Electronic Supplementary Material (ESI) for Nanoscale. This journal is © The Royal Society of Chemistry 2021

Supplementary Information

Electrochemically Reduced Ultra-high Mass Loading Three-Dimensional Carbon

Nanofibers Network: A Reproducible and Stable Cell Voltage of 2.0 V and High Energy

Density Symmetric Supercapacitor

Gunendra Prasad Ojha^{a, b}, Bishweshwar Pant^{a,b}, Jiwan Acharya^{a, b}, and Mira Park^{a b*,}

^aCarbon Composite Energy Nanomaterials Research Center, Woosuk University, 443 Samnye-ro, Samnye-eup, Wanju-gun, Chonbuk, Jeollabuk-do 55338, Republic of Korea.

^bWoosuk Institute of Smart Convergence Life Care (WSCLC), Woosuk University, 443 Samnye-ro, Samnye-eup, Wanju-gun, Chonbuk, Jeollabuk-do 55338, Republic of Korea

*Corresponding Author: <u>wonderfulmira@woosuk.ac.kr</u> (M. Park)



*Figure S1. Digital images of 3D mat during NaBH*₄ *treatment.*



Figure S2. Digital image of 3D mat after freeze-dry.



Figure S3. Digital photographs showing their representative mass loading (35, 20, and 10 mgcm^{-2.}) of the fabricated electrodes



Figure S4. Cross-section image showing thickness of the Na⁺-LBL 3D-CNF-35 electrode



Figure S5. Cross-section showing thickness of the Na⁺-LBL 3D-CNF-10 electrode



Figure S6. Cross-section showing thickness of the Na⁺-LBL 3D-CNF-20 electrode.



Figure S7. Surface morphology of pristine 2D-CNFs.



Figure S8. Color mapping of Na⁺-LBL 3D-CNF-35 electrode.



Figure S9. TEM (A) and SAED images of LBL 3D-CNF-35 before electroreduction technique.



Figure S10. Survey spectrum of LBL 3D-CNF-35 electrode before and after electroreduction technique.



Figure S11. Linear sweep voltammetry test of Na^+ -LBL 3D-CNF-35 electrode at 10 mV/s scan rate, which indicates the standard potential for hydrogen (H₂) evolution.



Figure S12. CV(*A*) and *GCD*(*B*) curves of Na⁺-LBL 3D-CN-10 electrode at different scan rates and current denisties.



Figure S13. CV (A) and GCD (B) curves of Na⁺*-LBL 3D-CN-20 electrode at different scan rates and current denisties.*

Table S1. A comparative study showing electrochemical performances of Na+-LBL 3D-CNF-35 electrode with reported literatures

S.N.	Materials	Loading	Specific	Working	Reference
		mass (mg)	capacitance (Fg ⁻¹)	potential (V)	S
1	Activated wood carbon	30	118 Fg ⁻¹	-0.9 - 0.0	1
2	Carbon//metal oxide composite	13.4	2098 mFcm ⁻ ₂	0-0.6	2
3	Activated carbon fibers	10 ± 1	161 Fg ⁻¹	0 - 1	3
4	CNT/MnO2/graphene	9.1	3.38 Fcm ⁻²	0 - 1	4
5	N-doped layered porous carbon	17.7	161 Fg ⁻¹	-1 - 0	5
6	A PPyNP/f-CNT	14	176.3 mFcm ⁻³	-0.2 - 0.6	6
7	3D lower structured graphene	11.16	103.6 Fg ⁻¹	0 – 1	7
8	Highly dense mesoporous carbon	11.5	186 Fg ⁻¹	0 – 1	8
9	Electrolyte-mediated chemically converted graphene	10	1570 mFcm ⁻ ₂	0-1	9
10	Na ⁺ -LBL 3D-CNF	35	170 Fg ⁻¹ (5.93 Fcm ⁻²)	-1.25 - 0	This work



Figure S14. *Relation between aerial (A) and gravimetric (B) capacitances and current density.*



Figure S15. IR drop of *Na*⁺-*LBL* 3*D*-*CNFs* electrodes at different mass loading 10, 20, and 35 mgcm⁻² at a current density of 1 mAcm⁻².



Figure S16. Relation between specific capacitance and current density of symmetric device

No.	Electrode materials	Current density (mA/cm2 or A/g)	Energy density (1922 µWhcm ⁻² /Whkg ⁻¹)	Power density (1922 µWcm ⁻² /W/kg)	Weight of electrod e material s (mg/cm ²)	Ref.
1	ZTC-300	1.25 A/g	7.5	625	-	10
2	800 AC	0.3 A/g	3	220		11
3	PCN-900	0.1 A/g	8.02	250		12
4	NOCS-1/10	0.5 A/g	4.3	250		13
5	a-CSN/EG-10	0.3 A/g	7.3	500		14
6	ACG-200	1 A/g	7.5	200		15
7	200-HTC-800-3	0.4 A/g	8.11	400		16
8	N-OMCN@GN	1 A/g	6.68	250		17
9	MOLC	0.5 A/g	3.85	27.7	8	18
10	HGOCN-A	0.5 A/g	4.8	5000		19
11	CPC	1 A/g	6.45		20	20
12	L-CAs	1 A/g	26.25	1000		21
13	BHNC	1 A/g	6.1	26000		22
14	RCFs	0.2 A/g	6.1	1600		23
15	N/O-CNS	0.5 A/g	6.5	80		24
16	N/S/O-3D PC	0.3	8.4	-		25
17	PCNFs	1 A/g	24.4	8800		26
18	NPSO	0.5 A/g	23.17	500		27
19	NHPCs-800	0.5 A/g	23.8	402		
20	N/S-PCNS1-1	0.2 A/g	21	180		28

Table S2: A comparative study showing Energy and Power density of the carbon-based symmetric supercapacitors.

23	Na ⁺ -LBL 3D- CNF-35//Na ⁺ - LBL 3D-CNF- 35	1 mAcm ⁻²	1922 µWhcm ⁻² (27 Whkg ⁻¹)	3979 1922 µWcm ⁻² (57 Wkg ⁻¹⁾	35 mgcm ⁻²	This work
22	PDD-DCNT	1 A/g	19.1	800		30
21	PCAs	0.5 A/g	19.74	500		29

References

- C. Chen, Y. Zhang, Y. Li, J. Dai, J. Song, Y. Yao, Y. Gong, I. Kierzewski, J. Xie and L. Hu, *Energy & Environmental Science*, 2017, 10, 538-545.
- X. Zhang, J. Luo, P. Tang, X. Ye, X. Peng, H. Tang, S.-G. Sun and J. Fransaer, *Nano Energy*, 2017, **31**, 311-321.
- M. Vijayakumar, R. Santhosh, J. Adduru, T. N. Rao and M. Karthik, *Carbon*, 2018, 140, 465-476.
- L. Lyu, K.-d. Seong, J. M. Kim, W. Zhang, X. Jin, D. K. Kim, Y. Jeon, J. Kang and Y. Piao, *Nano-Micro Letters*, 2019, 11, 88.
- L.-z. Sheng, Y.-y. Zhao, B.-q. Hou, Z.-p. Xiao, L.-l. Jiang and Z.-j. Fan, *New Carbon Materials*, 2021, **36**, 179-188.
- J. Parayangattil Jyothibasu, M.-Z. Chen and R.-H. Lee, ACS Omega, 2020, 5, 6441-6451.
- L. Chang, W. Wei, K. Sun and Y. H. Hu, *Journal of Materials Chemistry A*, 2015, 3, 10183-10187.
- L. Chang, K. Sun and Y. H. Hu, ACS Applied Materials & Interfaces, 2018, 10, 33162-33169.
- 9. X. Yang, C. Cheng, Y. Wang, L. Qiu and D. Li, *Science*, 2013, **341**, 534.
- 10. C. Teng, Y. Han, G. Fu, J. Hu, H. Zheng, X. Lu and J. Jiang, Journal of Materials

Chemistry A, 2018, **6**, 18938-18947.

- W. Liu, S. Zhang, S. U. Dar, Y. Zhao, R. Akram, X. Zhang, S. Jin, Z. Wu and D. Wu, *Carbon*, 2018, **129**, 420-427.
- G. Zhu, L. Ma, H. Lv, Y. Hu, T. Chen, R. Chen, J. Liang, X. Wang, Y. Wang, C. Yan, Z. Tie, Z. Jin and J. Liu, *Nanoscale*, 2017, 9, 1237-1243.
- R. Zhang, X. Jing, Y. Chu, L. Wang, W. Kang, D. Wei, H. Li and S. Xiong, *Journal of Materials Chemistry A*, 2018, 6, 17730-17739.
- Y. Liu, X. Qiu, X. Liu, Y. Liu and L.-Z. Fan, *Journal of Materials Chemistry A*, 2018, 6, 8750-8756.
- Z. Chen, K. Liu, S. Liu, L. Xia, J. Fu, X. Zhang, C. Zhang and B. Gao, *Electrochimica Acta*, 2017, 237, 102-108.
- Y. Wu, J.-P. Cao, X.-Y. Zhao, Z.-Q. Hao, Q.-Q. Zhuang, J.-S. Zhu, X.-Y. Wang and X.-Y. Wei, *Electrochimica Acta*, 2017, 252, 397-407.
- M. Zhao, X. Cui, Y. Xu, L. Chen, Z. He, S. Yang and Y. Wang, *Nanoscale*, 2018, 10, 15379-15386.
- M. Shaibani, S. J. D. Smith, P. C. Banerjee, K. Konstas, A. Zafari, D. E. Lobo, M. Nazari, A. F. Hollenkamp, M. R. Hill and M. Majumder, *Journal of Materials Chemistry A*, 2017, 5, 2519-2529.
- 19. Z. Jia, C. Chen, G. Xu, X. Wei and L. Yang, *ChemElectroChem*, 2018, 5, 546-557.
- W. Tian, J. Zhu, Y. Dong, J. Zhao, J. Li, N. Guo, H. Lin, S. Zhang and D. Jia, *Carbon*, 2020, 161, 89-96.
- Y. Zhang, C. Zhao, W. K. Ong and X. Lu, ACS Sustainable Chemistry & Engineering, 2019, 7, 403-411.
- 22. W. Tian, Q. Gao, Y. Tan, K. Yang, L. Zhu, C. Yang and H. Zhang, Journal of Materials

Chemistry A, 2015, **3**, 5656-5664.

- 23. Y. Zhou, Z. Zhu, C. Zhao, K. Zhang, B. Wang, C. Zhao and G. Chen, *ACS Sustainable Chemistry & Engineering*, 2019, 7, 5095-5102.
- Z. Song, D. Zhu, L. Li, T. Chen, H. Duan, Z. Wang, Y. Lv, W. Xiong, M. Liu and L. Gan, *Journal of Materials Chemistry A*, 2019, 7, 1177-1186.
- W. Yang, W. Yang, A. Song, L. Gao, L. Su and G. Shao, *Journal of Power Sources*, 2017, **359**, 556-567.
- T. F. Zhang, Q. X. Xia, Z. Wan, J. M. Yun, Q. M. Wang and K. H. Kim, *Chemical Engineering Journal*, 2019, 360, 1310-1319.
- S. Lei, L. Chen, W. Zhou, P. Deng, Y. Liu, L. Fei, W. Lu, Y. Xiao and B. Cheng, Journal of Power Sources, 2018, 379, 74-83.
- 28. Y. Li, G. Wang, T. Wei, Z. Fan and P. Yan, *Nano Energy*, 2016, **19**, 165-175.
- P. Hao, Z. Zhao, J. Tian, H. Li, Y. Sang, G. Yu, H. Cai, H. Liu, C. P. Wong and A. Umar, *Nanoscale*, 2014, 6, 12120-12129.
- 30. Y. He, X. Yang, N. An, X. Wang, Y. Yang and Z. Hu, *New Journal of Chemistry*, 2019,
 43, 1688-1698.