Experimental

***Synthesis of HNCNSs*:** The commercial EDTA calcium disodium salt was annealed at 3 °C min^{-1} from room temperature to 700 °C and kept at this temperature for 1 h under an argon flow to achieve high content of nitrogen doped carbon nanosheets (HNCNSs). And then the resulting products were immersed in a certain concentration of hydrochloric acid solution (37 wt %) to remove possible insolvable inorganic compounds. Finally, the pH was adjusted to neutral with distilled water, and the HNCNSs were dried at 80 °C in vacuum overnight.

***Characterization*:** The scanning electron microscopy (SEM) images were taken with a JEOLJSM-6700F field emission scanning electron microscope (15 kV). The transmission electron microscopy (HR-TEM) images were taken on a JEOL 2010 high-resolution transmission electron microscope performed at 200 kV. The specimen of HR-TEM measurement was prepared via spreading a droplet of ethanol suspension onto a copper grid, coated with a thin layer of amorphous carbon film, and allowed to dry in air. N_2 adsorption−desorption was measured with Micromeritics Instrument Corp.Gemini VII 2390 V1.03. The X-ray diffraction (XRD) patterns of the products were recorded with Rigaku D/max Diffraction System using a Cu Kα source (λ = 0.15406 nm). The Fourier transform IR (FTIR) spectroscopy used an FTIR spectrometer (Nicolet, Magna IR560). The Raman spectra were collected on a Raman spectrometer (Labram-010) using 632nm laser. The thermogravimetric analysis was carried out by a STA449C instrument with a heating rate of 5 °C in Ar. X-ray photoelectron spectroscopic (XPS) measurements were performed on an ESCALAB 250Xi using a monochromic Al X-ray source (200 W, 20 eV).

***Electrochemical Measurements*:** An electrochemical analysis (CHI 760E, CH Instrument, USA) was carried out with a three electrode cell to measure the electrocatalytic performance of HNCNSs electrode. A rotating ring disk electrode apparatus (RRDE-3A, ALS, Japan) in a three-electrode cell by using a platinum wire was used as counter electrode and saturated calomel electrode (SCE) as reference electrode, and a rotating ring/disk electrode with glass carbon disk (4 mm in diameter) as the working electrode. 5.0 mg HNCNSs were immersed in 2.5 mL ethanol and 0.1 mL Nafion solution (5 wt%, Sigma Aldrich, USA), and then ultrasonicated for 45 min to give a homogeneous ink. To prepare the working electrode, 10 μL of the ink was dropped onto the glassy carbon. A commercial Pt/C electrode (20%) was also prepared for comparison. Electrolyte was saturated with oxygen by bubbling O_2 prior to the start of each experiment. The ORR performance was first investigated by cyclic voltammetry (CV) in O_2-saturated 0.1 M KOH at room temperature, with a sweep rate of 10 mV s^{-1}. Linear sweep voltammetry (LSV) was conducted in the potential range of 0 to -1.2 V vs. SCE. During the recording process, a flow of O_2 or $N_2$was maintained over the electrolyte in order to ensure its continued O_2 or N_2 saturation.

The Koutecky－Levich equation was used to determine the transferred electron number (n) per O_2:

$$\frac{1}{j} = \frac{1}{j_k} + \frac{1}{B\omega^{0.5}} \qquad B = 0.2nF(D_{O_2})^{2/3}\upsilon^{-1/6}C_{O_2}$$

where *n* represents the transferred electron number per oxygen molecule. *F* is the Faraday constant (F = 96 485 C mol^{-1}). D_{O2} is the diffusion coefficient of O_2 in 0.1M KOH (1.9×10^{-5} cm^2 s^{-1}). *v* is the kinetic viscosity (0.01 cm^2 s^{-1}). C_{O2} is the bulk concentration of O_2 (1.2×10^{-6} mol cm^{-3}). The constant 0.2 is adopted when the rotation speed is expressed in rpm.

The RRDE technique also was used to determine the electron transfer number (n) per O_2:

$$n = 4 \times \frac{I_d}{I_d + I_r / N}$$

where I_d is the disk current, I_r is the ring current, and *N* is the current collection efficiency of the Pt ring. *N* was determined to be 0.42 from the reduction of $K_3Fe[CN]_6$.

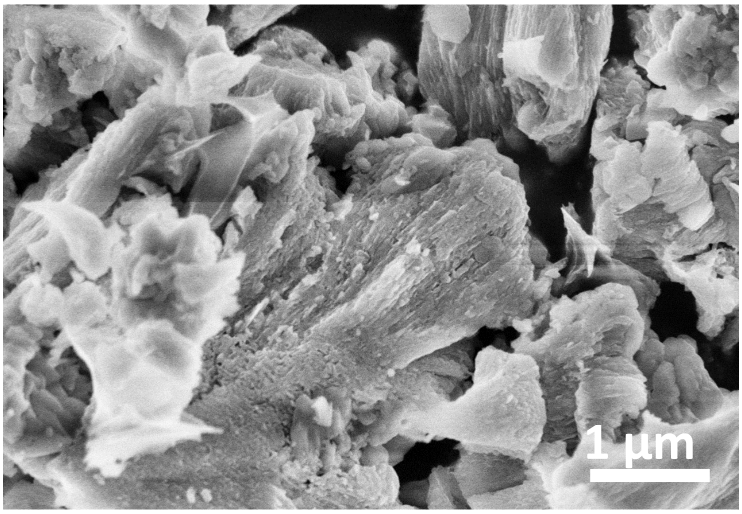

Fig. S1 SEM images of the unpickled products.

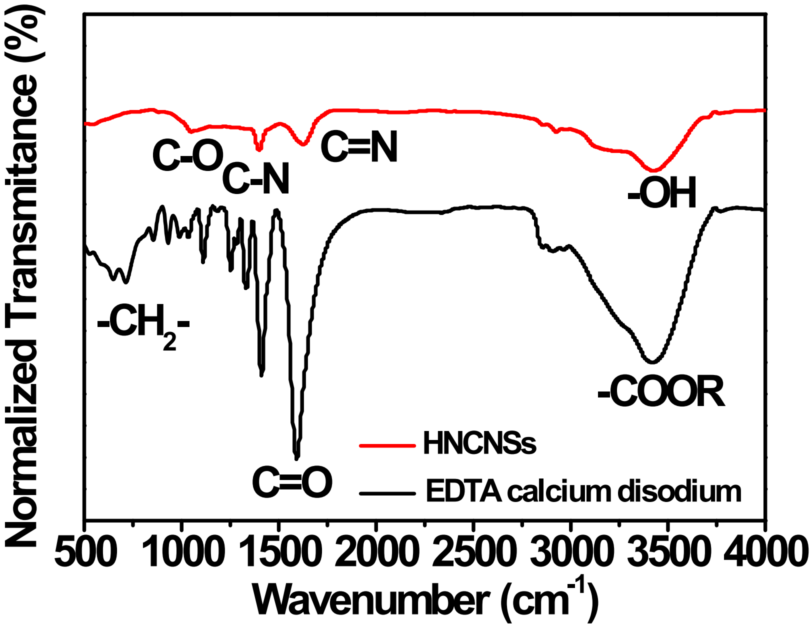

Fig. S2 FTIR spectra of the commercial EDTA calcium disodium salt and HNCNSs.

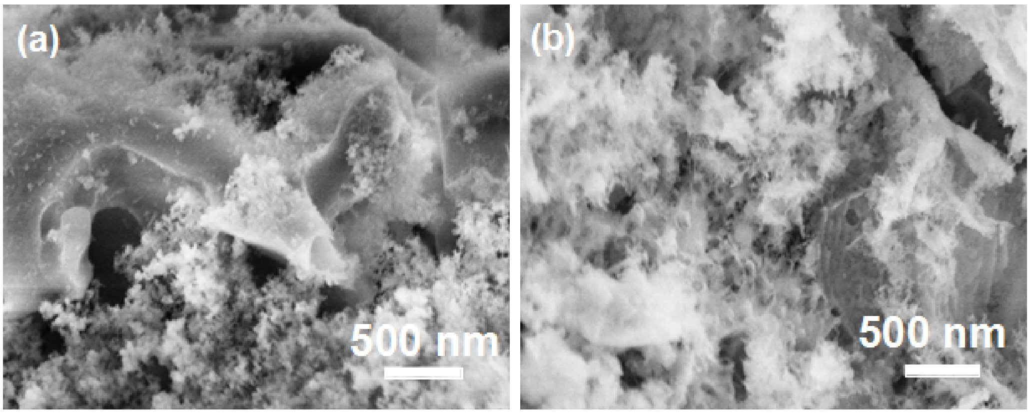

Fig. S3 SEM images of the samples synthetized at 600 (a) and 800 °C (b).

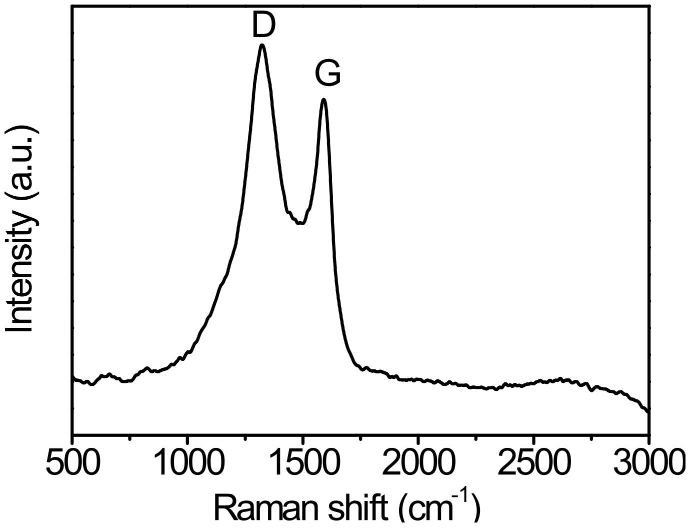

Fig. S4 The Raman spectra of HNCNSs.

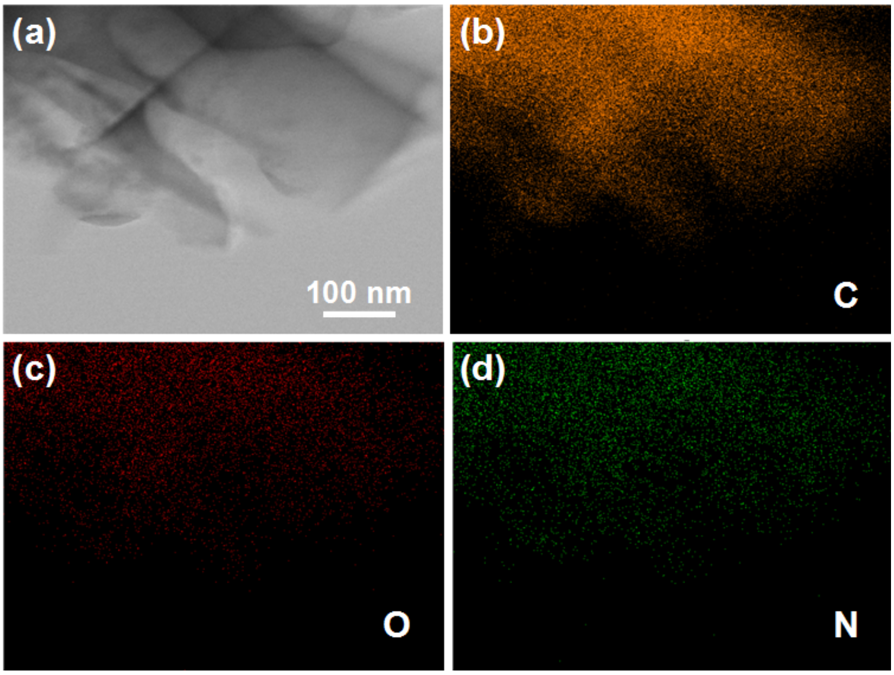

Fig. S5 EDX elemental maps of the HNCNSs. (a) TEM image, (b) C elemental map, (c) O elemental map, and (d) N elemental map.

Comment [I]: Change the "EDS" to the "EDX".

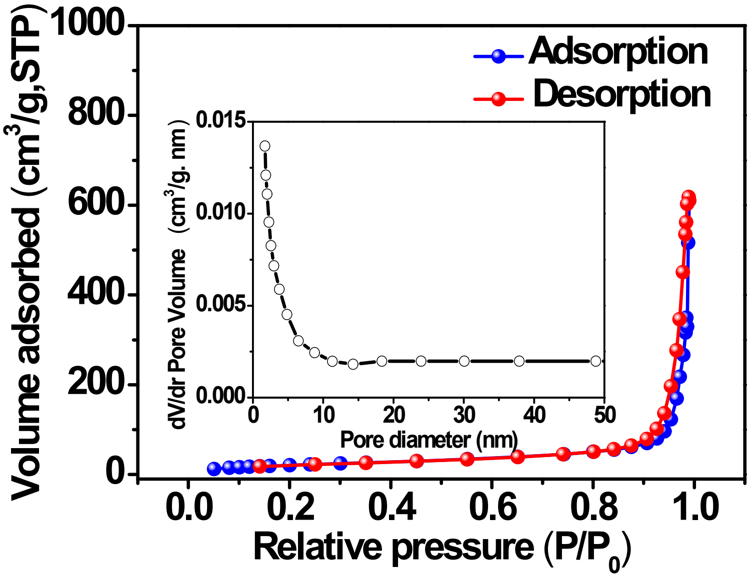

Fig. S6 Nitrogen absorption-desorption isotherms and porous size distributions of carbon honeycombs (inset).

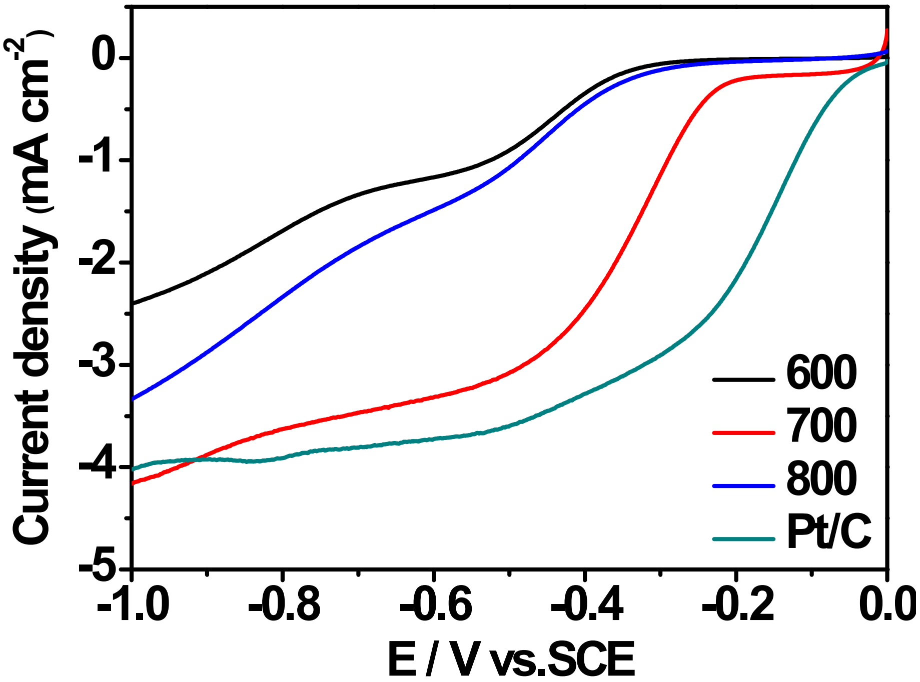

Fig. S7 Linear-sweep voltammetry (LSV) of samples obtained by different temperatures and the commercial Pt/C electrodes in an O_2-saturated 0.1 M aqueous KOH solution with a scan rate of 10 mV s^{-1} at 1600 rpm.

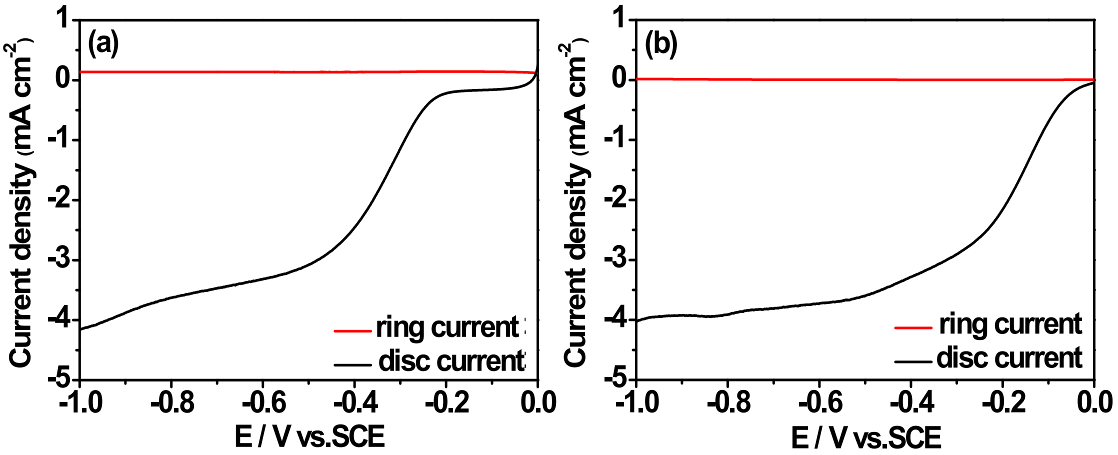

Fig. S8 The ring and disc currents of HNCNSs (a) and the commercial Pt/C (b) electrodes in an O_2-saturated 0.1 M aqueous KOH solution with a scan rate of 10 mV s^{-1} at 1600 rpm.