

Supplementary Information

Graphitic carbon nitride with thermal-induced nitrogen defects: an efficient process to enhance photocatalytic H₂ production performance

Guangzhi Dong^{a,b}, Yun Wen^a, Huiqing Fan^{a*}, Chao Wang^{a,c},
Zhenxiang Cheng^b, Mingchang Zhang^a, Jiangwei Ma^a, Shujun Zhang^b

a. State Key Laboratory of Solidification Processing, School of Materials Science and Engineering, Northwestern Polytechnical University, Xi'an, 710072, PR China.

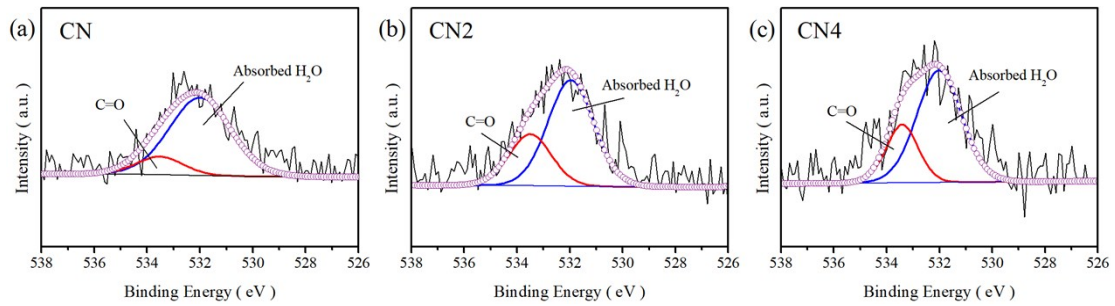
b. Institute for Superconducting and Electronic Materials, Australia Institute of Innovative Materials, University of Wollongong, Wollongong, 2522, Australia.

c. Department of Chemical Engineering, University College London, Torrington Place, London WC1E 7JE, United Kingdom.

* Corresponding author: hqfan@nwpu.edu.cn; hqfan3@163.com

Table S1. Distribution of N atoms in the samples.

Samples	C-N=C (N _{2C} ; %)	N-(C) ₃ (N _{3C} ; %)	-NH _x (N _{1C} ; %)	N _{2C} :N _{3C}
CN	78.22	11.82	9.96	6.61
CN2	78.73	13.90	7.37	5.66
CN4	80.72	12.18	7.10	6.63

**Fig. S1.** High resolution XPS spectra of O1s for (a) CN, (b) CN2, (c) CN4.**Table S2.** Variation of the N:C ratios and amino contents in samples.

Samples	N:C (from EA)	N:C (from XPS)	-NH _x
CN	1.767	2.327	9.96
CN2	1.711	2.288	7.37
CN4	1.710	2.252	7.10

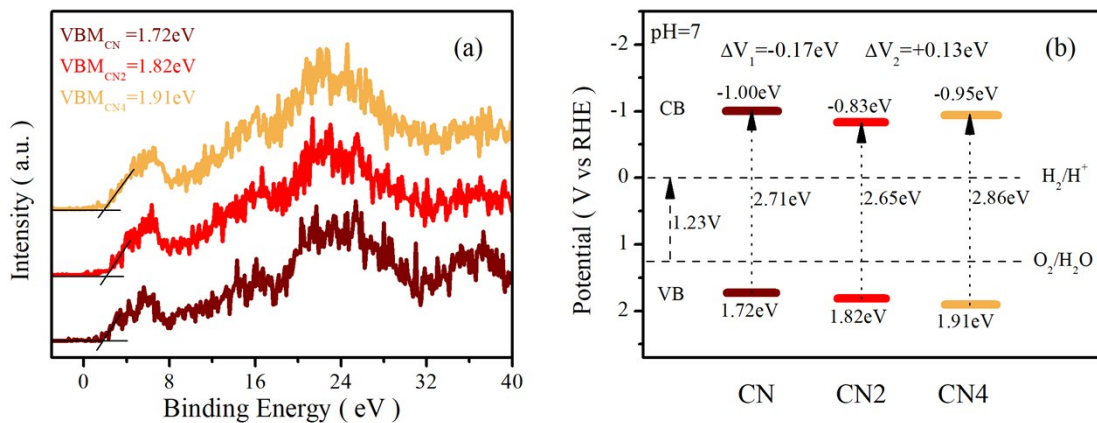
**Fig. S2.** (a) VB XPS spectra, and (b) corresponding diagrammatic band structure of CN, CN2 and CN4.

Table S3. Dynamics analysis of TRPL emission decay for CN, CN2, and CN4.

Samples	τ_1 (ns)	A_1	τ_2 (ns)	A_2	$\langle\tau\rangle$ (ns)
CN	1.10	24406.1	5.34	252.27	1.20
CN2	1.32	10989.5	5.42	293.73	1.72
CN4	1.10	22065.5	5.21	316.94	1.34

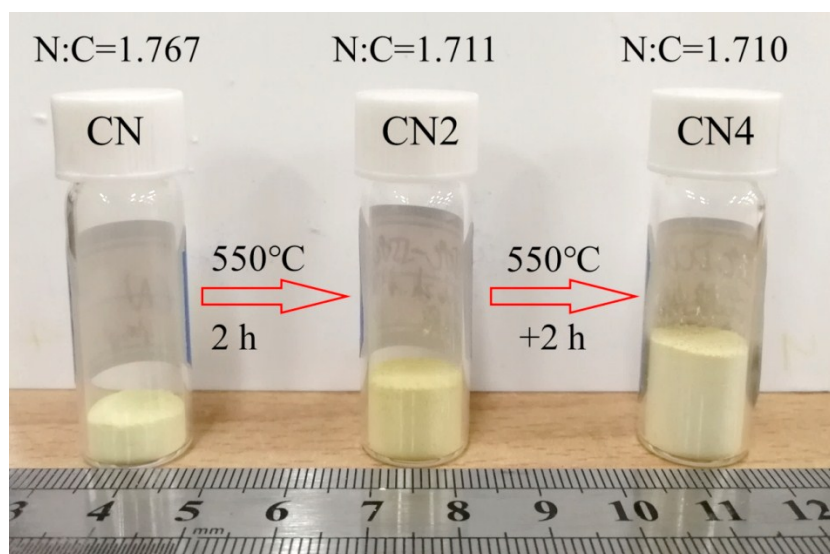


Fig. S3. Comparison of volume (300 mg) and color for CN, CN2, and CN4.

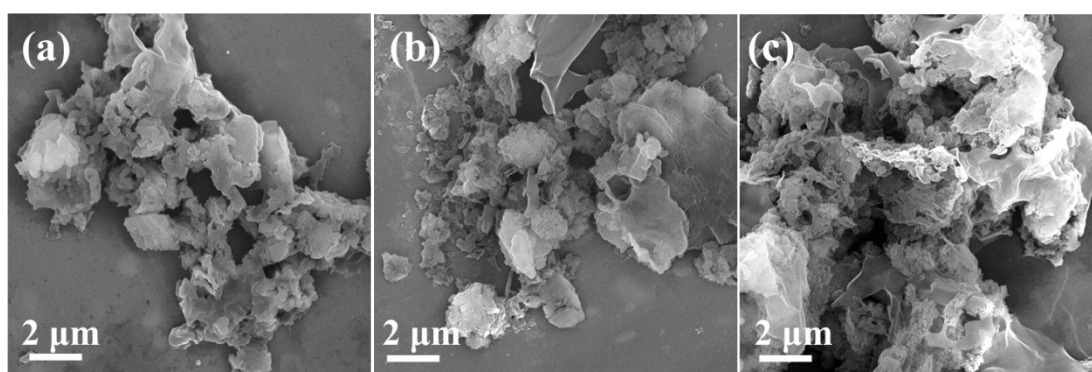


Fig. S4. SEM images of (a) CN, (b) CN2, and (c) CN4.

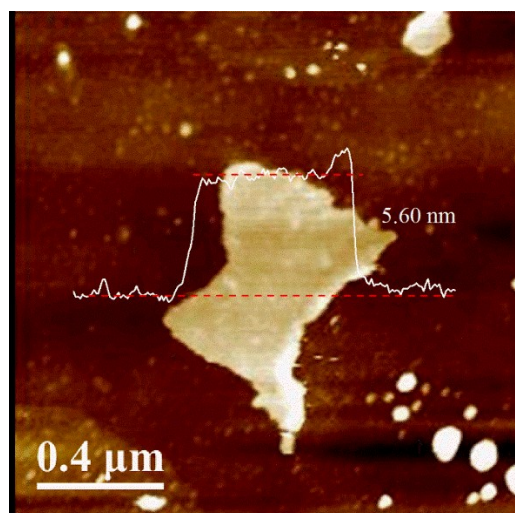


Fig. S5. AFM images of CN4 and the corresponding height curve.



Fig. S6. Status of 10 mg photocatalysts dispersing in 10 mL water after natural sedimentation for different days. (left: CN, middle: CN2, right: CN4).

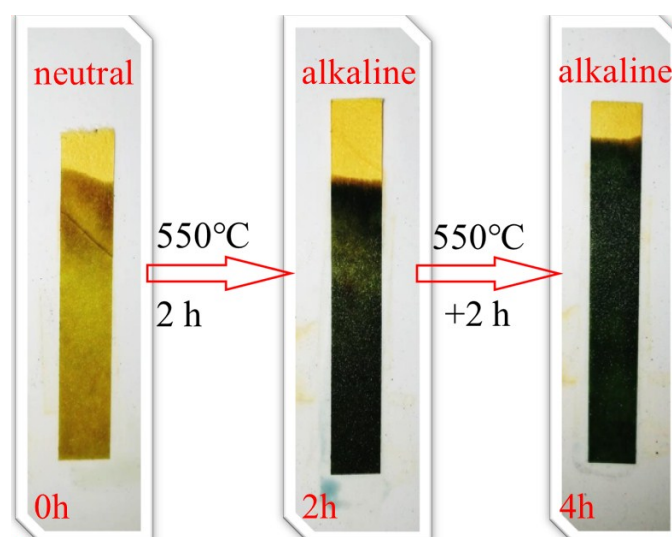


Fig. S7. Comparison of pH value for the tail gas solution obtained after the experiment finished.

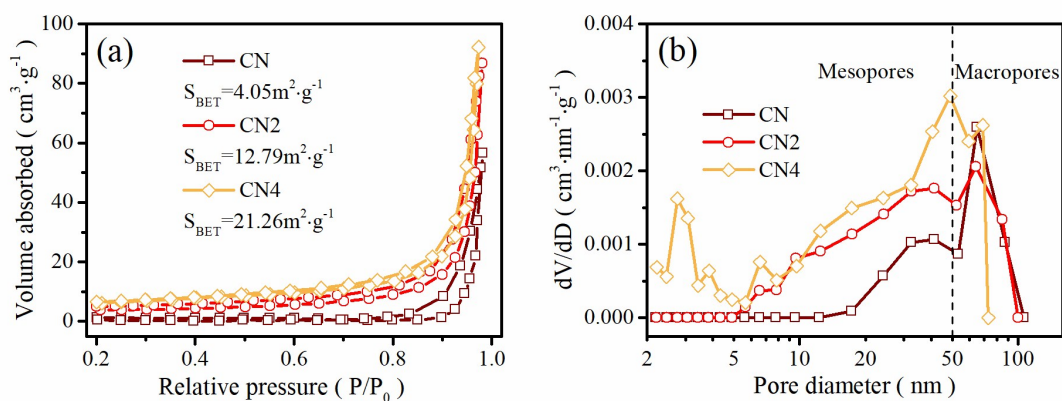


Fig. S8. (a) N_2 adsorption-desorption isotherms and BET surface area measured at 77K, and (b) corresponding pore size distribution curves for CN, CN2, and CN4.

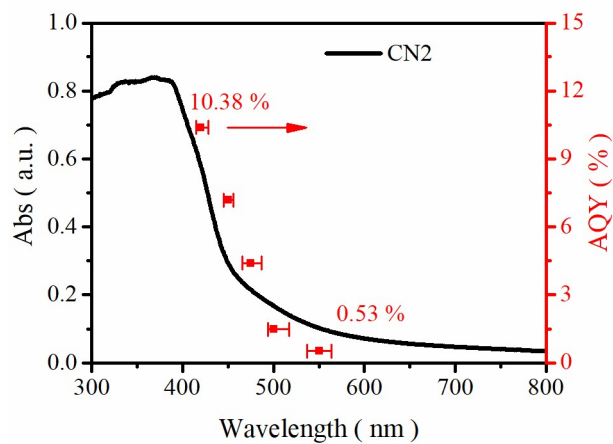


Fig. S9. Apparent quantum yields (AQYs) of CN2 under specific monochromatic light illuminating compared with the absorbance spectrum.