

Supplementary Information

**Toward High-Performance Flexible Micro-Supercapacitors: In-Situ
Construction of 2D Porous Carbon Nanosheets with A Unique Polycrystalline-
like Micro-Morphological Feature**

Wenyu Wu^{a,b}, Huaxin Ma^{a,b}, Zhao Zhang^{a,b}, Yu Gu^c, Jingjie Zhang^{a,b}, Shuo Li^{a,b},
Ruijun Zhang^{a,b,*}

^a State Key Laboratory of Metastable Materials Science and Technology, Yanshan University, Qinhuangdao 066004, China

^b Hebei key lab for optimizing metal product technology and performance, Yanshan University, Qinhuangdao 066004, China.

^c Academic Affairs Office, Tangshan Normal University, Tangshan 063000, China

* Corresponding author.

E-mail: zhangrj@ysu.edu.cn (R. Zhang).

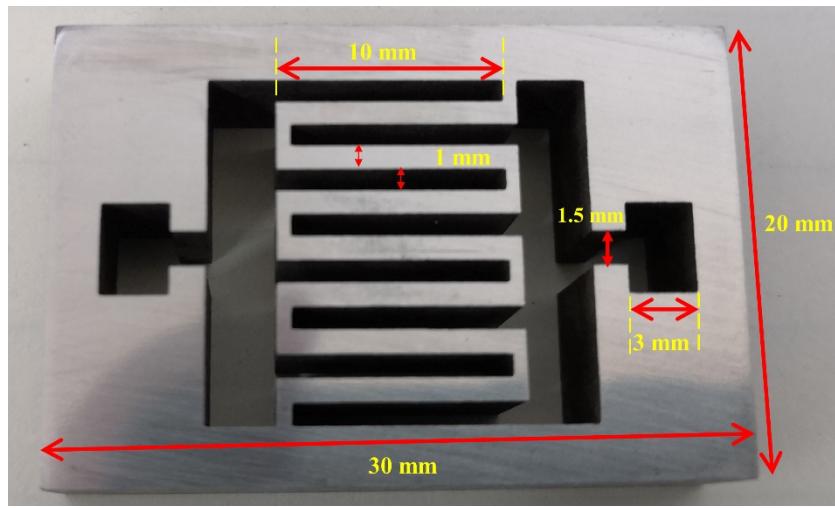


Figure S1 The optical photo of the interdigital mask.

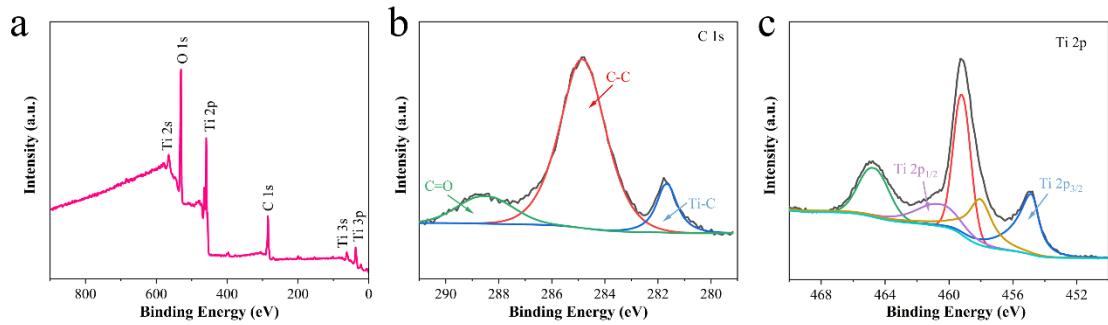


Figure S2. XPS spectrums of P-TiC: (a) survey spectrum; (b) C 1s; (c) Ti 2p.

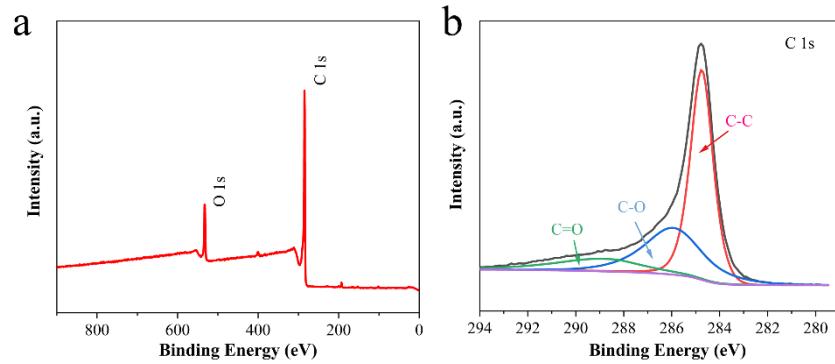


Figure S3. XPS spectrums of PL-PCN: (a) survey spectrum; (b) C 1s.

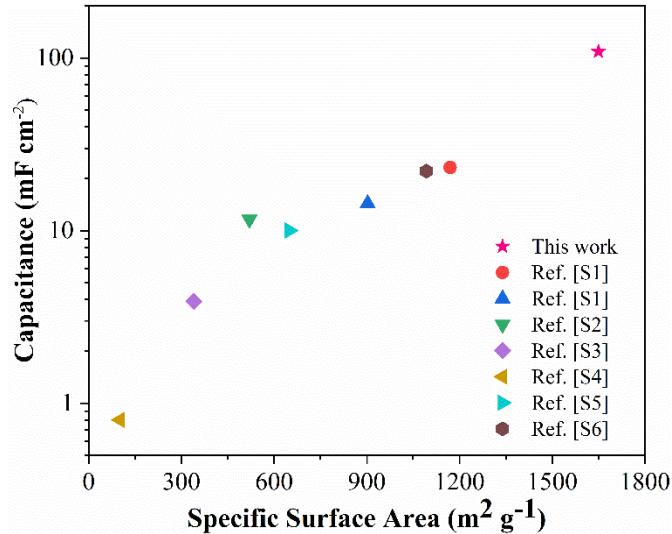


Figure S4. The specific capacitance as a function of the specific surface area of the electrodes summarized from the relevant literatures.

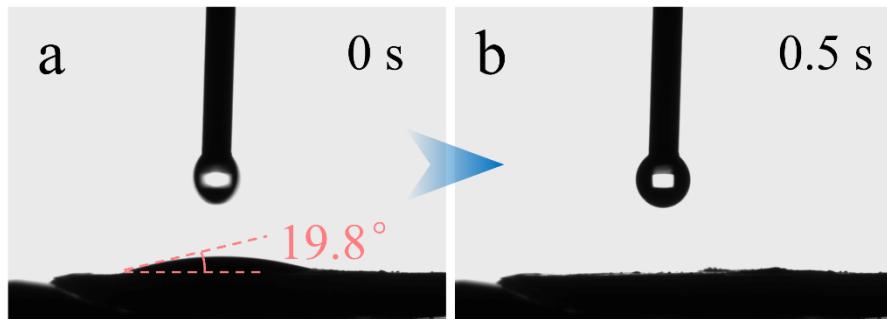


Figure S5. Dynamic contact angle test for PL-PCN with H_3PO_4 electrolyte.

Figure S5 shows the dynamic contact angle test for PL-PCN with H_3PO_4 electrolyte. It can be clearly seen that the contact angle has completely disappeared even within 0.5 s ($19.8^\circ \rightarrow 0^\circ$), suggesting a superior wettability.

Table S1. The comparison of electrochemical performance with other recently reported carbon-based MSCs works.

Electrode material	Electrolyte	Areal capacitance (mF cm^{-2})	References
O/S co-doped porous graphene	PVA/ H_2SO_4	53.2 (0.08 mA cm^{-2})	[S7]
Porous carbon nanosheets	PVA/ H_2SO_4	23.2 (0.2 mA cm^{-2})	[S1]
Porous carbon	PVA/ H_2SO_4	8.4 (100 mV s^{-1})	[S8]
3D porous graphene	PVA/ H_3PO_4	1.7 (20 mV s^{-1})	[S9]
Carbon nanotubes	PVA/ H_3PO_4	4.69 (50 mV s^{-1})	[S10]
Porous graphene	PVA/ H_2SO_4	37.95 (0.6 mA cm^{-2})	[S11]
Graphene/activated carbon	PVA/ H_2SO_4	12.5 (0.01 mA cm^{-2})	[S12]
Graphene quantum dots	EMIMBF4/AN	0.4681 (15 $\mu\text{A cm}^{-2}$)	[S13]
Fluorine-modified graphene	EMIMBF4/PVDF-HFP	17.4 (1 mV s^{-1})	[S14]
PL-PCN	PVA/ H_3PO_4	108.88 (0.1 mA cm^{-2})	This work

References

- [S1] R. Zhou, C. Xu, J. Yang, D. Guan, J. Cai, Heteroatom-doped Porous Carbon Nanosheets Derived from Green Deep Eutectic Solvents for High-performance Micro-supercapacitors, *Chemistry Letters* 49(5) (2020) 585-588.
- [S2] D. Pech, M. Brunet, H. Durou, P. Huang, V. Mochalin, Y. Gogotsi, P.L. Taberna, P. Simon, Ultrahigh-power micrometre-sized supercapacitors based on onion-like carbon, *Nature nanotechnology* 5(9) (2010) 651-654.
- [S3] J. Lin, Z. Peng, Y. Liu, F. Ruiz-Zepeda, R. Ye, E.L. Samuel, M.J. Yacaman, B.I. Yakobson, J.M. Tour, Laser-induced porous graphene films from commercial polymers, *Nature communications* 5 (2014) 5714.
- [S4] J.B. In, B. Hsia, J.-H. Yoo, S. Hyun, C. Carraro, R. Maboudian, C.P. Grigoropoulos, Facile fabrication of flexible all solid-state micro-supercapacitor by direct laser writing of porous carbon in polyimide, *Carbon* 83 (2015) 144-151.
- [S5] X. Ma, X. Hong, L. He, L. Xu, Y. Zhang, Z. Zhu, X. Pan, J. Zhu, L. Mai, High Energy Density Micro-Supercapacitor Based on a Three-Dimensional Bicontinuous Porous Carbon with Interconnected Hierarchical Pores, *ACS applied materials & interfaces* 11(1) (2019) 948-956.
- [S6] Y. Qiu, M. Hou, J. Gao, H. Zhai, H. Liu, M. Jin, X. Liu, L. Lai, One-Step Synthesis of Monodispersed Mesoporous Carbon Nanospheres for High-Performance Flexible Quasi-Solid-State Micro-Supercapacitors, *Small* 15(45) (2019) 1903836.

- [S7] M. Yuan, F. Luo, Y. Rao, Y. Wang, J. Yu, H. Li, X. Chen, Laser synthesis of superhydrophilic O/S co-doped porous graphene derived from sodium lignosulfonate for enhanced microsupercapacitors, *Journal of Power Sources* 513 (2021) 230558.
- [S8] H.-C. Huang, C.-J. Chung, C.-T. Hsieh, P.-L. Kuo, H. Teng, Laser fabrication of all-solid-state microsupercapacitors with ultrahigh energy and power based on hierarchical pore carbon, *Nano Energy* 21 (2016) 90-105.
- [S9] Y. Shao, J. Li, Y. Li, H. Wang, Q. Zhang, R.B. Kaner, Flexible quasi-solid-state planar micro-supercapacitor based on cellular graphene films, *Materials Horizons* 4(6) (2017) 1145-1150.
- [S10] W. Yu, H. Zhou, B.Q. Li, S. Ding, 3D printing of carbon nanotubes-based microsupercapacitors, *ACS applied materials & interfaces* 9(5) (2017) 4597-4604.
- [S11] X. Yun, Z. Xiong, L. Tu, L. Bai, X. Wang, Hierarchical porous graphene film: an ideal material for laser-carving fabrication of flexible micro-supercapacitors with high specific capacitance, *Carbon* 125 (2017) 308-317.
- [S12] H. Chen, S. Chen, Y. Zhang, H. Ren, X. Hu, Y. Bai, Sand-milling fabrication of screen-printable graphene composite inks for high-performance planar micro-supercapacitors, *ACS Applied Materials & Interfaces* 12(50) (2020) 56319-56329.
- [S13] W.W. Liu, Y.Q. Feng, X.B. Yan, J.T. Chen, Q.J. Xue, Superior micro - supercapacitors based on graphene quantum dots, *Advanced Functional*

Materials 23(33) (2013) 4111-4122.

[S14] F. Zhou, H. Huang, C. Xiao, S. Zheng, X. Shi, J. Qin, Q. Fu, X. Bao, X. Feng, K. Müllen, Electrochemically scalable production of fluorine-modified graphene for flexible and high-energy ionogel-based microsupercapacitors, Journal of the American Chemical Society 140(26) (2018) 8198-8205.