Electronic Supplementary Information (ESI)

Enhanced capacitive deionization of saline water using N-doped rod-like porous carbon derived from dual-ligand metal-organic frameworks

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Fig. S1 SEM images of the prepared (a) PC, (b) SNPC, and (c) ENPC; and TEM images of (d) PC, (e) SNPC, and (f) ENPC.



Fig. S2 Full XPS spectra of MNPC. The inset is the magnified area.



Fig. S3 High-resolution N1s spectrum of the SNPC and ENPC samples.



Fig. S4 Optical micrographs of the water contact angles on the surface of PC, SNPC, MNPC and ENPC electrodes as a function of contact time.

We tested the wettability of CDI electrodes. For PC, SNPC, MNPC and ENPC, we used the same electrode preparation method. The amount of binder added in the four electrodes was exactly the same, which is 10%. The final coating area of all the CDI electrodes was $60 \text{mm} \times 62 \text{mm}$. We made multiple measurements for every pair of electrodes, and finally get more reliable wettability data.



Fig. S5 Cyclic voltammograms of (a) PC, (b) SNPC, (c) MNPC and (d) ENPC electrodes at various scan rates. All the curves were obtained in a 0.5 M NaCl solution.



Fig. S6 Cyclic voltammograms of (a) various electrodes at a scan rates of 1 mV s⁻¹ and (b) MNPC electrodes at various scan rates in a 500 mg L^{-1} NaCl solution.



Fig. S7 GCD curves at various current densities of MNPC.



Fig. S8 Salt removal rates of PC, SNPC, MNPC and ENPC in a 50 mg L^{-1} NaCl solution at 1.2 V.



Fig. S9 Charge efficiency of MNPC electrodes in different concentration of NaCl solution at 1.2 V.

Samples	S _{BET}	S _{micro}	S_{micro}/S_{BET}	V _{pore}	V _{micro}	V _{micro} /V _{pore}
	$(m^2 g^{-1})$	$(m^2 g^{-1})$	(%)	$(cm^3 g^{-1})$	$(cm^3 g^{-1})$	(%)
PC	1185	582	49.1	0.930	0.258	27.7
SNPC	1198	723	60.3	0.905	0.309	34.1
MNPC	962	463	48.1	0.838	0.199	23.7
ENPC	356	38	10.8	0.302	0.015	5.0

 Table S1 Specific surface area and pore volume for the investigated samples.

Electrode materials	scan rate	NaCl	Specific capacitance	Ref.
	[mV s ⁻¹]	concentration	[F g ⁻¹]	
		[mol L ⁻¹]		
Dodecahedron-like	1	0.5	215.3	1
Carbon Frameworks				
Nitrogen-Doped	1	1	179	2
Hollow Mesoporous				
Carbon Spheres				
nitrogen-doped	1	1	199.0	3
cluster-like porous				
carbons				
PVDF-derived	1	1	198.9	4
porous carbon				
heterostructure with				
inserted carbon				
nanotube				
Boron-nitride-carbon	2	1	82.2	5
nanosheets				
Nitrogen-rich	1	1	221.3	6
microporous carbon				
free-standing	1	1	83	7
activated carbon				
electrodes				
MNPC	1	0.5	277.7	This work

 Table S2 Comparison of Specific capacitance of reported carbon materials.

Electrode materials	Applied	Applied Initial NaCl		Ref.
	voltage	concentration	percentage / %	
	[V]	[g L ⁻¹]		
carbon-based cell	1.0	0.5	19.2	8
Flow-electrode	2.4	10	75.6	9
sulfonated carbon	1.3	1	45.2	10
nanotubes				
Flow-electrodes	1.2	0.2	61	11
RGO and AC	1.2	0.05	55	12
composite				
MNPC	1.2	0.05	83	This work

Table S3 Comparison of salt removal percentage of reported carbon materials.

Reference

- Z. Wang, T. Yan, L. Shi and D. Zhang, In Situ Expanding Pores of Dodecahedron-like Carbon Frameworks Derived from MOFs for Enhanced Capacitive Deionization, ACS Appl. Mater. Interfaces, 2017, 9, 15068-15078.
- Y. Li, J. Qi, J. Li, J. Shen, Y. Liu, X. Sun, J. Shen, W. Han and L. Wang, Nitrogen-doped hollow mesoporous carbon spheres for efficient water desalination by capacitive deionization, *ACS Sustainable Chem. Eng.*, 2017, 5, 6635-6644.
- Y. Li, Y. Liu, J. Shen, J. Qi, J. Li, X. Sun, J. Shen, W. Han and L. Wang, Design of nitrogen-doped clusterlike porous carbons with hierarchical hollow nanoarchitecture and their enhanced performance in capacitive deionization, *Desalination*, 2018, 430, 45-55.
- 4. Y. Li, J. Qi, W. Zhang, M. Zhang and J. Li, Fabrication of polyvinylidene fluoride-derived porous carbon heterostructure with inserted carbon nanotube via phase-inversion coupled with annealing for capacitive deionization application. *J. Colloid Interface Sci.*, 2019, **554**, 353-361.
- 5. Y. Zhang, G. Wang, S. Wang, J. Wang and J. Qiu, Boron-nitride-carbon nanosheets with different pore structure and surface properties for capacitive deionization. *J. Colloid Interface Sci.*, 2019, **552**, 604-612.
- 6. D. Li, X.-a. Ning, Y. Huang and S. Li, Nitrogen-rich microporous carbon materials for high-performance membrane capacitive deionization. *Electrochim. Acta*, 2019, **312**, 251-262.
- 7. L. Wu, M. Liu, S. Huo, X. Zang, M. Xu, W. Ni, Z. Yang and Y.-M. Yan, Mold-casting prepared free-standing activated carbon electrodes for capacitive deionization. *Carbon*, 2019, **149**, 627-636.
- 8. O. u. Haq, D.-S. Choi, J.-H. Choi and Y.-S. Lee, Carbon electrodes with ionic functional groups for enhanced capacitive deionization performance. *J. Ind. Eng. Chem.*, 2019, DOI: 10.1016/j.jiec.2019.11.021.
- 9. J. Chang, F. Duan, H. Cao, K. Tang, C. Su and Y. Li, Superiority of a novel flow-electrode capacitive deionization (FCDI) based on a battery material at high applied voltage. *Desalination*, 2019, **468**, 114080.
- Y. Cai, X. Zhao, Y. Wang, D. Ma and S. Xu, Enhanced desalination performance utilizing sulfonated carbon nanotube in the flow-electrode capacitive deionization process. *Sep. Purif. Technol.*, 2019, DOI: 10.1016/j.seppur.2019.116381, 116381.
- 11. S.-i. Jeon, H.-r. Park, J.-g. Yeo, S. Yang, C. H. Cho, M. H. Han and D. K. Kim, Desalination via a new membrane capacitive deionization process utilizing flow-electrode. *Energy Environ. Sci.*, 2013, **6**, 1471.
- 12. H. Li, L. Pan, C. Nie, Y. Liu and Z. Sun, Reduced graphene oxide and activated carbon composites for capacitive Deionization. J. Mater. Chem., 2012, 22, 15556-15561.