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Electronic Supplementary Information

Boosting Deionization Capability by Effectively Improving Sodium-ion Storage

Capacity Based on Robust Interfacial Electronic Interaction within 3D

Na₄Ti₉O₂₀/N-doped Porous Carbon Heterostructures

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Experimental

Synthesis of graphene oxide (GO): Typically, 2 g graphite powder and 1 g NaNO₃ were added into a beaker with 50 mL H₂SO₄ successively. After the beaker was continuously stirred under the ice bath for 15 minutes, 5 g of KMnO₄ was slowly added and kept stirring under the ice bath for 2 hours. After the beaker rose to room temperature, it kept stirring under 40 °C for 2 hours. Then 100 mL DI was added into the beaker and continue the reaction for 30 minutes. Afterwards, 100 mL boiled DI was also added into the beaker and kept stirring for 10 minutes, following moderate hydrogen peroxide (H2O2) was added and stood for several hours. The precipitate was washed with DI by using centrifuge until the PH equals to 7. Finally, the precipitate was performed ultrasound and centrifuged for three times to obtain GO flakes.

Synthesis of Ag@rGO: 0.0472 g silver nitrate (AgNO₃) was added into a beaker with 60 mL GO (~1 mg/L) and kept ultrasound for 1 hour. Afterwards, a beaker with 15 ml sodium hydroxide (NaOH, 4 mol L⁻¹) was kept stirring under 85 °C and accompanied by adding the solution above drop by drop. Finally, the beaker was kept stirring for 30 minutes. The black precipitate was washed with DI by using centrifuge until the PH equals to 7.

Captions

Fig.S1. XRD pattern of NH₂-MIL-125 (a) and TiO₂/N-C (b). TEM images (c-e) and HR-TEM images (f) of 3D NTO/N-C-2.

Fig.S2. SEM images of NTO/N-C-1 (a,b), NTO/N-C-3 (c,d), NTO/N-C-4 (e,f).

Fig.S3. GCD curves of 3D NTO/N-C-x (x = 1, 2, 3, 4) at different current densities.

Fig.S4. XPS survey spectrum of NTO-2 and 3D NTO/N-C-2 (a). High-resolution spectra for N 1s (b) and C 1s (c) of 3D NTO/N-C-2. SEM images of GO (d) and Ag@rGO (e). XRD patterns for GO, rGO and Ag@rGO (f).

Fig.S5. XPS survey spectrum for GO and Ag@rGO (a). CV curves of 3D NTO/N-C-2 (b) and NTO-2 (c) at various scanning speeds. Linear relationship between the currents related to Na insertion/extraction in NTO-2 and scan rates in logarithmic format (d). GCD curves of NTO-2 at different current densities (e). EIS image of NTO-2 and 3D NTO/N-C-2 (f).

Fig.S6. CV curves of Ag@rGO at 40 mV·s⁻¹ (a). Conductivity of the desalination battery with different mass ratio of 3D NTO/N-C-2 versus Ag@rGO in NaCl solutions with 1000 mg·L⁻¹ at 1.4 V (b). The linear relationship between the conductivity and the concentration of NaCl solution (c). Desalination capacity vs time (d) and corresponding Ragone plots of SAR vs SAC (e) of the desalination battery with different mass ratio of 3D NTO/N-C-2 versus Ag@rGO in NaCl solutions with 1000 mg·L⁻¹ at 1.4 V. Transient conductivity of 3D NTO/N-C-2 electrodes in a 1000 mg·L⁻¹ NaCl solution at 1.0, 1.2, 1.4 V constant voltage (f).

Fig.S7. Schematic illustration of the desalination cell configuration.

Fig.S8. Conductivity transients of the desalination system at different initial concentration of NaCl solution (a-f).

TabS1. Surface composition of the samples derived from XPS analysis (in atomic %).



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Fig.S6. CV curves of Ag@rGO at 40 mV·s⁻¹ (a). Conductivity of the desalination battery with different mass ratio of 3D NTO/N-C-2 versus Ag@rGO in NaCl solutions with 1000 mg·L⁻¹ at 1.4 V (b). The linear relationship between the conductivity and the concentration of NaCl solution (c). Desalination capacity vs time (d) and corresponding Ragone plots of SAR vs SAC (e) of the desalination battery with different mass ratio of 3D NTO/N-C-2 versus Ag@rGO in NaCl solutions with 1000 mg·L⁻¹ at 1.4 V. Transient conductivity of 3D NTO/N-C-2 electrodes in a 1000 mg·L⁻¹ NaCl solution at 1.0, 1.2, 1.4 V constant voltage (f).



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Fig.S8. Conductivity transients of the desalination system at different initial concentration of NaCl solution (a-f).

TabS1. Surface composition of the samples derived from XPS analysis (in atomic %).

Sample	С	0	Ti	Na	Ν
3D NTO/N-C	46.29	34.51	11.62	5.86	1.72
NTO-2	31.34	44.59	15.95	8.13	0