

**Gram-scale Production of Holey Vertically Aligned Graphene Nanosheet Arrays Derived from Renewable Biomass Precursor by Facile Hydrothermal/Salt-assisted Pyrolysis Method for aqueous high-performance redox supercapacitors**

QinCheng Yang,<sup>‡1</sup> Shuaibing Liu,<sup>‡1</sup> Qianglin Li,<sup>\*2</sup> Ling Wu,<sup>\*3</sup> Binghua Zhou,<sup>4</sup> Zhipeng Wang,<sup>4</sup> Zheng-Hong Huang,<sup>5</sup> Hao Yang<sup>6</sup> and Ming-Xi Wang<sup>\*</sup>1

1.Key Laboratory of Biomass-based Materials for Environment and Energy in Petroleum & Chemical Industries, School of Chemical and Environmental Engineering, Wuhan Institute of Technology, Wuhan 430205, China.

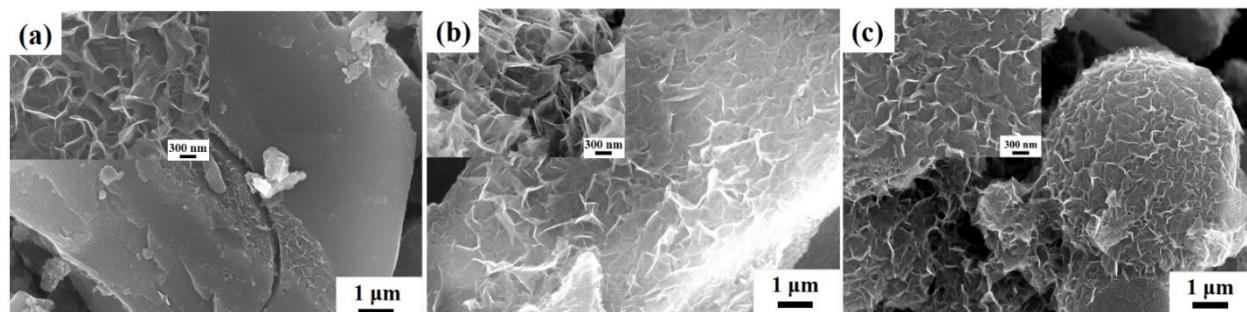
2. Department of Material and Environmental Engineering, Chengdu Technological University, Chengdu, 611730, China.

3. Hubei Province Key Laboratory of Coal Conversion and New Carbon Materials, School of Chemistry and Chemical Engineering, Wuhan University of Science and Technology, Wuhan 430081, China

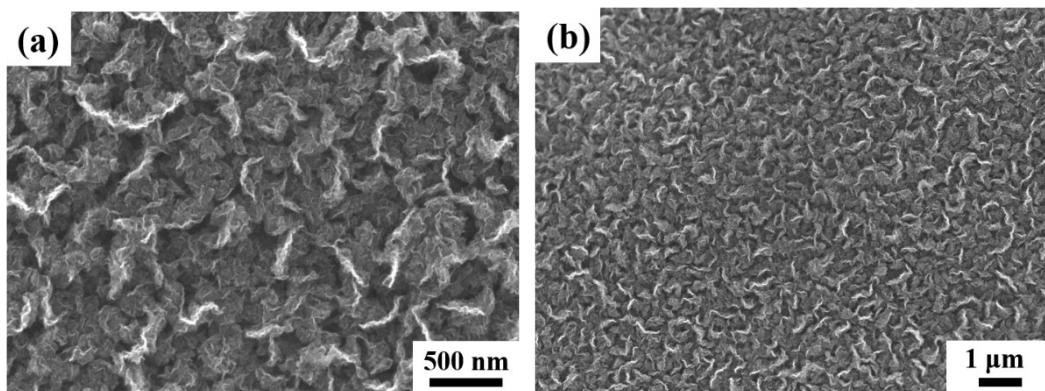
4. Institute of Advanced Materials, Jiangxi Normal University, 99 Ziyang Avenue, Nanchang 330022, China

5. Lab of Advanced Materials, School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

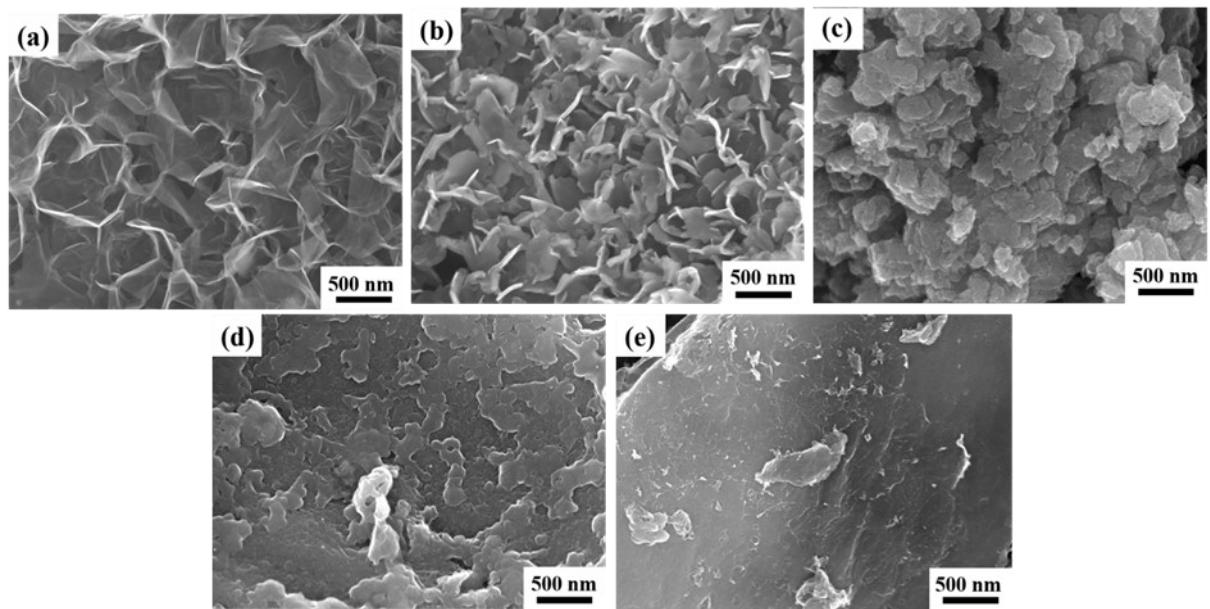
6. Key Laboratory for Green Chemical Process of Ministry of Education, School of Environmental Ecology and Biological Engineering, Wuhan Institute of Technology, Wuhan, 430205, China



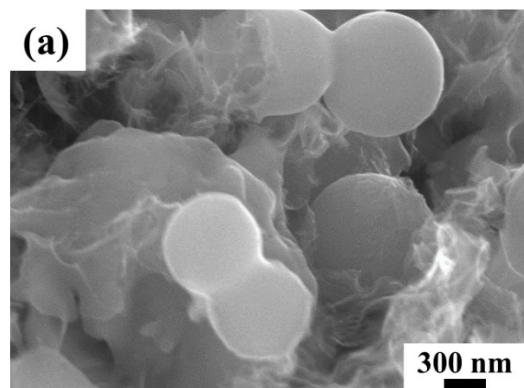
**Fig. S1** FE-SEM images of VAGNA-S-1000 , VAGNA-T-1000 and VAGNA-L-1000.



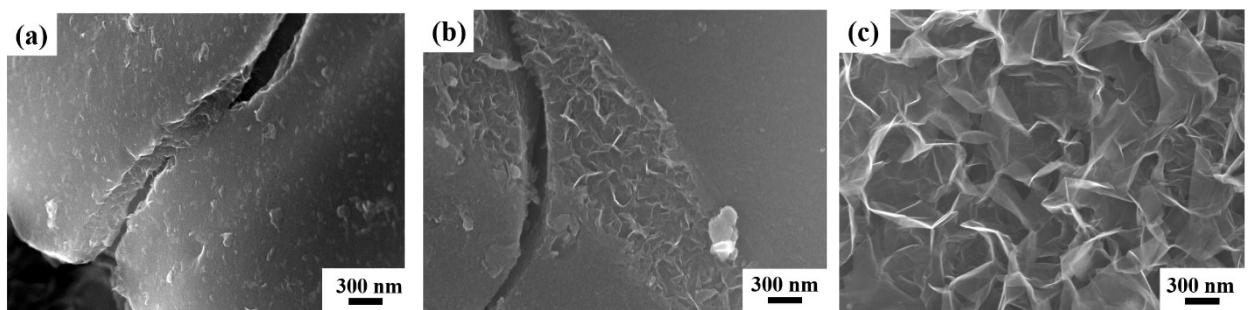
**Fig. S2** SEM diagram of VAGNAS prepared by PECVD method



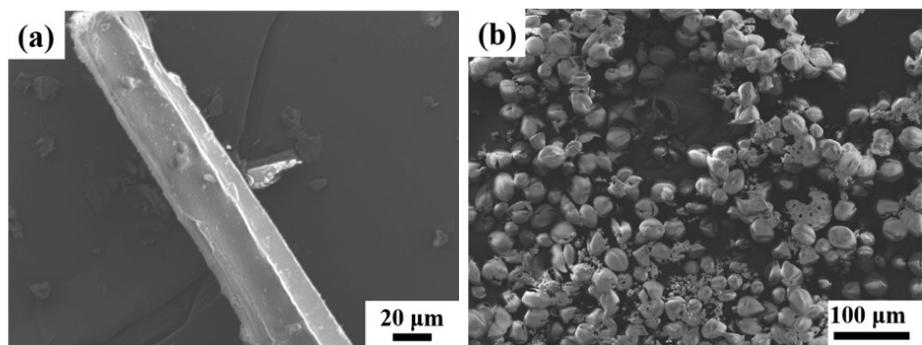
**Fig. S3** FE-SEM images of spruce bark-derived products when treated with different activators. (a) $\text{K}_2\text{CO}_3$ ; (b) $\text{Na}_2\text{CO}_3$ ; (c) $\text{Li}_2\text{CO}_3$ ; (d) $\text{KCl}$  ;(e) $\text{K}_3\text{PO}_4$ .



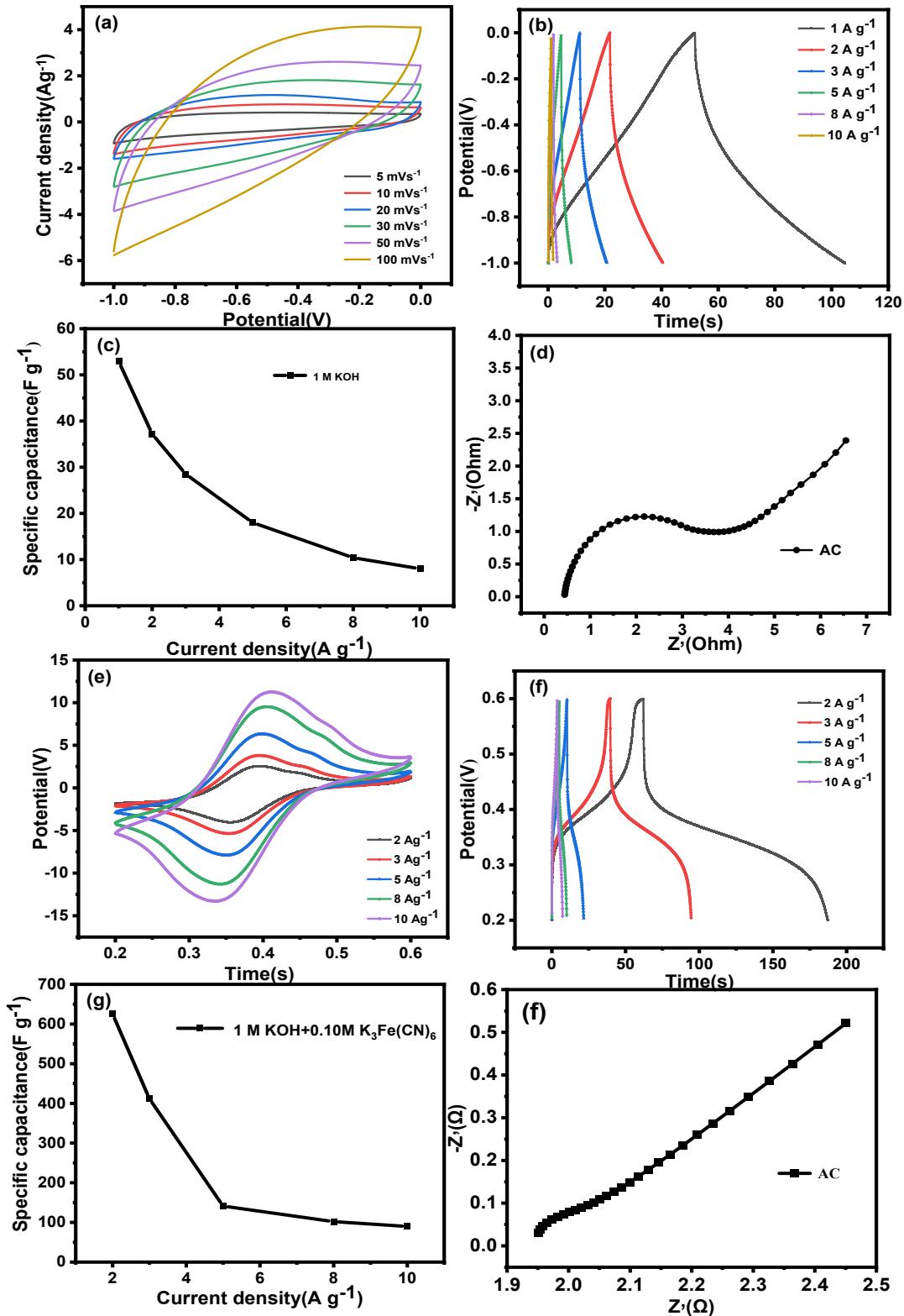
**Fig. S4** FE-SEM images of lotus pollen derived carbons without hydrothermal pretreatment at 1100°C



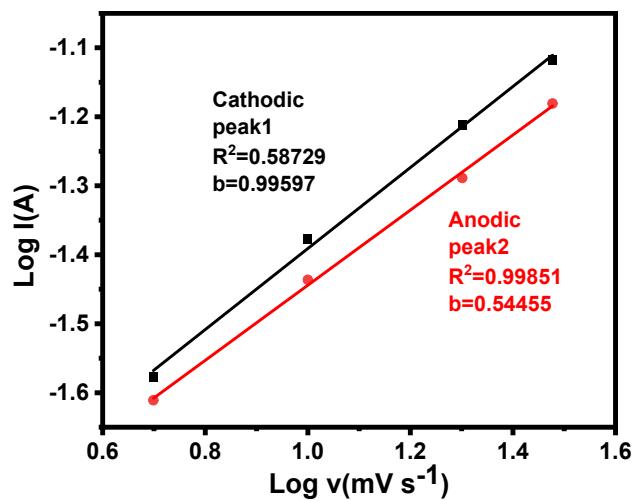
**Fig. S5** FE-SEM images of hydrothermal treated spruce bark-derived carbon materials at different pyrolysis temperatures:  
(a) 850°C ; (b) 900°C ; (c) 1000°C.



**Fig. S6** FE-SEM images of the hydrothermal product (a) spruce bark (b) lotus flower powder.



**Fig. S7** Electrochemical properties of AC in 1M KOH electrolyte: (a) Cyclic voltammetry curves at different scan rates; (b) Constant-current charge/discharge curves at different current densities; (c) Multiplicity curves with the addition; (d) Ac impedance curve. Electrochemical properties of AC in 1M KOH+0.10 M K<sub>3</sub>Fe(CN)<sub>6</sub> electrolyte: (e) Cyclic voltammetry curves at different scan rates; (f) Constant-current charge/discharge curves at different current densities; (g) Specific capacitance at different current densities; (h) EIS Nyquist plot.



**Fig. S8** Log i vs Log v plots