Electronic Supplementary Information

Edge-epitaxial growth of yellow g-C$_3$N$_4$ on red g-C$_3$N$_4$ nanosheets and superior photocatalytic activities

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1. Preparation of samples

Bulk g-C$_3$N$_4$ (named as BCN) was prepared at 500 ºC. 10g of melamine was added to a quartz boat and calcined at 500 ºC in argon atmosphere for 2h with a ramp rate of 5 ºC/min. The obtained sample was then washed with deionized water and ethanol for several times. Yellow g-C$_3$N$_4$ nanosheets (named as YCN) were prepared through polymerization of BCN. 10g of BCN was added to a quartz boat and calcined at 650 ºC in argon atmosphere for 2 h with a heat-treat rate of 5 ºC/min. Red g-C$_3$N$_4$ nanosheets were prepared using a molten salt assisted method.

10 g of melamine was mixed with 8 g of mixtures of NaCl and KCl (mole ratio = 1:1). The mixture was ground evenly by adding ethanol and water to obtain a powder sample. The obtained sample was then calcined at 700 °C for 1 h using a ramp rate of 5 °C/min in a muffle furnace. The product was purified using ethanol and H$_2$O and dried to obtain sample RCN.

The yellow/red g-C$_3$N$_4$ heterostructures (named as HCN-1 to HCN-4) were prepared by using red g-C$_3$N$_4$ as seeds. Typically, 0.4g of selected sample RCN was ground with 10g of bulk g-C$_3$N$_4$ for 2h. The sample was then added to a quartz boat and calcined at 650 ºC in argon atmosphere for 2h with a ramp rate of 5 ºC/min. Finally, sample was dispersed in water with sonication and selected separation. The amount of sample RCN was adjusted to 4, 8, 12, and 15 wt%, respectively.

2. Characterization

The PL spectra of samples were collected on a fluorescence spectrometer (Hitachi F-4600). 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$_3$N$_4$ 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. The Brunauer-Emmett-Teller (BET) specific surface area and pore diameter distribution of the samples was tested using an N_2 adsorber (MFA-140 of Beijing Builder Company). Nicolet 380 Fourier transform infrared spectroscopy (FTIR) spectrometer (Thermo, America) was used to obtain FTIR spectra. The PL efficiency of samples in solutions was estimated by comparing with a standard solution sample (a rhodamine 6G ethanol solution with PL efficiency of $\sim 95\%$).

3. Photocatalytic performance

A system consisting of a Pyrex top-irradiation reaction vessel and a glass-closed gas circulation system was used for photocatalytic H_2 production measurement. 10 mg of each of the g-C_3N_4 samples was dispersed in a 100 ml (triethanolamine: deionized water = 1:9) aqueous solution. The g-C_3N_4 sample solution was sonicated for 20 min and evacuated before light exposure and testing. The sample solution with constant stirring was then irradiated under a 300 W Xe lamp with a cutoff filter ($\lambda > 420$ nm). The amount of H_2 evolved was detected using Shimadzu GC-7920 gas chromatography with Argon as carrier gas. The hydrogen evolution activity data of the samples was collected in a 4 h cycle. The hydrogen evolution stability test was performed for 4 cycles.

The apparent quantum efficiency (AQE) of samples is calculated using the equation shown bellow.

$$\text{AQE} = \frac{N_e}{N_p} = \frac{2 \times M \times N_A \times h \times c}{S \times P \times t \times \lambda} * 100\% \tag{4}$$

where Ne and Np are the amount of reaction electrons and the incident photons, respectively. M is the amount of H_2 molecule, N_A the Avogadro constant, c is the speed of light, S is irradiation area, P is irradiation intensity, t is the time, and λ the wavelength of monochromatic light.

Photocatalytic CO_2 reduction measurement of samples was performed and the data were analyzed using gas chromatography (Shimadzu GC-7920). 20 mg of sample was added to 15 mL of water to

get a homogeneous suspension. CO_2 bubbling of the sample solutions was done with vigorous stirring of 20 min to remove air. A 300 W Xe lamp with a cut off filter of 420 nm was used as light source during CO_2 reduction.

4. Tables and Figures

Table S1 Specific surface area and pore size of the samples.

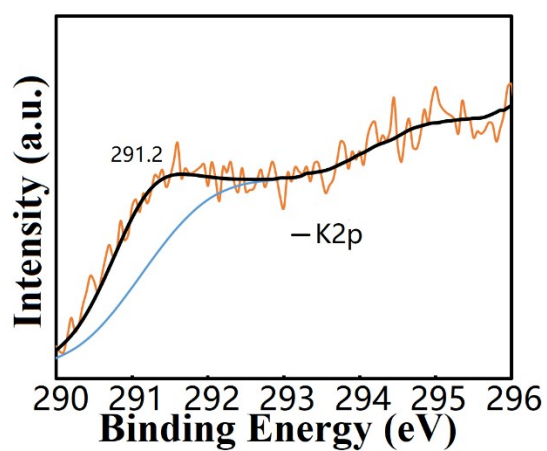| Sample	Composition	S (m^2/g)	Pore size (nm)
BCN	Bulk g-C_3N_4	57.7	6.2
RCN	Red g-C_3N_4	198.1	12.0
YCN	Yellow g-C_3N_4	82.1	6.4
HCN-1	Red/yellow g-C_3N_4	94.8	6.6
HCN-2	Red/yellow g-C_3N_4	1.3.5	7.1
HCN-3	Red/yellow g-C_3N_4	119.3	7.4
HCN-4	Red/yellow g-C_3N_4	135.1	7.8

Fig. S1 K 2p spectrum of yellow/red g-C_3N_4 heterostructures (sample HCN-3).

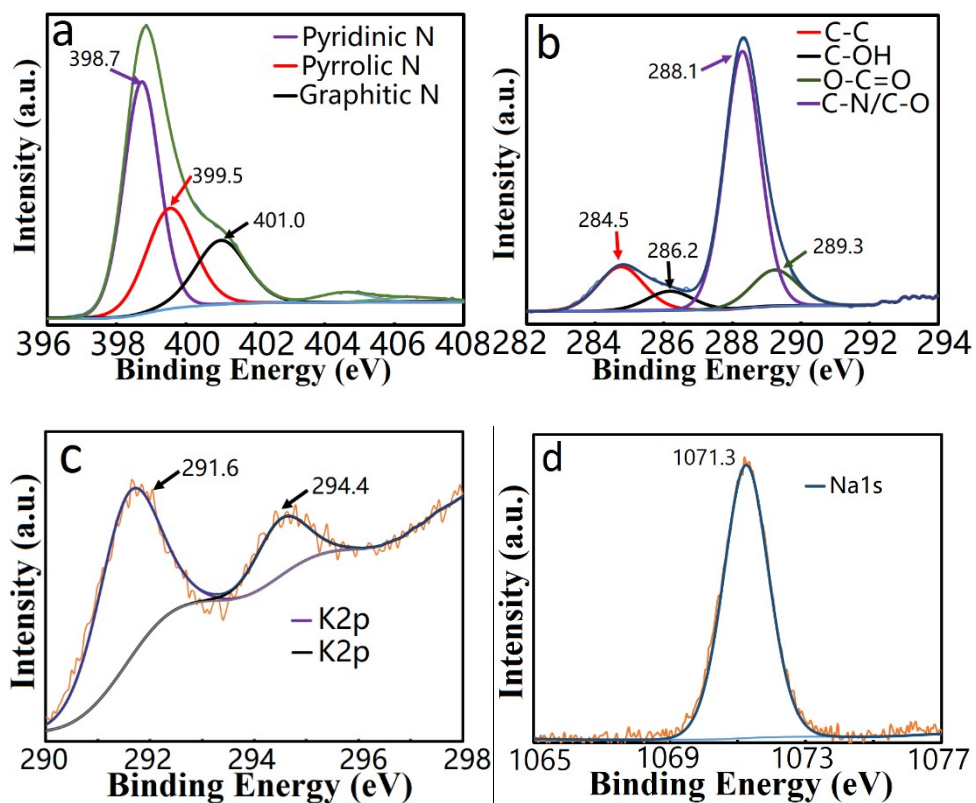**Fig. S2** XPS spectra of water-soluble red g-C$_3$N$_4$. (a), N 2p spectrum. (b) C 1S spectrum. (c), K 2p spectrum, (d), Na 1s spectrum.
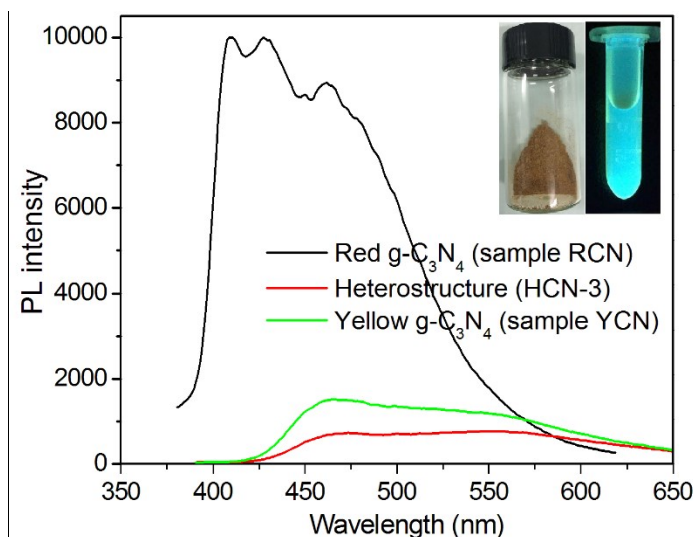

Fig. S3 Photoluminescence (PL) spectra of samples. Insets in figure show photographs of powder sample (left) and a colloidal solution of sample RCN irradiated under 365 UV light. Compared with red g-C$_3$N$_4$, the PL intensity of the heterostructures is the lowest, suggesting a sufficient photogenerated carrier separation.