

Supporting Information

Multiple Ordered Porous Honeycombed g-C₃N₄ with Carbon Ring In-Plane Splicing for Outstanding Photocatalytic H₂ Production

Xiaobo Wu^a, Huiqing Fan^{*a}, Weijia Wang^{*a}, Lin Lei^a, Xinye, Chang^a and Longtao Ma^{*b}

^aState Key Laboratory of Solidification Processing, School of Materials Science and Engineering, Northwestern Polytechnical University, Xi'an, 710072 (P. R. China)

^bFrontiers Science Center for Flexible Electronics, Institute of Flexible Electronics, Northwestern Polytechnical University, Xi'an, 710072 (P. R. China)

**Corresponding author: prof. Huiqing Fan*

E-mail: hqfan@nwpu.edu.cn

Photocatalytic Measurements for Water Splitting

50 mg of sample and 10 mL of triethanolamine (TOEA) were stirred and ultrasonically dispersed in 90 mL deionized water, then Pt co-catalyst (3 wt.%) was in-situ photo-deposited on the photocatalyst by injecting 2 g/L H_2PtCl_6 aqueous solution into preceding mixture solution. The whole system was vacuumized before photocatalytic testing and sustained at 10 °C by circulating condensed water. A 300 W Xeon lamp (MAX-302, Asahi Spectra, Torrance, CA, USA) is used as the radiation light source and the wavelength is controlled by a UV cut-off filter. The volume of evolved hydrogen was collected and tested by a gas chromatography instrument. The light intensity was detected by an irradiatometer (FZ-A, PEIFBNU, Beijing, China). AEQ was calculated using Eq. (1):

$$\text{AQE} = \frac{2 \times (\text{Number of hydrogen molecules})}{\text{Number of incident photons}} \times 100\% \quad (1)$$

Electrochemical Measurements

All electrochemical measurements were carried out in a three-electrode system by means of an electrochemical workstation (CHI660E, Chenhua, Shanghai, China). The photocatalyst paste was prepared by adding several drops of terpineol to the mixture (mass ratio of sample/ethyl cellulose/lauric acid = 5:3:2) and grinding it thoroughly. The working electrode was fabricated by coating paste onto exposed area (a circle of 8 cm in diameter) of FTO conductive glass. The glass is then calcined at 350 °C in a Muffle furnace for half an hour. Therein a square platinum sheet and an Ag/AgCl electrode (3 M KCl) served as the counter electrode and reference electrode, at the same time, 0.5 M Na_2SO_4 aqueous solution is selected as the electrolyte. Additionally, the EIS spectra

were measured by imposing an open-circuit voltage with an amplitude of 5 mV under a frequency range of 10^6 to 10^{-2} Hz.

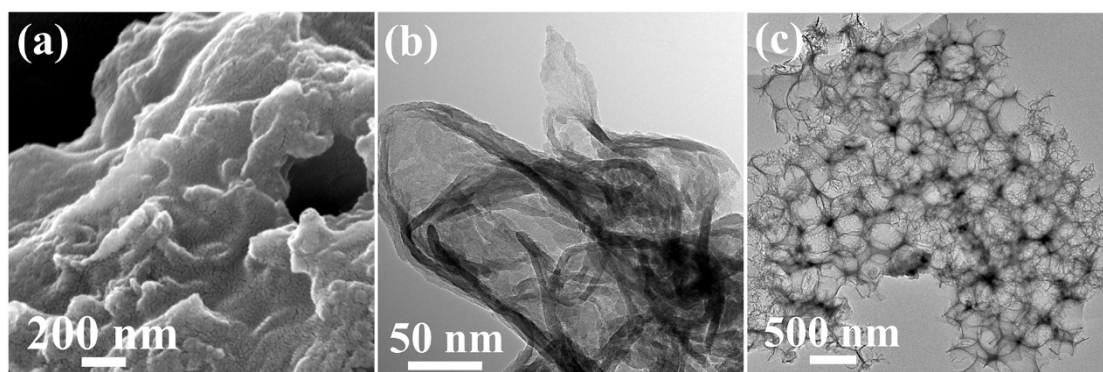


Fig. S1. (a) SEM image of CN; TEM images of (b) CN and (c) PHCN.

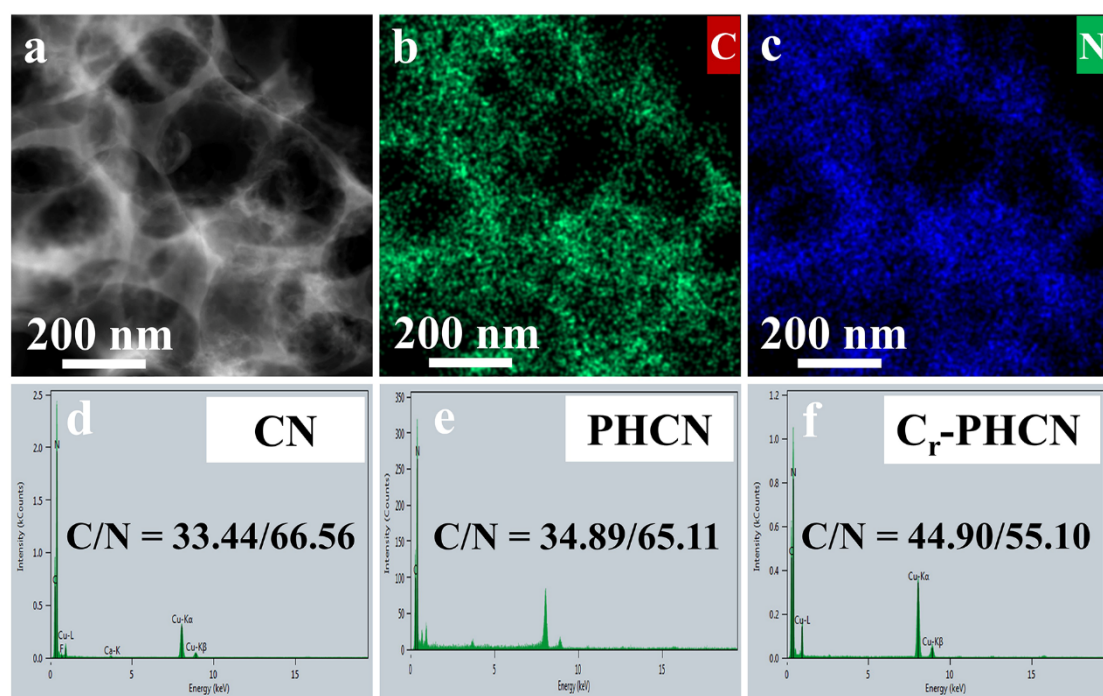


Fig. S2. (a), (b) and (c) Elements mapping on C_r-PHCN; STEM-EDS spectra and atomic fraction of (d) CN, (e) PHCN and (f) C_r-PHCN.

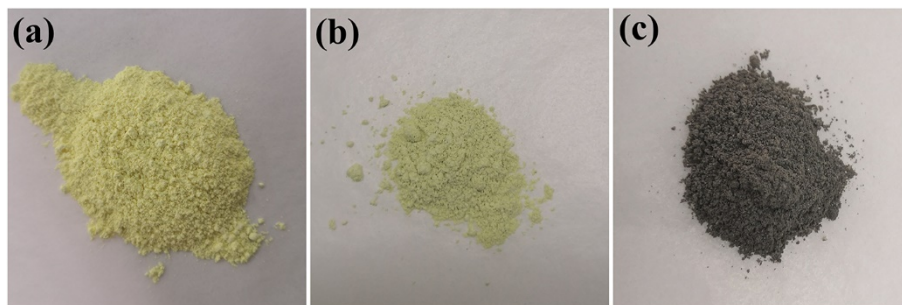


Fig. S3. The color of (a) CN, (b) PHCN and (c) C_r-PHCN.

Table S1. Fitting results for the lifetimes of photogenerated charge carriers of CN, PHCN and C_r-PHCN.

Sample	τ_{av} (ns)	Lifetime (ns)	Percentage (%)
CN	3.17	τ_1 : 1.40	82.2
		τ_2 : 5.32	17.8
CN-NT	3.81	τ_1 : 1.63	76.6
		τ_2 : 5.82	23.3
C _r -PHCN	4.58	τ_1 : 1.84	76.3
		τ_2 : 6.92	23.7

Table S2. AQE of CN, PHCN and C_r-PHCN under monochromatic lights with different wavelengths.

Sample	λ (nm) - AQE (%)				
	420	450	500	550	600
CN	0.94	0.29	0	0	0
PHCN	4.48	2.36	0.76	0.26	0
C _r -PHCN-	10.62	5.36	1.12	0.41	0

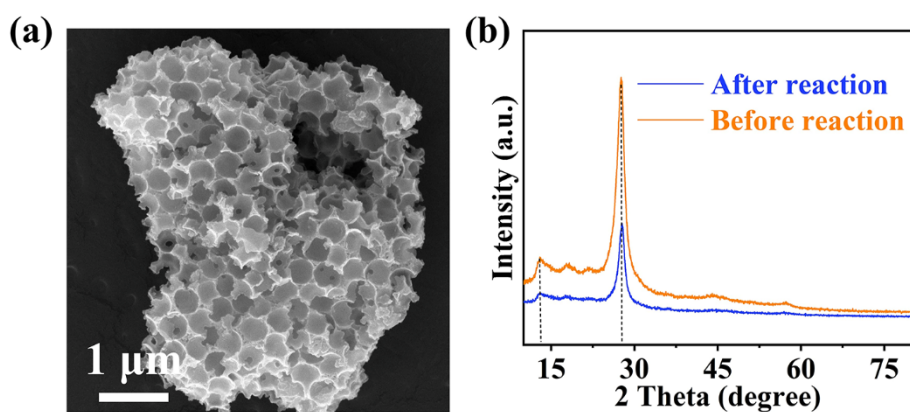


Fig. S4. (a) SEM image of C_r -PHCN after the photocatalytic reaction and (b) XRD patterns of C_r -PHCN before and after the photocatalytic reaction.

The powder under the SiO_2 templates during the CVD preparation process of C_r -PHCN was collected as a blank sample, which is carbon ring in-plane seamless splicing carbon nitride (denoted as C_r -CN). The time courses of H_2 evolution and the average hydrogen evolution rate (HER) are tested under the same conditions.

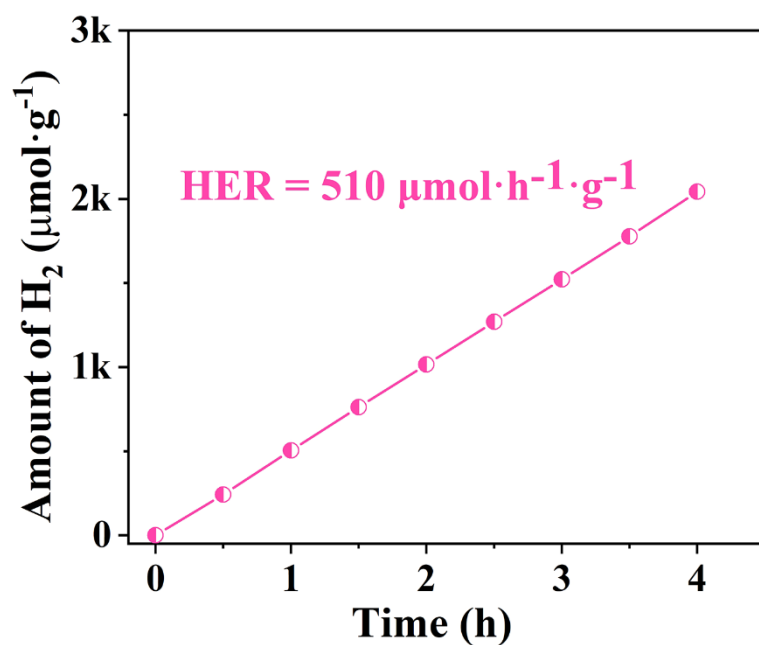


Fig. S5. Time courses of H_2 evolution and HER of C_r -CN.

Table S3. The comparison of photocatalytic hydrogen performance of the C_r-PHCN with other g-C₃N₄ based photocatalysts.

Photocatalyst	Method	AQE (%)	HER (μmol·h ⁻¹ g ⁻¹)	Ref
C _r -PHCN	In-air CVD	10.62	7581	This work
3DOM CN	Self-assembly	6.27	7900	1
CCN	Self-assembly		6400	2
CN-10	NaCl template	2.2	459	3
C _r -CN	Multistep heating	5	371	4
CN-Br-3	Co-pyrolysis	14.3	1354	5
CN-NT	In-air CVD	6.49	4605	6
R-CN	In-air CVD		1900	7
HOCN	Copolymerization	4.32	1140	8
CdZnS-HCN	In-situ precipitation		5145	9

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