Supporting Information

Assembly of electric double-layer capacitors with hardwood kraft lignin-based electrodes and separator together with ionic liquid electrolyte

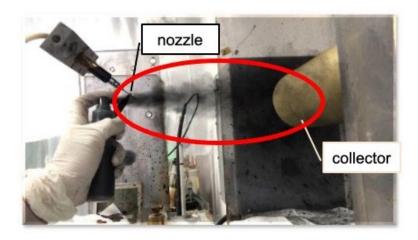
Nutthira Pakkang^a, Shiori Suzuki^b, Kengo Shigetomi^b, Yasumitsu Uraki^b*

	Table of Contents	Page #
Figure S1	A picture of spraying a 1 wt.% conductive carbon black (CB) suspension in acetone over an electrospun fiber mat during the electrospinning of the HKL-containing dope. The CB suspension was simply prepared by mixing a predetermined amount of CB powder and acetone with stirring at RT.	S2
Figure S2	2D and 3D surface images of the HKL-based electrodes: (a, b) E-S-CB-mat, (c, d) E-MS-CB-mat, and (e, f) E-S-CB-powder at a magnification ×2000.	S3
	How to calculate surface roughness (Sa)	S4
Figure S3	Average cyclic voltammograms (CVs) of HKL-based EDLCs and a reference EDLC at scan rate of $0.05~{\rm V~s^{-1}}$.	S5
Figure S4	Nyquist plots of HKL-based EDLCs and a reference EDLC.	S6
Figure S5	2D surface images of ACF mats and schematic illustrations of (a) a single thermostabilized fiber of tHKL separator and single carbon fibers of (b) S-CB-mat and (c) MS-CB-mat. These carbon fibers were activated by steam to develop micro- and mesopores in the fibers. CB aggregates are located both on the surface and inside of the S-CB-mat, while the aggregates positions only inside the MS-CB-mat in which CB fine powders would be dispersed in each single fiber.	S7
Figure S6	Cyclic stability of HKL-based EDLCs and a reference EDLC as shown by capacitance retention from GCD at current density of 1 A g ⁻¹ .	S8
Figure S7	Comparison of energy and power densities of lignin-based EDLCs.	S9
	References in Figure S7	S10-11

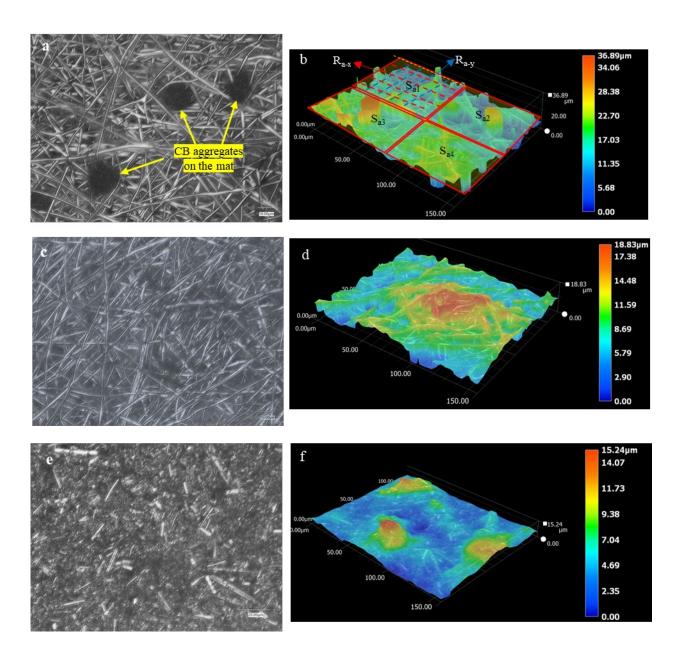
^a Graduate School of Agriculture, Hokkaido University, Sapporo 060-8589, Japan

^b Research Faculty of Agriculture, Hokkaido University, Sapporo 060-8589, Japan

^{*}Corresponding author: Yasumitsu Uraki; E-mail address: uraki@for.agr.hokudai.ac.jp

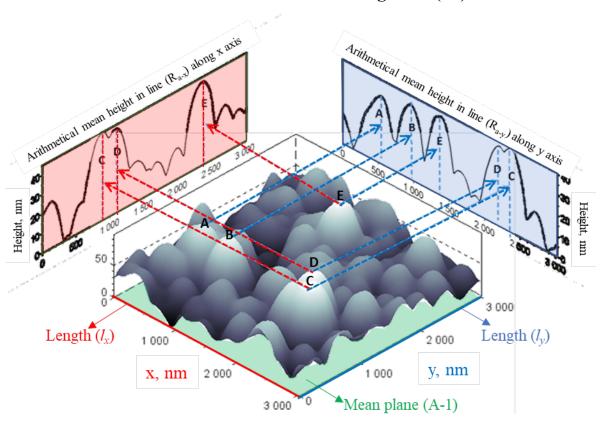


Supplementary Figure S1: A picture of spraying a 1 wt.% conductive carbon black (CB) suspension in acetone over an electrospun fiber mat during the electrospinning of the HKL-containing dope. The CB suspension was simply prepared by mixing a predetermined amount of CB powder and acetone with stirring at RT.



Supplementary Figure S2: 2D and 3D surface images of the HKL-based electrodes: (a, b) E-S-CB-mat, (c, d) E-MS-CB-mat, and (e, f) E-S-CB-powder at a magnification ×2000.

How to calculate surface roughness (Sa)



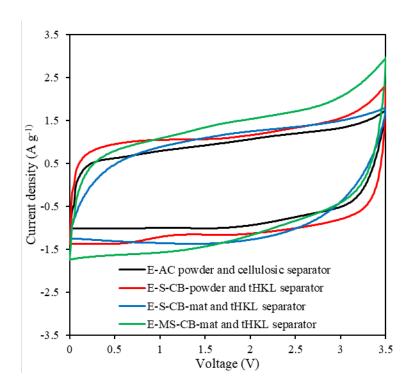
Arithmetical mean height in line (Ra)

along x axis: $R_{a-x} = \frac{1}{l_x} \int_0^{l_x} |z(x)| dx$

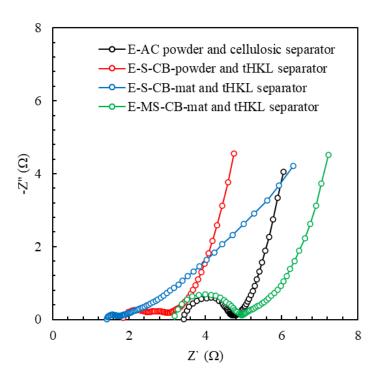
along y axis: $R_{a-y} = \frac{1}{l_y} \int_0^{l_y} |z(y)| dy$

Arithmetical mean height of the surface (Sa)

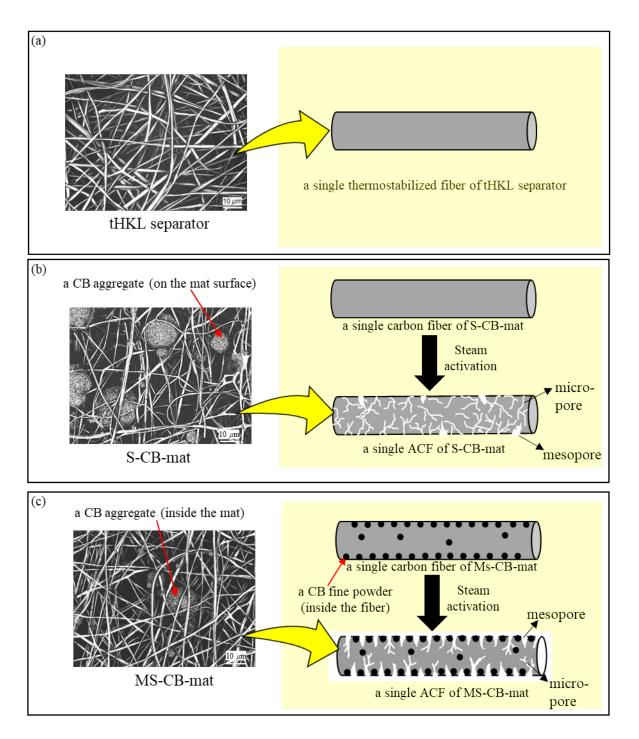
Mean plane: $S_{a1} = \frac{1}{A_1} \iint_{A_1} |z(x, y)| dx dy$



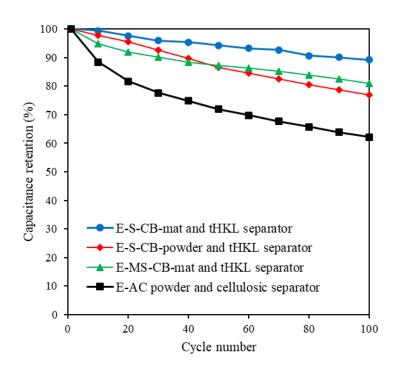
Supplementary Figure S3: Average cyclic voltammograms (CVs) of HKL-based EDLCs and a reference EDLC at scan rate of $0.05~\rm V~s^{-1}$.



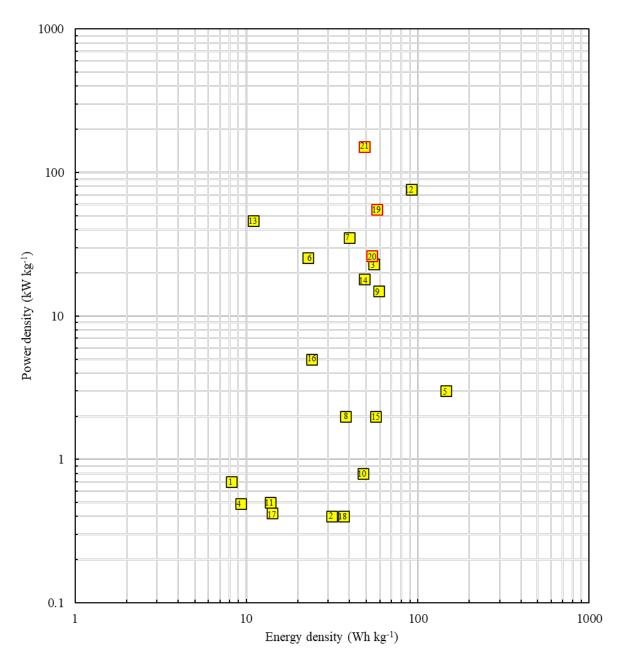
Supplementary Figure S4: Nyquist plots of HKL-based EDLCs and a reference EDLC.



Supplementary Figure S5: 2D surface images of ACF mats and schematic illustrations of (a) a single thermostabilized fiber of tHKL separator and single carbon fibers of (b) S-CB-mat and (c) MS-CB-mat. These carbon fibers were activated by steam to develop micro- and mesopores in the fibers. CB aggregates are located both on the surface and inside of the S-CB-mat, while the aggregates positions only inside the MS-CB-mat in which CB fine powders would be dispersed in each single fiber.



Supplementary Figure S6: Cyclic stability of HKL-based EDLCs and a reference EDLC as shown by capacitance retention from GCD at current density of $1\,\mathrm{A\,g^{-1}}$.



Supplementary Figure S7: Comparison of energy and power densities of lignin-based EDLCs.

References in Figure S7

- 1. Ajjan, F. N., Vagin, M., Rębiś, T., Aguirre, L. E., Ouyang, L. Q., and Inganäs, O. (2017). Scalable asymmetric supercapacitors based on hybrid organic/biopolymer electrodes. Adv. Sustain. Syst. 1: 1700054.
- 2. Cao, Q. P., Zhu, M. N., Chen, J. A., Song, Y. Y., Li, Y., and Zhou, J. H. (2020). Novel lignin-cellulose-based carbon nanofibers as high-performance supercapacitors. ACS Appl. Mater. Interfaces 12: 1210–1221.
- 3. Chen, W. M., Wang, X., Feizbakhshan, M., Liu, C. Z., Hong, S., Yang, P., and Zhou, X. Y. (2019). Preparation of lignin-based porous carbon with hierarchical oxygen-enriched structure for high-performance supercapacitors. J. Colloid and Interface Sci. 540: 524–534.
- 4. Dai, Z., Ren, P. G., Jin, Y. L., Zhang, H., Ren, F., and Zhang, Q. (2019). Nitrogen-sulphur co-doped graphenes modified electrospun lignin/polyacrylonitrile-based carbon nanofiber as high performance supercapacitor. J. Power Sources 437: 226937.
- 5. Das, S. and Ghosh, A. (2017). Solid Polymer Electrolyte Based on PVDF-HFP and Ionic Liquid Embedded with TiO2Nanoparticle for Electric Double Layer Capacitor (EDLC) Application. J. Electrochem. Soc. 164: F1348–F1353.
- 6. Guo, N. N., Li, M., Sun, X. K., Wang, F., and Yang, R. (2017). Enzymatic hydrolysis lignin derived hierarchical porous carbon for supercapacitors in ionic liquids with high power and energy densities. Green Chem. 19: 2595–2602.
- 7. Härmas, R., Palm, R., Härmas, M., Pohl, M., Kurig, H., Tallo, I., Tee, E., Vaas, I., Väli, R., Romann, T., Oll, O., Kanarbik, R., Liivand, K., Eskusson, J., Kruusma, J., Thomberg, T., Jänes, A., Miidla, P., and Lust, E. (2018). Influence of porosity parameters and electrolyte chemical composition on the power densities of non-aqueous and ionic liquid based supercapacitors. Electrochimica Acta 283: 931–948.
- 8. Jayawickramage, R. A. P. and Ferraris, J. P. (2019a). High performance supercapacitors using lignin based electrospun carbon nanofiber electrodes in ionic liquid electrolytes. Nanotechnology 30: 155402.
- 9. Jayawickramage, R. A. P., Balkus, K.J., and Ferraris, J.P. (2019b). Binder free carbon nanofiber electrodes derived from polyacrylonitrile- lignin blends for high performance supercapacitors. Nanotechnology 30: 355402.
- 10. Lei, D. Y., Li, X. D., Seo, M. K., Khil, M. S., Kim, H. Y., and Kim, B. S. (2017). NiCo2O4 nanostructure-decorated PAN/lignin based carbon nanofiber electrodes with excellent cyclability for flexible hybrid supercapacitors. Polymer 132: 31–40.
- 11. Li, F. F., Wang, X. L., and Sun, R. C. (2017). A metal-free and flexible supercapacitor based on redox-active lignosulfonate functionalized graphene hydrogels. J. Mater. Chem. A 5: 20643–20650.
- 12. Pakkang, N., Kumar, M., Taira, S., Koda, K., Shigetomi, K., and Uraki, Y. (2020). Preparation of kraft lignin-based activated carbon fiber electrodes for electric double layer capacitors using an ionic liquid electrolyte. Holzforschung 74: 577–588.

- 13. Senokos, E., Reguero, V., Cabana, L., Palma, J., Marcilla, R., and Vilatela, J. J. (2017). Large-Area, All-Solid, and Flexible Electric Double Layer Capacitors Based on CNT Fiber Electrodes and Polymer Electrolytes. Adv. Mater. Technol. 2: 1600290.
- 14. Shao, Q., Tang, J., Lin, Y., Li, J., Qin, F., Zhang, K., Yuan, J., and Qin, L. C. (2015). Ionic liquid modified graphene for supercapacitors with high rate capability. Electrochimica Acta 176: 1441–1446.
- 15. Wang, J., Tang, J., Xu, Y., Ding, B., Chang, Z., Wang, Y., Hao, X., Dou, H., Kim, J., Zhang, X., and Yamauchi, Y. (2016). Interface miscibility induced double-capillary carbon nanofibers for flexible electric double layer capacitors. Nano Energy 28: 232–240.
- 16. Yadav, N., Yadav, N., and Hashmi, S. (2020). Ionic liquid incorporated, redox-active blend polymer electrolyte for high energy density quasi-solid-state carbon supercapacitor. J. Power Sources 451: 227771.
- 17. You, X. Y., Koda, K., Yamada, T., and Uraki, Y. (2016). Preparation of high-performance internal tandem electric double-layer capacitors (IT-EDLCs) from melt-spun lignin fibers. J. Wood Chem. Technol. 36: 418–431.
- Zhu, M. N., Liu, H., Cao, Q. P., Zheng, H., Xu, D., Guo, H. Y., Wang, S. M., Li, and Y., Zhou, J. H. (2020). Electrospun lignin-based carbon nanofibers as supercapacitor electrodes, ACS Sustain. Chem. Eng. 8: 12831–12841.
- 19. This work (E-MS-CB-mat)
- 20. This work (E-S-CB-mat)
- 21. This work (E-S-CB-powder)