

Incorporation of Nonmetal Group Dopants into g-C₃N₄ Framework for Highly Improved Photocatalytic H₂ Production

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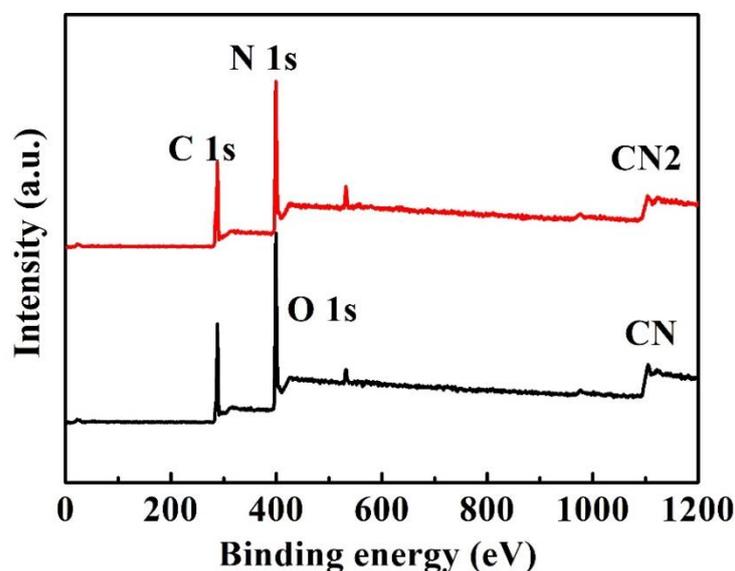


Figure S1. XPS survey spectra of CN and CN-2.



Figure S2. Digital images of CN, CN-1, CN-2 and CN-3.

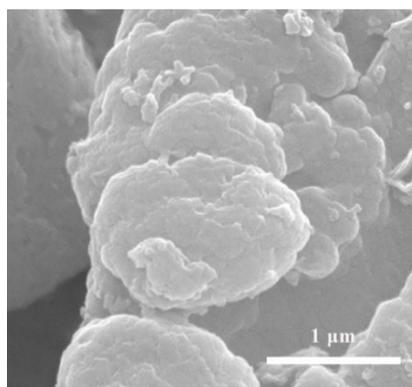


Figure S3. SEM image of CN.

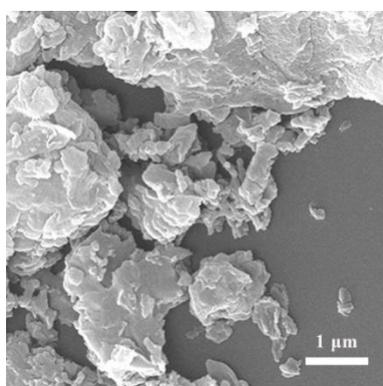


Figure S4. SEM image of CN-1.

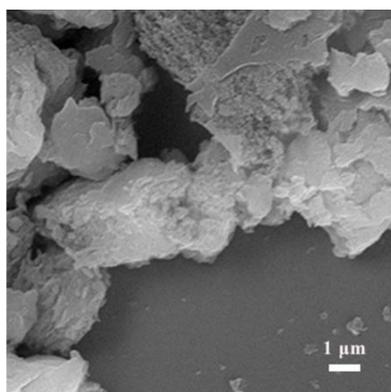


Figure S5. SEM image of CN-2.

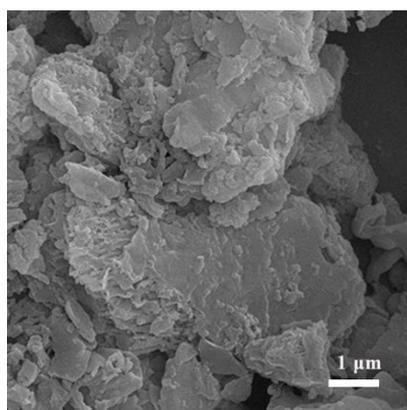


Figure S6. SEM image of CN-3.

Table S1. Elemental analysis of C and N content (wt.%) in CN, CN-1, CN-2 and CN-3.

Sample	N wt.%	C wt.%	C/N
CN	55.65	42.40	0.76
CN-1	53.59	43.32	0.81
CN-2	54.06	44.53	0.82
CN-3	53.81	44.47	0.83

Table S2. BET surface area and H₂ production rate of CN and CN-2 samples.

Sample	BET surface area (m ² g ⁻¹)	H ₂ production rate (μmol h ⁻¹ g ⁻¹)
CN	5.26	101.2
CN-2	22.74	830

Table 3. Hydrogen evolution of CN-2 and comparison with other reported g-C₃N₄ photocatalyst.

Sample	Light	H ₂ evolution (μmol h ⁻¹)	Ref.
CN-2	λ >400 nm	830	This work
g-C ₃ N ₄ nanosheets	λ >420 nm	230	[1]
g-C ₃ N ₄ /C ₆₀	λ >420 nm	266	[2]
PAN/ g-C ₃ N ₄	λ >400 nm	370	[3]
S doped g-C ₃ N ₄	λ >420 nm	525	[4]
C bridged g-C ₃ N ₄	λ >420 nm	529	[5]
Cl doped g-C ₃ N ₄	λ >420 nm	537	[6]

References

- Xu, J.; Zhang, L. W. Shi, R.; Zhu, Y. F. Chemical exfoliation of graphitic carbon nitride for efficient heterogeneous photocatalysis. *J Mater Chem A* **2013**, *1*, 14766–14772.
- Chen, X.; Chen, H.; Guan, L. J.; Zhen, J. M.; Sun, Z. J.; Du, P. W.; Lu, Y. L.; Yang, S. F. A facile mechanochemical route to covalently bonded graphitic carbon nitride (g-C₃N₄) and fullerene hybrid toward enhanced visible light photocatalytic hydrogen production. *Nanoscale* **2017**, *9*, 5615–5623.
- He, F.; Chen, G.; Yu, Y. G.; Hao, S.; Zhou, Y. S.; Zheng, Y. Facile approach to synthesize g-PAN/g-C₃N₄ composites with enhanced photocatalytic H₂ evolution activity. *Acs Appl Mater Inter* **2014**, *6*, 7171–7179.
- Yang, C.; Zhang, S. S.; Huang, Y.; Lv, K. L.; Fang, S.; Wu, X. F.; Li, Q.; Fan, J. J. Sharply increasing the visible photoreactivity of g-C₃N₄ by breaking the intralayered hydrogen bonds. *Appl Surf Sci* **2020**, *505*, 144654.

5. Li, H.L.; Li, F.P.; Wang, Z.Y.; Jiao, Y.C.; Liu, Y.Y.; Wang, P.X.; Zhang, Y.; Qin, X.Y.; Dai, Y.; Huang, B.B. Fabrication of carbon bridged g-C₃N₄ through supramolecular self-assembly for enhanced photocatalytic hydrogen evolution. *Appl Catal B-Environ* **2018**, *229*, 114–120.
6. Liu, C.Y.; Zhang, Y. H.; Dong, F.; Reshak, A.H.; Ye, L.Q.; Pinna, N.; Zeng, C.; Zhang, T.R.; Huang, H.W. Chlorine intercalation in graphitic carbon nitride for efficient photocatalysis. *Appl Catal B-Environ* **2017**, *203*, 465–474.