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SUPPORTING INFORMATION

Enabling High-Volumetric-Energy-Density Supercapacitors: Designing Open, Low-Tortuosity Heteroatoms-Doped Porous Carbon-Tube Bundles Electrode

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Experiment Section

Synthesis of PCTBs: Firstly, the dandelion fluffs were sifted with a sieve for wiping off the seeds, then the obtained dandelion fluffs were washed with acetone and deionized (DI) water to remove impurities. Then 5 g dried dandelion fluffs were carbonized at 800℃ for 2 h and cooled to ambient temperature naturally. The obtained carbon was named as CTBs. Subsequently, 0.1 g CTBs was mixed with 0.1 g KOH in 100 mL DI water and dried at 60℃. Ultimately, the solid was further carbonized under Ar at 850℃ for 1 h in a tube furnace. After cooling down to room temperature, the as-prepared carbon sample was washed with 1 M HCl and DI water several times, followed by drying at 60℃ overnight. The collected sample was named as PCTBs. For comparison, 0.1 g CTBs mixed with 0.2 g KOH (100 mL) in a 1:2 mass ratio was prepared to investigate the effect of higher KOH content on the tube bundled structure, the collected sample was named as PCTBs1-2.

Synthesis of B, N co-doping PCTBs: For preparation of the B, N co-doping carbon material, the boric and the urea were served as the B and N precursors, respectively. 50 ml urea (1.05 g) and boric (1.1 g) solution were mixed with 0.5 g PCTBs in 50 mL DI water with slightly stirring. After dry in 60^{o}C, the obtained sample was further pyrolyzed at 850℃ for 1 h under Ar atmosphere. The collected sample was named as B/N-PCTBs. For comparison, the boric was selected to be as the B precursor for single B-doping. The obtained sample was named as B-PCTBs. In a similar way, the urea was chosen to prepare the single N-doping carbon material, which was denoted as N-PCTBs. The AC as a comparison was purchased from Kurary company in Japan (YP-50F).

Characterization methods: The morphologies and elements distribution of the prepared samples were observed by scanning electron microscope (SEM, Hitachi Su8000), transmission electron microscopy (TEM, FEI Teccai G2 S-Twin, Philips) and energy-dispersive X-ray (EDX) equipment. The phase components of the materials were investigated by the X-ray diffraction (XRD) (Rigaku TTR III) with Cu Kα radiation in the 2θ range from 10° to 90°. Brunauer-Emmett-Teller (BET, Micromeritics TriStar 3020) nitrogen adsorption-desorption analysis was used to test the specific surface areas (SSA) of the electrode materials under 77 K. The functional group and element contents of the electrode materials were presented by XPS (Tryhermo ESCALAB 250). The defective characterization of the samples was carried out by Raman spectra with a Jobin-Yvon HR800 Raman spectrometer.

Electrochemical Measurements: Electrodes were prepared by mixing in accordance with active material, carbon black and PVDF in a mass ratio of 8: 1: 1. Then the mixture was coated into the foam nickel (1cm×1cm) then dried at 80℃ for 12 h. Figure S11a and b show the SEM images of the cross-section of the B/N-PCTBs electrode with mass loadings of 40 mg cm^{-2} and 2.6 mg cm^{-2}, respectively. The B/N-PCTBs was used as the working electrode, and the Ag/AgCl electrode and platinum sheet were used as the reference electrode and the counter electrode, respectively. The electrochemical performance of single electrode was conducted in a three-electrode system in 6 M KOH aqueous solution, including cyclic voltammetry (CV), galvanostatic charge/discharge tests and electrochemical impedance spectroscopy (EIS) on a VMP3/Z Bio-Logic electrochemical workstation. The cycling life was performed by a LAND measurement system. The specific capacitance (Cs) of the as-prepared samples can be calculated from galvanostatic charge/discharge curves based on the equation as follows:

$$\mathrm{Cs} = \frac{\mathrm{I}\Delta \mathrm{t}}{\mathrm{m}\Delta \mathrm{V}} \qquad (1)$$

where I is the discharge current, Δt is the discharge time, m is the mass of active electrode materials on single electrode and ΔV is the voltage change upon discharging after IR drop.

In addition, with the transform of the frequency, the capacitance C(w) can be described below:

$$\mathrm{C} = \mathrm{C}'(w) - \mathrm{j}\mathrm{C}''(w) \qquad (2)$$

$$C'(w) = \frac{-Z''(w)}{w\,|Z(w)|^2} \qquad (3)$$

$$C''(w) = \frac{Z'(w)}{w\,|Z(w)|^2} \qquad (4)$$

Taking into account the cathodic and anodic limits in the different electrolytes, it is well-known that the highest stability window can be attained in neutral solution. The symmetric supercapacitor was assembled with a fibrous separator and conducted in a two-electrode system with 1 M Na_2SO_4 aqueous solution. The thickness of the electrode is about 490 μm and the total volume of the symmetric cell is about 0.1 cm^3, including the cathode, separator and anode. The main parameters of the device, the power density (P: W L^{-1}) and energy density (E: Wh L^{-1}) were obtained with equations:

$$E = \frac{1}{7.2} \times Cs \times \Delta V^2 \qquad (5)$$

$$P = \frac{E}{\Delta t} \times 3600 \qquad (6)$$

where Cs (F g^{-1}) is the specific capacitance of the material, Δt is the discharge time (s) and ΔV is the range of the discharge potential.

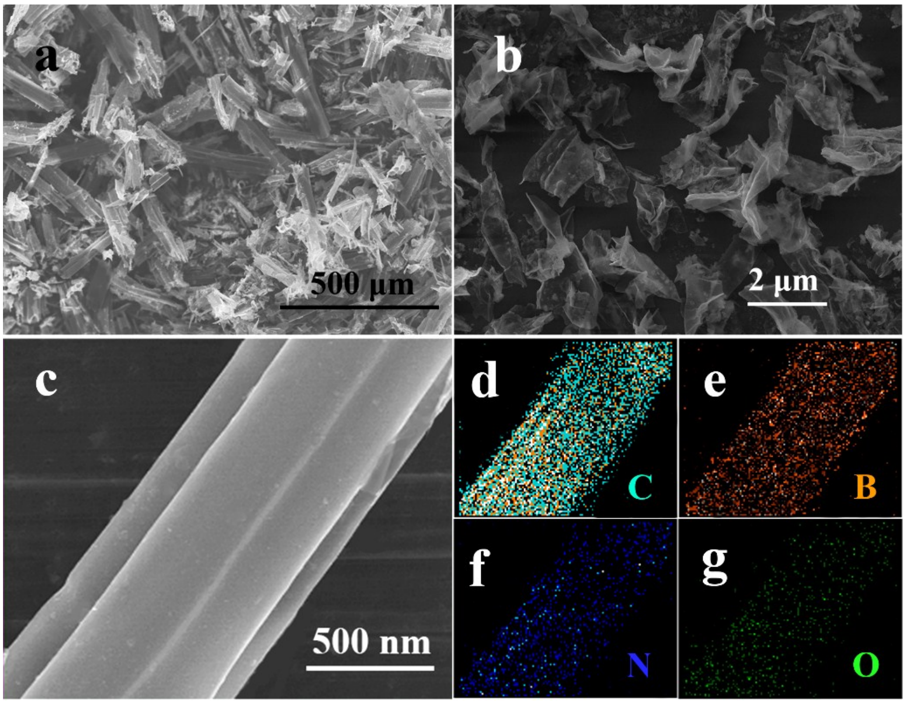

Figure S1. (a, b) SEM images of the PCTBs1-2. The SEM images demonstrate the morphology of the carbon tube bundle is destroyed by the KOH. (c-g) SEM image and corresponding C, B, N and O elemental mappings of the B/N-PCTBs, demonstrating the uniform distributions of B, N and O atoms.

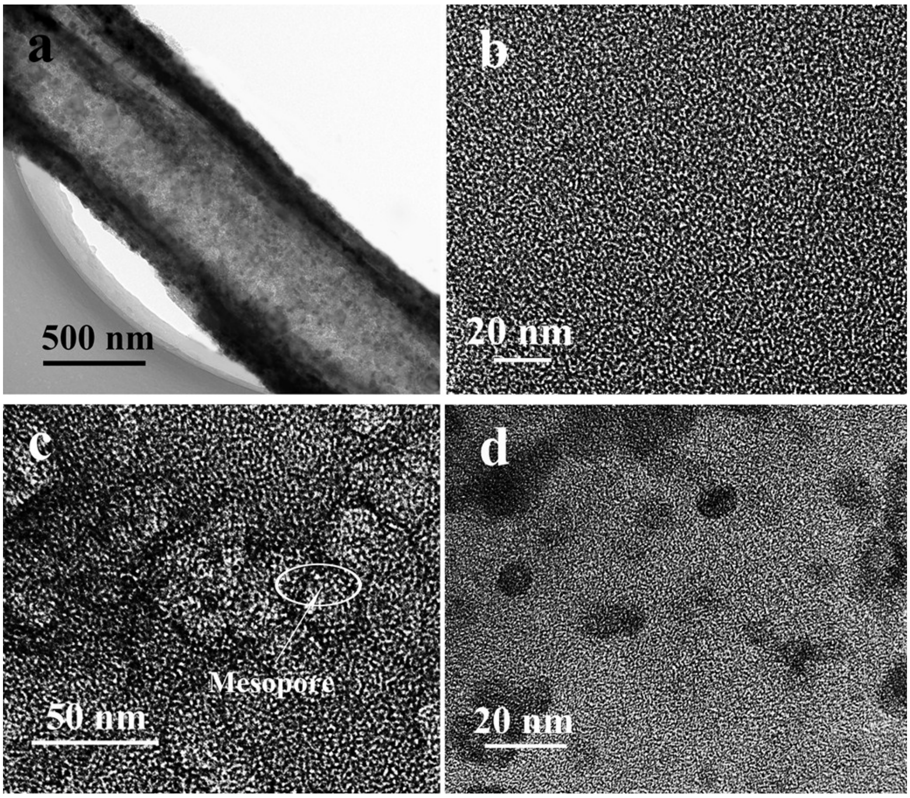

Figure S2. (a) TEM and (b) HRTEM images of the CTBs. (c, d) HRTEM images of the PCTBs, illustrating the abundant mesopores and micropores distribute uniformly inside the PCTBs.

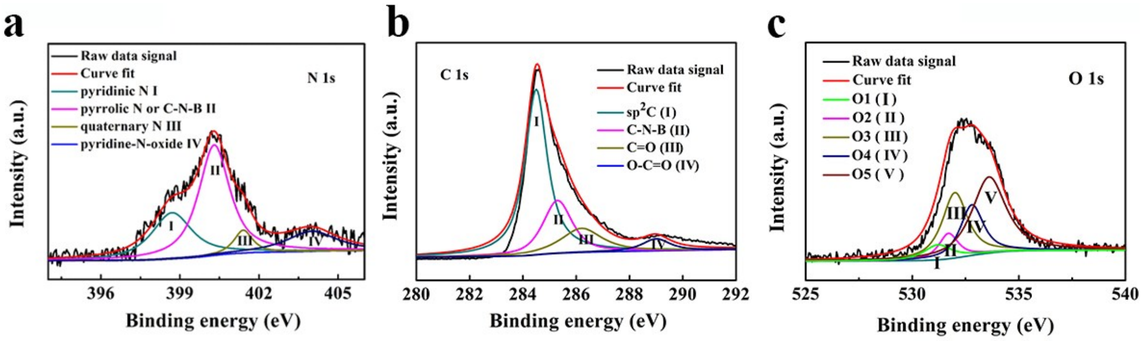

Figure S3. (a) High-resolution N1s and its fitting peaks of the N-PCTBs. (b) High-resolution C1s and its fitting peaks of the B/N-PCTBs. (c) High-resolution O1s and its fitting peaks of the B/N-PCTBs.

Table S1. Element contents of the CTBs, PCTBs, N-PCTBs, B-PCTBs and B/N-PCTBs.

Carbon Samples	**Composition (at%)**			
	C	**O**	**N**	**B**
CTBs	91.32	8.54	0.14	-
PCTBs2-1	91.12	8.78	0.10	-
PCTBs	90.62	9.27	0.11	-
N-PCTBs	90.04	7.08	2.88	-
B-PCTBs	92.62	5.06	0.12	2.2
B/N-PCTBs	88.02	5.18	2.2	4.6

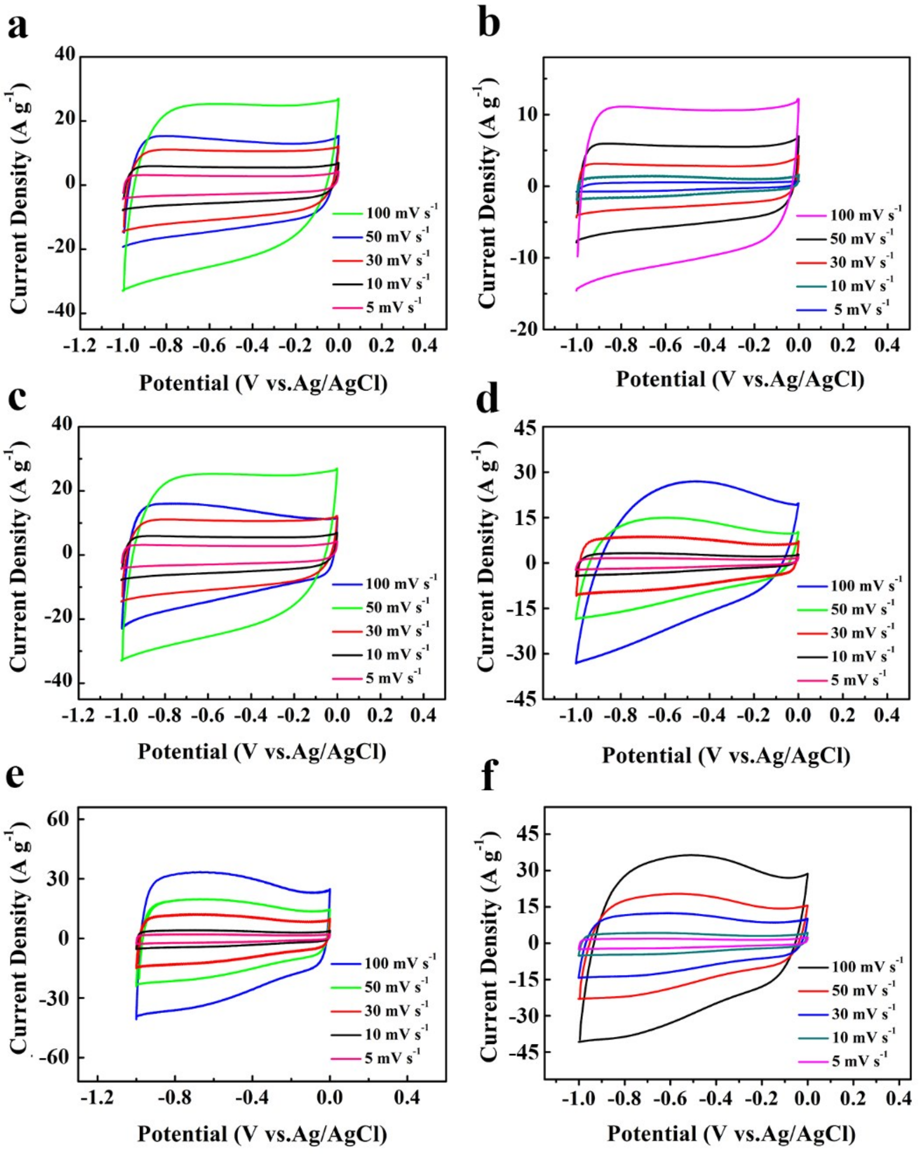

Figure S4. CV curves at different scan rates of 5, 10, 30, 50 and 100 mV s^{-1} of the (a) PCTBs, (b) CTBs, (c) PCTBs2-1, (d) N-PCTBs, (e) B-PCTBs and (f) B/N-PCTBs.

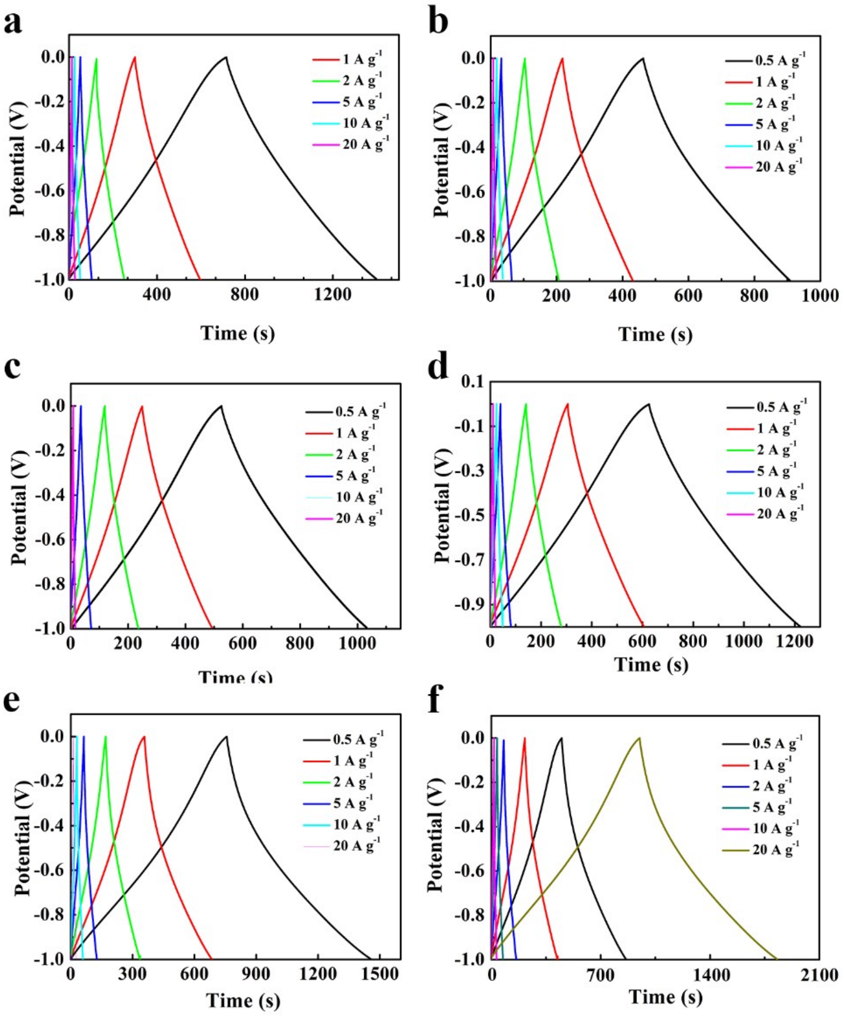

Figure S5. Galvanostatic charge/discharge curves versus various current densities (0.5-20 A g^{-1}) of the (a) PCTBs, (b) CTBs, (c) PCTBs2-1, (d) N-PCTBs, (e) B-PCTBs and (f) B/N-PCTBs.

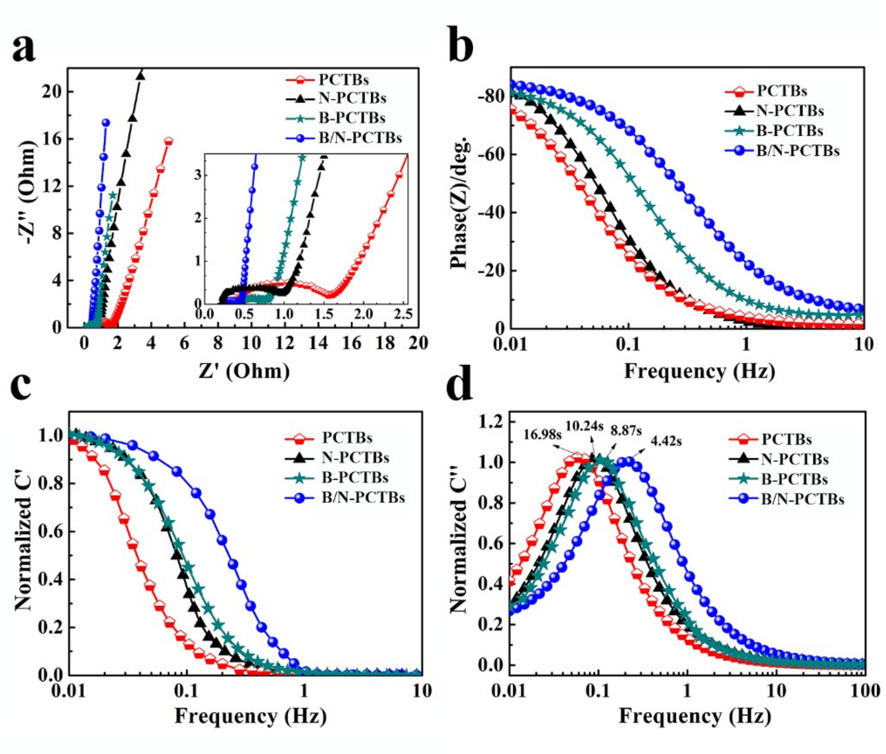

Figure S6. (a) Nyquist plots of the PCTBs, N-PCTBs, B-PCTBs and B/N-PCTBs. The inset is the high frequency region of the Nyquist curves. (b) Bode phase diagrams of the PCTBs, N-PCTBs, B-PCTBs and B/N-PCTBs. (c) Normalized real part of the PCTBs, N-PCTBs, B-PCTBs and B/N-PCTBs. (d) Normalized imaginary part of the PCTBs, N-PCTBs, B-PCTBs and B/N-PCTBs. The short characteristic relaxation time constant of the B/N-PCTBs manifests the better reversible charge/discharge rate than the other carbon samples.

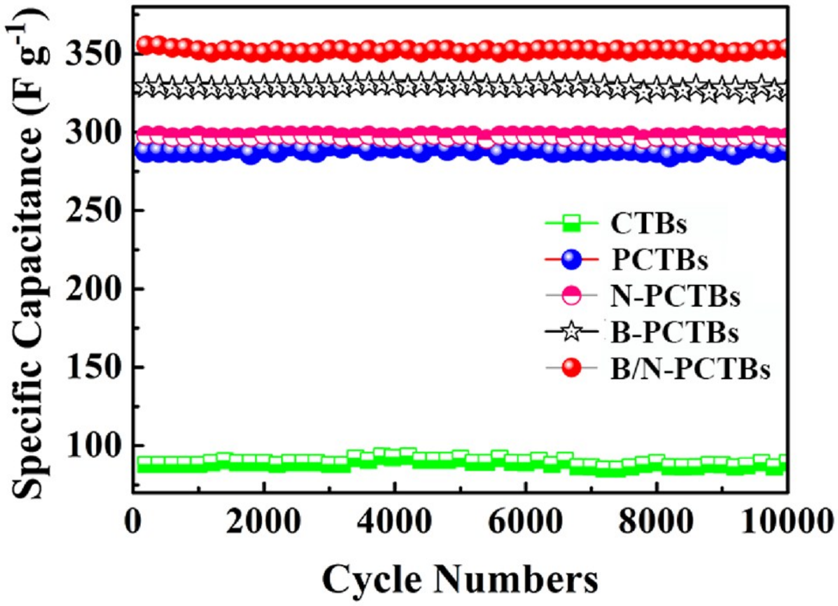

Figure S7. Cycling performance of the CTBs, PCTBs, N-PCTBs, B-PCTBs and B/N-PCTBs electrodes at a 1 A g^{-1} for 10,000 cycles.

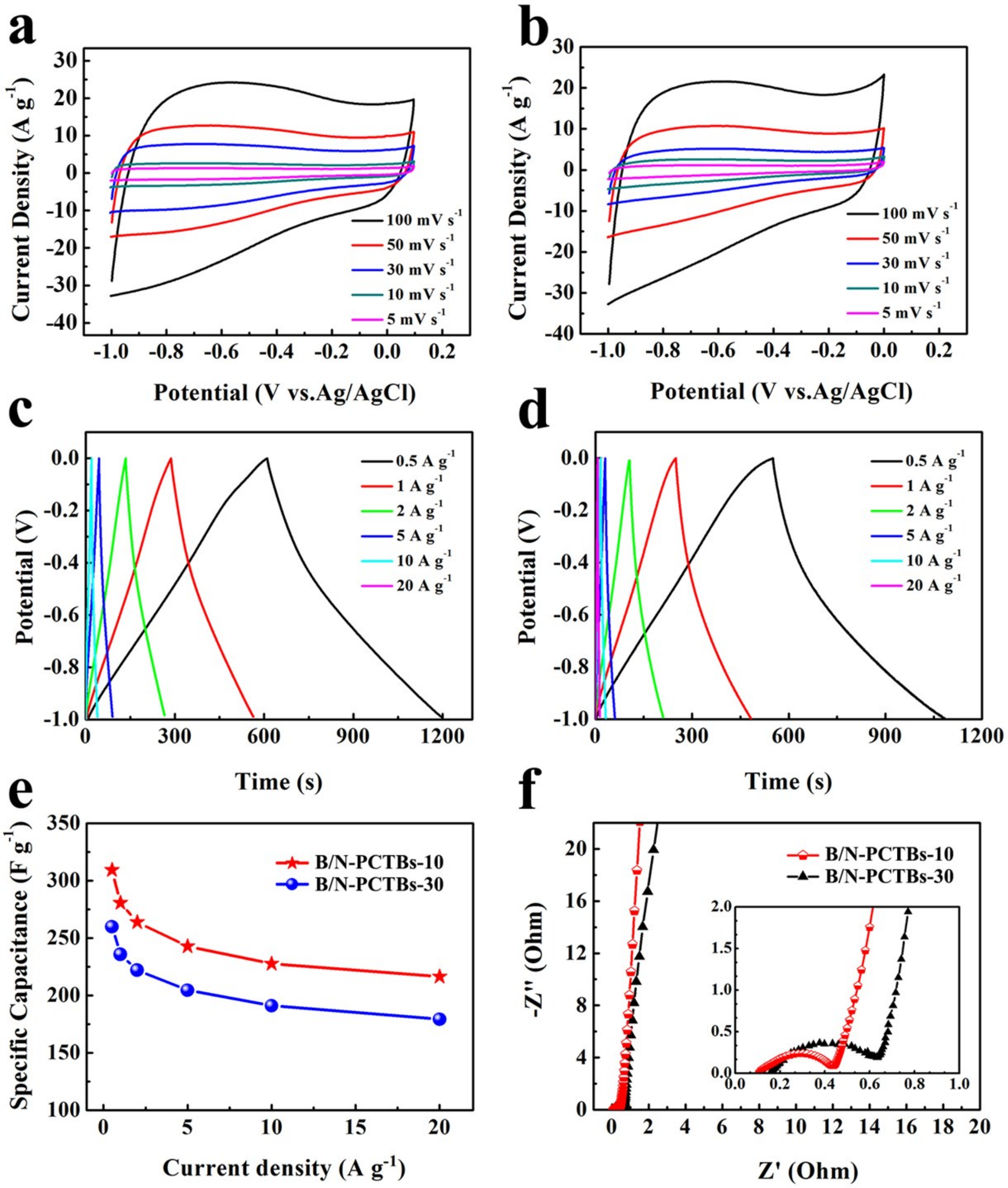

Figure S8. CV curves of the B/N-PCTBs electrodes with 10 mg cm^{-2} (B/N-PCTBs-10) (a) and 30 mg cm^{-2} (B/N-PCTBs-30) (b) at various scan rates of 5, 10, 30, 50 and 100 mV s^{-1}. Galvanostatic charge/discharge curves of the B/N-PCTBs-10 (c) and B/N-PCTBs-30 (d) electrodes at different current densities. (e) Specific capacitances of the B/N-PCTBs-10 and B/N-PCTBs-30 electrodes versus current densities. (f) Nyquist plots of the B/N-PCTBs-10 and B/N-PCTBs-30 electrodes.

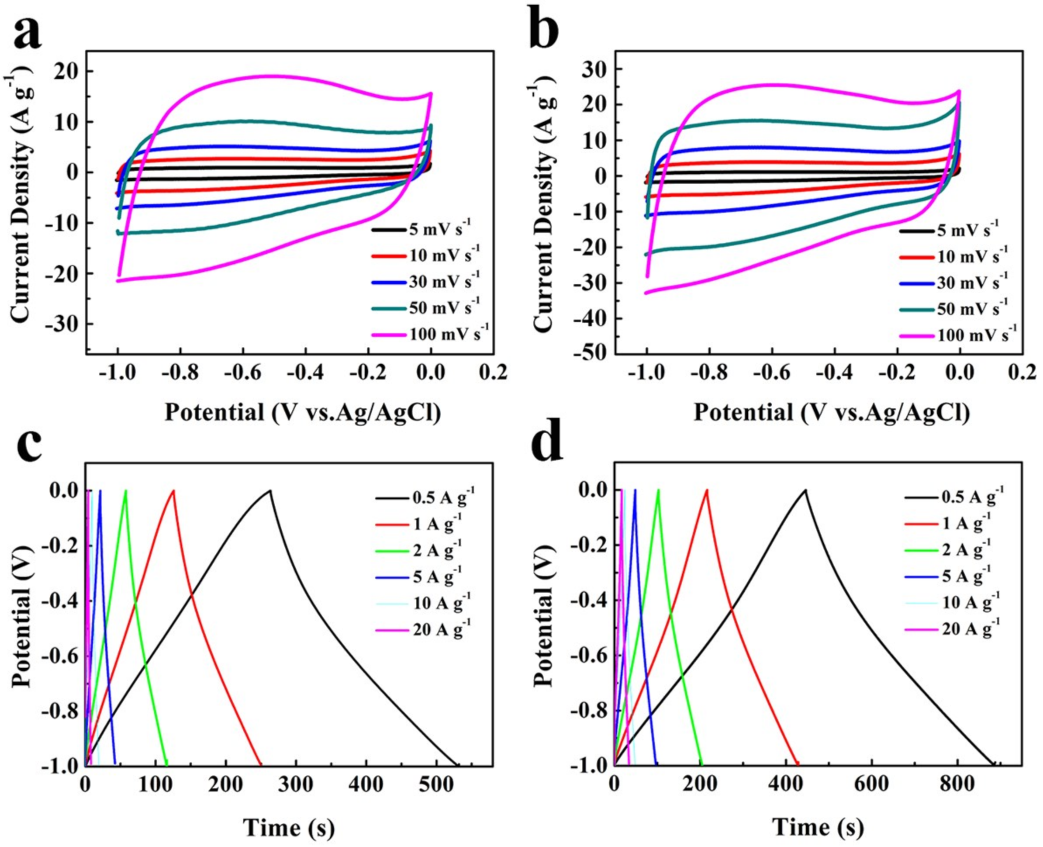

Figure S9. Electrochemical performance of the AC and B/N-PCTBs with a high mass loading of 40 mg cm^{-2}. CV curves at various scan rates of 5, 10, 30, 50 and 100 mV s^{-1} of the (a) AC and (b) B/N-PCTBs. Galvanostatic charge/discharge curves versus different current densities (0.5-20 A g^{-1}) of the (c) AC and (d) B/N-PCTBs.

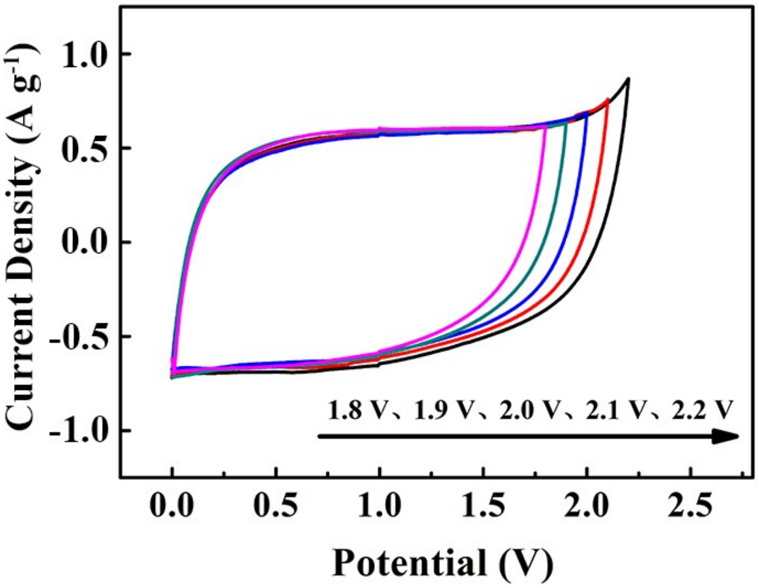

Figure S10. CV curves of the symmetric cell with a total active material mass loading of 80 mg cm^{-2} within different voltage ranges at 20 mV s^{-1}. The potential of 1.8 V was adopted as the operating voltage in this work.

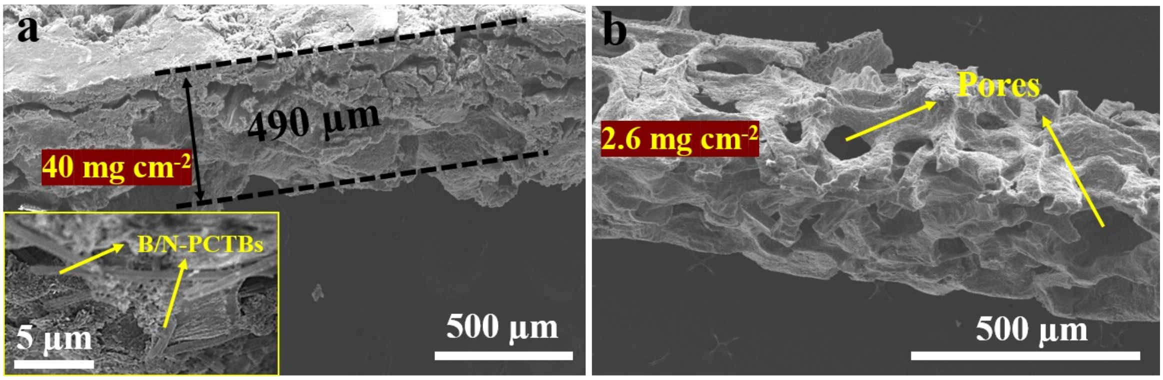

Figure S11. (a) SEM image showing the cross-section of the B/N-PCTBs electrode with a mass loading of 40 mg cm^{-2}. The enlarged SEM showing the B/N-PCTBs interconnect with each other (inset). (b) SEM image showing the cross-section of the B/N-PCTBs electrode with a mass loading of 2.6 mg cm^{-2}. There are many pores without filled with active materials.