## **Supporting Information**

**Cost-Effective Conversion of "Stones" into High-Performance Capacitor Carbon through Solid-Solid Inorganic Chemical Reaction**

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**Fig.** S6 Electrochemical performance of symmetrical SCs using  $TC_{1kg}$  as electrodes in

6.0 mol  $L^{-1}$  KOH/H<sub>2</sub>O electrolyte.

Fig. S7 Cycling stability and Coulombic efficiency of YP50F in KOH/H<sub>2</sub>O electrolyte.

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**Table S2** Collection for the  $R_s$ ,  $R_{\text{ct}}$ ,  $R_w$ , and  $R_{\text{ESR}}$  of TC and YP50F in aqueous and organic electrolytes.

**Table S3** Cost evaluation for the preparation of 1 kg of TC.



**Fig. S1** The corresponding fine spectra of Si 2p of TC.



Fig. S2 XRD patterns of TC treated by 5 mol L<sup>-1</sup> H<sub>2</sub>SO<sub>4</sub> and 10 mol L<sup>-1</sup> NaOH solutions, respectively.



**Fig. S3** Component characterization of TC. a) FTIR analysis; b) Raman spectra. The data of YP50F are provided for comparison.

FT-IR spectral data of TC show two major adsorption bands at 1241 cm-1 and 1589 cm-1 , corresponding to the backbone vibration of C=C and the stretching vibration of C-O-C, respectively. The presence of oxygen-containing groups on the surface of TC is demonstrated, which can improve the hydrophilicity of TC in aqueous/organic electrolytes. Typical D-peak  $(\sim 1345 \text{ cm}^{-1})$  and G-peaks  $(\sim 1586 \text{ cm}^{-1})$ can be observed, and the positions of the peaks approximately overlap with YP50F.



**Fig. S4** Electrochemical performance of symmetrical SCs using TC as electrodes in aqueous and organic electrolytes, respectively. a, b) CV curves at  $0.01$ - $0.5$  V s<sup>-1</sup> in KOH/H<sub>2</sub>O; c, d) CV curves at 0.02-0.5 V s<sup>-1</sup> in Et<sub>4</sub>NBF<sub>4</sub>/AN; e, f) GCD curves at different current densities (i.e., 1-50 A  $g^{-1}$ ) in KOH/H<sub>2</sub>O, respectively; g, h) GCD curves at 1-50 A  $g^{-1}$  in Et<sub>4</sub>NBF<sub>4</sub>/AN. The data of YP50F are also provided for comparison.



**Fig.** S5 Characterizations of average particle size distribution. a) TC; b)  $TC_{1kg}$ . The inset is the corresponding SEM image.



Fig. S6 Electrochemical performance of symmetrical SCs using TC<sub>1kg</sub> as electrodes in 6.0 mol L<sup>-1</sup> KOH/H<sub>2</sub>O electrolyte. a) CV curves at 0.01-0.5 V s<sup>-1</sup>); b) GCD curves at  $0.2\n-10 \text{ A g}^{-1}$ ; c) Nyquist plots.



**Fig.** S7 Cycling stability and Coulombic efficiency of YP50F at 5 A  $g^{-1}$  in 6.0 mol L<sup>-1</sup> KOH/H2O electrolyte.



Table S1 Comparison of TC with carbon derived from CaC<sub>2</sub>, CaCO<sub>3</sub> and others on parameters and capacitance properties.

Samples	$R_{\rm s}(\Omega)$	$R_{\rm ct}(\Omega)$	$R_{\rm w}(\Omega)$	$R_{\mathrm{ESR}}(\Omega)$
TC(KOH/H <sub>2</sub> O)	0.27	0.67	1.88	2.82
$TC_{1kg}$ (KOH/H <sub>2</sub> O)	0.28	1.74	3.18	5.20
YP50F (KOH/H <sub>2</sub> O)	0.21	2.44	2.82	5.47
TC (Et <sub>4</sub> NBF <sub>4</sub> /AN)	1.94	0.64	1.72	4.30
$TC_{1kg}$ (Et <sub>4</sub> NBF <sub>4</sub> /AN)	1.72	0.69	3.61	6.03
YP50F (Et <sub>4</sub> NBF <sub>4</sub> /AN)	2.74	2.21	8.14	13.09

**Table S2** Collection for the  $R_s$ ,  $R_{\text{ct}}$ ,  $R_w$ , and  $R_{\text{ESR}}$  of TC and YP50F in aqueous and organic electrolytes.

**Table S3** Cost evaluation for the preparation of 1 kg of TC.

	Materials	Consumption	Price (RMB)	Total (RMB)	
Raw materials	CaC <sub>2</sub>	$1.3 \text{ kg}$	2.7	$\overline{4}$	
	CaCO <sub>3</sub>	$1 \text{ kg}$	0.5		
Energy	Ball milling	44 kWh	0.49		
	Drying	$10$ kWh	0.49	26.4	
Auxiliary	HC <sub>1</sub>	$25 \text{ kg}$	0.4	15.6	
materials	N <sub>2</sub>	1.25L	4.5		

## **References**

1. K. Zhang, S. Tao, X. Xu, H. Meng, Y. Lu and C. Li, Preparation of mesoporous carbon materials through mechanochemical reaction of calcium carbide and transition metal chlorides, *Ind. Eng. Chem. Res.*, 2018, **57**, 6180- 6188.

- 2. Y. Li, S. Li, X. Xu, J. Gu, X. He, H. Meng, Y. Lu and C. Li, Converting  $CO<sub>2</sub>$ into an oxygenated alkynyl carbon material with high electrochemical performance through a mechanochemical reaction with CaC<sub>2</sub>, *ACS Sustainable Chem. Eng.*, 2021, **9**, 9221-9229.
- 3. J. Yang, Y. Chen, J. Peng, J. Zeng, G. Li, B. Chang, Y. Shen, X. Guo, G. Chen, X. Wang and L. Zheng, Green preparation and supercapacitive behaviors of calcium carbide derived porous carbon based on solvent free mechanochemical route, *J. Energy Storage*, 2022, **51**, 104473.
- 4. L. Zheng, Y. Wang, X. Wang, X. Wang, H. An and L. Yi, The effects of surface modification on the supercapacitive behaviors of carbon derived from calcium carbide, *J. Mater. Sci.*, 2010, **45**, 6030-6037.
- 5. C. Dai, X. Wang, Y. Wang, N. Li and J. Wei, Synthesis of nanostructured carbon by chlorination of calcium carbide at moderate temperatures and its performance evaluation, *Mater. Chem. Phys.*, 2008, **112**, 461-465.
- 6. R. Liu, S. Yao, Y. Shen, Y. Tian and Q. Zhang, Preparation of N doped layered porous carbon and its capacitive deionization performance, *Materials*, 2023, **16**, 1435.
- 7. Z. Xiao, Y. Zhu, H. Yi and X. Chen, A simple CaCO<sub>3</sub> assisted template carbonization method for producing nitrogen containing nanoporous carbon

spheres and its electrochemical improvement by the nitridation of azodicarbonamide, *Electrochim. Acta*, 2015, **155**, 93-102.

- 8. F. Shen, M. Qiu, Y. Hua and X. Qi, Dual functional templated methodology for the synthesis of hierarchical porous carbon for supercapacitor, *ChemistrySelect*, 2018, **3**, 586-591.
- 9. Z. Sun, W. Wang, J. Zhang, G. Wang, K. Wang, X. Liu, G. Ni and Y. Jiang, Nitrogen rich hierarchical porous carbon materials with interconnected channels for high stability supercapacitors, *New J. Chem.*, 2019, **43**, 1864- 1873.
- 10. C. Li, X. Zhang, K. Wang, X. Sun, G. Liu, J. Li, H. Tian, J. Li and Y. Ma, Scalable self propagating high temperature synthesis of graphene for supercapacitors with superiorpower density and cyclic stability, *Adv. Mater.*, 2016, **29**, 1604690.
- 11. H. Liang, R. Shi, Y. Zhou, W. Jiang, T. Sun, Z. Zhang, L. Sun, J. Lian, H. Li and Y. Bu, Removing cost barriers to template carbon synthesis for high performance supercapacitors by establishing a zero emission chemical cycle from CO2, *ACS Energy Lett.*, 2022, **7**, 4381-4388.
- 12. Y. K. Kim, J. H. Park and J. W. Lee, Facile nano templated  $CO<sub>2</sub>$  conversion into highly interconnected hierarchical porous carbon for high performance supercapacitor electrodes, *Carbon*, 2018, **126**, 215-224.
- 13. L. Wang, G. Mu, C. Tian, L. Sun, W. Zhou, P. Yu, J. Yin and H. Fu, Porous graphitic carbon nanosheets derived from cornstalk biomass for advanced supercapacitors, *ChemSusChem*, 2013, **6**, 880-889.
- 14. A. Guo, X. Zhang, B. Shao, S. Sang and X. Yang, Catalytic graphitization assisted synthesis of Fe<sub>3</sub>C/Fe/graphitic carbon with advanced pseudocapacitance, *RSC Adv.*, 2022, **12**, 7935-7940.
- 15. X. Zhang, B. Liu, X. Yan, X. Zhao, Y. Zhang, Y. Wei and Q. Cao, Design and structure optimization of 3D porous graphitic carbon nanosheets for high performance supercapacitor, *Microporous Mesoporous Mater.*, 2020, **309**, 11058.
- 16. Z. Li, H. Mi, L. Liu, Z. Bai, J. Zhang, Q. Zhang and J. Qiu, Nano sized ZIF-8 anchored polyelectrolyte decorated silica for nitrogen rich hollow carbon shell frameworks toward alkaline and neutral supercapacitors, *Carbon*, 2018, **136**, 176-186.
- 17. L. F. Chen, Y. Lu, L. Yu and X. W. Lou, Designed formation of hollow particle based nitrogen doped carbon nanofibers for high performance supercapacitors, *Energy Environ. Sci.*, 2017, **10**, 1777-1783.
- 18. Q. L. Zhu, P. Pachfule, P. Strubel, Z. Li, R. Zou, Z. Liu, S. Kaskel and Q. Xu, Fabrication of nitrogen and sulfur Co-doped hollow cellular carbon nanocapsules as efficient electrode materials for energy storage, *Energy Storage Mater.*, 2018, **13**, 72-79.
- 19. E. Frackowiak, K. Metenier, V. Bertagna and F. Beguin, Supercapacitor electrodes from multiwalled carbon nanotubes, *Appl. Phys. Lett.*, 2000, **77**, 2421-2423.
- 20. L. Zhang and G. Shi, Preparation of highly conductive graphene hydrogels for fabricating supercapacitors with high rate capability, *J. Phys. Chem. C*, 2011, **115**, 17206-17212.
- 21. Y. Xu, Z. Lin, X. Zhong, X. Huang, N. O. Weiss, Y. Huang and X. Duan, Holey graphene frameworks for highly efficient capacitive energy storage, *Nat. Commun.*, 2014, **5**, 4554.
- 22. C. Wang, Y. Zhou, L. Sun, Q. Zhao, X. Zhang, P. Wan and J. Qiu, N/P-Co doped thermally reduced graphene for high performance supercapacitor applications, *J. Phys. Chem. C*, 2013, **117**, 14912-14919.
- 23. J. Yan, T. Wei, B. Shao, F. Ma, Z. Fan, M. Zhang, C. Zheng, Y. Shang, W. Qian and F. Wei, Electrochemical properties of graphene nanosheet/carbon black composites as electrodes for supercapacitors, *Carbon*, 2010, **48**, 1731- 1737.