Supporting Information

# Multifunctional Carbon Aerogels with Hierarchical Anisotropic Structure Derived from Lignin and Cellulose Nanofibers for $CO_2$ Adsorption and Energy Storage

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**Table S1.** Porosity and density of the LTCAs.

| Sample coding | Porosity (%) | Density (g $cm^{-3}$) |
|---|---|---|
| LTCA8 | 91.6 | 0.175 |
| LTCA10 | 91.9 | 0.169 |
| LTCA12 | 93.4 | 0.138 |

**Figure S1.** Contact angle demonstration in which one droplet of distilled water was deposited on the surface of LTCA12. The image was taken from the first second after the deposition.

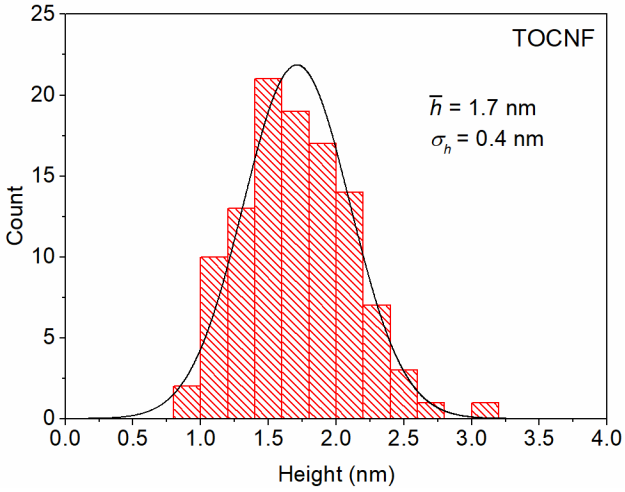


**Figure S2**. Histogram of the TOCNF width measured from AFM height images.

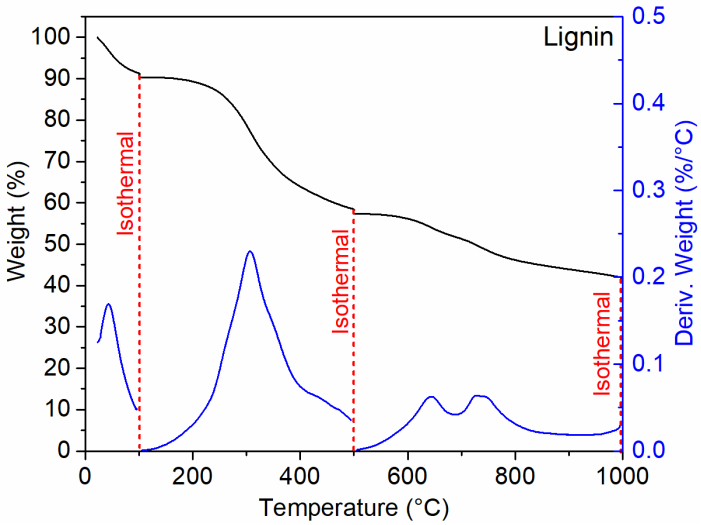


**Figure S3**. TGA curves of lignin used in this study.

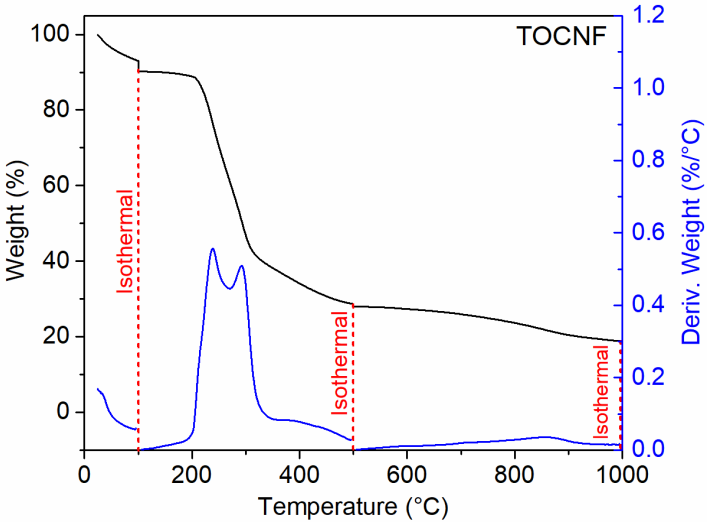


**Figure S4**. TGA curves of TOCNF used in this study.

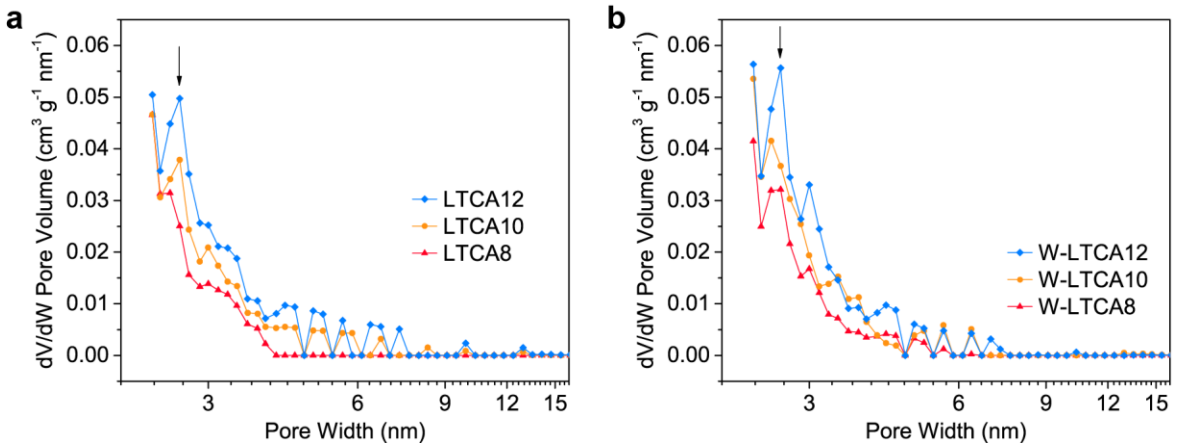


**Figure S5**. Pore size distribution of (a) LTCAs and (b) W-LTCAs calculated using non-local density functional theory (NLDFT) for $N_2$ adsorption at 77 K.

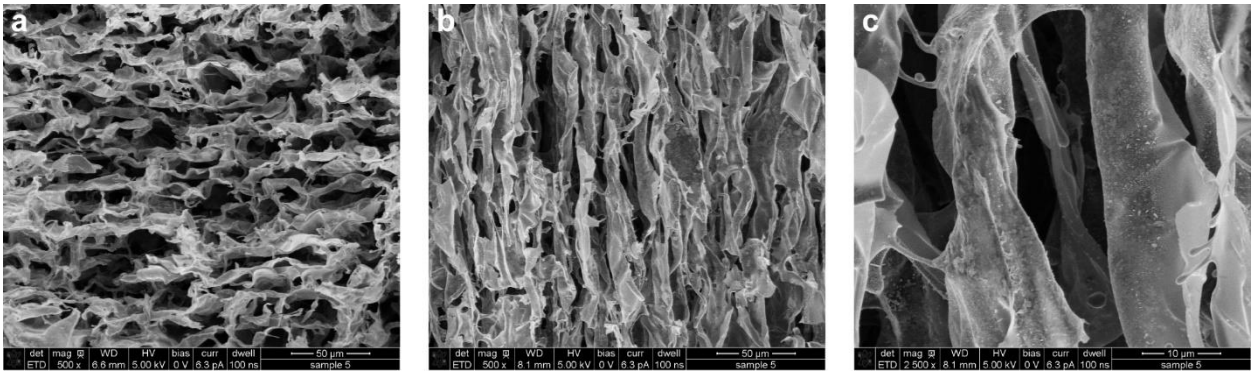


**Figure S6**. SEM images of LTCA8 from the (a) cross-section, (b) longitudinal section and (c) magnified view.

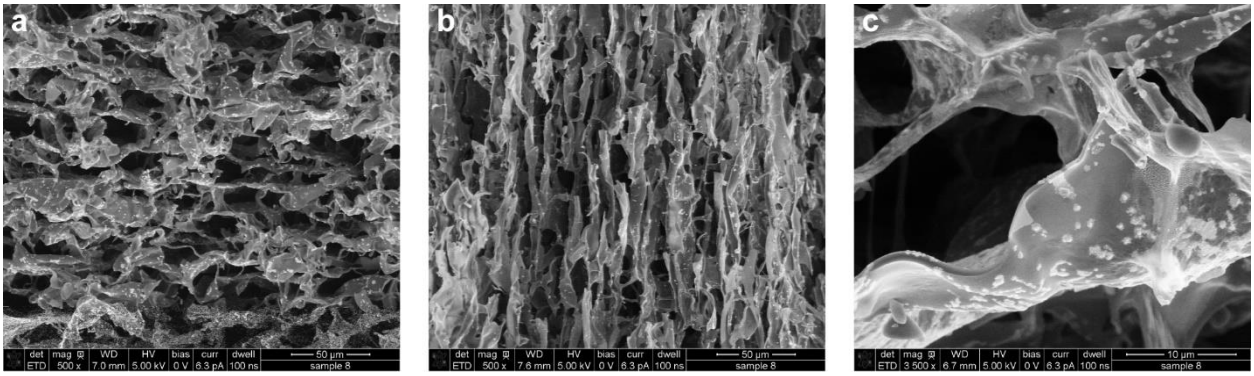


**Figure S7**. SEM images of LTCA10 from the (a) cross-section, (b) longitudinal section and (c) magnified view.

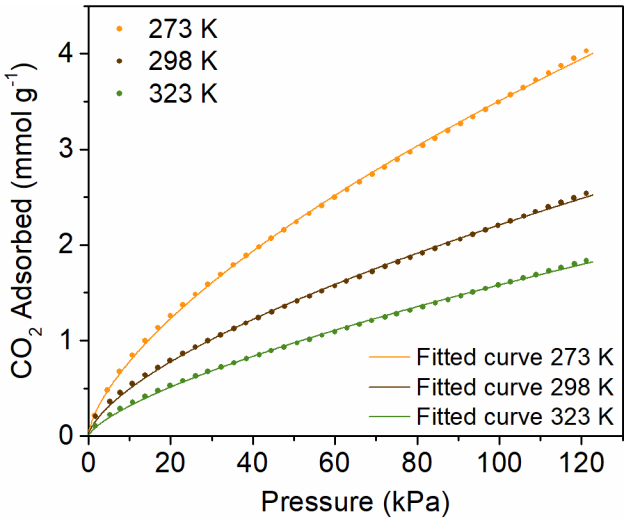


**Figure S8**. $CO_2$ adsorption isotherms of LTCA10 at different temperatures and the related Langmuir fitted curves.

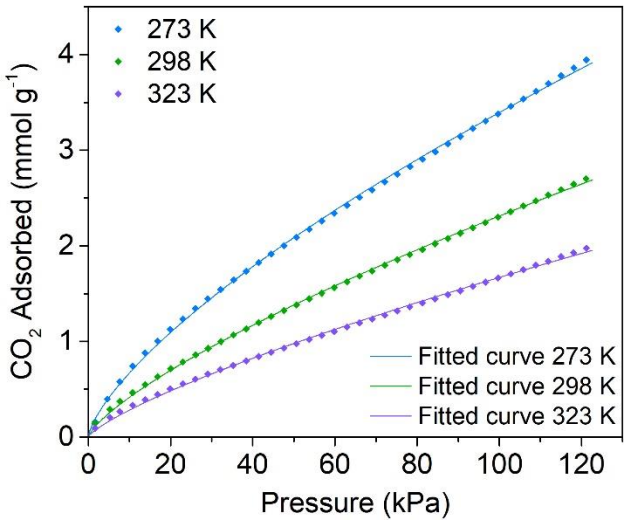


**Figure S9**. $CO_2$ adsorption isotherms of LTCA12 at different temperatures and the related Langmuir fitted curves.

**Table S2**. The data collected from the BET surface analysis.

| Sample | BET surface area ($m^2\ g^{-1}$) | t-Plot micropore area ($m^2\ g^{-1}$) | Pore volume* ($cm^3\ g^{-1}$) | Pore size (nm) |
|---|---|---|---|---|
| LTCA8 | 413 | 289 | 0.20 | 1.92 |
| LTCA10 | 704 | 458 | 0.34 | 1.93 |
| LTCA12 | 806 | 487 | 0.39 | 1.95 |
| W-LTCA8 | 617 | 476 | 0.27 | 1.72 |
| W-LTCA10 | 839 | 542 | 0.40 | 1.91 |
| W-LTCA12 | 1101 | 638 | 0.55 | 1.99 |

*Total pore volume of the pores having the diameter less than 200 nm

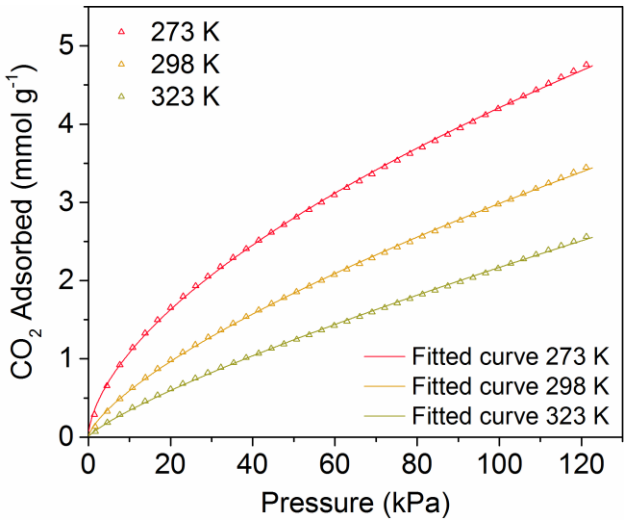


**Figure S10**. $CO_2$ adsorption isotherms of W-LTCA8 at different temperatures and the related Langmuir fitted curves.

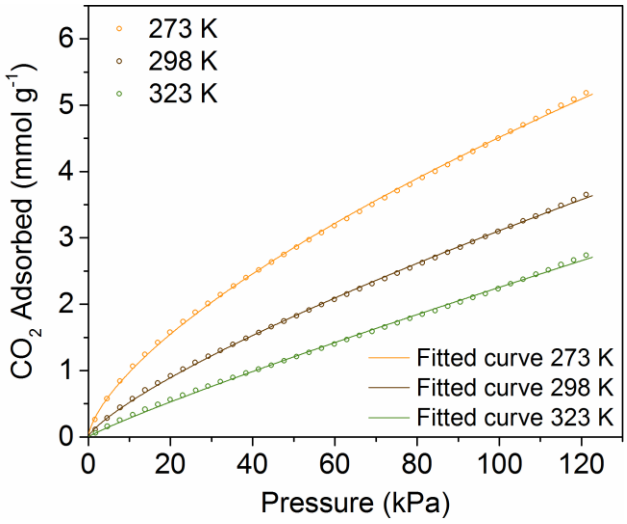


**Figure S11**. $CO_2$ adsorption isotherms of W-LTCA10 at different temperatures and the related Langmuir fitted curves.

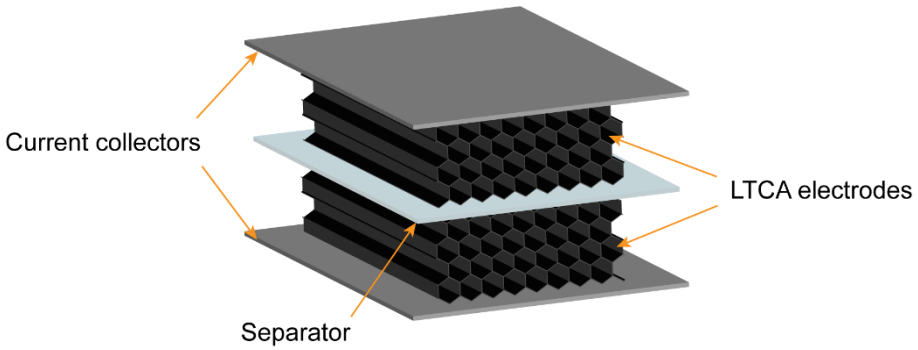


**Figure S12**. Schematic of the two-electrode setup used in the electrochemical tests.

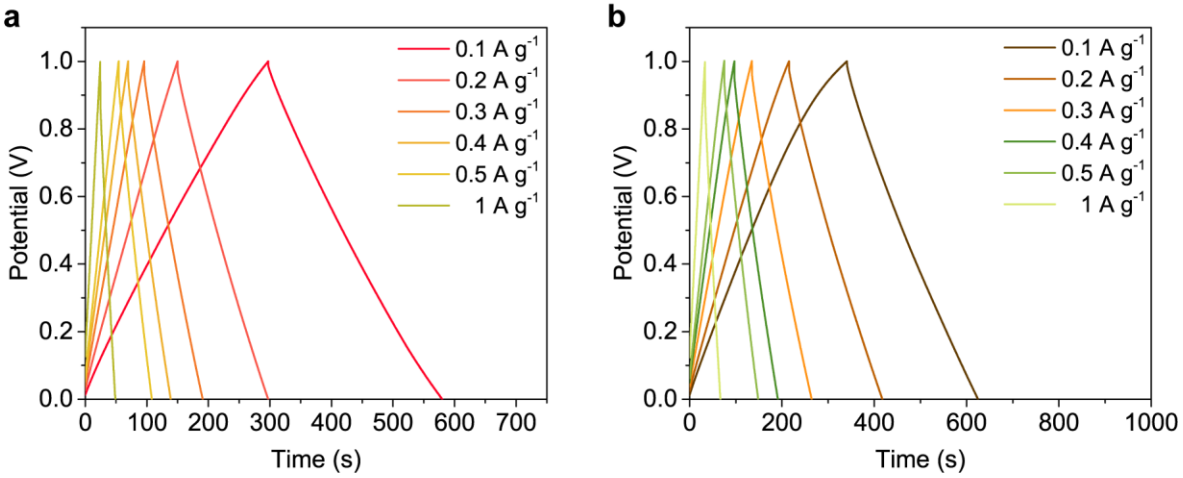


**Figure S13**. GCD curves of the SCs with (a) LTCA8 and (b) LTCA10 electrodes tested at different current densities.

**Table S3.** Electrochemical properties of the SCs with LTCA electrodes.

| Sample | Current density | | $C_g$ | $C_{cell}$* | $C_a$ | $E_d$ | $P_d$ |
|---|---|---|---|---|---|---|---|
| | (A $g^{-1}$) | (mA $cm^{-2}$) | (F $g^{-1}$) | (F $g^{-1}$) | (F $cm^{-2}$) | (Wh $kg^{-1}$) | (W $kg^{-1}$) |
| LTCA8 | 0.1 | 3.1 | 113.2 | 28.3 | 1.74 | 3.93 | 50 |
| | 0.2 | 6.1 | 110.0 | 27.5 | 1.69 | 3.82 | 100 |
| | 0.3 | 9.2 | 106.4 | 26.6 | 1.63 | 3.69 | 150 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | 0.4 | 12.3 | 103.5 | 25.9 | 1.59 | 3.59 | 200 |
| | 0.5 | 15.3 | 100.9 | 25.2 | 1.55 | 3.50 | 250 |
| | 1 | 30.7 | 91.1 | 22.8 | 1.40 | 3.16 | 500 |
| | 2 | 61.4 | 76.4 | 19.1 | 1.17 | 2.65 | 1000 |
| | 3 | 92.1 | 56.0 | 14.0 | 0.86 | 1.94 | 1500 |
| | 4 | 122.8 | 42.7 | 10.7 | 0.65 | 1.48 | 2000 |
| | 5 | 153.5 | 31.1 | 7.8 | 0.48 | 1.08 | 2500 |
| LTCA10 | 0.1 | 3.0 | 113.5 | 28.4 | 1.71 | 3.94 | 50 |
| | 0.2 | 6.0 | 113.3 | 28.3 | 1.71 | 3.93 | 100 |
| | 0.3 | 9.0 | 108.8 | 27.2 | 1.64 | 3.78 | 150 |
| | 0.4 | 12.1 | 105.6 | 26.4 | 1.59 | 3.67 | 200 |
| | 0.5 | 15.1 | 102.9 | 25.7 | 1.55 | 3.57 | 250 |
| | 1 | 30.1 | 93.2 | 23.3 | 1.41 | 3.24 | 500 |
| | 2 | 60.3 | 77.1 | 19.3 | 1.16 | 2.68 | 1000 |
| LTCA12 | 0.1 | 1.9 | 123.4 | 30.8 | 1.20 | 4.28 | 50 |
| | 0.2 | 3.9 | 124.4 | 31.1 | 1.21 | 4.32 | 100 |
| | 0.3 | 5.8 | 120.7 | 30.2 | 1.18 | 4.19 | 150 |
| | 0.4 | 7.8 | 119.9 | 30.0 | 1.17 | 4.16 | 200 |
| | 0.5 | 9.7 | 118.1 | 29.5 | 1.15 | 4.10 | 250 |
| | 1 | 19.5 | 111.8 | 27.9 | 1.09 | 3.88 | 500 |
| | 2 | 39.0 | 100.5 | 25.1 | 0.98 | 3.49 | 1000 |
| | 3 | 58.4 | 92.3 | 23.1 | 0.90 | 3.20 | 1500 |
| | 4 | 77.9 | 85.5 | 21.4 | 0.83 | 2.97 | 2000 |
| | 5 | 97.4 | 80.6 | 20.2 | 0.79 | 2.80 | 2500 |

*Cell capacitance ($C_{cell}$) of the SCs was calculated using:

$$C_{cell} = \frac{I \cdot \Delta t}{\Delta V \cdot m} = C_g/4 \qquad \text{Eq. S1}$$