Electronic Supplementary Information

Construction of 2D g-C_3N_4 lateral-like homostructures and their photo- and electro-catalytic activities

Xiao Zhang, Jean-Pierre Veder, Shuai He, and San Ping Jiang*

Fuels and Energy Technology Institute and Western Australia School of Mines: Minerals, Energy and Chemical Engineering, Curtin University, Perth WA6845, Australia, E-mail: S.Jiang@curtin.edu.au.

1. Synthesis of samples

Amorphous g-C_3N_4 was prepared at 650 ºC named as CN-650. Typically, 10g of melamine was added to a quartz boat and calcined at 650 ºC in argon atmosphere for 2h with a heat-treat rate of 5 ºC/min. After that, the obtained sample was washed with deionized water and ethanol for several times. Crystalline g-C_3N_4 was prepared using a slow heat-treated procedure at 750 ºC named as CN-750. Namely, 10g of melamine was added to a quartz boat and calcined at 350 ºC in argon atmosphere for 2 h with a heat-treat rate of 5 ºC/min. The temperature was then increased to 750 ºC for 3 h with a rate of 1 ºC/min. The as-prepared sample was sonicated in water to be dispersed and selected by controlling centrifuged speed. Finally, the suspension with small specimen was precipitated and washed with water and ethanol for several times.

For the preparation of crystalline/amorphous g-C_3N_4 homostructures named as CN-750@650, 1g of selected CN-750 was ground with 2g of melamine for 2h. The sample was then added to a quartz boat and calcined at 650 ºC in argon atmosphere for 2h with a heat-treat rate of 5 ºC/min. finally, sample was dispersed in water with sonication and selected separation.

To deposit Au or Pt nanoparticles, 10 mg of as-prepared samples was uniformly dispersed in 5 ml of deionized water. 10 μl of 50 M $HAuCl_4$ or $HPtCl_4$ aqueous was then added and stirred for 5 min. The solution was sonicated for a certain time. To control the size of nanoparticles, the reaction time was changed. The as-prepared precipitate was washed for three times with water and ethanol, and dried. For comparison, Au and Pt nanoparticles were deposited via a reflux process using $NaBHO_4$ as the reductant. A $NaBHO_4$ solution was added to the solution with g-C_3N_4 sample and Au or Pt ion solution. After refluxing for 10 to 30 min, the as-prepared samples were precipitated and washed.

2. Characterization

UV-visible spectrophotometer (U-4100, Hitachi) was used for measuring UV-visible diffuse reflectance and absorption spectra. The X-ray diffraction (XRD) patterns of the C_3N_4 samples were taken by an X-ray diffractometer (Bruker D8, Germany) with Cu Kα radiation source recorded in 2θ range 10-80°. Transmission electron microscopy (TEM) (FEI Titan G2 80-200 TEM/STEM with Chemi STEM Technology, FEI company, US) was used for morphology observation. Hitachi F-4600 Fluorescence spectrometer was used to measure photoluminescence (PL) spectra at room temperature (with 400 nm as the excitation wavelength). X-ray photoelectron spectroscopy (XPS) was performed on Kratos Axis Ultra DLD spectrometer with a monochromatic AlKα (1486.6eV) X-rays operating at 150 W.

3. Photocatalytic and electrocatalytic performance

3.1 Photocatalytic H_2 production

A system of Pyrex top-irradiation reaction vessel connected to a glass-closed gas circulation was used for photocatalytic H_2 production measurement. 10 mg of each of the g-C_3N_4 sample was dispersed in a 100 ml (triethanolamine: deionized water = 1:9) aqueous solution. The mixture solution was sonicated for 15 min before light exposure and was evacuated before testing. The solution with constant stirring was then irradiated under a 300 W Xe lamp with a cutoff filter ($\lambda > 420$ nm). Finally the amount of H_2 evolved was detected using Shimadzu GC-7920 gas chromatography with Argon as carrier gas.

3.2 Photocatalytic degradation of organic pollutants

Photocatalysis investigation was carried out to degrade rhodamine (RhB) under visible light. 30 mg of sample was mixed with RhB solution (50ml, 10mgL/l). The mixture was stirred in dark for 60 minutes to get the equilibrium absorption state. Measurement of absorbance of RhB was performed on a UV-VIS spectrophotometer using 300W Xe arc lamp with a UV-cut off filter ($\lambda \geq 420$ nm) as the light source.

3.3 Electrochemical Measurement.

Electrochemical measurements were performed on a three-electrode system using CHI 660E electrochemical analyzer (with graphite rod as the counter electrode, an Ag/AgCl electrode as the reference electrode, and a glassy carbon electrode with diameter of 3.0 mm as the working electrode). The sample was dispersed in a mixed solution of 5 wt% Nafion, water and alcohol (V_{Nafion}:V_{water}:$V_{alcohol}$ = 0.025:4:1) to form a 2 mg mL^{-1} uniform catalyst ink solution. 6 μL of the prepared ink (0.17 mg cm^{-2}) was pipetted onto the working electrode. Linear sweep voltammetry (LSV) curve was recorded in a 0.5 M H_2SO_4 solution with 5 mV s^{-1} scan rate. The measured potentials are corrected to reversible hydrogen electrode (RHE) using the following equation:

$E_{(RHE)} = E\ (Ag/AgCl) + 0.059pH + 0.197$

4. Tables and Figures

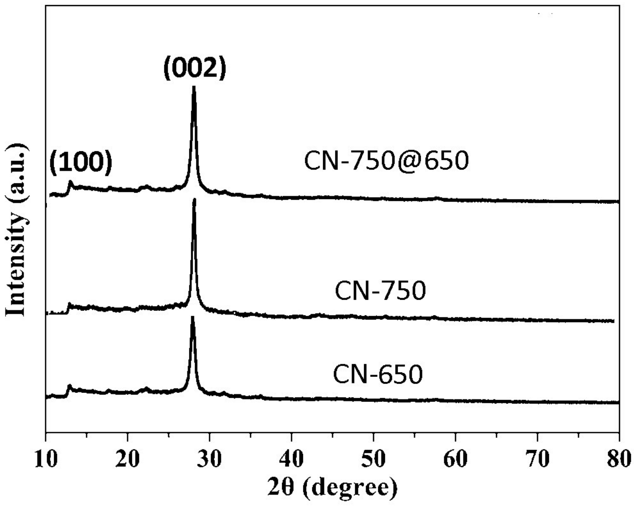

Fig. S1 XRD patterns of samples

Table S1. Peak intensity ratios of samples in C1s spectra.

Sample	C-C	C-O-C& C-OH	C=N	C-N	Sat
CN-650	2.0	1.0	5.6	11.6	0.87
CN-750	1.8	1.0	2.0	5.1	0.2
CN-750@650	2.9	1.0	4.9	22.3	1.8

Table S2. Peak intensity of samples in N1s spectra.

Sample	C-N=C (Pyridinic)	N-$(C)_3$ (Pyrrolic)
CN-650	44.84	25.45
CN-750	52.44	22.15
CN-750@650	46.81	29.22

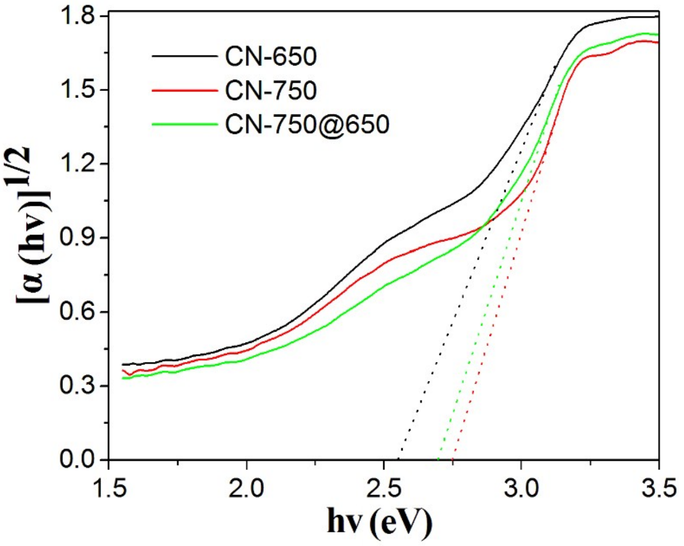

Fig. S2 Plots of $(\alpha h\nu)^{1/2}$ versus photon energy of samples CN-650, CN-750, and CN-750@650.

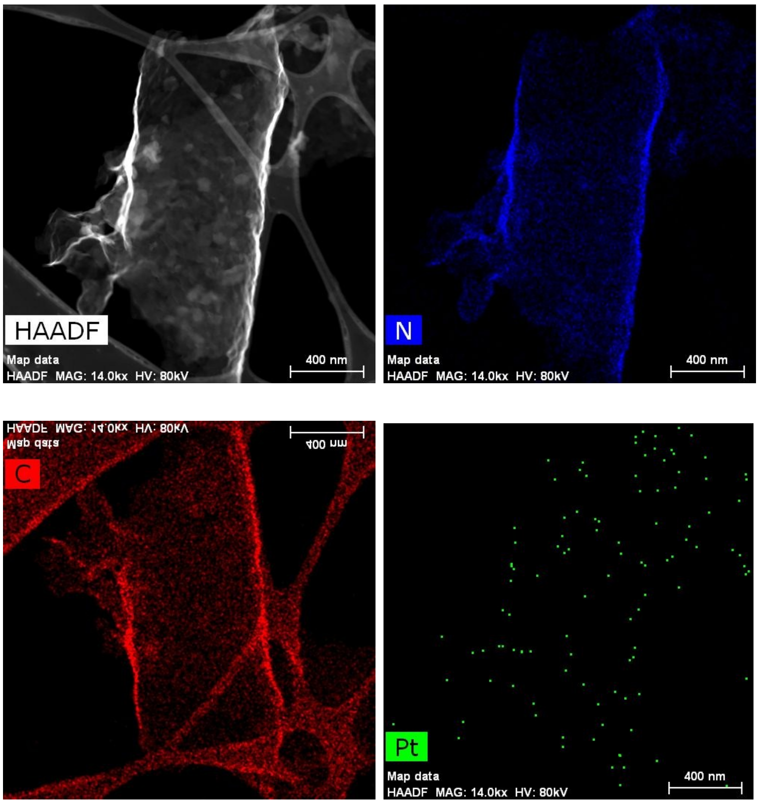

Fig. S3 (a) HAADF-STEM images and N (b), C (c), and Pt (d) mapping of Pt-deposited CN-750@650-1.

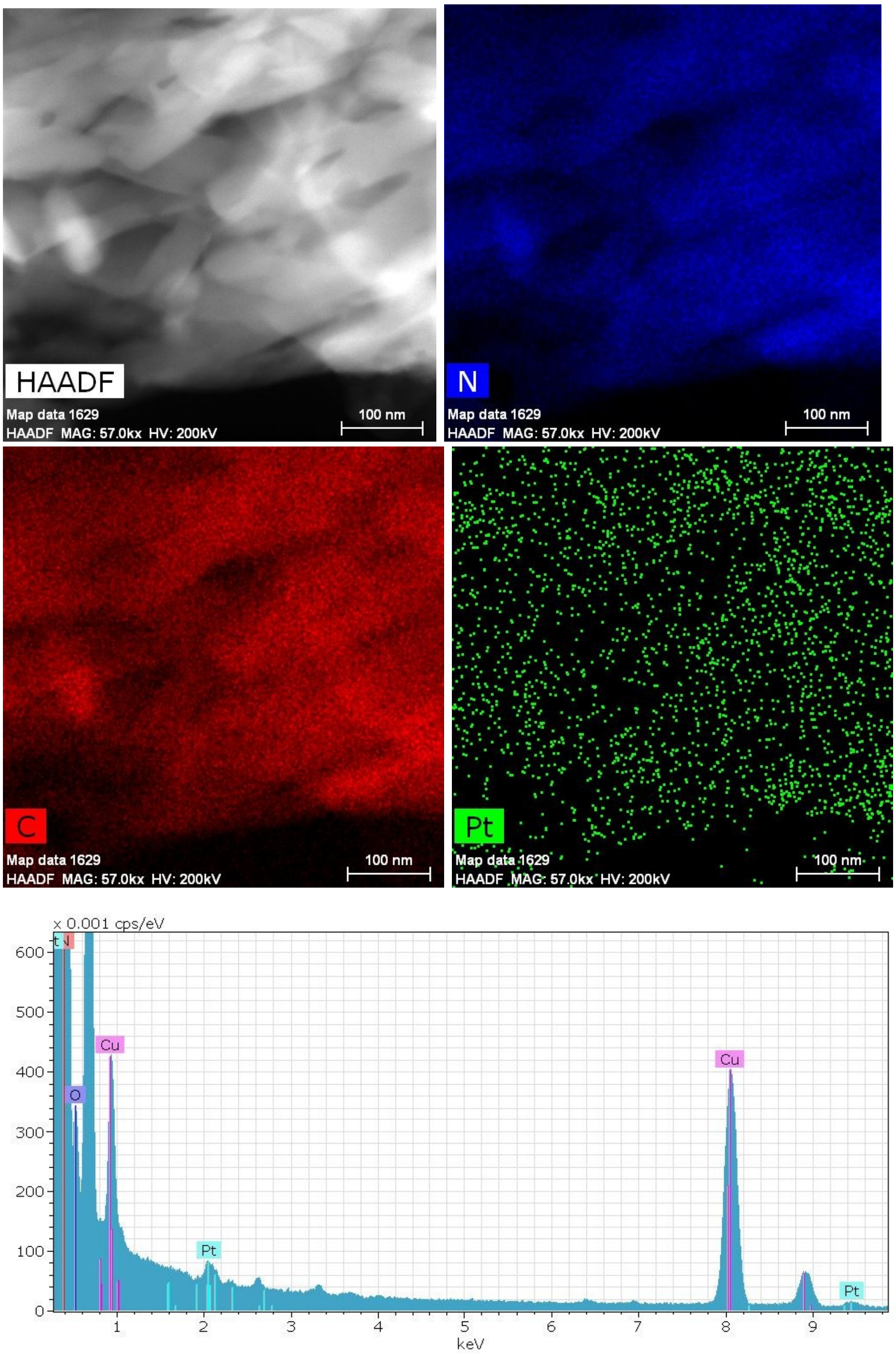

Fig. S4 (a) HAADF-STEM images and N (b), C (c), Pt (d) mapping and EDS spectrum of Pt-deposited CN-750@650-2.

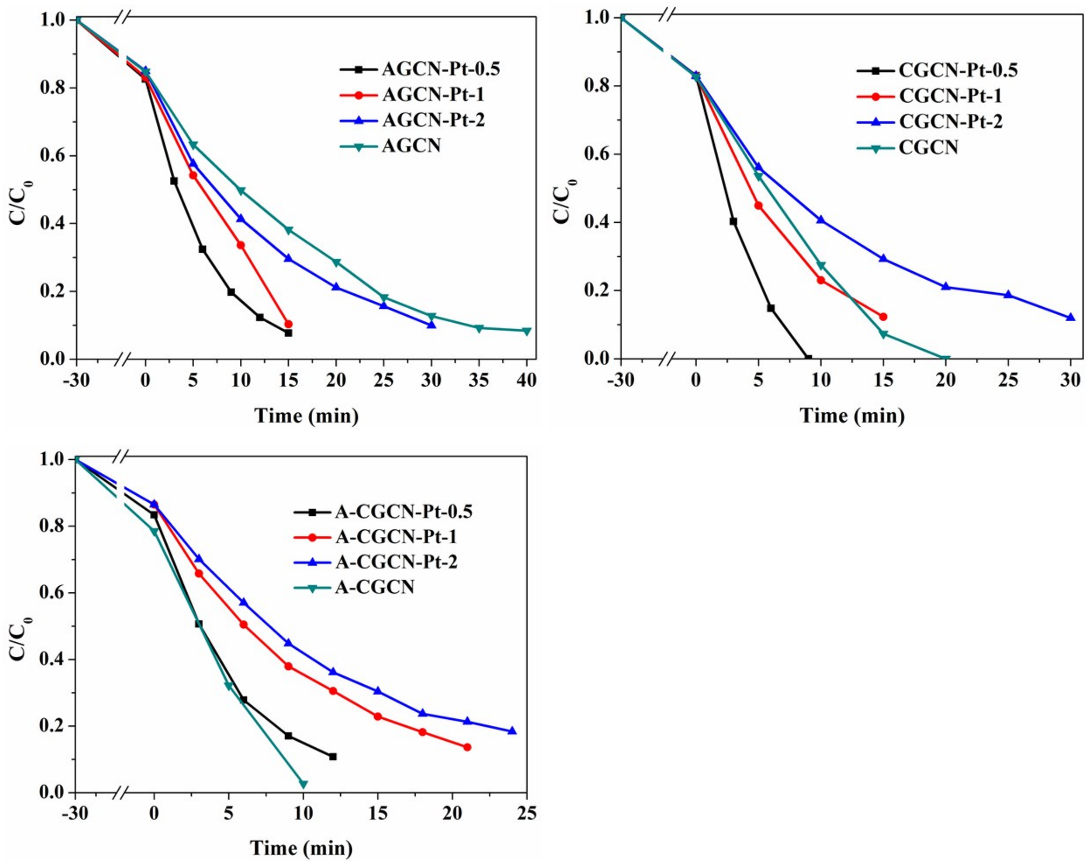

Fig. S5 Photocatalytic degradation plots of RhB with time for g-C_3N_4 samples. (a) CN-650. (b) CN-750. (c) CN=750@650. Pt-deposited sample with short time of 0.5 h revealed the best performance.

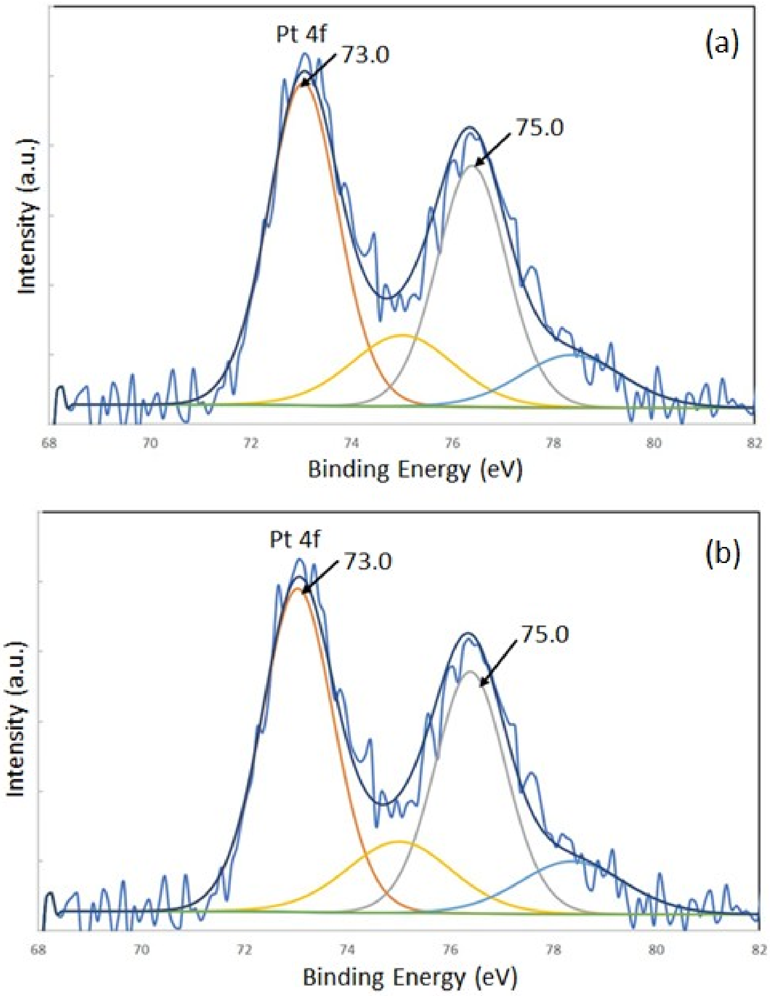

Fig. S6 XPS spectra of Pt for Pt-modified CN-750 Pt-C_3N_4 with deposition time of 0.5 (a) and 2 h (b).

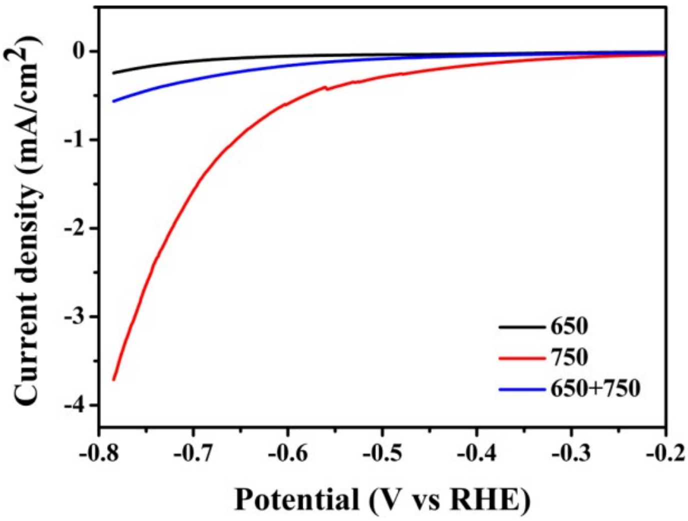

Fig. S7 HER polarization curves of g-C_3N_4 samples under visible light irradiation with a scan rate of 5 mV s^{-1}.