Supplementary Information

High-Frequency Fabrication of Discrete and Dispersible Hollow Carbon Spheres with Hierarchical Porous Shell by Using Secondary-crosslinking Pyrolysis

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Experimental

The monodisperse core-shell structure poly (styrene-co-divinylbenzene) sphere (CSPS) was synthesized according to previous work.¹ 0.3 g of dried CSPS-30 was put into CCl₄ (30 mL) in a 100 mL three-necked flask in a thermostatted water bath of 40 °C. After the mixture was vigorously stirred at a speed of 160 rpm for 10 h, the product was collected and purified by centrifugation and redispersion in acetone by ultrasonic, and then dried at 45 °C. The dried product (HPC-30) was prepared for Nitrogen adsorption-desorption measurement. In addition, the upper transparent solution was also collected. And the product obtained by drying the upper transparent solution was prepared for GPC measurements.

Characterization

The average molecular weight and its polydispersity (PD) of the solution (Table S1) were determined by a PL-GPC50 Integrated GPC (gel permeation chromatographic) System equipped with PL gel 10 μ m MIXED-B, 300×7.5 mm

*Corresponding author Email: chenjf@zzu.edu.cn; qunxu@zzu.edu.cn Fax: +86 0371 67767827 Tel: +86 0371 67767827 columns from Polymer Laboratories Ltd, UK. Signals generated from a differential refractive index detector. Polymer samples were dissolved in tetrahydrofuran (THF) at a concentration of 5-10 mg/mL. THF was used as the mobile phase at a flow rate of 1 mL/min and column temperature was maintained at 40 °C. Calibration was made using PS standards. Nitrogen adsorption/desorption measurements were carried out in a Quantachrome NovaWin2 Instruments at 77 K. Before the sorption tests, the samples were degassed at 100°C under vacuum for 1 h. The specific surface area (SSA) and the pore size distribution (PSD) of HPC-30 were, respectively, calculated by the Brunauer-Emmett-Teller (BET) method and the Barrett-Joyner-Halenda (BJH) method

The specific capacitance (C) was calculated according to the following equation:

$$C = \frac{1}{m\nu\Delta V} \int \mathbf{i}(V) \mathrm{d}v$$

where *m* is the mass of active materials, *v* is the potential scan rate, and ΔV is the potential window in CV, and $\int i(V)dv$ is the integral area of the whole CV loop.²

Results and Discussion



Fig. S1 (a) Nitrogen adsorption-desorption isotherm of HPC-30 (b) PSD for HPC-30.



Fig. S2 SEM photo of the HCSs prepared by carbonizing the HHPS-T-30 without the

secondary hypercrosslinking reaction.



Fig. S3 t-plots of the samples.



Fig. S4 Calculated Rouquerol plot of the samples (Inset: enlarged plot turning part) along with pressure ranges used for the BET surface area calculations.



Fig. S5 BET plots of the samples from nitrogen isotherms at 77 K. The selected points are located in the relative pressure ranges (between 0.01 and 0.12) determined according to the Rouquerol plots (see Fig. S3). Correlation coefficient R was 0.99942 (HCS-20), 0.99965 (HHPS-T-30), 0.99968 (HHPS-T-D-30), 0.99913 (HCS-30), 0.99952 (HCS-40).



Fig. S6 (a) Cyclic voltammetry (CV) curves of HCS-20 at the scan rates of 5, 25, 50, 100, 200 and 400 mV s⁻¹. (b) Chronopotentiometry (CP) curves of HCS-20 at current densities of 0.5, 1.0 and 2.0 A g⁻¹. (c) Nyquist plot of HCS-20 electrode material.



Fig. S7 (a) Cyclic voltammetry (CV) curves of HCS-40 at the scan rates of 5, 25, 50, 100, 200 and 400 mV s⁻¹. (b) Chronopotentiometry (CP) curves of HCS-40 at current densities of 0.5, 1.0 and 2.0 A g⁻¹. (c) Nyquist plot of HCS-40 electrode material.



Fig. S8 (a) Cyclic voltammetry (CV) curves of SCS-30 at the scan rates of 5, 25, 50, 100, 200 and 400 mV s⁻¹. (b) Nyquist plot of SCS-30 electrode material.

Table S1. Average molecular weight and its polydispersity of the solution

$M_{ m p}$	M _n	$M_{ m w}$	Mz	$M_{ m w}/M_{ m n}$
16055	9763	18390	30979	1.8836

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