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# Hydrothermal Synthesis of Carbon-rich Graphitic Carbon Nitride Nanosheets for Photoredox Catalysis

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## S1. Experimental section

### *Materials synthesis.*

All chemicals were reagent grade and used without further purification. In a typical procedure, 5.5 mmol melamine (MA, Purity: 99.95%) and 16.5 mmol glocose (Purity: > 99%) powders were put into a 100 ml Teflon-lined autoclave, which was then filled with distilled water up to 60 % of the total volume. The mixture was stirred for 12 h, and then the autoclave containing nickel foam substrate was sealed and maintained at 180 $^{o}$C for 4 hours. After heating at certain temperature, the autoclave was allowed to cool to room temperature naturally. The obtained product was sequentially washed with distilled water and absolute ethanol (Purity: 92.1%) several times to remove residual impurities and then the resulting powder was dried at 60 $^{o}$C for 12 h to get the final products. Meanwhile, the nickel foam substrate was then etched away by HCl solution (3M, Purity: 37%) at 80 °C for 12 h to obtain the pure products named as CCN for simplicity.

As a reference sample, 3 g melamine was placed into a crucible with a cover and heated to 550$^{o}$C for 4 h in a muffle furnace to induce the bulk condensation of melamine. Products synthesized from the bulk condensation were denoted as CN.

### *Characterization:*

The morphologies of the as-prepared nanofibers were observed by the scanning electron microscope (SEM; XL-30 ESEM FEG, Micro FEI Philips), energy dispersive X-ray (EDX) and transmission electron microscopy (TEM; Hitachi 600). X-ray diffraction (XRD) measurements were carried out using a D/max 2500 XRD spectrometer (Rigaku) with Cu Kα line of 0.1541 nm. X-ray photoelectron spectroscopy (XPS) was accomplished using a PHI-5702 multi-functional X-ray photoelectron spectrometer with pass energy of 29.35 eV and an Mg Ka line excitation source. The UV-vis diffuse reflectance (DR) spectroscopy of the samples was recorded on a Cary 500 UV-vis-NIR spectrophotometer.

### *Electrochemical Measurements:*

Electrochemical impedance spectroscopy was carried out using the conventional three electrode setup connected to an electrochemical station (CH Instrument 660C, Shanghai Chenhua, China). This assembly has C/FTO, CN/FTO and CCN/FTO (effective area was 1 $cm^2$) as working electrodes, and a Pt wire and an Ag/AgCl (saturated KCl) electrode were used as the counter electrode and reference electrode, respectively. The electrolyte was 0.1mol $L^{-1}$ $Na_2SO_4$ aqueous solution. The measurements were carried out at a constant potential of +1.0 V at the working photoanode. Incident-photon-to-current-conversion eficiency (IPCE) spectra were measured on a QE/IPCE Measure-ment Kit (Crowntech QTest Station 1000AD) with a tungsten halogen lamp (CT-TH-150), a calibrated silicon diode and a monochromator (Crowntech QEM24-S 1/4 m).

### *Photocurrent and photocatalytic measurements:*

Visible light and UV light photocurrent measurements were performed in a conventional three electrode cell, using a Pt wire and an Ag/AgCl electrode (saturated KCl) as counter electrode and reference electrode, respectively. This assembly has C/FTO, CN/FTO and CCN/FTO (effective area was 1 $cm^2$) as working electrodes, and the electrolyte was 0.1mol $L^{-1}$ $Na_2SO_4$ aqueous solution. Before any measurement, the film electrode was immersed in a nitrogen purged sodium sulphate electrolyte solution (0.2 M) for 60s. The visible light was generated by a 500w xenon lamp (Beijing Changtuo, CHF-XM500) with a 400nm cut-off filter, and was chopped manually (≈0.20 Hz). The UV-Visible photocatalytic activity of C, CN and CCN was evaluated from the degradation of 4-nitrophenol (4-NP). In the photodegradation of 4-NP dye test, 0.05g photocatalyst was suspended in 100 mL 4-NP solution with the initial concentration of 10 ppm in a beaker. A 150 W xenon lamp was used as the UV-visible light source. At given intervals of illumination, 3 mL reacting solutions in series were taken out and analyzed. The concentrations of 4-NP in the reacting solutions were analyzed by a Cary 500 UV-vis NIR spectrophotometer.

| **Glucose (mmol)** | **Melamine (mmol)** | **Reaction Condition** | **Results** | **Molar ratio C/N*** |
|---|---|---|---|---|
| 16.5 | 0 | Hydrothemal | C | / |
| 0 | 50 | Calcined | CN | 0.77 |
| 16.5 | 5.5 | Hydrothemal | CCN | 0.8 |
| 0 | 5.5 | Hydrothemal | Melamine | / |

*Determined by XRD using the Scherrer equation.

*

**Table S1.** Experimental details of different samples.

| Samples | $R_b$ (Ω) | Warburg (Ω) | $R_{ct}$ (Ω) |
|---|---|---|---|
| C | 5.21 | 21.3 | 203.2 |
| CCN | 8.13 | 27.1 | 282.49 |
| CN | 10.11 | 33.4 | 390 |

**Table S2.** The EIS simulation parameters of C, CN and CCN electrodes

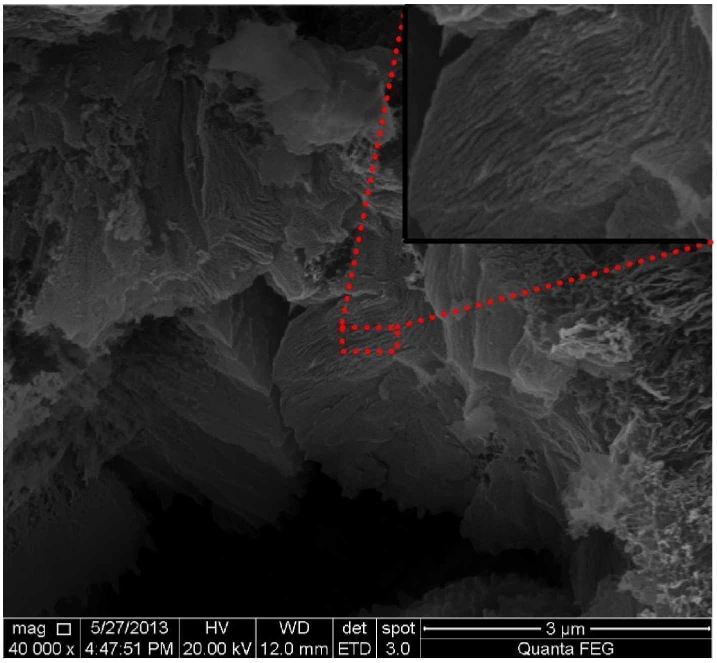
mag □
40 000 x
5/27/2013
4:47:51 PM
HV
20.00 kV
WD
12.0 mm
det
ETD
spot
3.0
3 µm
Quanta FEG

**Figure S1.** SEM image of CN.

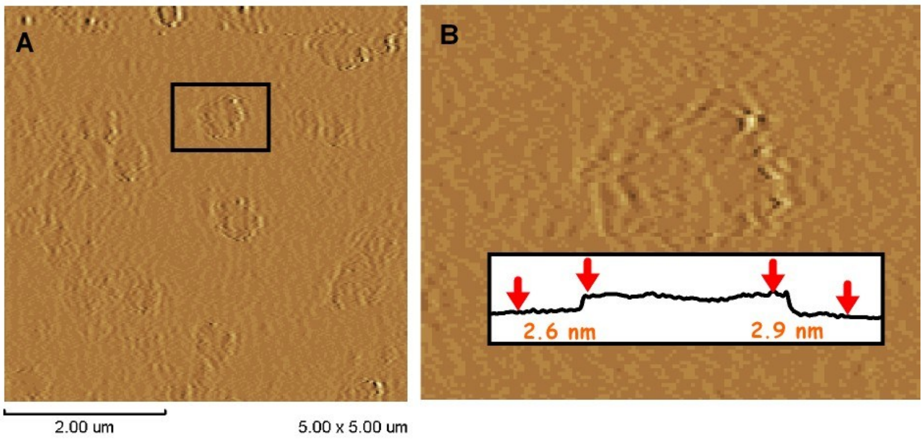

**Figure S2.** (A) A typical AFM image of CCN on SiO2 substrate; (B) AFM image of a single CCN with smooth planar structure

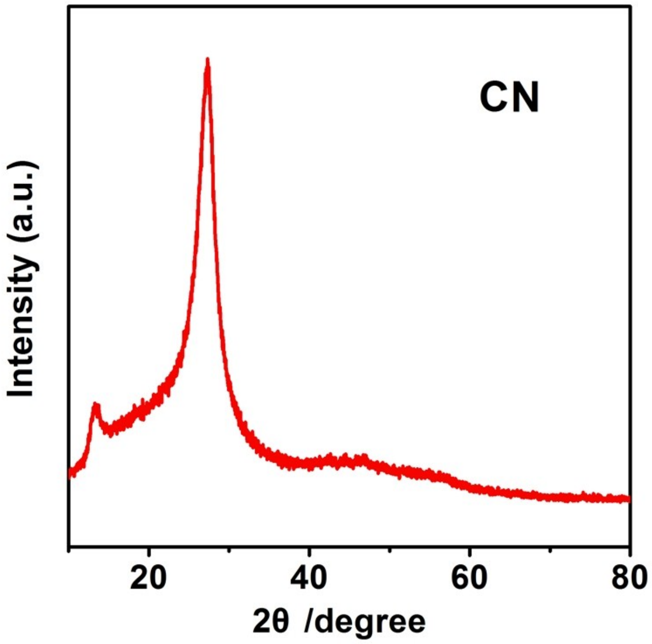

**Figure S3.** XRD partten of CN.

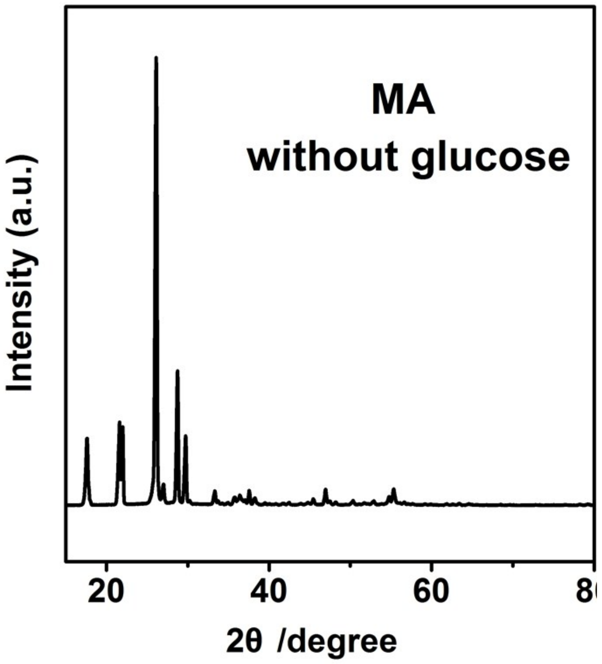
MA
without glucose
Intensity (a.u.)
20
40
60
2θ /degree

**Figure S4.** XRD partten of the sample (Reaction condition: MA without glucose).

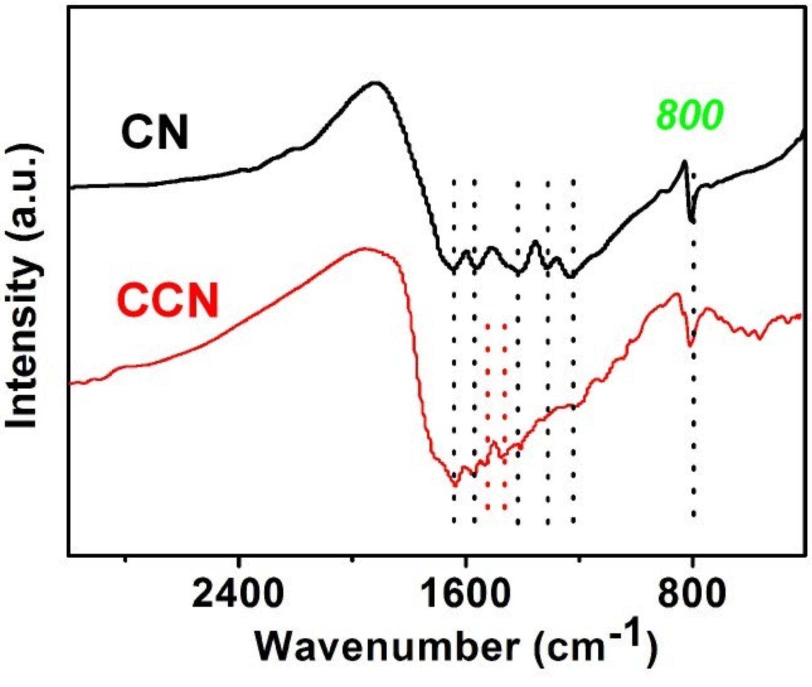

**Figure S5.** FTIR spectra of CN and CCN.

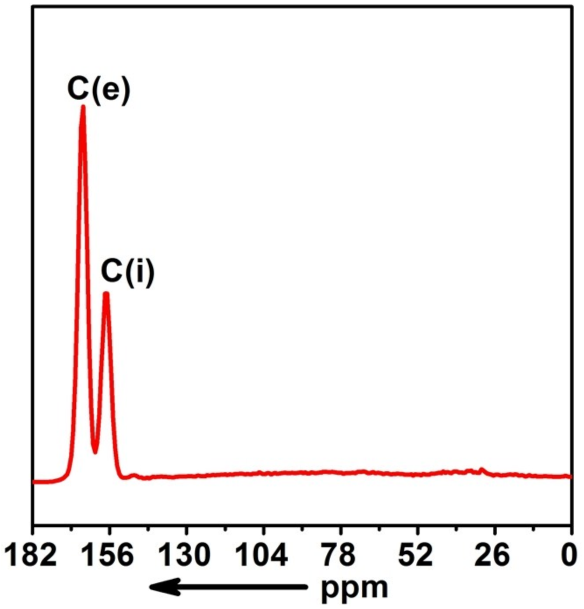

**Figure S6.** $^{13}$C MAS NMR analysis of CCN.

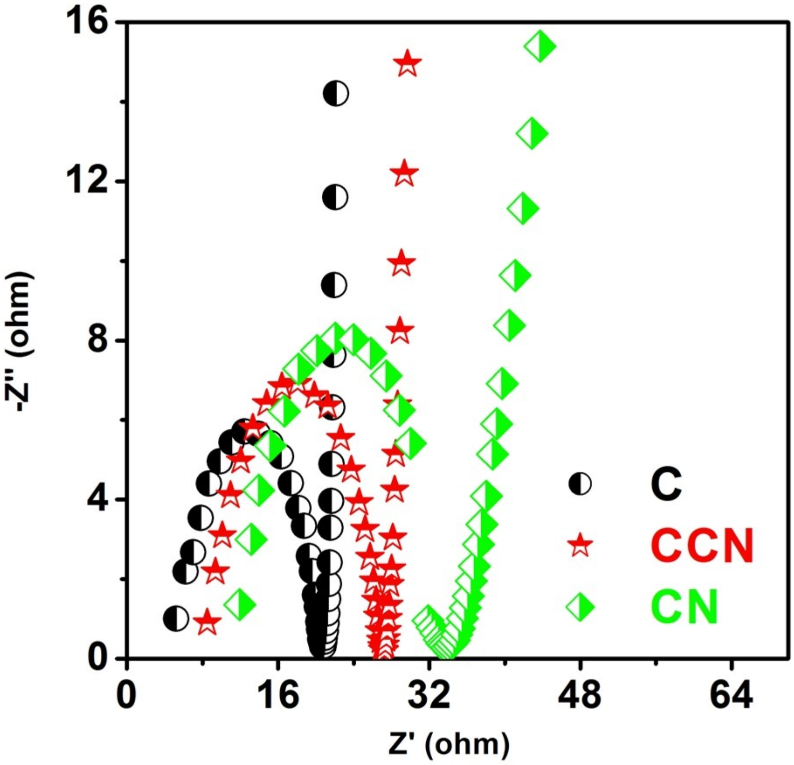
-Z'' (ohm)
0
4
8
12
16
Z' (ohm)
0
16
32
48
64
C
CCN
CN

**Figure S7.** EIS Nyquist plots of C, CN and CCN electrodes.

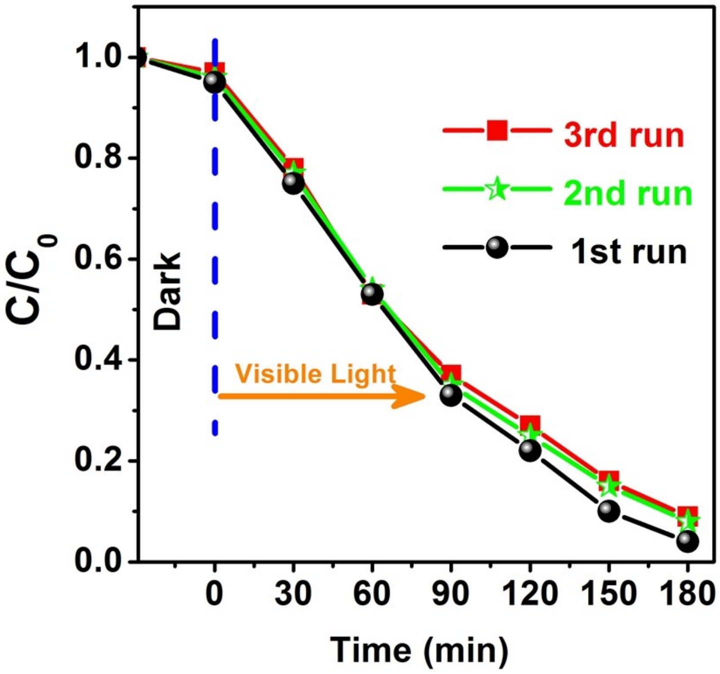

**Figure S8.** Photocatalytic activity of the CCN with three times of cycling uses.

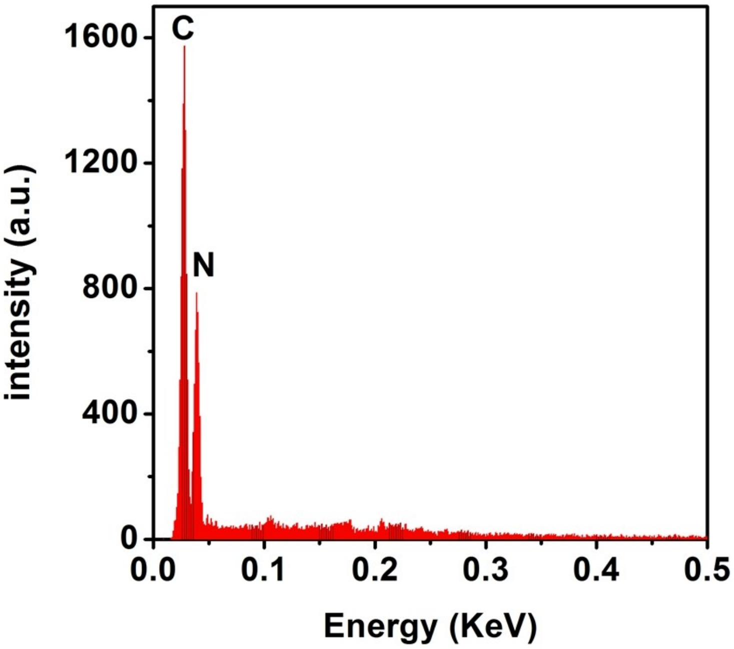

**Figure S9** EDX data of CCN.