**Supporting Information**

**Paper flower-derived porous carbons with high-capacitance by chemical and physical activation for sustainable applications**

Pitchaimani Veerakumar,a,b,\* Thandavarayan Maiyalagan,c,\* Balasubramaniam Gnana Sundara Raj,cKuppuswamy Guruprasad,c Zhongqing Jiangdand King-Chuen Lina,b,\*

aDepartment of Chemistry, National Taiwan University, Taipei 10617, Taiwan

bInstitute of Atomic and Molecular Sciences, Academia Sinica, Taipei 10617, Taiwan

cSRM Research Institute, Department of Chemistry, SRM University, Kattankulathur-603203, India.

dDepartment of Physics, Key Laboratory of ATMMT Ministry of Education, Zhejiang Sci-Tech University, Hangzhou 310018, China

\*Corresponding author E-mail: [spveerakumar@gmail.com](mailto:spveerakumar@gmail.com) (P. Veerakumar); [kclin@ntu.edu.tw](mailto:kclin@ntu.edu.tw) (K.-C Lin); maiyalagan@gmail.com (T. Maiyalagan).

D:\##Submitted Paper work\###PFC work_Dr. T.Maiyalagan\SY Ads\Figure-S3.tif

**Figure S1.** Photographs of SY dye before and after adsorption.

D:\##Submitted Paper work\###PFC work_Dr. T.Maiyalagan\SY Ads\Figure-S4.tif

**Figure S2.** (a) N2 adsorption-desorption isotherm and (b) pore size distribution of PFC-800 after SY adsorption.

D:\##Submitted Paper work\###PFC work_Dr. T.Maiyalagan\##_PFC_Manuscript\Figures\Figure-S3.tif

**Figure S3.** (a-b) FE-TEM image of PFC-400 sample with different magnification.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Table S1.** Comparison of supercapacitor performance between PFC-800-modified electrode and other electrodes. | | | | |
| Electrode | *S*BET  (m2 g-1) | Electrolyte | Speciﬁc capacitance  (F g-1) | Ref. |
| Treated food-waste | 535.80 | KOH | 106 at 10 mV s-1 | [1] |
| Bamboo | 1025 | Et4NBF4 | 60 at 1.0 mV s-1 | [2] |
| Banana fiber | 1097 | Na2SO4 | 74 at 100 mV s-1 | [3] |
| Bamboo | 1251 | H2SO4 | 68 at 40 mA cm-2 | [4] |
| Nano-porous RuO2 | - | H2SO4 | 50 at 20 mV s-1 | [5] |
| RuO2 thin film | - | H2SO4 | 73 at 1.8 mF cm−2 | [6] |
| SnO2/CNT | - | KCl | 90 at 2 mV s−1 | [7] |
| Fe2O3/CB | - | KCl | 22.35 at 10 mV s−1 | [8] |
| ZnO/Activated carbon | 818.9 | KOH | 117 at 0.5 A g-1 | [9] |
| Ni/rGO | - | KOH | 100 at 0.5 A g-1 | [10] |
| ZnO/Carbon | - | KNO3 | 21.7 at 50 mV s-1 | [11] |
| ZnO/Graphene | - | KCl | 109 at 5 mV s-1 | [12] |
| PFC-800 | 1801 | H2SO4 | 118 at 1 A g-1 | This work |

There are many reports in the literature describing supercapacitors fabricated with different carbon based electrodes [1-12]. A list for comparing specific capacitance values among these different electrodes, including the present study, is summarized in Table SI, along with their BET specific surface areas. Note that the speciﬁc capacitance calculated to be 60 and 74 F g-1 for bamboo and banana fiber derived carbons [13] were much smaller than our carbon materials (see Table SI). This is due to the fact the specific surface area of carbons developed provides higher electrical double-layer capacitance [14,15].

|  |  |
| --- | --- |
| **Table S2.** Chemical structure and some properties of SY dye used in this study | |
| Common name | Sunset Yellow FCF (SY) |
| IUPAC name | Disodium 6-hydroxy-5-[(4-sulfophenyl)azo]-2-naphthalenesulfonate |
| Appearance | Dark red/orange powder |
| Empirical formula | C16H10N2Na2O7S2 |
| Solubility in water (mg L-1) | 20×103 |
| Melting point | 300 oC |
| Molecular weight (g mol-1) | 452.36 |
| λmax (nm) | 482 nm |
| Molecular structure | https://www.sigmaaldrich.com/content/dam/sigma-aldrich/structure3/090/mfcd00036437.eps/_jcr_content/renditions/mfcd00036437-large.png |

**Dye removal experiments**

The food dye solution was prepared by dissolving an appropriate amount of sunset yellow (SY) in ultrapure water (MilliQ, 18.25 MΩ•cm), which was used as a stock solution. Adsorption studies were carried out by the batch method to obtain both the adsorption rate and equilibrium data. The batch adsorption experiments were performed with a rotary shaker using 100 mL screw-cap conical flasks under fixed conditions (stirring rate 200 rpm; at RT). The SY solution concentration of 50 mg L−1 was prepared before addition of 25 mg of PFCs. The batch adsorption equilibrium isotherms were also carried out on a rotary shaker using equal amount (25 mg) of PFCs, which were thoroughly mixed with 100 mL SY solution. The isotherm studies were performed by varying the initial SY concentrations from 10 to 50 mg L−1 at RT.

**Analytical Procedure**

After the kinetic and equilibrium studies, the resultant mixture solution was filtered through a 0.45 μm membrane filter, followed by filtrate analyses. The concentrations of SY in the solutions derived from all runs were measured by UV-vis spectroscopy (Thermo Scientific evolution 220). The absorbance signals were measured at a wavelength λ = 485 nm. The absorption amount at equilibrium (*q*e) and sorption efficiency (%) were calculated according to the equations [16]:

where~~,~~ *q*e represent equilibrium adsorption amount (in mg g−1), *C*0 and *C*e are the initial and equilibrium concentrations (in mg L−1), *V* is the solution volume (in L), and *m* is the mass of the carbon material (in mg).

The isotherm of adsorption indicates the quantities of target species that are distributed between the liquid and solid phases when a balance was reached for the adsorption process. It is employed to estimate the maximum adsorption capacity of adsorbents, which is expressed in terms of quantity of PFCs adsorbed per unit of mass (in mg g−1). In the present study, the data were correlated with a suitable isotherm using the Langmuir and Freundlich equation [17]:

where *K*L is the Langmuir equilibrium constant for the adsorption (in L mg−1), *Q*o is the maximum adsorption capacity (in mg g−1), *q*e is the amount adsorbed at equilibrium (in mg g−1), and *C*e is the equilibrium concentration (in mg L−1)

Alternatively, the Freundlich isotherm equation was used to describe the exponential distribution of active centers, which are characteristics of systems with heterogeneous surface or indefinite surface coverage [18]:

The equation may be rewritten as:

where *K*F and *n* are the isotherm constants of the Freundlich equation.

|  |  |  |  |
| --- | --- | --- | --- |
| **Table S3.** Comparison of PFCs adsorption capacity with different adsorbents. | | | |
| Adsorbent | Conditions | *Q*o, (mg g−1) | Reference |
| MOFs*a*/GO*b* | Adsorbent (10 mg); SY dye (5 mL, 45.24 mg L-1; | 81.28 | [19] |
| MPMWCNT*c* | Adsorbent (2 mg); SY dye (20 mL, 100 mg L-1, 25 ◦C 6 h | 85.47 | [20] |
| Mangrove barks | Adsorbent (1 g, 100 mg L-1), SY (50 mL, 100 mg L-1), pH 2, 30 ◦C. 5 h | 12.72 | [21] |
| Lady finger stem | Adsorbent (2.5 g), SY (100mL, 1000 mg L-1) pH 5, 30 ◦C, 0.7 h. | 15.92 | [22] |
| PPy*d*/mw | Adsorbent (0.007 g), SY (30 mL) pH 2, 25 ◦C, 5 h. | 212.1 | [23] |
| PFC-400 | Adsorbent (25 mg), SY (20 mL, 100 mg L-1), 25◦C, 3 h. | 65.7 | This work |
| PFC-700 | Adsorbent (25 mg), SY (20 mL, 100 mg L-1), 25◦C, 2 h. | 253.4 | This work |
| PFC-800 | Adsorbent (25 mg), SY (20 mL, 100 mg L-1), 25◦C, 2 h. | 273.6 | This work |
| *a*Metal organic framework. *b*Graphene oxide. *c*Magnetic polymer multi-wall carbon nanotube. *d*Polypyrrole. | | | |

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Table S4**. Textural properties of the PFC-800 before and after SY adsorption. | | | | | |
| **Sample** | ***S*Total**  **(m2 g−1)** | ***S*Micro**  **(m2 g−1)** | ***V*Total**  **(cm3 g−1)** | ***V*Micro**  **(cm3 g−1)** | ***D***  **(nm)** |
| PFC-800  (Before SY adsorption) | 1801 | 348.8 | 1.161 | 0.553 | 3.28 |
| PFC-800  (After SY adsorption) | 1765.7 | 309.9 | 1.041 | 0.468 | 3.24 |

**References**

[1] C.K. Sim, S.R. Majid, N.Z. Mahmood, Electrochemical performance of activated carbon derived from treated food-waste, Int. J. Electrochem. Sci. 10 (2015) 10157–10172.

[2] C. Kim, J.-W. Lee, J.-H. Kim, K.-S. Yang, Feasibility of bamboo-based activated carbons for an electrochemical supercapacitor electrode, Korean J. Chem. Eng. 23 (2006) 592–594.

[3] V. Subramaniam, C. Luo, A.M. Stephan, K.S. Nahm, S. Thomas, B. Wei, Supercapacitors from activated carbon derived from banana fibers, J. Phys. Chem. C. 111 (2007) 7527–7531.

[4] Y.J. Kim, B.-J. Lee, H. Suezaki, T. Chino, Y. Abe, T. Yanagiura, K.C. Park, M. Endo, Preparation and characterization of bamboo-based activated carbons as electrode materials for electric double layer capacitors, Carbon 44 (2006) 1592–1595.

[5] V.D. Patake, C.D. Lokhande, Chemical synthesis of nano-porous ruthenium oxide (RuO2) thin films for supercapacitor application, Appl. Surf. Sci. 254 (2008) 2820–2824.

[6] U.M. Patil, S.B. Kulkarni, V.S. Jamadade and C.D. Lokhande, Chemically synthesized hydrous RuO2 thin films for supercapacitor application. J. Alloys Compounds 509 (2011) 1677–1682.

[7] C.-H. Xu, Y.-F. Chiu, P.-W. Yeh, J.-Z. Chen, Screen-printed SnO2/CNT quasi-solid-state gel-electrolyte supercapacitor, Mater. Res. Express 3 (2016) 085002.

[8] M. Nasibi, M.A. Golozar, G. Rashed, Nano iron oxide (Fe2O3)/carbon black electrodes as electrode material for electrochemical capacitors: Effect of the nanoparticles dispersion quality, Mater. Chem. Phys. 139 (2013) 12–16.

[9] Y. Li, X. Liu, Activated carbon/ZnO composites prepared using hydrochars as Intermediate and their electrochemical performance in supercapacitor, Mater. Chem. Phys. 148 (2014) 380–386.

[10] H. Huang, Y. Tang, L. Xu, S. Tang, Y. Du, Direct formation of reduced graphene oxide and 3D lightweight nickel network composite foam by hydrohalic acids and its application for high-performance supercapacitors, ACS Appl. Mater. Interfaces 6 (2014) 10248–10257.

[11] M. Jayalakshmi, M. Palaniappa and K. Balasubramanian, Single step solution combustion synthesis of ZnO/carbon composite and its electrochemical characterization for supercapacitor application, Int. J. Electrochem. Sci. 3 (2008) 96–103.

[12] A. Ramadoss, S.J. Kim, Facile preparation and electrochemical characterization of graphene/ZnO nanocomposite for supercapacitor applications, Mater. Chem. Phys. 140 (2013) 405–411.

[13] K. Kinoshita Carbon: Electrochemical and physicochemical properties. New York: Wiley; (1988).

[14] C.T. Hsieh, H. Teng, Influence of oxygen treatment on electric double-layer capacitance of activated carbon fabrics, Carbon 40 (2002) 667–674.

[15] Y. Huang, J. Liang, Y. Chen, An overview of the applications of graphene-based materials in supercapacitors, Small 8 (2012) 1805–1834.

[16] P. Veerakumar, T. Jeyapragasam, M. Ramakrishnan, I. Panneer Muthuselvam, K.-C. Lin, Graphene oxide nanosheets as an efficient and reusable sorbents for eosin yellow dye removal from aqueous solutions. ChemistrySelect, 2 (2017) 3598–3607.

[17] F.P.D. Sá, B.N. Cunha, L.M. Nunes, Effect of pH on the adsorption of Sunset Yellow FCF food dye into a layered double hydroxide (CaAl-LDH-NO3), Chem. Eng. J. 215 (2013) 122–127.

[18] H.J. Gao, T.T. Kanc, S.Y. Zhao, Y.X. Qian, X.Y. Cheng, W.L. Wu, X.D. Wang, L.Q. Zheng, Removal of anionic azo dyes from aqueous solution by functional ionic liquid cross-linked polymer. J. Hazard. Mater. 261 (2013) 84–90.

[19] L. Li, Z. Shi, H. Zhu, W. Hong, F. Xie, K. Sun, Adsorption of azo dyes from aqueous solution by the hybrid MOFs/GO, Water. Sci. Tech. 73 (2016) 1728–1737.

[20] H.J. Gao, S.Y. Zhao, X.Y. Cheng, X.D. Wang, L.Q. Zheng, Removal of anionic azo dyes from aqueous solution using magnetic polymer multi-wall carbon nanotube nanocomposite as adsorbent, Chem. Eng. J. 223 (2013) 84–90.

[21] T.L. Seey, M.J.N.M. Kassim, Acidic and basic dyes removal by adsorption on chemically treated mangrove barks, Int. J. Appl. Sci. Technol. 2 (2012) 270–276.

[22] A. Abbas, R. Rehman, S. Murtaza, U. Shafique, A. Zahid, R. Ayub, Adsorptive removal of congo red and sunset yellow dyes from water systems by lady finger stem, J. Chem. Soc. Pak. 34 (2012) 1241–1247.

[23] R.S. Aliabadi, N.O. Mahmoodi, Synthesis and characterization of polypyrrole, polyaniline nanoparticles and their nanocomposite for removal of azo dyes; sunset yellow and Congo red J. Clean. Prod. 179 (2018) 235–245.