

Supporting information

Fabrication of a high-efficiency hydrogen generation photocatalysis Pd/C₃N₅-K,I through synergistic effects of planar and spatial carrier separation

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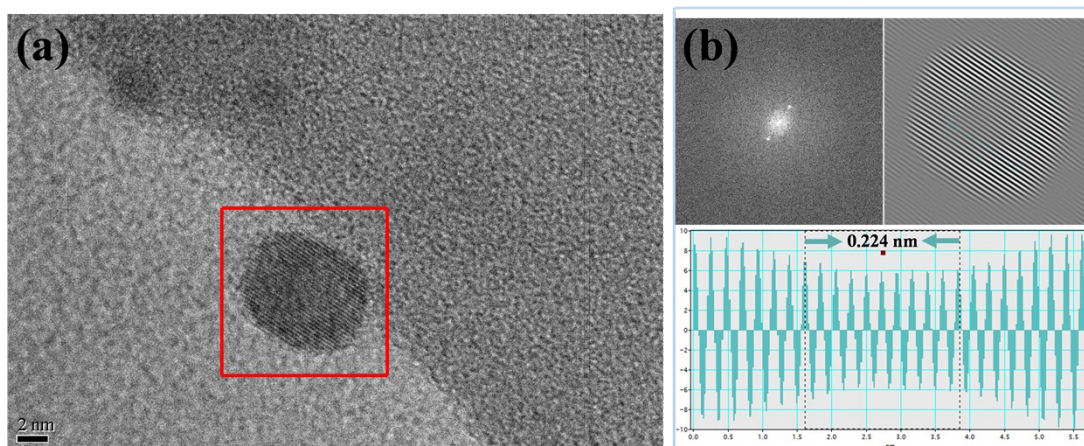


Figure S1. lattice spacing of Pd nanoparticles.

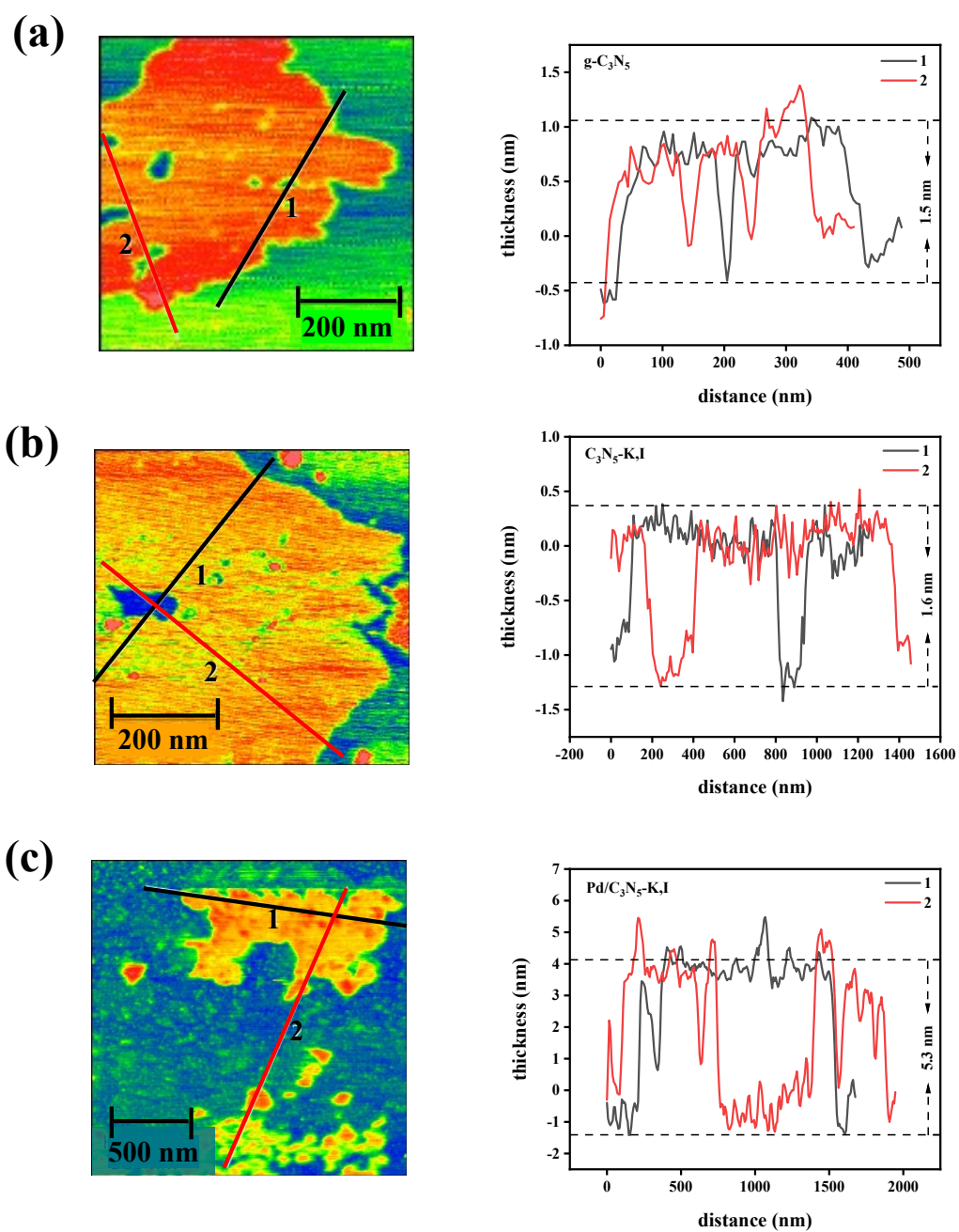


Figure S2. AFM images of (a) g-C₃N₅, (b) C₃N₅-K,I, and (c) Pd/C₃N₅-K,I

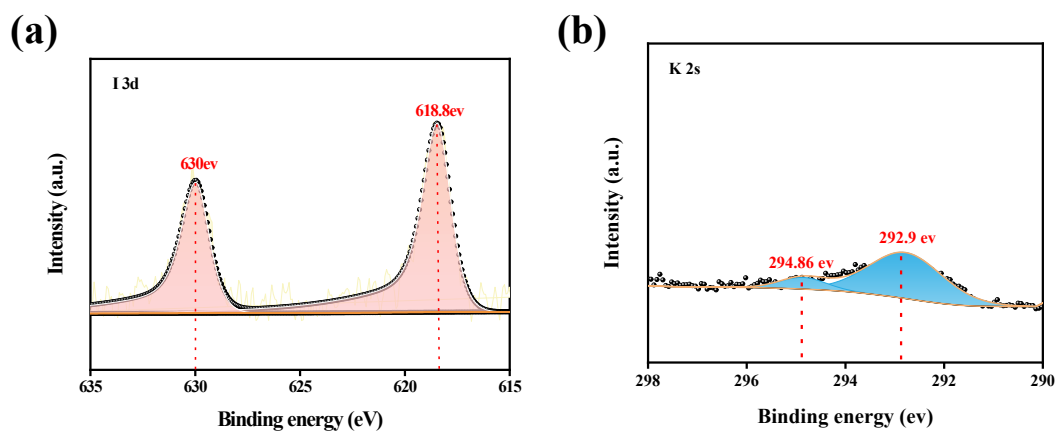


Figure S3. (a) XPS I 3d spectrum and (b) XPS K 2s spectrum of Pd/C₃N₅-K,I.

The K 2s peaks of Pd/C₃N₅-K,I are located at around 292.9 and 294.86 eV, demonstrating that K ions doped into Pd/C₃N₅-K,I.

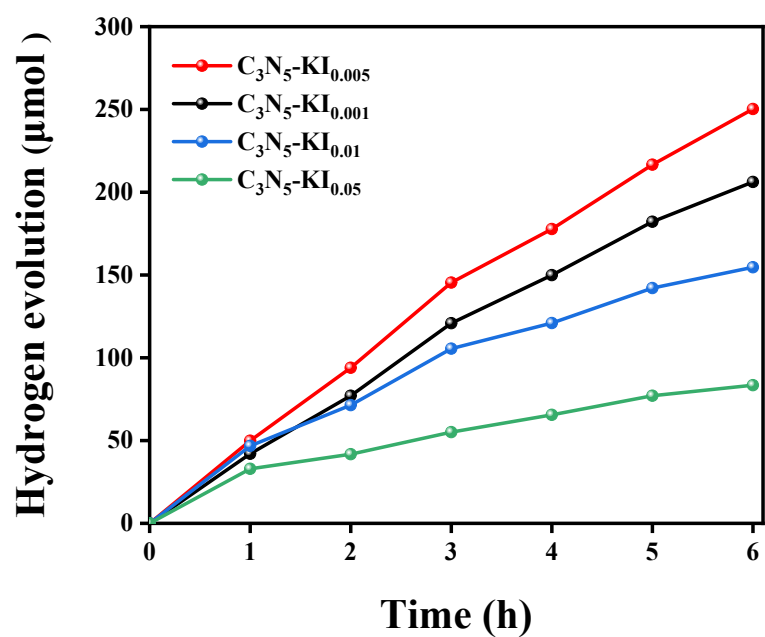


Figure S4. Photocatalytic H₂ evolution amounts of C₃N₅-K,I at different KI-to-C₃N₅ molar ratios.

The C₃N₅-K,I_{0.005} (200 mg 3-amino-1,2,4-triazole and 60mg KI) shows the best H₂-evolution activity (250.3 μmol), which is used to synthesize Pd/C₃N₅-K,I.

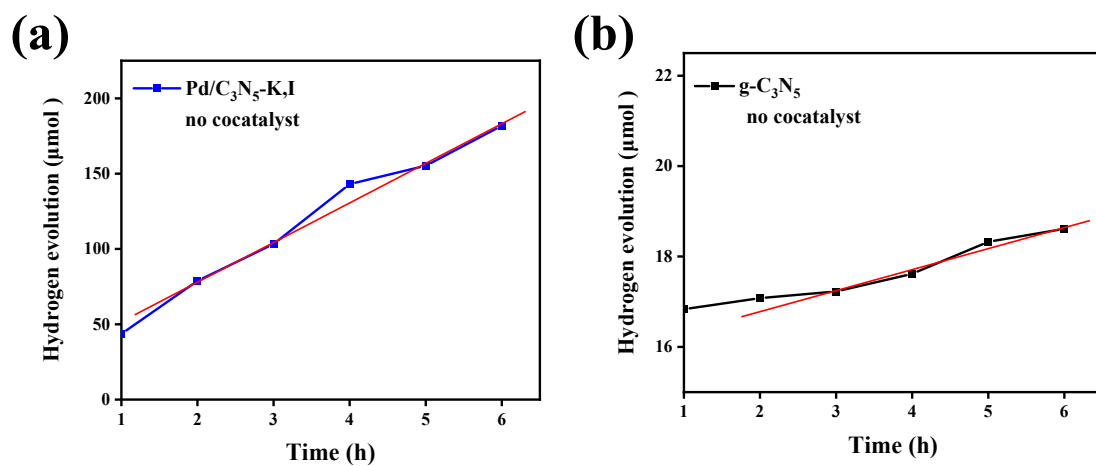


Figure S5. H_2 generation amounts for the $Pd/C_3N_5-K,I$ (a), and $g-C_3N_5$ (b) without Pt.

The $Pd/C_3N_5-K,I$ and $g-C_3N_5$ without Pt exhibit a partially linear growth trend in H_2 production.

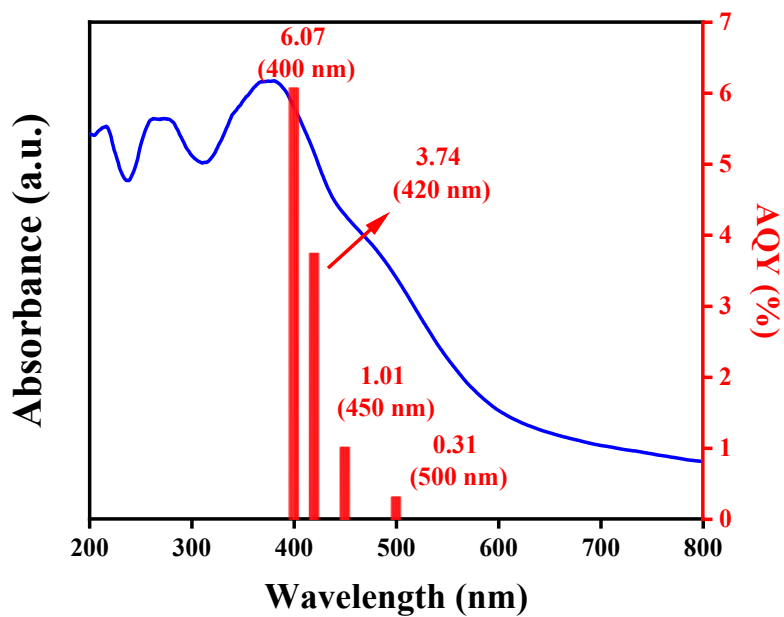


Figure S6. Wavelength dependence of AQY on H₂ evolution performance of Pd/C₃N₅-K,I.

The AQY of Pd/C₃N₅-K,I is 6.07% (400 nm), 3.74% (420 nm), 1.01% (450 nm) and 0.31% (500 nm), respectively, indicating a good correlation with the light absorption properties.

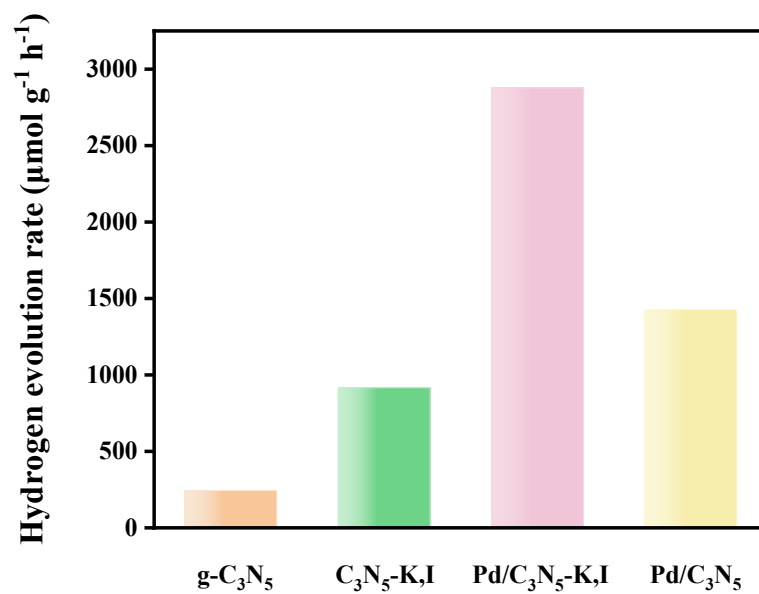
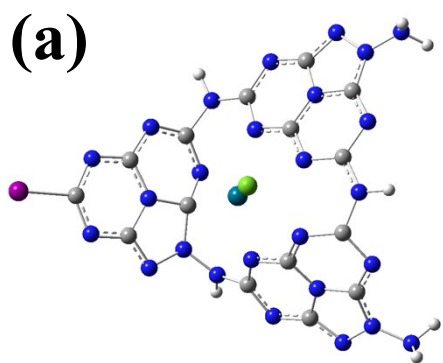
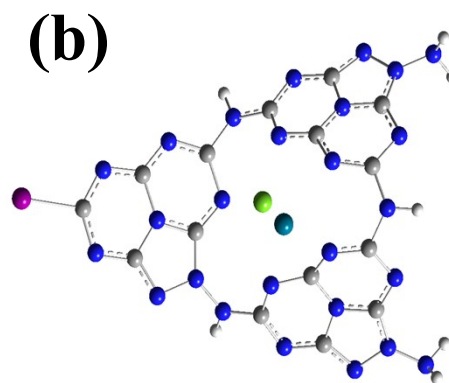


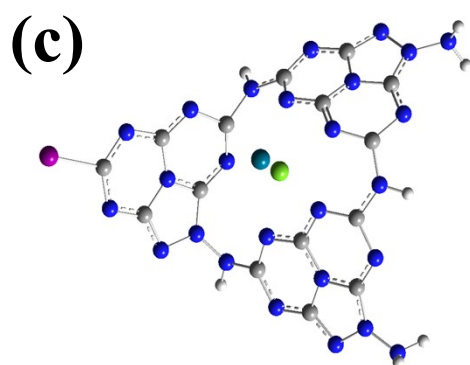
Figure S7. Photocatalytic H₂ evolution rates of g-C₃N₅, C₃N₅-K,I, Pd/C₃N₅ and Pd/C₃N₅-K,I.



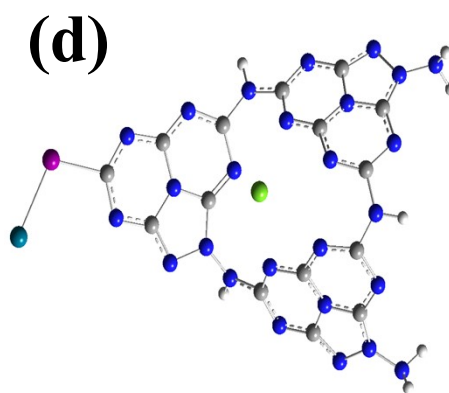
$$E = -9647.33092617 \text{ ev}$$



$$E = -9647.32674931 \text{ ev}$$



$$E = -9647.32110571 \text{ ev}$$



$$E = -9647.31835398 \text{ ev}$$

Figure S8. Structural energies of the Pd atoms at different positions of the CN. Gray, blue, white, bright green, purple and blackish green represent C, N, H, K, I and Pd, respectively.

Table S1. Specific BET surface area and pore volume of g-C₃N₅, C₃N₅-K,I and Pd/C₃N₅-K,I.

Sample	S_{BET}(m² g⁻¹)	V_p(cm³ g⁻¹)
g-C₃N₅	2.9438	0.0161
C₃N₅-K,I	6.1582	0.0277
Pd/C₃N₅-K,I	6.4064	0.0275

Table S2. Elemental composition of 2.25% Pd/C₃N₅-K,I obtained from ICP analysis.

Catalyst	I(wt%)	Pd(wt%)	K(wt%)
2.25% Pd/C₃N₅-K,I	1.07	2.25	0.37

Table S3. Comparison of photocatalytic H₂ evolution of Pd/C₃N₅-K,I with literature reported g-C₃N₅-based catalysts.

Catalysts	Pt Cocatalyst	H ₂ evolution rate (μmol g ⁻¹ h ⁻¹)	AQY (%)	Year	Ref.
mesoporous S-C ₃ N ₅	3 wt%	1370	/	2024	[1]
C ₃ N ₅ /Ti ₃ C ₂	1 wt%	2581.23	3.81 (420 nm)	2024	[2]
NiO/C ₃ N ₅	/	112.2	5.94 (400 nm)	2024	[3]
S-C ₃ N ₅	3 wt%	486	/	2023	[4]
C ₃ N ₅ /TiO ₂	/	1833.86	1.93 (420 nm)	2023	[5]
Pt SA/C ₃ N ₅	0.5 wt%	1776.8	0.47 (420 nm)	2023	[6]
CdS/g-C ₃ N ₅	3 wt%	502.11	/	2023	[7]
g-C ₃ N ₅ /poly	1 wt%	2326.8	2.25 (420 nm)	2022	[8]
NH ₂ -UiO-66/C ₃ N ₅	2 wt%	3936	6.78 (420 nm)	2022	[9]
C ₃ N ₅ nanosheets	/	28.97	/	2020	[10]
Pd/C ₃ N ₅ -K,I	0.36 wt%	2878	6.07 (400 nm) 3.74 (420 nm)	This work	

References

- [1] X. Guan, X. Zhang, Z. Li, S. Deshpande, M. Fawaz, N.P. Dharmarajan, C.-H. Lin, Z. Lei, L. Hu, J.-K. Huang, P. Kumar, Z. Sun, S. Chakraborty, A. Vinu, *Chem. Mater.*, 2024, 36, 4511-4520.
- [2] P. Yu, S. Chen, Y. Wang, J. Li, Z. Zhang, S. Zhao, Y. Zhang, Y. Zhou, *Int. J. Hydrogen Energy*, 2024, 58, 1266-1276.
- [3] J. Hu, J. Li, Z. Pu, W. Xiao, H. Yu, Z. Zhang, F. Yu, C. Liu, Q. Zhang, *J. Colloid Interface Sci.*, 2024, 665, 780-792.
- [4] X. Guan, M. Fawaz, R. Sarkar, C.-H. Lin, Z. Li, Z. Lei, P.D. Nithinraj, P. Kumar, X. Zhang, J.-H. Yang, L. Hu, T. Wu, S. Chakraborty, J. Yi, A. Vinu, *J. Mater. Chem. A*, 2023, 11, 12837-1284.
- [5] S. Chen, X. Sheng, Y. Wang, P. Yu, C. Yang, X. Zhang, H. Fang, Y. Zhang, Y. Zhou, *Appl. Surf. Sci.*, 2024, 643, 158600.
- [6] X. Du, Q. Liu, M. Cheng, R. Wang, J. Hu, T. Wei, Y. Ling, Y. Cui, W. Li, B. Liu, *Int. J. Hydrogen Energy*, 2024, 53, 353-363.
- [7] H.-T. Vuong, D.-V. Nguyen, L.P. Phuong, P.P.D. Minh, B.N. Ho, H.A. Nguyen, *Carbon Neutralization*, 2023, 2, 425-457.
- [8] Z. Xiong, Y. Liang, J. Yang, G. Yang, J. Jia, K. Sa, X. Zhang, Z. Zeng, *Sep. Purif. Technol.*, 2023, 306, 122522.
- [9] B. Wu, T. Sun, N. Liu, L. Lu, R. Zhang, W. Shi, P. Cheng, *ACS Appl. Mater. Interfaces*, 2022, 14, 26742-26751.
- [10] T. Liu, G. Yang, W. Wang, C. Wang, M. Wang, X. Sun, P. Xu, J. Zhang, *Environ. Res.*, 2020, 188, 109741.