO<sub>2</sub>/C@NMC and other reported electrode materials.

Electrode materials	Voltage (V)	NaCl concentration (mg L <sup>-1</sup> )	SAC (mg g <sup>-1</sup> )	Ref.
rGO/T2	1.2	300	16.4	1
rGO-15TiO <sub>2</sub>	1.2	75	24.58	2
TiO <sub>2</sub> @CNTs	1.4	25-800	4.0	3
TiO <sub>2</sub> /carbon	1.2	280	17.4	4
MoS <sub>2</sub> /graphene	1.2	500	19.4	5
MnO <sub>x</sub> nanofiber	1.2	877	27.8	6
Open and interconnected porous architectures	1.2	500	14.35	7
Nitrogen-doped activated carbon	1.2	468	24.7	8
Sugarcane Biowaste-Derived Biochars	1.2	600	21.8	9
ZIF-8@PZS-C	1.2	500	22.19	10
TiO <sub>2</sub> @COF-2	1.4	200	26.0	11
TiO <sub>2</sub> /C@NMC	1.2	250	27.73	This work
TiO <sub>2</sub> /C@NMC	1.6	250	35.54	This work

## References

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