Facile Synthesis of Novel Bowl-like Hollow Carbon Spheres by Combination of

Hydrothermal Carbonization and Soft Templating

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Experiments

Synthesis of Hollow Bowl-like Carbon spheres: In a typical experiment, 0.4 g ascorbic acid (VC) was dissolved in deionized water (20 ml). Then 0.15 g ferrocene and 0.4 g trioctylamine (TOA) were added to the VC solution. The mixture of reactants was sonic-treated for 15 min to emulsify until the appearance of a light yellow opaque color. Then the emulsion was quickly transferred to 50 ml Teflon-lined autoclave and sealed for 24 h at 130 °C. After the reaction, the autoclave was cooled to room temperature naturally. The resulting precipitates were harvested by centrifugation with water and ethanol for several times and finally vacuum-dried at 60 °C for 6 h.

Electrochemical measurements: Electrochemical measurements were conducted in an electrochemical working station (CHI660E, Shanghai, China) with a threeelectrode system in 6 M KOH electrolyte at room temperature. The electrodes were prepared by pressing the mixture 80 wt.% of active material (BHCSs) and 10 wt.% of acetylene black and 10 wt.% of PTFE packaged with two-piece nickel foam and then pressed under 10 MPa for 10 s. A 7-9 mg amount was loaded on an area of 1 cm². Hg/HgO electrode was employed as the reference electrode and Pt as the counter electrode.

The identity and the phase of the as-synthesized HBC were verified by the X-ray powder diffraction (XRD) using a Rigaku D/max-RB12 X-ray diffraction meter with Cu-Ka radiation. The morphology of the products was examined by scanning electron microscopy (SEM, JEOL, JSM-7001F). The transmission electron microscopy (TEM) images were obtained on a microscope (JEOL, JEM-2010). The Raman spectra were obtained on a microscopic confocal Raman spectrometer (Renishaw RM2000) using Ar+ laser excitation with a wavelength of 514.5 nm. The Fourier transform infrared spectra (FT-IR) were recorded using a Nicolet 740 FT-IR spectrometer. X-ray photoelectron spectra (XPS) were recorded with an ESCALAB 250 spectrometer (PerkinElmer) to characterize the surface composition. Dynamic light scattering (DLS) measurements of polyplexes were performed with a Zetasizer Nano ZS (Malvern Instruments, Southborough, MA) equipped with a laser of wavelength 633 nm at a 173° scattering angle. The morphology of the emulsion droplets was examined by a confocal laser scanning microscope (SP8, Leica, Germany).



Figure S1XRD pattern of BHCSs. The broad peak around $2\theta=23^{\circ}$ indicates amorphous carbon for BHCSs.



Figure S3 SEM image of the product prepared without TOA



Figure S4 SEM image of the product prepared under the concentration of TOA was lower



Figure S5 SEM image of the product prepared under the concentration of TOA was higher



Figure S6 SEM image of small solid spheres separated by low centrifugal speed



Figure S7 SEM image of the product prepared under ultra high temperature of HTC



Figure S8 CV curves at sweep rates 0.1 V/s - 0.2 V/s of carbonized solid spheres



Figure S9 CV curves at sweep rates V/s - 2 V/s of carbonized solid spheres



Figure S10 Electrochemical impedance spectra of carbonized solid spheres

Table S1 Properties of reported carbon materials as supercapacitor electrode materials

Types of materials	Electrolyte	Specific capacitance (F/g)	References
Graphite oxide	H ₂ SO ₄ aqueous	60	1
Carboxylic		155	
functionalized SWNT	H_2SO_4 aqueous	155	2
Carboxylic		77	
functionalized MWNT			
Ester functionalized		41	
SWNT			
Ester functionalized		43	
MWNT			
Pristine MWCNT		23	
Pristine SWCNT		38	
Commercial activated	KOH aqueous	76	3
carbon			
Hollow carbon	Na ₂ SO ₄ aqueous	107.8	4
BHCSs	KOH aqueous	143.8	our work

Supplementary References

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