

Supporting Information

Hierarchical structure N, O co-doped porous carbon/carbon nanotubes composite derived from coal for supercapacitors and CO₂ capture

Jian Hao^{*a}, Xiu Wang^a, Yanxia Wang^a, Xiaoyong Lai^a, Qingjie Guo^a, Jiupeng Zhao^b, Yu Yang^b, Yao Li^c

^a. State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering, College of Chemistry & Chemical Engineering, Ningxia University, Yinchuan, 750021, China. E-mail: haojian@nxu.edu.cn.

^b. School of Chemical Engineering and Technology, Harbin Institute of Technology, 150001, Harbin, China.

^c. Centre for Composite Material, Harbin Institute of Technology, Harbin, 150001, China.

Experimental Section

Electrochemical data calculation

The gravimetric specific capacitances (C_m) based on galvanostatic charge-discharge (GCD) for the three-electrode and two-electrode systems were calculated according to Eqs. (1) and (2), respectively:

$$C = I t / m \Delta V \quad (1)$$

$$C = 2 I t / m \Delta V \quad (2)$$

where I is the discharge current (A), t is the discharge time (s), m is the mass of the active material (g) in a single electrode, and ΔV is the discharge potential range (V).

In the two-electrode device, the energy density (E , Wh kg⁻¹) and power density (P , W kg⁻¹) were calculated from discharge curves at various current densities according to Eqs. (3) and (4),

respectively:

$$E = C (\Delta V)^2 / 8 * 3.6 \quad (3)$$

$$P = 3600E/t \quad (4)$$

Where ΔV is the working voltage after ohmic drop (V) and t is the discharge time (s).

Results and discussion

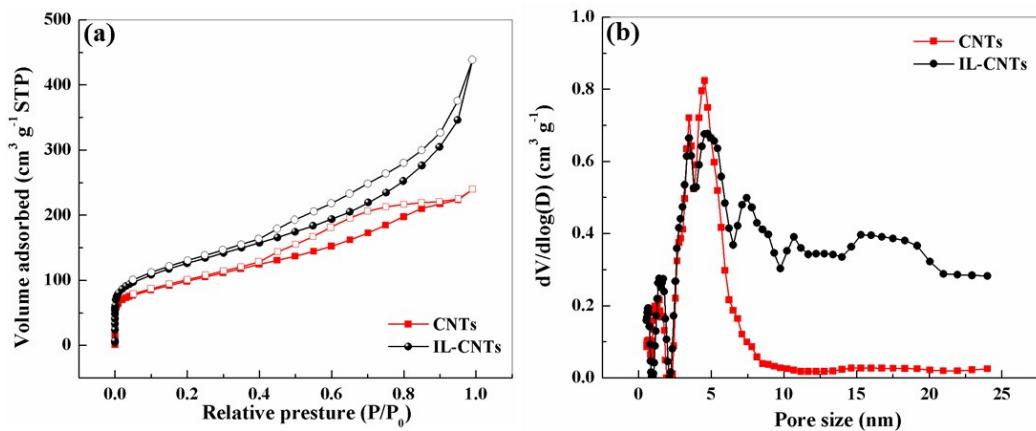


Fig.S1 (a) Nitrogen adsorption desorption isotherms, and (b) pore size distribution of CNTs and IL-CNTs.

Table S1. XPS Analysis of the Samples

Sample	C (at%)	O (at%)	N (at%)	S (at%)
PC	90.98	7.25	1.41	0.36
N, O-PC	88.00	3.61	8.07	0.32
N, O-PC-CNTs	88.71	2.97	8.03	0.29

Table S2. The fitting peak areas of C 1s, N 1s and O 1s spectra of all the samples form XPS analysis.

Sample	Peak area										
	C 1s				N1s				O1s		
	C=C	C-N	C-O	C=O	N-6	N-5	N-Q	N-X	quinone	C=O	C-OH
PC	41951		9036	14489					7552	8345	6087
N, O-PC	34574	9966	7656	5514	6731	7196	2137	119	1452	5617	3782
N, O-PC-CNTs	35217	22143	9767	11466	9844	10675	1263	3767	4929	5225	2619

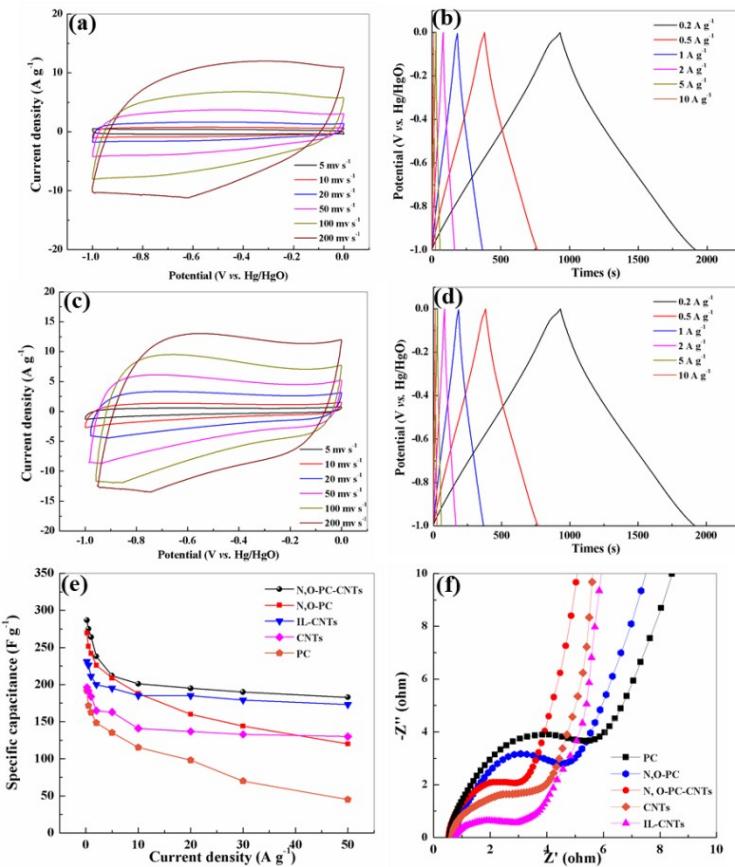


Fig.S2 Electrochemical performance tested by a three-electrode system in 6 mol L⁻¹ KOH, (a) CV curves of CNTs, (b) GCD curves of CNTs under different constant currents, (c) CV curves of IL-CNTs, (d) GCD curves of IL-CNTs under different constant currents, (e) Specific capacitance of as-prepared samples at different current densities, and (h) Nyquist plots of as-prepared samples.

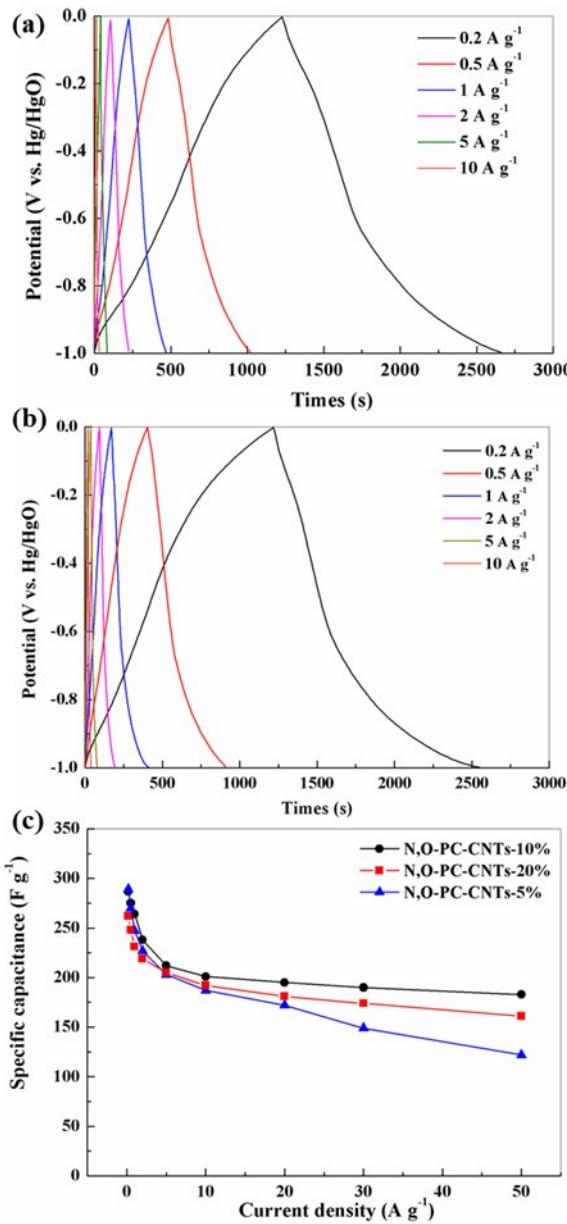


Fig.S3 (a) GCD curves of N, O-PC-CNTs-5% under different constant currents, (b) GCD curves of N, O-PC-CNTs-20% under different constant currents, (c) Specific capacitance of N, O-PC-CNTs-5%, N, O-PC-CNTs-10% and N, O-PC-CNTs-20% at different current densities.

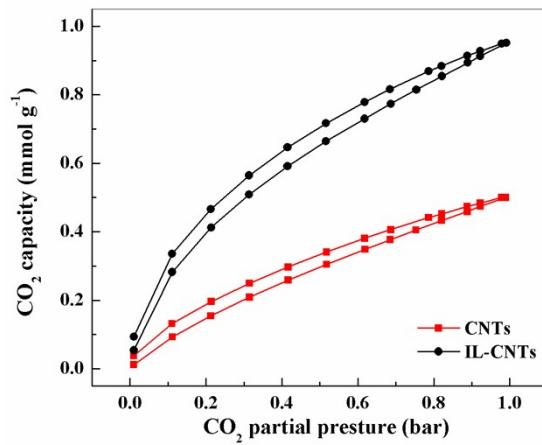


Fig.S4 CO₂ adsorption isotherms for CNTs and IL-CNTs at 298 K.

Table S3 Comparision of the supercapacitors performance in three-electrode cell of porous carbons in the literatures.

Samples	Synthetic method (activation agent)	S_{BET} (m ² g ⁻¹)	Specific capacitance (F g ⁻¹)	Current density (A g ⁻¹)	Electrolyte	Ref.
Activated carbon850-1	KOH	1968	223	0.1 A g ⁻¹	6 mol L ⁻¹ KOH	[1]
N,S,O-doped PC	KOH	1593.8	285	0.5 A g ⁻¹	6 mol L ⁻¹ KOH	[2]
PICNs(NiSSC-140-2-0.45)	KOH	2372.18	312	0.5 A g ⁻¹	6 mol L ⁻¹ KOH	[3]
Nanohexahedron PC	Carbonizing ZIF-8	1142	187	0.5 A g ⁻¹	2 mol L ⁻¹ KOH	[4]
C-N bond PC	KOH	3965	342	1 A g ⁻¹	6 mol L ⁻¹ KOH	[5]
NPCM-600	KOH	1778	298	1 A g ⁻¹	6 mol L ⁻¹ KOH	[6]
AC	H ₃ PO ₄	633.43	234.4	1 A g ⁻¹	1 mol L ⁻¹ KOH	[7]
WP carbon	KOH	416.59	160	1 mA cm ⁻²	6 mol L ⁻¹ KOH	[8]
AC-35	KOH	2312	342.8	0.5 A g ⁻¹	KOH/LiOH	[9]
BL-ACs	KOH	3557	188	1 mA cm ⁻²	0.1 mol L ⁻¹ H ₂ SO ₄	[10]
AHC-4	KOH	849	264	0.25 A g ⁻¹	6 mol L ⁻¹ KOH	[11]
N-doped porous carbon	CO ₂	1414.97	179	1 A g ⁻¹	6 mol L ⁻¹ KOH	[12]
3D-MP-CFW	KOH	1270	240	1 A g ⁻¹	6 mol L ⁻¹ KOH	[13]
LC-550-1	NaNH ₂	1087	266	0.5 A g ⁻¹	6 mol L ⁻¹ KOH	[14]
OAC-4	KOH	2869	287	0.5 A g ⁻¹	6 mol L ⁻¹ KOH	[15]
PNPC-4	KOH	2599.61	287.1	1 A g ⁻¹	6 mol L ⁻¹ KOH	[16]
N, O-PC-CNTs	KOH-IL	2164	287	0.2 A g ⁻¹	6 mol L ⁻¹ KOH	This work

Table S4 Comparision of the CO₂ adsorption performance at 25°C of porous carbons in the literatures

Samples	Synthetic method (activation agent)	S_{BET} (m ² g ⁻¹)	CO ₂ uptake (mmol g ⁻¹)	Ref.
C-char -800	CO ₂ -ammonia	610	2.26	[17]
ANCs-3-700	KOH	3401	4.7	[18]
C-KU-600	KOH/Urea	1087	3.5	[19]
BGC-1-700	KOH	1258	3.46	[20]
SMLK-1	LiCl/KCl	951	3.00	[21]
MB	KOH	1379	2.50	[22]
NAC	KOH	1593	3.20	[23]
CRF	K ₂ CO ₃	595-683	2.3-3.0	[24]
MMCs	CO ₂	1192	3.6 0	[25]
AC	KOH	1503	3.15	[26]
PC-2:1-700	KOH	1433	3.68	[27]
BC	CO ₂	809	2.20	[28]
OTSS-3-350	NaNH ₂	779	3.50	[29]
CN-600-3	K ₂ CO ₃ /CN	1082	3.71	[30]
CAC-S	NaOH	1149	4.28	[31]
WTP-PVA	Annealing	783-1384	2.62-2.91	[32]
AC-900-800-1-H	N ₂ /CO ₂	798	2.94	[33]
N, O-PC-CNTs	KOH-IL	2164	3.7	This work

References

- J. Serafin, M. Baca, M. Biegun, E. Mijowska, R. J. Kalenczuk, J. Srensek-Nazzar and B. Michalkiewicz, *Appl Surf Sci*, 2019, **497**, 143722.
- D. Zhang, Y. C. Xue, J. L. Chen, X. M. Guo, D. D. Yang, J. C. Wang, J. H. Zhang, F. Zhang and A. H. Yuan, *J Nanosci Nanotechno*, 2020, **20**, 2728-2735.
- R. X. Xu, Y. P. Zhao, G. H. Liu, J. S. Zhu, R. Y. Wang, J. P. Cao and X. Y. Wei, *J Colloid Interf Sci*, 2020, **558**, 211-219.
- J. Wu, X. P. Zhang, F. X. Wei, Y. W. Sui and J. Q. Qi, *Mater Lett*, 2020, **258**, 211-219.
- P. Han, M. S. Cheng, D. H. Luo, W. Cui, H. C. Liu, J. G. Du, M. L. Wang, Y. P. Zhao, L. Chen, C. Z. Zhu and J. Xu, *Energy Storage Mater*, 2020, **24**, 486-494.
- D. M. Xue, S. C. Qi, X. Liu, Y. X. Li, X. Q. Liu and L. B. Sun, *J Ind Eng Chem*, 2019, **80**, 568-575.
- M. Sivachidambaram, J. J. Vijaya, L. J. Kennedy, R. Jothiramalingam, H. A. Al-Lohedan, M. A.

- Munusamy, E. Elanthamilane and J. P. Merlin, *New J Chem*, 2017, **41**, 3939-3949.
8. D. Kalpana, S. H. Cho, S. B. Lee, Y. S. Lee, R. Misra and N. G. Renganathan, *Journal Of Power Sources*, 2009, **190**, 587-591.
9. X. J. He, Y. J. Geng, J. S. Qiu, M. D. Zheng, S. A. Long and X. Y. Zhang, *Carbon*, 2010, **48**, 1662-1669.
10. S. G. Lee, K. H. Park, W. G. Shim, M. S. Balathanigaimani and H. Moon, *J Ind Eng Chem*, 2011, **17**, 450-454.
11. W. J. Si, J. Zhou, S. M. Zhang, S. J. Li, W. Xing and S. P. Zhuo, *Electrochimica Acta*, 2013, **107**, 397-405.
12. E. Lei, W. Li, C. H. Ma, Z. Xu and S. X. Liu, *Appl Surf Sci*, 2018, **457**, 477-486.
13. Y. Li, X. Wang and M. H. Cao, *J CO₂ Util*, 2018, **27**, 204-216.
14. S. F. Liu, P. P. Yang, L. L. Wang, Y. L. Li, Z. Z. Wu, R. Ma, J. Y. Wu and X. Hu, *Energ Fuel*, 2019, **33**, 6568-6576.
15. Y. T. Li, Y. T. Pi, L. M. Lu, S. H. Xu and T. Z. Ren, *Journal Of Power Sources*, 2015, **299**, 519-528.
16. Y. B. Zhou, J. Ren, Y. Yang, Q. J. Zheng, J. Liao, F. Y. Xie, W. J. Jie and D. M. Lin, *J Solid State Chem*, 2018, **268**, 149-158.
17. X. Zhang, S. H. Zhang, H. P. Yang, Y. Feng, Y. Q. Chen, X. H. Wang and H. P. Chen, *Chemical Engineering Journal*, 2014, **257**, 20-27.
18. H. M. Wei, J. Chen, N. Fu, H. J. Chen, H. L. Lin and S. Han, *Electrochimica Acta*, 2018, **266**, 161-169.
19. W. Z. Shen, T. P. Hu, P. Y. Wang, H. Z. Sun and W. B. Fan, *Chempluschem*, 2014, **79**, 284-289.
20. A. Chithra, P. Wilson, R. Rajeev and K. Prabhakaran, *Mater Res Express*, 2018, **5**, 115606.
21. A. Rehman and S. J. Park, *J CO₂ Util*, 2019, **34**, 656-667.
22. J. Serafin, U. Narkiewicz, A. W. Morawski, R. J. Wrobel and B. Michalkiewicz, *J CO₂ Util*, 2017, **18**, 73-79.
23. G. Sethia and A. Sayari, *Carbon*, 2015, **93**, 68-80.
24. J. Du, W. C. Li, Z. X. Ren, L. P. Guo and A. H. Lu, *J Energy Chem*, 2020, **42**, 56-61.
25. J. Phuriragpitikhon, P. Ghimire and M. Jaroniec, *J Colloid Interf Sci*, 2020, **558**, 55-67.
26. C. Quan, X. Y. Jia and N. B. Gao, *Int J Energ Res*, 2019, DOI: 10.1002/er.5017.
27. G. Singh, I. Y. Kim, K. S. Lakhi, P. Srivastava, R. Naidu and A. Vinu, *Carbon*, 2017, **116**, 448-455.
28. P. C. Vilella, J. A. Lira, D. C. S. Azevedo, M. Bastos-Neto and R. Stefanutti, *Ind Crop Prod*, 2017, **109**, 134-140.
29. Y. Zhang, L. Liu, P. X. Zhang, J. Wang, M. Xu, Q. Deng, Z. L. Zeng and S. G. Deng, *Chemical Engineering Journal*, 2019, **355**, 309-319.
30. L. M. Yue, Q. Z. Xia, L. W. Wang, L. L. Wang, H. DaCosta, J. Yang and X. Hu, *J Colloid Interf Sci*, 2018, **511**, 259-267.
31. Y. F. Guo, C. Tan, J. Sun, W. L. Li, J. B. Zhang and C. W. Zhao, *Chemical Engineering Journal*, 2020, **381**, 122736.
32. L. Vazhayal, P. Wilson and K. Prabhakaran, *Chemical Engineering Journal*, 2020, **381**, 122628.
33. A. E. Ogungbenro, D. V. Quang, K. A. Al-Ali, L. F. Vega and M. R. M. Abu-Zahra, *J Environ Chem Eng*, 2018, **6**, 4245-4252.