

**Electronic Supplementary Information**

**Synergistically boosting electrochemical performance of  
polypyrrole coated-activated carbon derived from carbon dots for  
high-performance supercapacitor**

Lanxin Wang,<sup>‡ a,b</sup> Qingxin Zeng,<sup>‡ a,b</sup> Qihong Chen,<sup>a,b</sup> ChangMing Li,<sup>a,b\*</sup> Jiucun Chen<sup>a,b\*</sup>

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<sup>a</sup>Key Laboratory of Luminescence Analysis and Molecular Sensing (Southwest University), Ministry of Education, School of Materials and Energy, Southwest University, Chongqing 400715, China. E-mail: chenjc@swu.edu.cn

<sup>b</sup>Chongqing Key Laboratory for Advanced Materials & Technologies of Clean Energies, Chongqing 400715, China

<sup>‡</sup> These authors contributed equally to this work.

## **Materials and Methods**

### **Materials and reagents**

Citric acid (CA), urea, pyrrole (Py), potassium hydroxide (KOH), and anhydrous iron trichloride ( $\text{FeCl}_3$ ) were obtained from Aladdin Chemical Co., Ltd. Carbon black and polytetrafluoroethylene emulsion (PTFE) were purchased from Guangdong Klude New Energy Technology Co., Ltd. All chemicals can be used without further purification.

### **Synthesis of carbon dot-based activated carbons (AC-CDs)**

The citric acid (CA) and urea were mixed in a solid phase with a weight ratio of 3:2, and then the mixture was heated at  $180^\circ\text{C}$  for 10 min using a heating table until the mixture became a black solid, and carbon dots (CDs) were obtained. The carbon dots (CDs) and KOH were dispersed in deionized water at a mass ratio of 2:1, ultrasonicated and stirred, and then dried at  $80^\circ\text{C}$  for 24 h. The resulting product was heated in a tube furnace under argon at  $600^\circ\text{C}$  for 2 hours at a heating rate of  $2^\circ\text{C min}^{-1}$ . Then, the product was washed with dilute HCl solution and distilled water, and dried in an oven at  $80^\circ\text{C}$  for 24 hours to obtain carbon dot-based activated carbons (AC-CDs).

### **Synthesis of carbon dot-based activated carbon/polypyrrole (AC-CDs/PPy)**

Disperse the sample carbon-based activated carbons (AC-CDs) in  $\text{FeCl}_3$  ethanol solution ( $50\text{mg ml}^{-1}$ ), soak for 5 minutes, filter out, and dry at  $80^\circ\text{C}$  for 3 hours, then put it and pyrrole (Py) in a sealed container, keep for 2 hours under normal temperature and pressure, wash the residual  $\text{Fe}^{3+}$  with deionized water, and finally dry it in an oven at  $80^\circ\text{C}$  for 24 hours to obtain a carbon dot-based activated carbon/polypyrrole (AC-CDs/PPy) composite material. In fact, we synthesized a series of AC-CDs/PPy composite samples by fumigating at different times. According to different fumigation times (0h, 1h, 2h, 3h), the names of the hybrid materials after synthesis are AC-CDs/PPy-0h, AC-CDs/PPy-1h, AC-CDs/PPy-2h and AC-CDs/PPy-3h.

### **Characterization**

Use scanning electron microscope (FESEM, JEOL JSM-6510LV) and transmission electron microscope (TEM, JEOL JEM-2100) to observe the surface morphology and structure of the sample. Use X-ray diffractometer (Shimadzu XRD-7000) to record the XRD patterns of all samples. Raman spectroscopy was obtained with HORIBA LabRAM spectrometer. X-ray photoelectron spectroscopy (XPS) measurement was performed on a Thermo Fisher ESCALAB 250Xi spectrometer. Use the Thermo Scientific Nicolet 6700 spectrometer and Shimadzu UV-

2550 spectrophotometer to record the Fourier transform infrared (FT-IR) and ultraviolet-visible (UV-Vis) spectra of the sample. The photoluminescence (PL) spectra were characterized with spectrofluorometer (RF-5310C, Shimadzu). Zeta potential tester (Nano ZS90) was used to characterize Zeta potential. Nitrogen adsorption specific surface and pore size analyzer (NOVA-1200e) was used to obtain nitrogen adsorption isotherms.

### **Electrochemical measurement**

The slurry is obtained by mixing active material, carbon black and polytetrafluoroethylene emulsion (PTFE) in ethanol at a mass ratio of 8:1:1. Then the slurry was pressed onto the foamed nickel collector (1 cm×1 cm) and dried at 80°C for 12 h. The nickel foam was sonicated in acetone for 30 minutes, and then in deionized water for 30 minutes. The above operation was repeated three times, rinsed with deionized water and absolute ethanol, and dried in a vacuum oven at 80°C for 24 hours. The mass load of the electrode material is 1~3 mg cm<sup>-2</sup>. In a conventional three-electrode battery, 3M KOH was used as the electrolyte for electrochemical characterization. Platinum foil and Ag/AgCl were used as counter electrode and reference electrode, respectively. All electrochemical measurements are performed using CHI 760E electrochemical workstation (Shanghai Chenhua Instrument Co., Ltd., China). The voltage window for CV and GCD tests is set in the range of -1~0 V (relative to SCE). The electrochemical impedance spectroscopy (EIS) was measured using a 5 mV AC amplitude recorded in the frequency range of 10 mHz to 100 kHz. At the same time, using a two-electrode capacitor system, through the CV and GCD tests, the potential window is set in the range of 0 to 1 V to evaluate the electrochemical performance of the supercapacitor. The symmetrical supercapacitor uses 3M KOH aqueous solution as the electrolyte and glass fiber as the diaphragm to assemble a two-electrode button capacitor. The difference in the mass of the active material between the two electrodes does not exceed 0.3mg. The specific capacitance of the three electrodes is calculated from the charge/discharge curve by the following formula:

$$C = \frac{I \times \Delta t}{m \times \Delta V} \quad (1)$$

Where C (F g<sup>-1</sup>) is the specific capacitance based on the mass of the active material, I is the current density, ΔV is the potential change within the discharge time Δt, Δt is the discharge time measured in seconds, and m(g) is the activity The mass of the substance, ΔV is the potential window (V).

The specific capacitance of a symmetrical capacitor is calculated from the charge/discharge curve by the following formula:

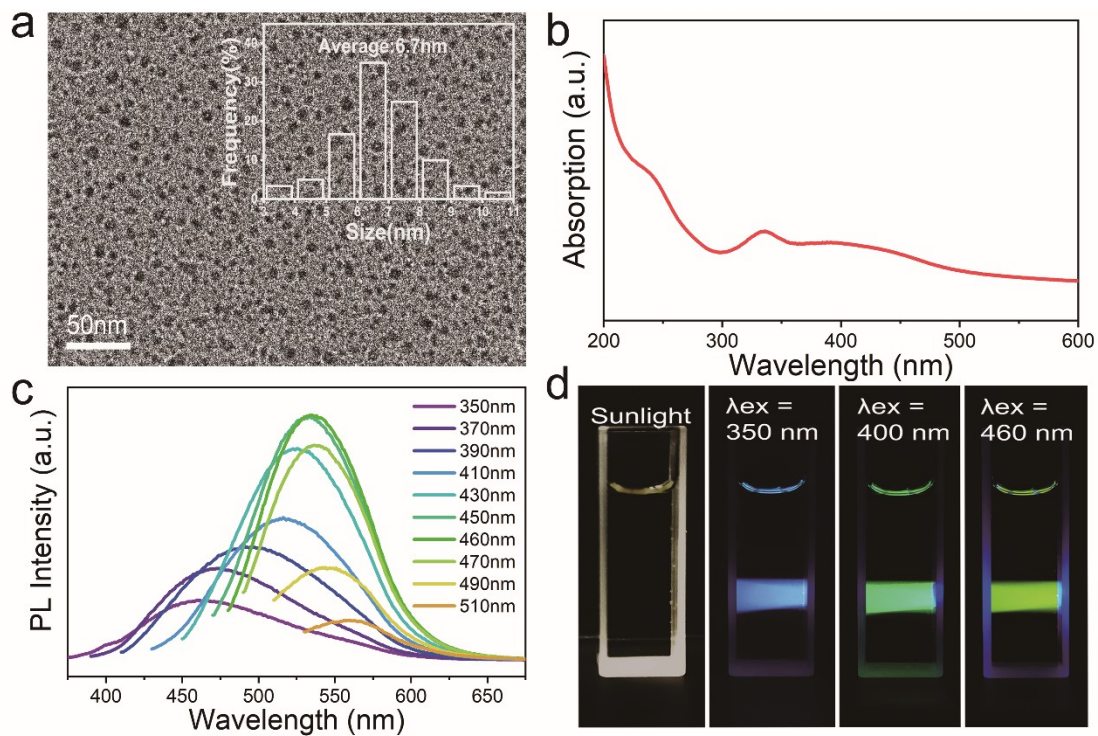
$$C_{cell} = \frac{I \times \Delta t}{m_{total} \times \Delta V} \quad (2)$$

Where  $C_{cell}$  ( $F g^{-1}$ ) is the specific capacitance based on the mass of the active material,  $I$  is the current density,  $\Delta V$  is the potential change within the discharge time  $\Delta t$ ,  $\Delta t$  is the discharge time measured in seconds, and  $m(g)$  is the total mass of active material on two electrodes,  $\Delta V$  is the potential window (V).

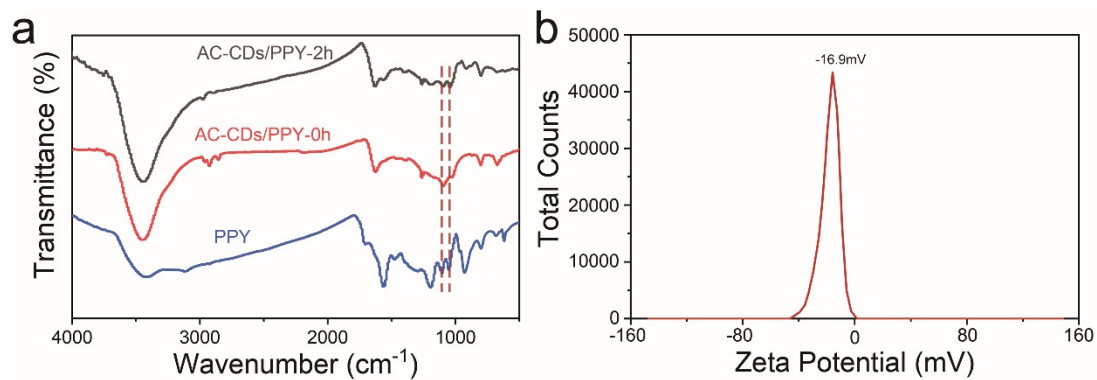
The energy density (E) and power density (P) of the capacitor can be calculated according to the following equations:

$$E = 0.5 \times C_{cell} \times \Delta V^2 \quad (3)$$

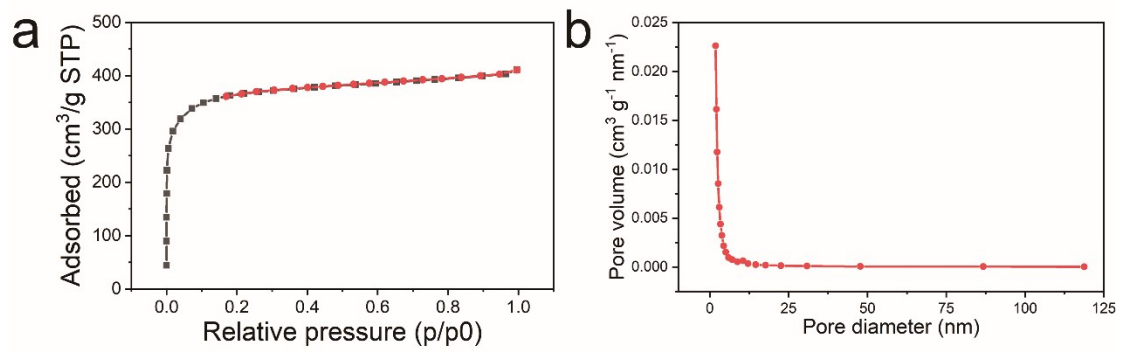
$$P = \frac{E}{\Delta t} \quad (4)$$



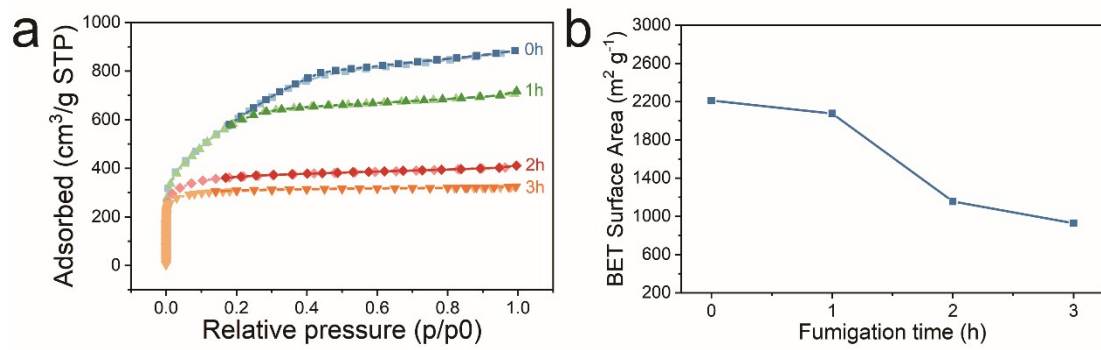
**Figure S1** (a) TEM images of CDs (inset: the lateral size distribution); (b) UV-vis absorption spectra of the CDs; (c) Emission spectra of the CDs; (d) Digital photo of an aqueous solution of CDs excited by sunlight and different emission wavelengths.



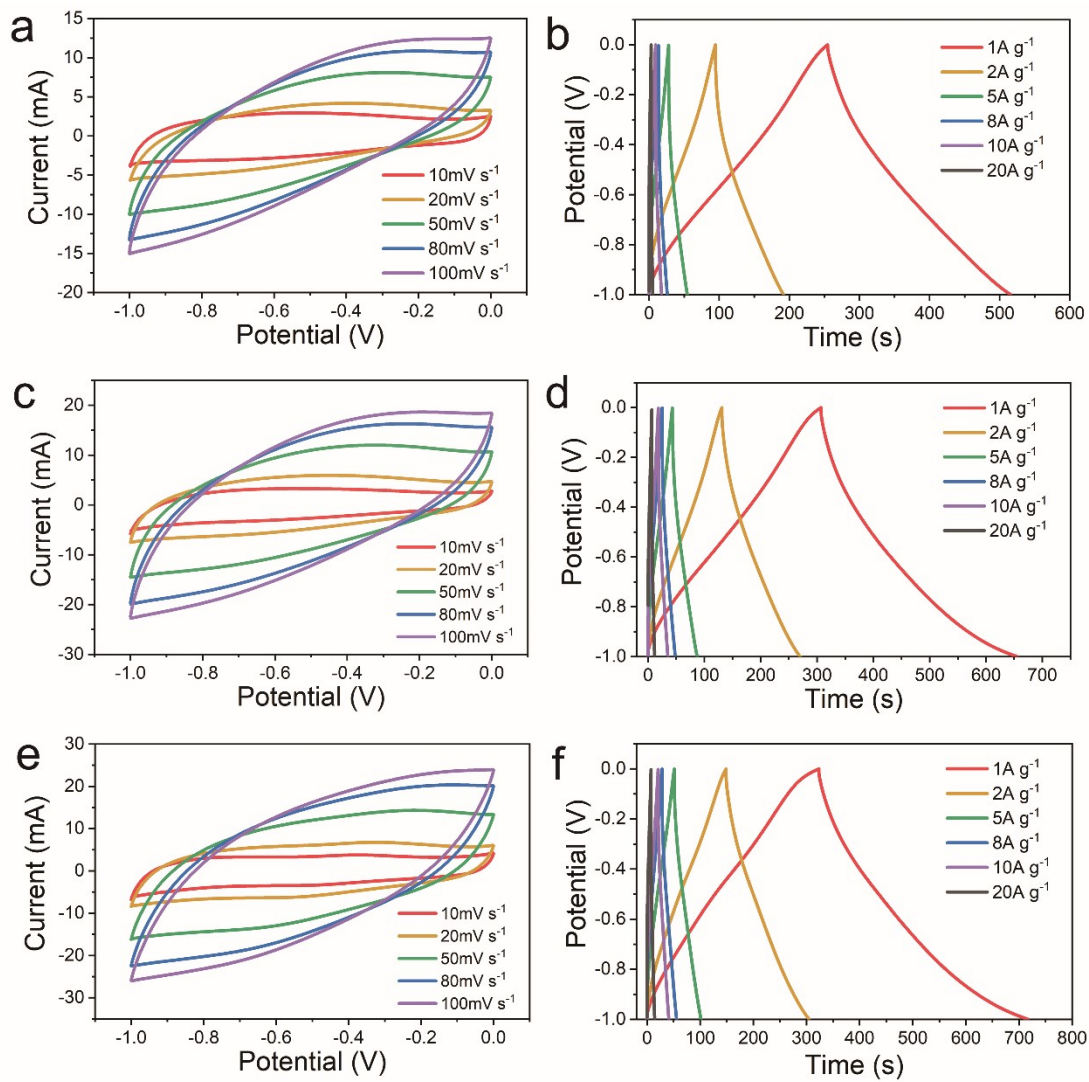
**Figure S2** (a) FT-IR spectra of PPY, AC-CDs/PPy-0h, AC-CDs/PPy-2h; (b) Zeta potential distribution of the CDs.



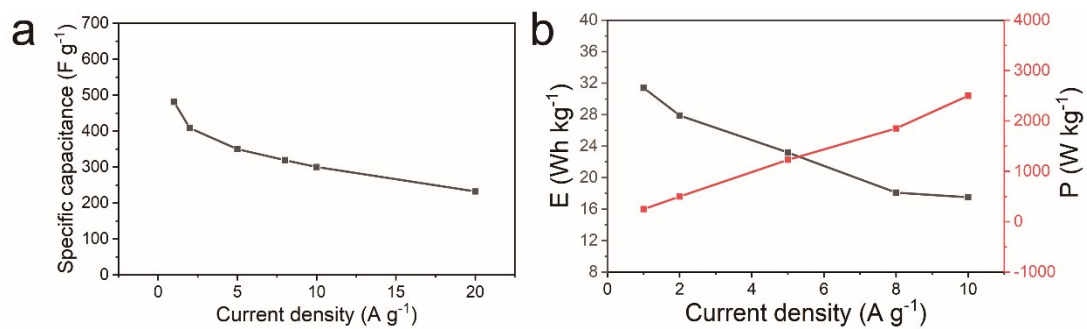
**Figure S3** (a)  $N_2$  adsorption/desorption isotherms of AC-CDs/PPy-2h; (b) The pore size distribution of AC-CDs/PPy-2h.



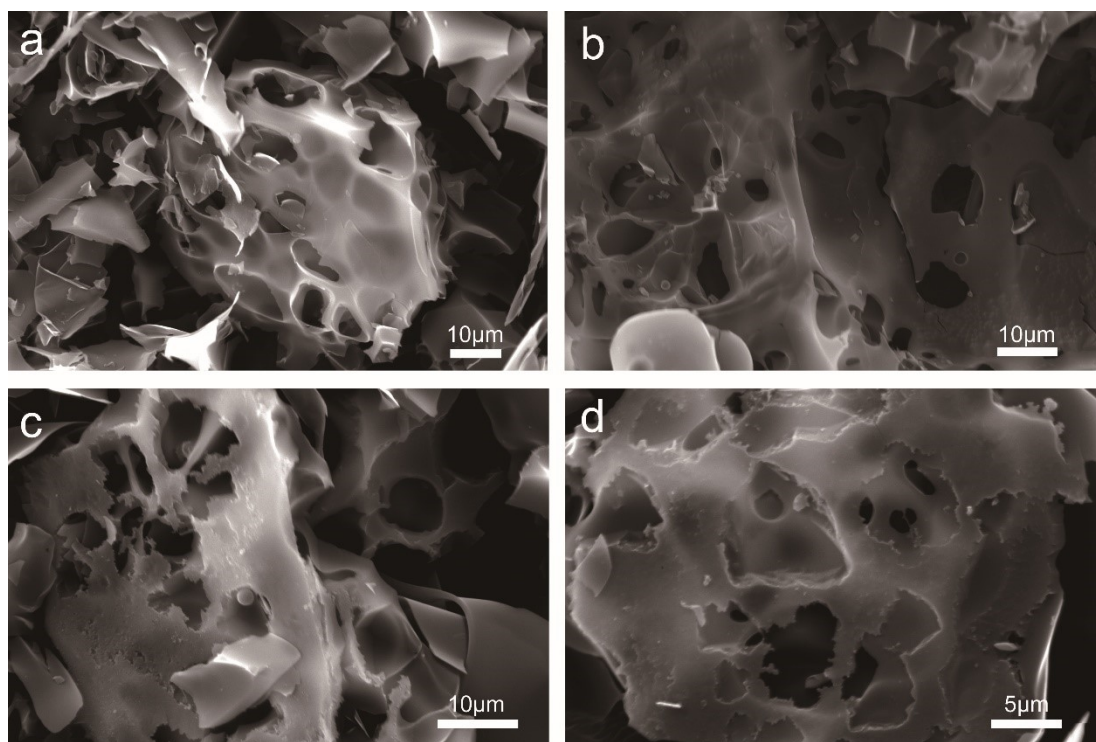
**Figure S4** (a)  $N_2$  adsorption/desorption isotherms of AC-CDs/PPy-0h, AC-CDs/PPy-1h, AC-CDs/PPy-2h and AC-CDs/PPy-3h; (b) The BET surface area of AC-CDs/PPy-0h, AC-CDs/PPy-1h, AC-CDs/PPy-2h and AC-CDs/PPy-3h.



**Figure S5** (a) CV curves of AC-CDs/PPy-0h detected at various scan rates; (b) GCD curves of AC-CDs/PPy-0h at different current densities; (c) CV curves of AC-CDs/PPy-1h detected at various scan rates; (d) GCD curves of AC-CDs/PPy-1h at different current densities; (e) CV curves of AC-CDs/PPy-3h detected at various scan rates; (f) GCD curves of AC-CDs/PPy-3h at different current densities in a three-electrode system.



**Figure S6** (a) Capacitances of AC-CDs/PPy-2h at different current densities; (b) The energy density (E) and power density (P) of the capacitor at different current densities.



**Figure S7** SEM images of AC-CD/PPy-2h after (a) 0, (b) 5000, (c) 7500 and (d) 10000 cycles.



**Table S1** Comparison of electrochemical performances of AC-CD/PPy to other carbon-based hybrids

<b>Electrode material</b>	<b>Current density (A g<sup>-1</sup>)</b>	<b>Specific capacitance (C, F g<sup>-1</sup>)</b>	<b>Electrolyte</b>	<b>Ref.</b>
CA/PPy	—	231	1 M H <sub>2</sub> SO <sub>4</sub>	S1
PPy/N-PCM	1	237.5	1 M H <sub>2</sub> SO <sub>4</sub>	S2
PPy@rGOH	1	340	1 M KNO <sub>3</sub>	S3
rGO/PPy	0.5	304	6 M KOH	S4
CB@CNF/PANI	1	298	1 M H <sub>3</sub> PO <sub>4</sub>	S5
CNTs@NC	0.5	310	1 M Na <sub>2</sub> SO <sub>4</sub>	S6
PD2ET-g-SWCNTs	1	399	1 M KCl	S7
AC-CDs/PPy	1	481	3 M KOH	This work

## References

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