Supporting Information

Tunable N-doped hollow carbon spheres induced by ionic liquid for energy storage application

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Experimental Section

Synthesis of N-HCS samples

Resorcinol (0.215 g) was added quickly into the solution of 1-alkyl-3-methylimidazolium bromide ([C_{18}Mim]Br, 0.7 g) in H_2O (200 mL) and EtOH (100 mL) under vigorous stirring. Ethyl orthosilicate (TEOS, 2.15 mL) was added to the reaction mixture and subsequently stirred for 5 min and formaldehyde (37 wt%, 0.3 mL) was added. The mixed solution was subsequently

stirred at ambient temperature overnight. The as-synthesized suspension was centrifuged and dried at 70 ^{o}C overnight. For carbonization, the obtained product was heated at 800 ^{o}C for 3 h with a heating rate of 3 ^{o}C min^{-1} under N_2 atmosphere. Finally, the N-HCS were obtained, after removing the silica with 10 wt% HF solution for 24 h. The EtOH/H_2O ratios of 6/24, 10/20, 14/16, 16/14 and 20/10 are used to prepare N-HCS-6, N-HCS-10, N-HCS-14, N-HCS-16 and N-HCS-20 respectively.

Characterizations

The morphology and microstructure of samples were investigated by transmission electron microscopy (TEM, JEOL JEM-2100). Nitrogen adsorption-desorption isotherms were carried out on a Micromeritics TriStar 3020 instrument at -196 °C. The Brunauer-Emmett-Teller (BET) method was employed to calculate the specific surface area , while the Barrett-Joyner-Halenda (BJH) method was applied to analyze the pore size distribution using the desorption branch of isotherm. The total pore volume was obtained from the amount of N_2 adsorbed at the relative pressure (P/P_0 = 0.97). X-ray photoelectron spectroscopy (XPS) was conducted on a Thermo Scientific ESCALab 250Xi system using an Al-$K\alpha$ radiation under a vacuum of 3×10^{-10} mbar. The composition of C, H and N content was evaluated using elemental analysis (CHN) on the Flash EA 1112.

Electrochemical Measurements

The working electrode was prepared by coating the viscous slurry (samples, carbon black and polytetrafluoroethylene with the mass ratio of 8:1:1 in ethanol) onto Ni foam current collector. The mass of active material loaded on each working electrode was 4~5 mg after drying at 100 °C for 24 h. Electrochemical measurements were carried out in both three-electrode and two-electrode system using an electrochemical workstation (CHI 760E, Chenhua Instruments, China) with 6 M KOH solution as the electrolyte. For three-electrode system, a Pt wire and Hg/HgO was used as the counter and reference electrodes. For the fabrication of supercapacitor devices, two slices of electrode were immersed in 6 M KOH and were separated by a filtration

paper, then tested by the current collector. Electrochemical performances were evaluated by cyclic voltammetry (CV), galvanostatic charge-discharge (GCD) and electrical impedance spectroscopy (EIS) analysis. For the two-electrode system, the specific capacitances (C, F g^{-1}), energy density (E, Wh kg^{-1}) and power density (P, W kg^{-1}) were calculated by the following equations: $C=4\ I\Delta t/\Delta Vm$, $E=0.5\ C(\Delta V)^2/4/3.6$ and $P=3600\ E/\Delta t$, where I (A), Δt (s), ΔV (V) and m (g) are GCD current, discharge time, voltage window, and mass of active material, respectively. In three-electrode system, the specific gravimetric capacitance according to the GCD measurements: $C=I\Delta t/\Delta Vm$.

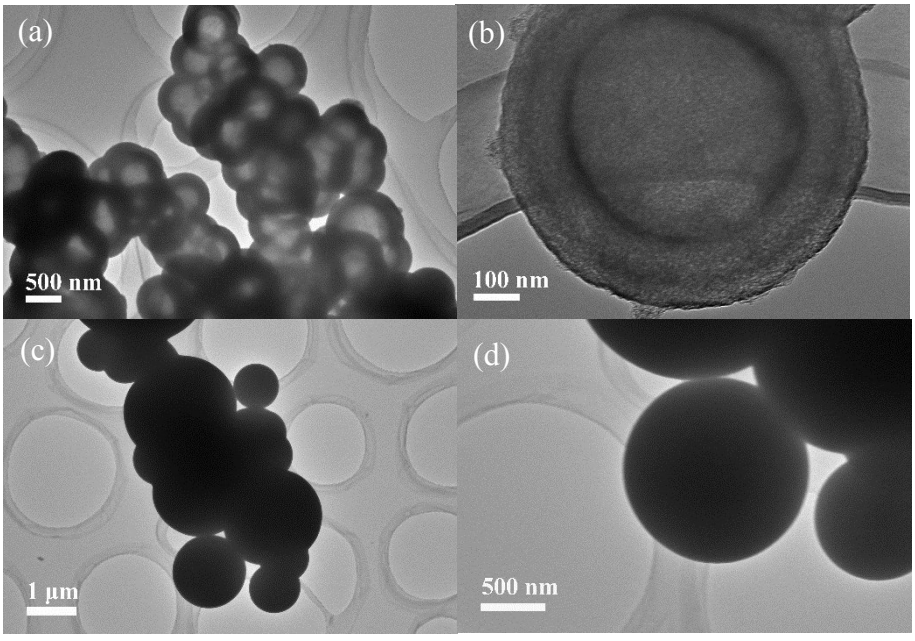

Fig. S1. TEM images of N-HCS-16 (a, b) and N-HCS-20 (c, d).

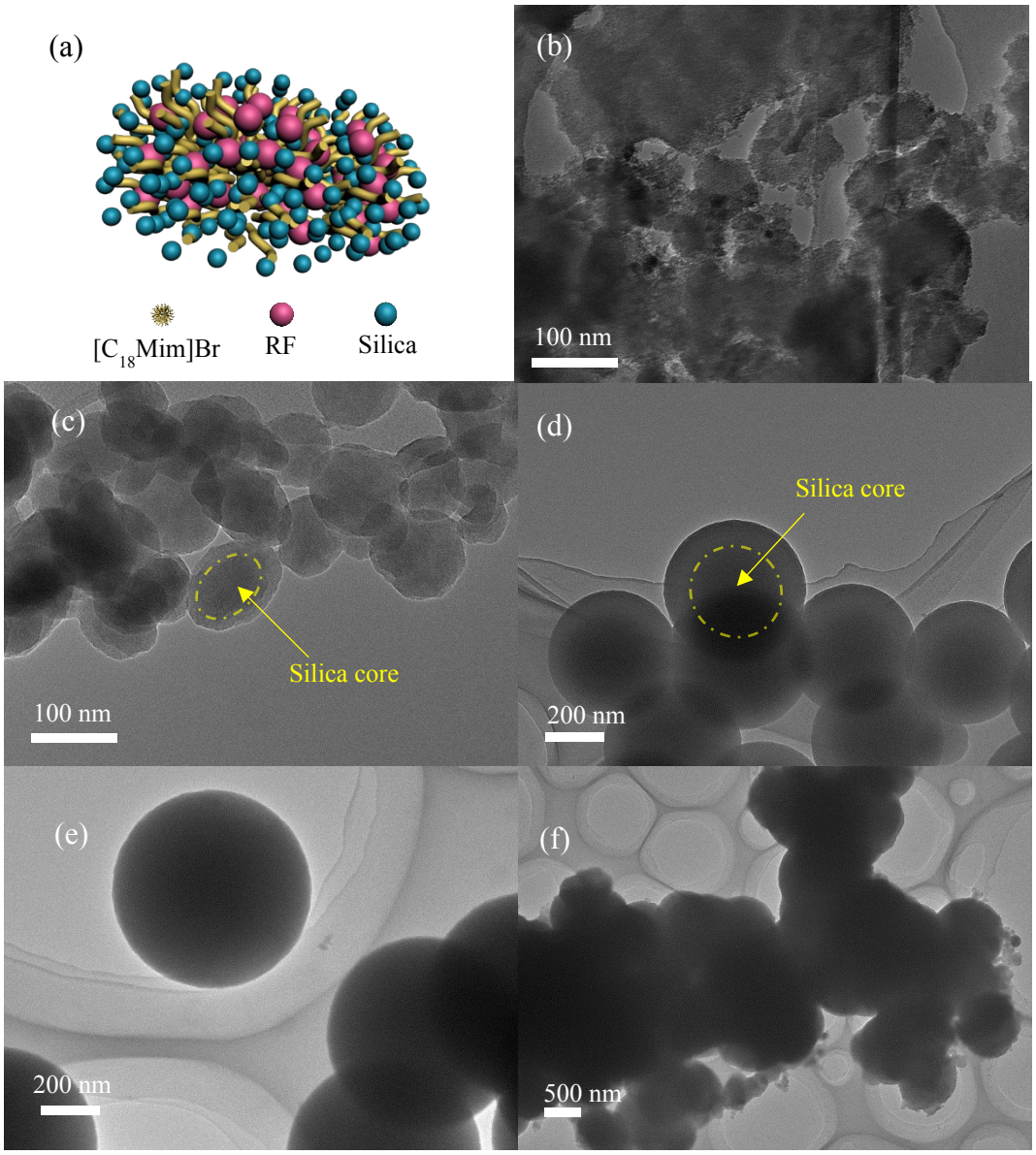

Fig. S2. Model of hybridization of silica/resin (a) and TEM images of hybridization of silica/resin precursor of N-HCS-6 (b), N-HCS-10 (c), N-HCS-14 (d), N-HCS-16 (e) and N-HCS-20 (f).

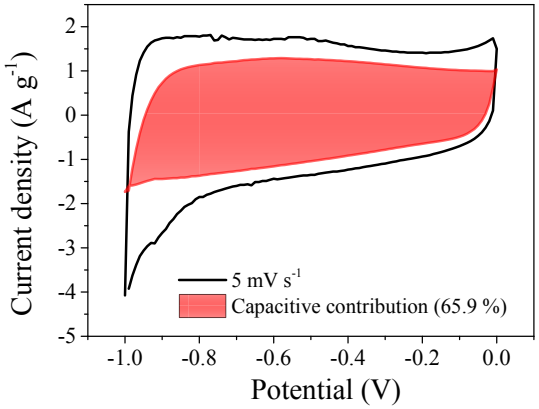

Fig. S3. Capacitive contribution of N-HCS-14 to the total charge storage at 5 mV s^{-1}.

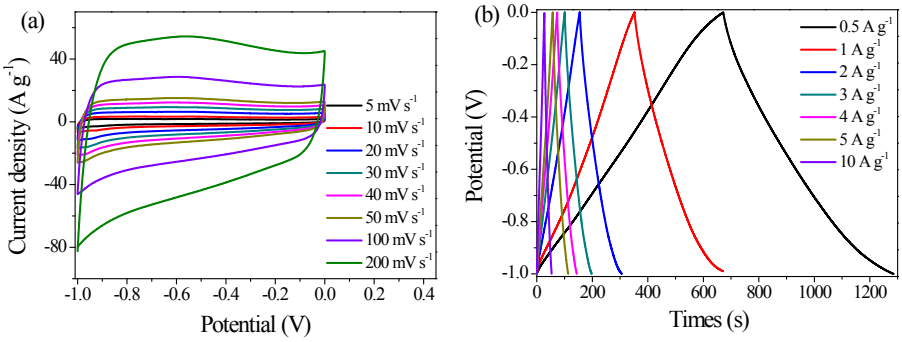

Fig. S4. CV curves at different scan rates (a), GCD curves at different current densities (b) of N-HCS-14 in three-electrode system.

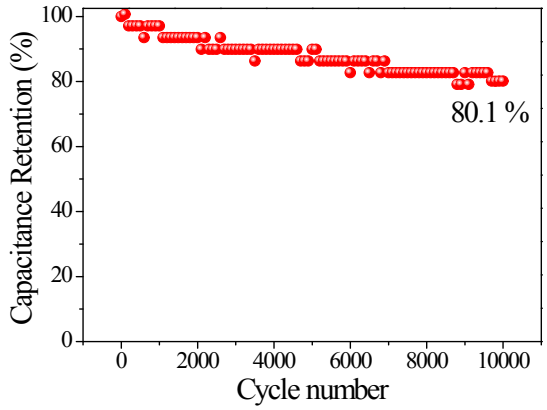

Fig. S5. Cycle test of N-HCS-14 in current density of 2 A g^{-1} in two-electrode system.