Supporting Information

Construction of 3D Nanostructure Hierarchical Porous Graphitic Carbon by Charge-induced Self-assembly and Nanocrystal-assisted Catalytic Graphitization for supercapacitors

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Experimental Section

Preparation of 3D HPGCs

Chinese grapefruits were purchased in a local fruit market. The peels were stripped with a knife and cut into small pieces, then were dried in an oven at at 100 °C. 2.5 g of dried grapefruit peels was subjected to extraction by adding 100 ml distilled water under continuous stirring for 12 h at room temperature. The pectin solution (Fig. S1) was obtained by filtrating the mixture using stainless steel mesh (200 mesh). The extraction rate of pectin from grapefruit peels was about 50%. 6g of magnesium acetate tetrahydrate ($C_4H_6O_4Mg\cdot 4H_2O$) were dissolved in the above pectin solution and stirring for 12 h. The mixed solution was dried at 100 °C to obtain the solid, and then heated in a quartz tube at 800 °C for 1 h with a heating rate of 5 °C min⁻¹ under a nitrogen atmosphere. The carbonized sample was washed with 1 M HCl solution and washed several times with deionized water to completely remove magnesium oxide (MgO) template. The carbon materials were collected by centrifugation and dried at 120 °C overnight to obtain 3D HPGCs, which was named as HPGC₀₋₈₀₀. For comparison, the solid obtained at 100 °C was first heated at 500 °C for 1 h under a nitrogen atmosphere, and then well mixed with 0.6 g KOH. The mixtures were heated at 700 °C for 1 h with a heating rate of 5 °C min⁻¹ under a nitrogen atmosphere.

Characterization

Scanning electron microscopy (SEM) micrographs were obtained from a Hitachi S-4800 instrument. Transmission electron microscopy (TEM) was carried out with a JEM-2100 electron microscope (JEOL, Japan). XRD analysis was conducted on Rigaku D/max-IIIB with copper radiation (CuK α , 0.15406 nm). Raman spectra were performed on a Jobin Yvon HR 800 micro-Raman spectrometer at 458 nm. The XPS measurements were performed on a VG ESCALAB MK II (VGScientific, UK). The nitrogen adsorption and desorption isotherms were measured at -196 °C with a Quadrachrome AUTOSORB-1-MP Adsorption Instrument. The specific surface area was calculated by Brunauer-Emmett-Teller (BET) method and the pore size distribution was determined by the Barrett-Joyner-Halenda (BJH) theory using the desorption branch data.

Electrochemical measurements

The electrochemical measurements were carried out on a CHI 660E electrochemical workstation (Shanghai Chenhua, China) in 6 M KOH solution and 1 M Na₂SO₄ solution. To prepare the testing electrode, CHHPCs were mixed with acetylene black and poly(tetrafluoroethylene) at a mass ratio of 80:10:10, pressed onto nickel foam (the active mass is about 2.0 mg cm⁻²). In a three-electrode system, the above prepared electrode, platinum foil (2.5×2.5 cm²) and Ag/AgCl were used as working electrode, counter electrode and reference electrode, respectively. Impedance measurements were conducted over a frequency range from 100 kHz to 10 mHz with the amplitude of 5mV. The specific gravimetric capacitance was calculated from the discharge process according to:

$$C_s = \frac{I\Delta t}{m\Delta V}$$

where I (A) is the charge/discharge current, Δt (s) is the discharge time, m (g) is the mass of active material, and ΔV (V) is the voltage change excluding the IR drop during the discharge process.

In a two-electrode system, the two symmetrical electrodes and a porous polypropylene separator were sandwiched together in a poly(tetrafluoroethylene) cell for a two-electrode cell. The specific capacitance for the single electrode was calculated according to:

$$C_s = \frac{4I\Delta t}{m\Delta V}$$

where I(A), $\Delta t(s)$, m(g), $\Delta V(V)$ are the current, the discharge time, the total mass of active material in both electrodes, and the voltage change excluding the IR drop during the discharge process, respectively. The energy density and power

density of symmetrical supercapacitor systems were calculated according to:

$$E_t = \frac{C_t \left(\Delta V\right)^2}{2 \times 3.6}$$
$$P_t = \frac{E_t}{t} \times 3600$$

Where E_t (W h kg⁻¹) is the specific energy density, P_t (W kg⁻¹) is the specific power density, C_t (F g⁻¹) is the specific capacitance of the total symmetrical system, ΔV (V) is the cell voltage for charging and discharging excluding the IR drop during the discharge process, and Δt (s) is the discharge time.



Fig. S1 The digital photograph of pectin solution extracted from grapefruit peel.



Fig. S2 The TEM images of (a) HPGC₀₋₈₀₀ and (b) HPGC_{0.6-700}.



Fig. S3 XRD pattern of $HPGC_{0-800}$ without removal of MgO.





Fig. S5 The digital photograph of the pH determination of pectin solution.



Fig. S6 Fourier transforms infrared spectra of pectin extracted from grapefruit peel.



Fig. S7 Thermogravimetric analysis curves of pectin/ magnesium acetate composite.



Fig. S8 SEM image of carbon material obtained using soluble starch as carbon sources with the same preparing process like as HPGCs.



Table S1 The C, O, and N contents of HPGCs						
Sample	Elemental analysis			XPS		
	C(wt%)	O(wt%)	N(wt%)	C(wt%)	O(wt%)	N(wt%)
HPGC ₀₋₈₀₀	86.32	10.21	3.47	90.9	7.1	2.0
HPGC _{0.6-700}	80.33	16.02	3.65	85.4	11.9	2.7



Fig. S10 Galvanostatic discharge curves of HPGCs at 20 A g^{-1} .



Fig. S11 Cycling stability of HPGC_{0.6-700} cell at a current density of 2 A g⁻¹ in 6 M KOH.