**Green synthesis of free standing cellulose/graphene oxide/polyaniline aerogel electrode for high performance flexible all-solid-state supercapacitors**

**Yueqin Li1,2\*, Zongbiao Xia1,2, Qiang Gong1,2, Xiaohui Liu1,2, Yong Yang1,2, Chen Chen 2, Changhao Qian 2**

1 Co-Innovation Center of Efficient Processing and Utilization of Forest Resources, Nanjing Forestry University, Nanjing 210037, China

2 College of Chemical Engineering, Jiangsu Key Lab for the Chemistry & Utilization of Agricultural and Forest Biomass, Nanjing Forestry University, Nanjing 210037, China

**\*** Correspondence: yueqinli@njfu.edu.cn; Tel.: +86-025-85427024



**Fig. S1.** The cellulose/GO3.5/PANI film was folded and used as a wire to light up a red LED.





**Fig. S2.** (a) CV curves of the cellulose/GO1.0/PANI, cellulose/GO2.0/PANI, cellulose/GO3.5/PANI and cellulose/GO5.0/PANI samples at 10 mV/s. (b) CV curves of cellulose/PANI sample. (c) GCD curves of cellulose/PANI sample. (d) Nyquist plot of the cellulose/PANI and cellulose/GO/PANI with an equivalent circuit in the inset.



**Fig. S3.** (a) CV curves of the cellulose/GO3.5/PANI0.5M, cellulose/GO3.5/PANI0.75M, cellulose/GO3.5/PANI1.0M (namely cellulose/GO3.5/PANI sample in the manuscript) and cellulose/GO3.5/PANI1.25M samples at 10 mV/s. (d) GCD curves of the cellulose/GO3.5/PANI0.5M, cellulose/GO3.5/PANI0.75M, cellulose/GO3.5/ PANI1.0M and cellulose/GO3.5/PANI1.25M samples.

**Table S1** Comparison of electrochemical performance of various electrodes based on conducting fillers/cellulose composites.

|  |  |  |  |
| --- | --- | --- | --- |
| Materials | Maximum CS (mF/cm2) | Cyclic stability | Reference |
| Graphene/cellulose paper | 81 (1 mV/s) | 99.1 % (5000) | [[1](#_ENREF_1)] |
| SWCNT/PANI/cellulose | 330 (0.2 mA/cm2) | 79 % (1000) | [[2](#_ENREF_2)] |
| Graphite /PANI/paper | 355.6 (0.5 mA/cm2) | - | [[3](#_ENREF_3)] |
| GO/PPy | 387.6 (0.2 mA/cm2) | 84.8 % (5000) | [[4](#_ENREF_4)] |
| CNT/PANI hydrogel | 680 (1 mA/cm2) |  | [[5](#_ENREF_5)] |
| PANI/Graphite paper | 176 (0.2 mA/cm2) | - | [[6](#_ENREF_6)] |
| PANI/RGO film | 718 (0.45 A/g) | 74 % (500) | [[7](#_ENREF_7)] |
| Graphene/PANI/Graphene | 190.6 (0.5 mA/cm2) | 96 % (1000) | [[8](#_ENREF_8)] |
| RGO/PPy CCFs paper | 363 (0.5 mA/cm2) | - | [[9](#_ENREF_9)] |
| Carbon cloth-PANI-rGO | 471 (0.5 mA/cm2) | 75.5 % (10000) | [[10](#_ENREF_10)] |
| PANI/CNT/Graphene | 465 (1 mA/cm2) | 84 % (1000) | [[11](#_ENREF_11)] |
| PANI/GO/CNT | 510.5 (1 A/g)  | - | [[12](#_ENREF_12)] |
| CNFs-RGO/PPy | 334  (0.1 mA/cm2) | 100% (2000) | [[13](#_ENREF_13)] |
| Cellulose/GO3.5/PANI  | 1218 (1.0 mA/cm2) | 83.5 % (1000) | This work |

References

[1] Z. Weng, Y. Su, D.-W. Wang, F. Li, J. Du, H.-M. Cheng, Graphene-Cellulose Paper Flexible Supercapacitors, Advanced Energy Materials, 1 (2011) 917-922.

[2] D. Ge, L. Yang, L. Fan, C. Zhang, X. Xiao, Y. Gogotsi, S. Yang, Foldable supercapacitors from triple networks of macroporous cellulose fibers, single-walled carbon nanotubes and polyaniline nanoribbons, Nano Energy, 11 (2015) 568-578.

[3] B. Yao, L. Yuan, X. Xiao, J. Zhang, Y. Qi, J. Zhou, J. Zhou, B. Hu, W. Chen, Paper-based solid-state supercapacitors with pencil-drawing graphite/polyaniline networks hybrid electrodes, Nano Energy, 2 (2013) 1071-1078.

[4] J. Cao, Y. Wang, J. Chen, X. Li, F.C. Walsh, J.-H. Ouyang, D. Jia, Y. Zhou, Three-dimensional graphene oxide/polypyrrole composite electrodes fabricated by one-step electrodeposition for high performance supercapacitors, Journal of Materials Chemistry A, 3 (2015) 14445-14457.

[5] S. Zeng, H. Chen, F. Cai, Y. Kang, M. Chen, Q. Li, Electrochemical fabrication of carbon nanotube/polyaniline hydrogel film for all-solid-state flexible supercapacitor with high areal capacitance, Journal of Materials Chemistry A, 3 (2015) 23864-23870.

[6] K. Li, X. Liu, S. Chen, W. Pan, J. Zhang, A flexible solid-state supercapacitor based on graphene/polyaniline paper electrodes, Journal of Energy Chemistry, 32 (2019) 166-173.

[7] M. Kim, C. Lee, J. Jang, Fabrication of Highly Flexible, Scalable, and High-Performance Supercapacitors Using Polyaniline/Reduced Graphene Oxide Film with Enhanced Electrical Conductivity and Crystallinity, Adv. Funct. Mater., 24 (2014) 2489-2499.

[8] K. Chi, Z. Zhang, J. Xi, Y. Huang, F. Xiao, S. Wang, Y. Liu, Freestanding graphene paper supported three-dimensional porous graphene-polyaniline nanocomposite synthesized by inkjet printing and in flexible all-solid-state supercapacitor, Acs Appl Mater Interfaces, 6 (2014) 16312-16319.

[9] S. Lyu, H. Chang, F. Fu, L. Hu, J. Huang, S. Wang, Cellulose-coupled graphene/polypyrrole composite electrodes containing conducting networks built by carbon fibers as wearable supercapacitors with excellent foldability and tailorability, J. Power Sources, 327 (2016) 438-446.

[10] P. Du, Y. Dong, H. Kang, X. Yang, Q. Wang, J. Niu, S. Wang, P. Liu, Graphene-Wrapped Polyaniline Nanowire Array Modified Functionalized of Carbon Cloth for High-Performance Flexible Solid-State Supercapacitor, ACS Sustainable Chemistry & Engineering, 6 (2018) 14723-14733.

[11] B. Vedhanarayanan, T.-H. Huang, T.-W. Lin, Fabrication of 3D hierarchically structured carbon electrode for supercapacitors by carbonization of polyaniline/carbon nanotube/graphene composites, Inorganica Chimica Acta, 489 (2019) 217-223.

[12] Q. Jiang, Y. Shang, Y. Sun, Y. Yang, S. Hou, Y. Zhang, J. Xu, A. Cao, Flexible and multi-form solid-state supercapacitors based on polyaniline/graphene oxide/CNT composite films and fibers, Diamond Relat. Mater., 92 (2019) 198-207.

[13] M. Mo, C. Chen, H. Gao, M. Chen, D. Li, Wet-spinning assembly of cellulose nanofibers reinforced graphene/polypyrrole microfibers for high performance fiber-shaped supercapacitors, Electrochim. Acta, 269 (2018) 11-20.