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## **Supporting Information**

Nanocellulose supported hierarchical structured

polyaniline/nanocarbon nanocomposite electrode via layer-by-layer

assembly for green flexible supercapacitors

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Figure S1. TEM images of (a) PANi, (b) CMWCNT, and (c) GO. (d) Particle-size distributions of PANi, CMWCNT, and GO.



Figure S2. Representative photographic image of (a) hydrogels in wet state after assembly and (b, c) the

compressed aerogel film.



Figure S3. TEM images of (a) pure CNF nanofibril, (b) PC<sub>10</sub> and (c) PG<sub>10</sub> assembled on CNF nanofibril.



**Figure S4.** (a) Pore size distributions of the CNF,  $CPC_{10}$ , and  $CPG_{10}$  aerogels. (b) XPS spectra of the CNF,  $CPC_{10}$ , and  $CPG_{10}$  aerogels. (c) Atomic percentages in the CNF,  $CPC_{10}$ , and  $CPG_{10}$  aerogels. (d) High-resolution XPS C1s spectrum of the CNF aerogel, and its deconvolution. High-resolution XPS N1s spectra of the (e)  $CPC_{10}$  aerogel and (f)  $CPG_{10}$  aerogel, and their deconvolutions.



**Figure S5.** (a) CV curves for the CPC<sub>10</sub> and CPR<sub>10</sub> electrodes at a scan rate of 5 mV s<sup>-1</sup> under three-electrode testing conditions in 1 M H<sub>2</sub>SO<sub>4</sub>. (b) Graphs of log *I* vs. log *v* for the CPC<sub>10</sub> and CPR<sub>10</sub> electrodes used to obtain *b* values.



Figure S6. (a) Mass loadings and (b) conductivities of the  $CPC_n$  and  $CPR_n$  electrodes as functions LbL assembly cycles.



**Figure S7.** Bode plots of the (a) lgZ and (b) phase angle versus frequency in the three-electrode test in 1 M H<sub>2</sub>SO<sub>4</sub>. (c) Nyquist plots for the CPC<sub>10</sub> and CPR<sub>10</sub> electrodes under two-electrode test with PVA/H<sub>3</sub>PO<sub>4</sub> gel electrolyte. Bode plots of the (d) lgZ and (e) phase angle versus frequency in the two-electrode test with PVA/H<sub>3</sub>PO<sub>4</sub> gel electrolyte.



Figure S8. CV curves for (a)  $CPC_{10}$  and (b)  $CPR_{10}$  electrodes during cycle-stability testing. All data were obtained under two-electrode testing conditions with  $PVA/H_3PO_4$  gel electrolyte.



**Figure S9.** Optical images of the (a) front and the (c) back sides of electrodes. Optical images of supercapacitor device showing (b) working area and (d) current collector



**Figure S10.** (a) Coulombic efficiency of the two assembled supercapacitors. All data were obtained under two-electrode testing conditions with  $PVA/H_3PO_4$  gel electrolyte. (b) Stress-strain curves of the  $CPC_{10}$  and  $CPR_{10}$  electrodes.



**Figure S11.** Ragone plots of power density vs. energy density for the two assembled supercapacitors calculated by the mass (a) or volume (b) of the device.