Electronic Supplementary Information

A Seeded Synthetic Strategy for Uniform Polymer and Carbon Nanospheres with

Tunable Sizes for High Performance Electrochemical Energy Storage

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1. Methods

All chemicals were analytical reagents purchased from Sinopharm Chemical Co., Ltd. and were used without further purification. A mixture of 0.063 g (0.5 mmol) phloroglncinol, 0.05 g (0.375 mmol) terephthalaldehyde and 28 mL distilled water was stirred at 70°C (15-480 min) to obtain colloidal seeds. Into the sol, 0.165 g (1.5 mmol) resorcinol was added and this was stirred for 30 min. Then 0.1 mL ammonia solution (25 wt%) and 0.225 mL formalin (37 wt%) containing 0.090 g formaldehyde (3.0 mmol) was added to the reaction system and stirred for 24 h at 25°C, followed by heating at 100°C for 24 h in a Teflon-lined autoclave to obtain polymer nanospheres. After centrifugation and drying at 100°C, the solid product of polymer nanospheres was recovered. These TJU-x samples were directly carbonized at 850°C (5 °C/min) for 4 h under N_2 flow to produce carbon nanospheres (denoted as C-TJU-x).

A sample of carbon nanospheres (C-TJU-60) was activated by KOH (C-TJU-60/KOH=1:3 w/w) at 800°C for 2 h in a N_2 flow to produce porous carbon nanospheres (denoted as PC-TJU-60) for electrochemical energy storage.

2. Characterization Instruments

TEM images were obtained using a JEOL 2100 microscope operating at 200 kV. Samples for TEM observation were suspended in ethanol and supported on a carbon-coated copper grid. SEM was obtained using a Hitachi S-4800. N_2 adsorption and desorption analysis was measured at -196 °C by using Micromeritics Tristar 3000 gas adsorption analyzer. Dynamic light scattering (DLS) tests were carried out at 25 °C on a Malvern Zetasizer ZEN 3690 instrument.

3. Capacitor Construction

All electrochemical measurements were carried out in a two-electrode cell (capacitor) with 6.0 M KOH aqueous solution as electrolyte. For the preparation of one electrode, 90wt% active materials was mixed with 10 wt% polytetrafluoroethylene binder (PTFE 60 wt% dispersion in ethanol) to form slurry. The slurry was spread onto a nickel-foam (1 cm^2), followed by pressure under 25 MPa and drying at 100 °C to prepare electrode. After that, two electrodes with same weights of materials were put together, and a polypropylene (PP) membrane was sandwiched between them as a separator. Two identical electrodes were adopted as cathode and anode for the cell configuration.

4. Electrochemical Measurements

All the electrochemical experiments were performed at ambient temperature. Before the measurements, the capacitor cell was pumped for 30 min so that the active material was soaked fully by the electrolyte. Cyclic voltammograms (CV) and galvanostatic charge–discharge (GCD) measurements were both carried out on a CHI 660D workstation. The specific capacitance (C, F g^{-1}) is calculated according to the following equation (1):

$$C = \frac{\Delta Q}{2 \times \Delta V \times m \times r} \qquad (1)$$

Where ΔQ is the charge integrated from the whole voltage range, ΔV is the whole range of voltage window, and m is the total mass of carbon on two electrodes, r is the scan rate of CV measurement.

GCD measurements at various current densities were performed with a voltage range of 0 to 1 V. Specific capacitance of each electrode was calculated according to the following equation (2):

$$C = \frac{2 \times I \times \Delta t}{\Delta V \times m} \qquad (2)$$

Where I is the discharge current, Δt is the discharge time from 0-1 V, ΔV is the voltage difference within the discharge time Δt, and m is the total mass of carbon on two electrodes. The factor of two in these equations comes from the fact that the total capacitance measured from the test cells is the sum of two equivalent single electrode capacitors in series. [1]
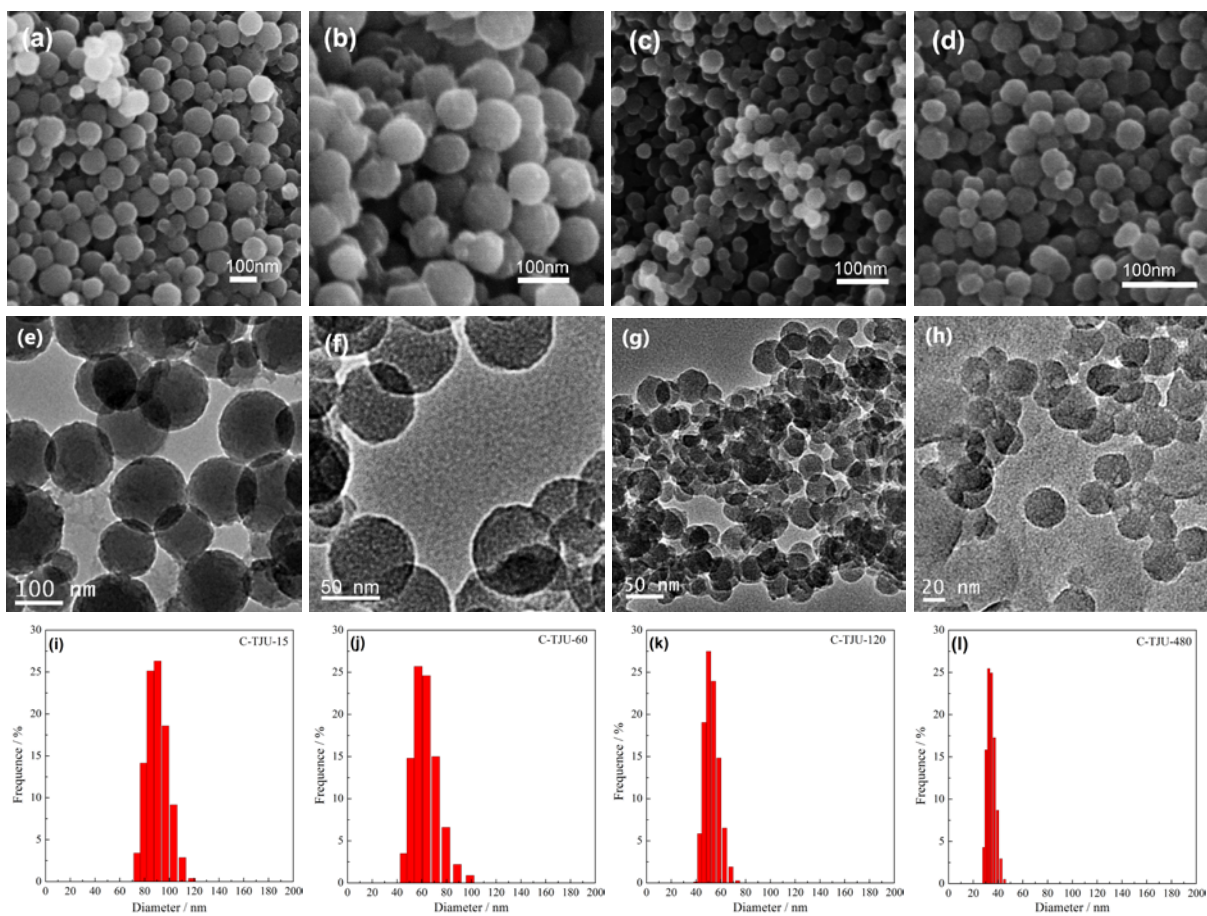
Reference

[1] B. Liu, H. Shioyama, T. Akita, Q. Xu, *J. Am. Chem. Soc.* **2008**, *130*, 5390.
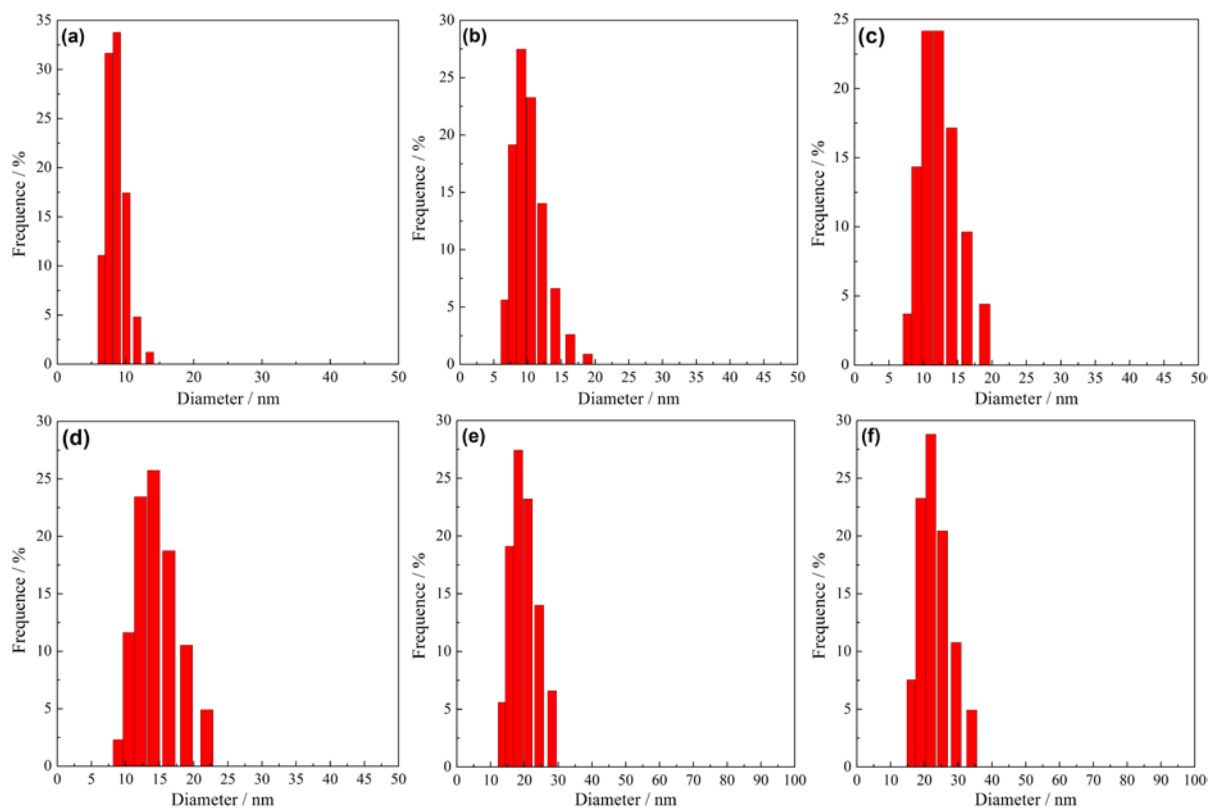
Fig. S1 DLS datum for colloidal seeds obtained by controlled polymerization of phloroglucinol and terephthalaldehyde for 15 min (a), 30 min (b), 60 min (c), 120 min (d), 240 min (e) and 480 min (f).
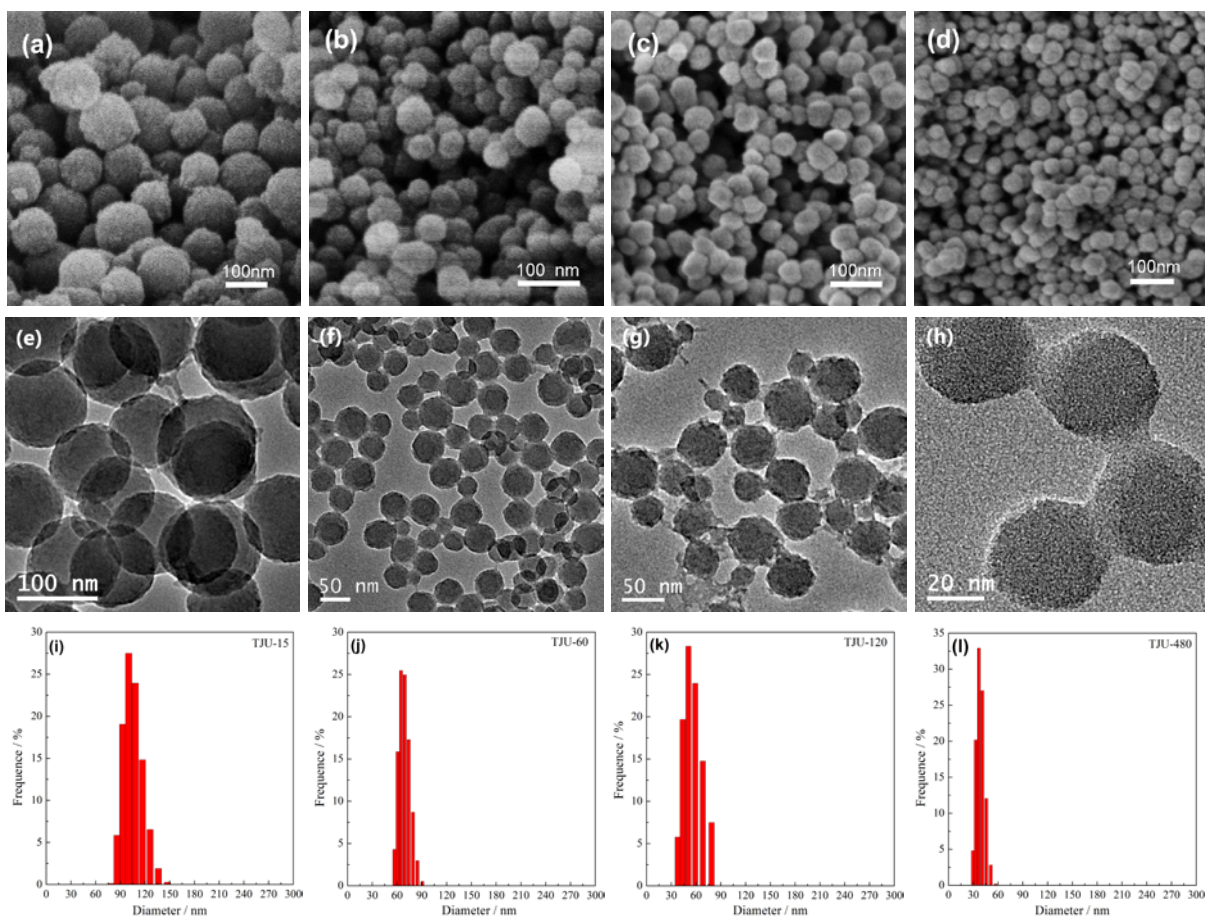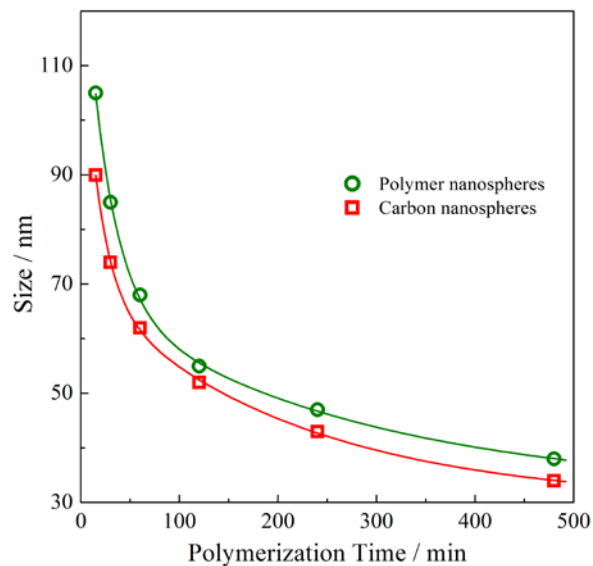
Fig. S2 SEM images, TEM images and DLS datum for TJU-15 (a, e, i), TJU-60 (b, f, j), TJU-120 (c, g, k) and TJU-480 (d, h, l).

Fig. S3 SEM images, TEM images and DLS datum for C-TJU-15 (a, e, i), C-TJU-60 (b, f, j), C-TJU-120 (c, g, k) and C-TJU-480 (d, h, l).
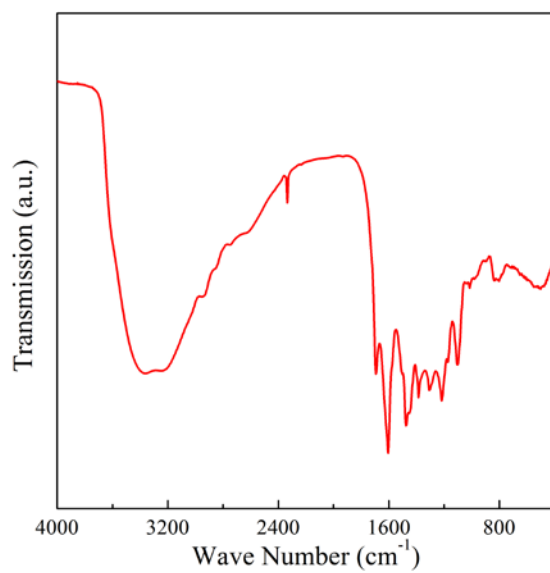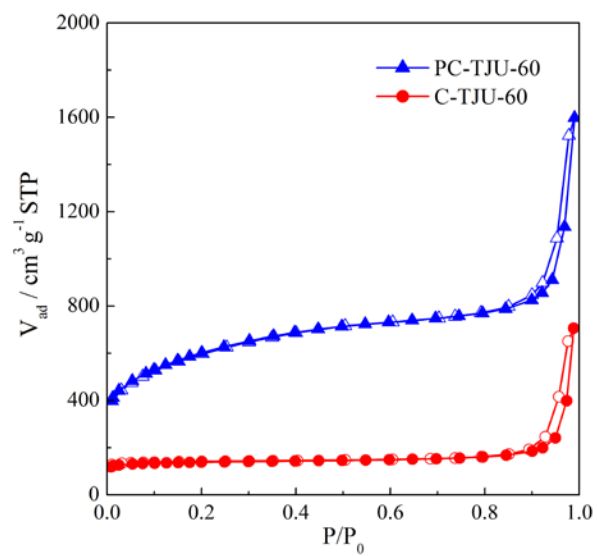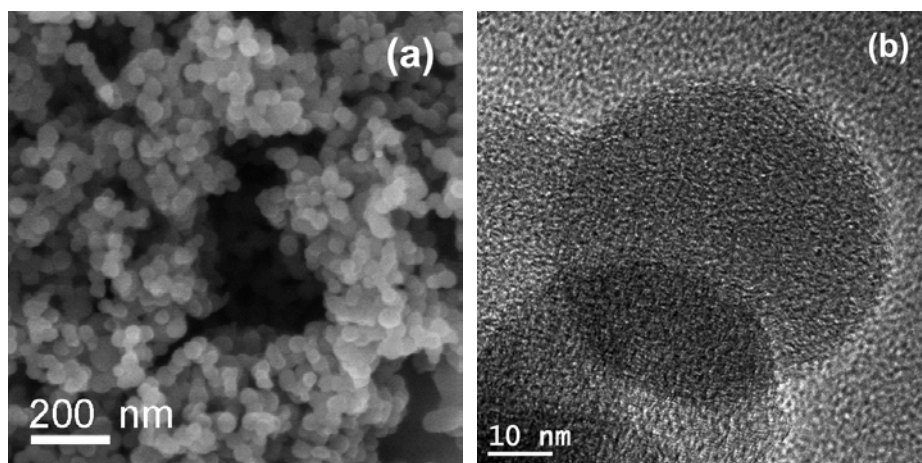
Fig. S4 The FT-IR spectrum of TJU-60.

Fig. S5 Relationship between the polymerization time of phloroglucinol and terephthalaldehyde and the particle sizes of polymer and carbon nanospheres (using 10-20 nm seeds).
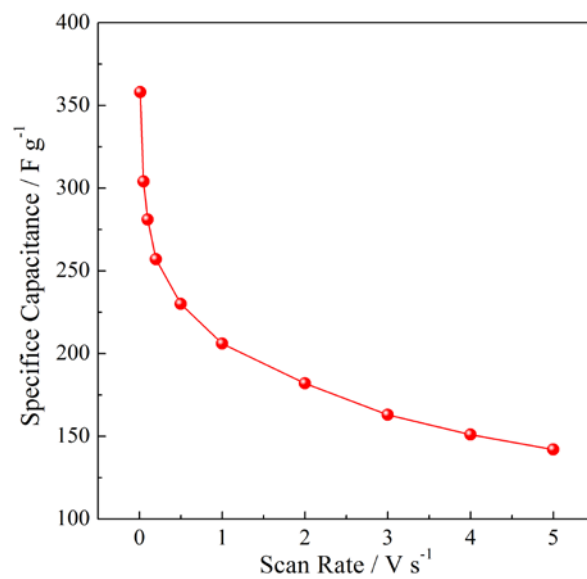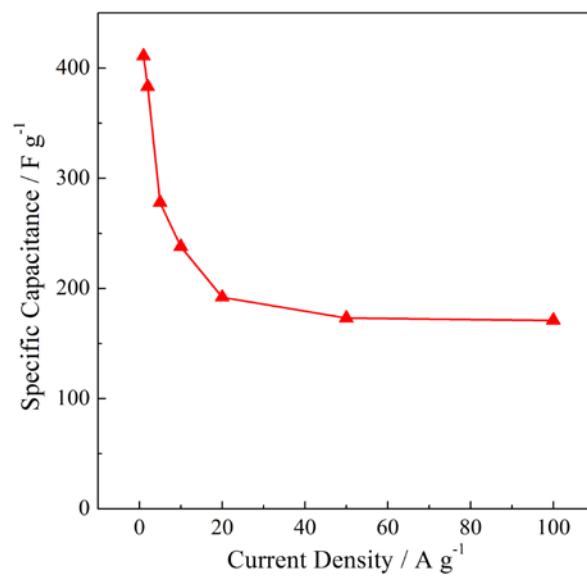
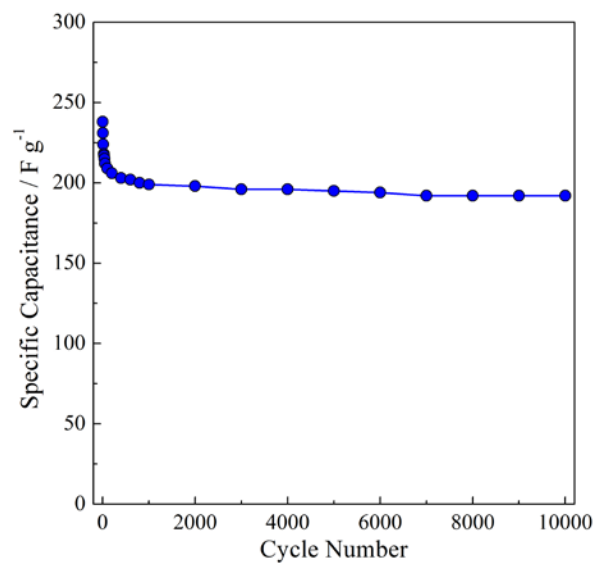**Fig. S6** N_2 adsorption-desorption isotherms of C-TJU-60 and PC-TJU-60.

Fig. S7 SEM (a) and TEM (b) images of PC-TJU-60.

Fig. S8 The specific capacitances of PC-TJU-60 under different scan rates.

Fig. S9 The specific capacitances of PC-TJU-60 under different current densities.

Fig. S10 Long-term stability estimation of PC-TJU-60 at 10 A g^{-1}.