Supporting information

Candle Soot as Supercapacitor Electrode Material

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Characterization:

High-Resolution transmission electron microscopy (HRTEM) measurements were conducted on a JEM-2010 microscope operated at 200 kV, to reveal the structures of the MS, BS and MnO₂@CS samples. Powder X-ray diffraction (SA-XRD) patterns were recorded by a X'Pert PRO powder X-ray diffractometer using Cu Kα radiation. Nitrogen adsorption–desorption isotherm measurements were performedon on a Micromeritics ASAP 2020 volumetric adsorption analyzer at 77 K. The Brunauer–Emmett–Teller (BET) method was utilized to calculate the specific surface area of each sample and the pore size distribution was derived from the adsorption branch of the corresponding isotherm using the Barrett–Joyner–Halenda (BJH) method. Thermal stability was determined with a thermogravimetric analyzer (TGA) (Netzsch, STA 449 C) over a temperature range of 25–800 °C at a heating rate of 10 °C/min under air atmosphere. FT-IR was recorded on a TENSOR 27 instrument (BRUCKER).

Electrochemical measurements in two-electrode system

Two symmetric capacitors (BS/BS supercapacitor and MnO₂@CS/MnO₂@CS supercapacitor) were fabricated for comparison. The electrodes were prepared using nickel foam as the current collector; electrode total contained $5 \sim 10$ mg of electrochemical active material and had a geometric surface area of about 4 cm². The electrochemical measurements of the symmetric supercapacitors were carried out in 6 M KOH (BS) and 1 M Li₂SO₄(MnO₂@CS) aqueous electrolytes using the electrochemical working station in a two-electrode cell at room temperature.

The specific capacitance of the supercapacitor cell can be evaluated from the charge/discharge test together with the following equation:

$$Cs = 4 \frac{I\Delta t}{Um} \tag{1}$$

Where *I* is the current, Δt the discharge time, *U* the potential rang, and *m* the total mass of the active materials on work electrodes.

The charge efficiency of supercapacitor cell can be evaluated from the charge/discharge test together with the following equation:

$$C_{\text{eff}} = \frac{\Delta t_{\text{discharge}}}{\Delta t_{\text{charge}}} \qquad (2)$$

Where E is the charge efficiency, $\Delta t_{discharge}$ the discharge time, Δt_{charge} the discharge time.

More characterizations for MS, BS and MnO₂@CS



Figure S1: Raman spectroscopy is widely used for analysis of composite and nano structures of carbonaceous materials. Characteristic Raman peaks for carbon materials are the disorder-induced D band at ~ 1350 cm⁻¹ and the graphitic G band at ~ 1580 cm⁻¹. The ratios between the intensities of the G and D bands were 1.01 for candle soot collected by a Pt sheet. The high content of G bands proves there is numbers sp² carbon atoms exist in candle soot.



Figure S2: TG curves of MS at a heating rate of 10 $^{\circ}$ C min⁻¹ in air flow. The lower weight loss prove BS obtained under 450 $^{\circ}$ C treatment may just lost transparent shell than MS.



Figure S3: XRD patterns of BS. The diffraction peaks of the as-synthesized BS are similar to carbon nanotubes (CNTs), while the most intensive peak of CNTs at around $2\theta=25.7^{\circ}$ and 42.5° corresponds to (002) and (100) reflection, respectively.



Figure S4. FTIR spectra of MS and BS. The FTIR spectra were employed to further confirm the transparent shell lost in BS. We can see the 1713cm⁻¹ peak splits into 1747cm⁻¹ and 1713cm⁻¹ after heat treatment of MS. It's likely -CO- combusts to -COOH dimer. Meanwhile, 1631cm⁻¹ peak shifts to 1596cm⁻¹. possiblely owing to -CO-C₆H₄-NH₄ translating to -CONH₂.



Figure S5: XRD Patterns of BS and MnO₂@CS. As displayed in Figure S5, the diffraction peaks of the as-synthesized MnO₂@CS are similar to α -MnO₂, marching JCPDS 44-0141 (a=9.7845 Å, c=2.8630 Å), while the most intensive peak of α -MnO₂ at around 20=37.5° corresponds to (211) reflection. Meanwhile, the (002) reflection peak of layered TS has almost disappeared. Our results correlate well with previous studies that the diffraction peaks become weakened or even disappear when the regular stacks of carbon nanoparticle are covered, and α -MnO₂ maintain their amorphous characteristics, which is said to have better capacitance performance.



Figure S6: Chronopotentiometry (CP) measurements on the capacitance performance of TS with a two-electrode system. Typical CP curves of $MnO_2@CS$ at a current of 10 mA, 20 mA, 50 mA, 100 mA. The typical CP curves and the calculated specific capacitances versus discharge current densities are presented in Figure S6. The operation voltages are optimized by adjusting the potential range to keep the triangular form of the CP curves, which is 1V for all the samples. Significantly, the specific capacitance is highly decreased from 309 to $106Fg^{-1}$, for the pseudocapacitors performance increased by α -MnO₂.