Supplementary Material

3D ordered honeycomb-like nitrogen-doped micro-mesoporous carbon for brackish water desalination using capacitive deionization
Materials and methods

Materials


Methods

Synthesis polystyrene spheres (PS) and 3D ordered PS template

The polystyrene spheres (PS) have an average diameter of ~100 nm. Firstly, 65mL of styrene was first washed thoroughly with 20 mL of 10 wt% NaOH solution and washed with deionized water for several times. Then all washed styrene was added into a mixture of deionized water (500 mL) and PVP (2.5 g). Next, the solution was bubbled with nitrogen for 15 min, the mixture was stirred magnetically at 75 ℃ for 30 min. Then, 50 mL of aqueous solution containing 1g K$_2$S$_2$O$_8$ was quickly added into the mixture, which was kept stirring for 24h at 75 ℃.

3D ordered PS template was simply achieved by filtration. First, the PS colloidal solution was poured on to a filter funnel with traditional vacuum filtration. After 24 h, burly filter cakes were obtained, which were further dried at 60 ℃ overnight.

Synthesis of 3D ZIF-8@PS template and 3D ZIF-8

The 3D ZIF-8@PS template was synthesis by typical method. First, put a piece of PS template into 45 mL of methanol solution with dissolved 8.15 g of Zn(NO$_3$)$_2$•6H$_2$O and 6.75 g of 2-methylimidazole, and kept 1 h at room temperature. Then the solution was treated with vacuum degassing for 10 min. After drying at 50 ℃ for several hours, white solid was obtained. Then the obtained white solid was immersed into a mixed solution with 20 mL of
CH₃OH and 20 mL of NH₃·H₂O at room temperature and further treated with vacuum degassing for 3 min. Consequently, put this mixture at room temperature and atmospheric pressure for 24 h. Then the obtained solid was soaked with tetrahydrofuran and repeated several times to ensure the complete removal of PS template. Finally, the obtained white powder was dried at 110 °C overnight, named as 3D ZIF-8.

**Synthesis of 3D ordered Hive-like nitrogen-doped micro-mesoporous carbon (3D HPC)**

The 3D HPC was synthesized by traditional methods. Successfully, the temperature was steadily increased from room temperature to 800 °C at a rate of 3 °C/min and kept for 3 h. The obtained product was washed with aqueous hydrochloric acid for 12 hours, and then washed with deionized water to neutrality, named 3D HPC. Besides, the 3D HPC-700 and 3D HPC-900 were obtained at pyrolysis temperature of 700 °C and 900 °C.

**Synthesis of ZIF-8 and porous carbon (PC)**

The preparation method of ZIF-8 was the same as that of 3D HPC, except that there was no addition of PS template and no vacuum degassing treatment. The PC was obtained by pyrolysis of ZIF-8 at 800 °C at a rate of 3 °C/min and kept for 3 h.
Fig. S1 (a), (b), (c) SEM image of PC. (d), (e) TEM image of PC. (f), (g) HRTEM image of PC.
Fig. S2 (a), (b) HRTEM image of 3D HPC.
Fig. S3 (a) CV curves at a scan rate of 1-150 mV s\(^{-1}\) of PC electrode and (b) AC electrode. (c) Galvanostatic charge-discharge curves at current densities of 0.5 to 10 A g\(^{-1}\) of PC and (b) AC electrode. All curves were obtained in 1 M NaCl solution.
Fig. S4 (a), (b) Water contact angles of the PC and 3D HPC.
Fig. S5 (a), (b) SEM image of 3D HPC-700 and 3D HPC-900.

Fig. S6 SEM image of 3D HPC after 20 cycles of CDI desalination tests.

Fig. S7 XRD patterns of ZIF-8@PS and PC.
Fig. S8 XRD patterns of ZIF-8.

Fig. S9 Nitrogen adsorption desorption isotherms of 3D ZIF-8@PS and ZIF-8.
Fig. S10 XPS survey spectrum for 3D HPC-700 and 3D HPC-900.

Fig. S11 Thermogravimetric analysis (TGA) plots of 3D ZIF-8@PS.
Fig. S12 CV curves at a scan rate of 1-150 mV s\(^{-1}\) of 3D HPC electrode in 500 mg L\(^{-1}\) NaCl solution (inset showed the specific capacitance of 3D HPC at different scan rate).

Fig. S13 3D HPC’s Long-term galvanostatic charge-discharge curves at current densities of 10 A g\(^{-1}\).
Fig. S14 Variation of solution conductivity vs. time of 3D HPC-700 and 3D HPC-900 at 1.2 V with an initial concentration of 500 mg/L.

Fig. S15 The changes of SAC (mg g⁻¹) at different concentration of NaCl solution.
Fig. S16 Nitrogen adsorption desorption isotherms of 3D HPC-700 and 3D HPC-900.

Table S1. Comparison between samples and other carbon electrodes.

<table>
<thead>
<tr>
<th>Electrode material</th>
<th>Applied voltage (V)</th>
<th>Initial concentration (mg·L⁻¹)</th>
<th>Adsorption capacity (mg·g⁻¹)</th>
<th>Ref.</th>
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<tr>
<td>N-HMCSs</td>
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<tr>
<td>(N, Ti)-HMCs</td>
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Table S2 The BET specific surface area and nitrogen content calculated by XPS of 3D HPC-700 and 3D HPC-900.

<table>
<thead>
<tr>
<th>Electrode material</th>
<th>$S_{\text{BET}}$ (m²·g⁻¹)</th>
<th>Nitrogen content (%)</th>
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<tr>
<td>3D HPC-700</td>
<td>742</td>
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<td>3D HPC</td>
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<td>3D HPC-900</td>
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References


