Supporting Information

Densification-Activation Strategy toward Hierarchical Porous Self-Supporting Thick Carbon Electrodes for High-Power Supercapacitors

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1 Experimental

1.1 Materials

Balsa wood (O. pyramidale), was obtained from Haolin Industrial Co., Ltd, China. KOH (AR) was purchased from Sinopharm Chemical Reagent Co, Ltd, China.

1.2 Preparation of hot-pressed wood-derived thick carbon electrode

Balsa wood chips (20 cm × 10 cm × 10 cm) were cut and hot-pressed at 60°C, 10 MPa for at least 6 hours to ensure that the volume was compressed to a stable state. After that, the hot-pressed wood block precursor was heated to 900 °C at a heating rate of 5 °C min^{-1} in N_2 atmosphere for 120 min. This sample was named PWC. In addition, wood blocks directly carbonized without hot pressing treatment were named WC.

1.3 Activation of hot-pressed wood-derived thick carbon electrode.

The PWC was immersed in different concentrations of equal volume KOH solution for 6 hours, and the carbon-alkali ratio was controlled to be 1:1, 1:2, 1:3, respectively. After that, the dried sample was heated to 900 °C at a heating rate of 5 °C min^{-1} in N_2 atmosphere and held for 120 min. Finally, the sample was washed with 2 M HCl and water. The samples were named PWCK1 (1:1), PWCK2 (1:2), PWCK3 (1:3) according to the carbon-alkali ratio. All samples were ground to 10 mm × 10 mm × 0.8 mm.

2 Characterizations and electrochemical measurements

The morphologies of the materials were characterized by scanning electron microscopy (Hitachi, Regulus 8100) and transmission electron microscopy (JEOL, JEM-1400). The specific surface area

and pore size analysis were carried out by automatic surface area and porosity analyzer (Micromeritics, ASAP2460). The structures and disorder information were characterized by X-ray diffraction (Rigaku, XRD Ultima IV) and laser raman spectrometer (Themor, DXR532).

Cyclic voltammetry (CV), Galvanostatic charge discharge (GCD) and Electrochemical impedance spectroscopy (EIS), and cycling stability were performed using an electrochemical workstation (Chenhua, Shanghai, China) in a 3 M KOH electrolyte solution. Pt plate and Hg/HgO electrode served as the counter and reference electrode, respectively.

The areal specific capacitances (C_s, mF cm^{-2}) was calculated from discharge curves. The C_s of a single electrode was obtained from GCD curves according to equations (1).

$$Cs = \frac{I\Delta t}{s\Delta V} \qquad (1)$$

Where I (A) is the discharge current, Δt (s) is the galvanostatic discharge time (s), s represents the area (cm^2) of carbon monolith in a single electrode. ΔV (V) is potential window of the discharge process. That is, I/s (mA cm^{-2}) refers to the areal current density.

The areal energy density (E_s), areal power density (P_s) of electrodes were based on the calculated capacitance values and evaluated according to the following equation:

$$Es = \frac{1}{2} C_s V^2 \qquad (2)$$

$$PS = \frac{E_S}{\Delta t} \qquad (3)$$

Tab. S1 Textural parameters of all samples.

Samples	S_{BET}[a] (m^2 g)	S_{mic}[b] (m^2 g)	V_t[c] (m^3 g)	D_P[d] (nm)
WC	135	108	0.09	1.59
PWC	189	155	0.08	1.62
PWCK1	1280	1198	0.56	1.71
PWCK2	1475	1339	0.67	1.78
PWCK3	1347	1234	0.6	1.81

[a] BET surface area.

[b] Micropore area by t-Plot method.

[c] Total pore volume.

[d] Desorption average pore diameter by BET method.

Tab. S2 Areal capacitance of all samples with various current densities.

Current density (mA/cm^2)	WC (F/cm^{-2})	PWC (F/cm^{-2})	PWCK1 (F/cm^{-2})	PWCK2 (F/cm^{-2})	PWCK3 (F/cm^{-2})
2	1.01	7.04	6.13	7.55	5.50
5	0.93	6.46	5.75	6.64	5.28
10	0.85	5.91	5.48	6.38	5.08
20	0.76	5.09	5.21	6.17	4.83
30	0.70	3.87	4.66	5.75	4.43
50	0.61	2.75	3.90	5.02	4.02

Tab. S3 Parameters of PWCK2 and other reported electrodes

Samples	Areal Capacitance (F cm^{-2})	Mass loading (mg cm^{-2})	Electrolyte	Ref.
PWCK2	7.55 (2 mA cm^{-2})	35.2	3 M KOH	This work
3D-printed carbon	4.2 (2 mA cm^{-2})	30.2	1 M H_2SO_4	[1]
(CNC/PIL)-15L	1.4 (0.25 mA cm^{-2})	15	1 M H_2SO_4	[2]
CWZ-3	5.2 (2.5 mA cm^{-2})	21.2	3 M KOH	[3]
SF-3D GA	1.6 (5 mA cm^{-2})	12.8	0.5 M KNO_3	[4]
PCF@MnO2-2h	3.1 (10 mA cm^{-2})	6.8	6 M KOH	[5]
PWC/MnO_2/GQDs	2.7 (1 mA cm^{-2})	15	1 M Na_2SO_4	[6]
balsa carbon/CNTs	1.9 (1 mA cm^{-2})	-	6 M KOH	[7]
YRA	2.2 (1 A g^{-1})	10	1 M $TEABF_4$/AN	[8]

Tab. S4 Parameters of this SSC device and other reported SSCs or ASCs based on self-supporting electrodes.

	Mass Loading mg cm^{-2}	Energy density mwh/cm^2	Power density Wh/cm^2	Electrolyte	Cycling stability	Ref.
This work	35.2	0.71	1.19	3M KOH	50000/94%	This

						work
Co-EG/SV-MoS2	1.5	0.51	0.84	1 M K_2SO_4	10000/100%	[9]
MWCNT	30.2	0.1	0.56	1 M H_2SO_4	5000/89%	[10]
APA-3 PVA	-	0.07	4.96	5 M LiCl/PVA	5000/95%	[11]
FVO/rGO	2	0.17	4	6M KOH	8000/90%	[12]
OW/P/K2	35	0.63	3.91	3M KOH	50000/96%	[13]
C-ZIF-8@ACW	39.8	0.55	0.5	2M KOH	10000/97%	[14]
NCS/CNT@CWS	6.75	0.27	3.75	PVA-KOH	10000/93%	[15]

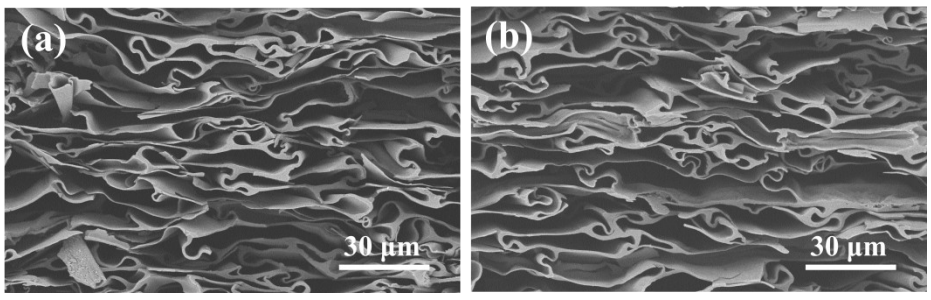

Fig. S1 SEM images of PWCK1 (a) and PWCK3 (b).

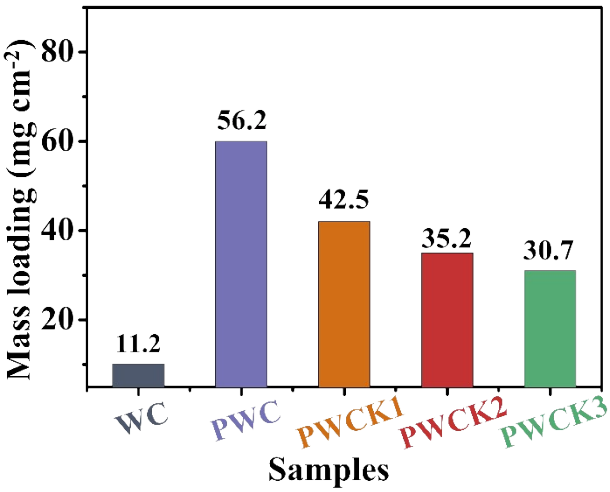

Fig. S2 Mass loading of all samples.

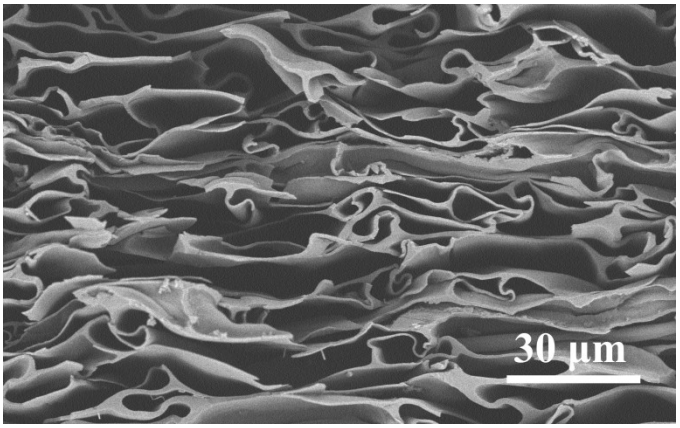

Fig. S3 SEM image of PWCK2 after the cycling tests.

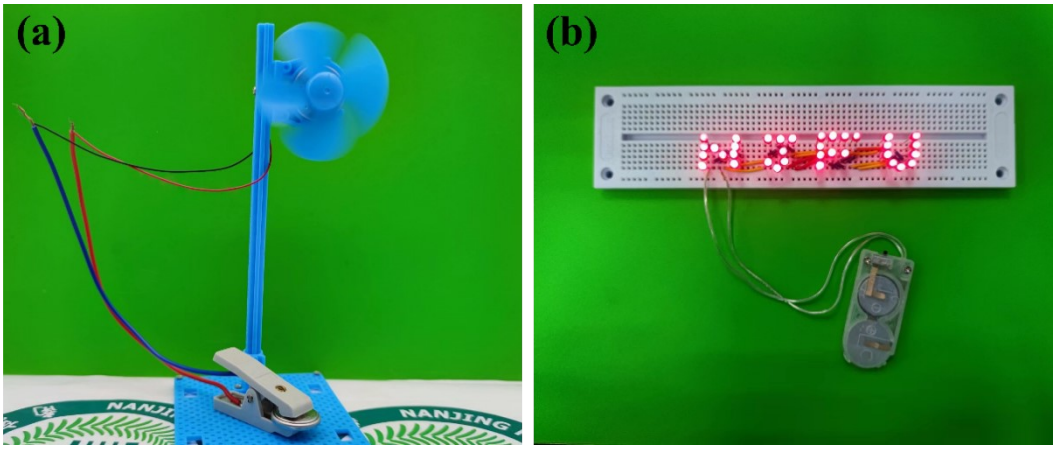

Fig. S4 Photos of fan powered by one SSC device (a) and LED beads powered by two devices in series (b).

Reference

1. G. Zhou, M. C. Li, C. Liu, C. Liu, Z. Li and C. Mei, *Adv Sci (Weinh)*, 2023, **10**, 2206320.
2. H. Huang, Y. Zhao, T. Cong, C. Li, N. Wen, X. Zuo, Y. Guo, H. Zhang, Z. Fan and L. Pan, *Advanced Functional Materials*, 2022, **32**, 2110777.
3. Z. Zhang, S. Deng, D. Wang, Y. Qing, G. Yan, L. Li and Y. Wu, *Chemical Engineering Journal*, 2023, **454**, 140410.
4. B. Yao, S. Chandrasekaran, H. Zhang, A. Ma, J. Kang, L. Zhang, X. Lu, F. Qian, C. Zhu, E. B. Duoss, C. M. Spadaccini, M. A. Worsley and Y. Li, *Adv Mater*, 2020, **32**, e1906652.
5. T. Liu, Z. Zhou, Y. Guo, D. Guo and G. Liu, *Nat Commun*, 2019, **10**, 675.
6. W. Zhang, Y. Yang, R. Xia, Y. Li, J. Zhao, L. Lin, J. Cao, Q. Wang, Y. Liu and H. Guo, *Carbon*, 2020, **162**, 114-123.
7. Q. He, R. He, A. Zia, G. Gao, Y. Liu, M. Neupane, M. Wang, Z. Benedict, K. K. Al-Quraishi, L. Li, P. Dong and Y. Yang, *Small*, 2022, **18**, 2200272.
8. B. Asbani, G. Buvat, J. Freixas, M. Huvé, D. Troadec, P. Roussel, T. Brousse and C. Lethien, *Energy Storage Materials*, 2021, **42**, 259-267.
9. L. Kang, S. Liu, Q. Zhang, J. Zou, J. Ai, D. Qiao, W. Zhong, Y. Liu, S. C. Jun, Y. Yamauchi and J. Zhang, *ACS Nano*, 2024, **18**, 2149-2161.
10. G. Zhou, M.-C. Li, C. Liu, C. Liu, Z. Li and C. Mei, *Advanced Science*, 2023, **10**, 2206320.
11. Y. Zhao, Y. Alsaid, B. Yao, Y. Zhang, B. Zhang, N. Bhuskute, S. Wu and X. He, *Advanced Functional Materials*, 2020, **30**, 1909133.
12. B. Joshi, B.-y. Kim, E. Samuel, J. Huh, A. Aldalbahi, M. El-Newehy, H.-S. Lee and S. S. Yoon, *Chemical Engineering Journal*, 2024, **481**, 148617.
13. Z. Tian, G. Duan, F. Wang, Y. Wang, H. Hou, C. Zhang, S. He, J. Han, X. Han and S. Jiang, *Chemical Engineering Journal*, 2024, **484**, 149615.
14. W. Zhang, M. Li, L. Zhong, J. Huang and M. Liu, *Materials Today Energy*, 2022, **24**, 100951.
15. X. Wang, Y. Deng, Z. Wang, Z. Li, L. Wang, J. Ouyang, C. Zhou and Y. Luo, *ACS Applied Energy Materials*, 2022, **5**, 7400-7407.