

## Supporting Information

# Three-dimensional Assembly of Carbon Nitride Tubes as Nanoreactors for Enhanced Photocatalytic Hydrogen Production

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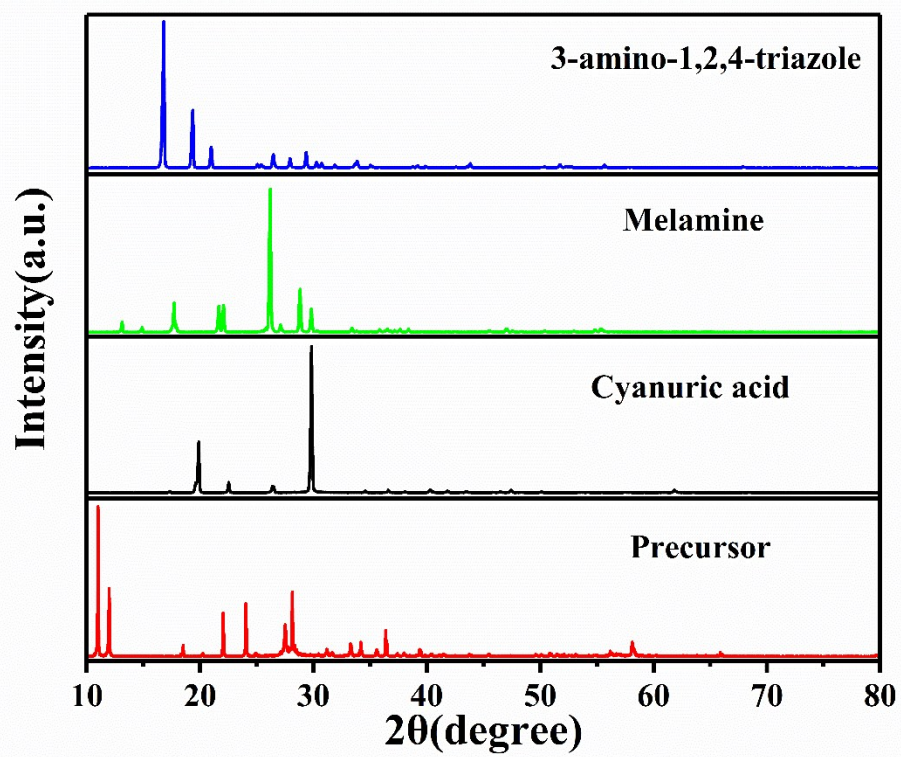
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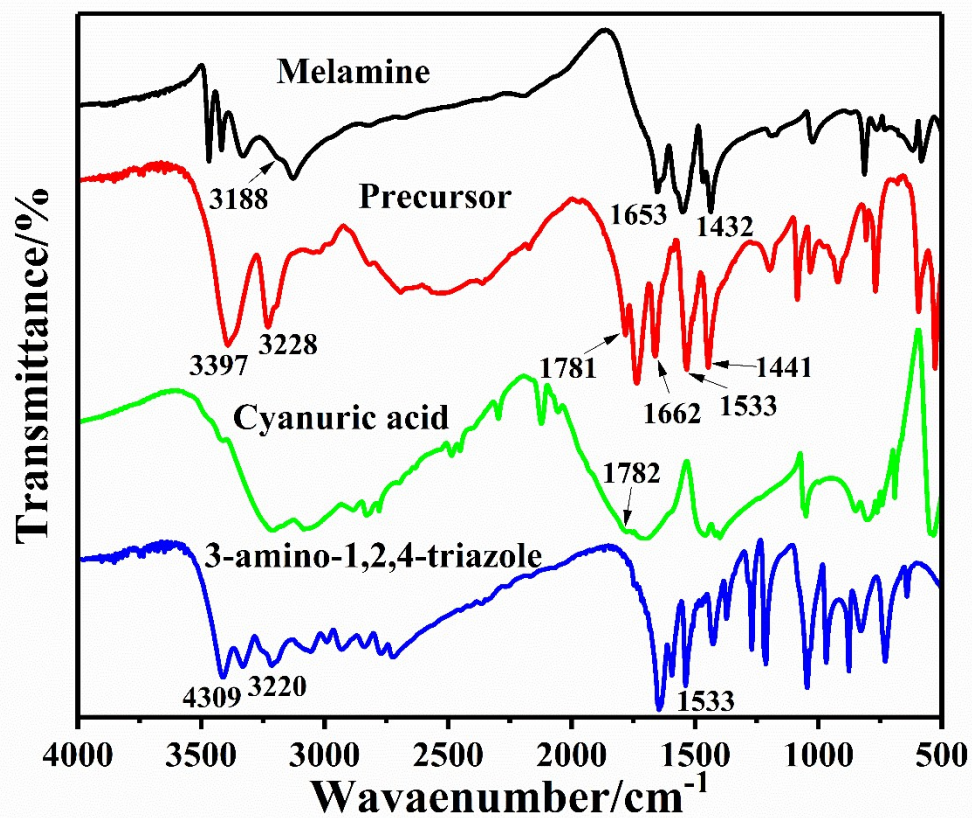
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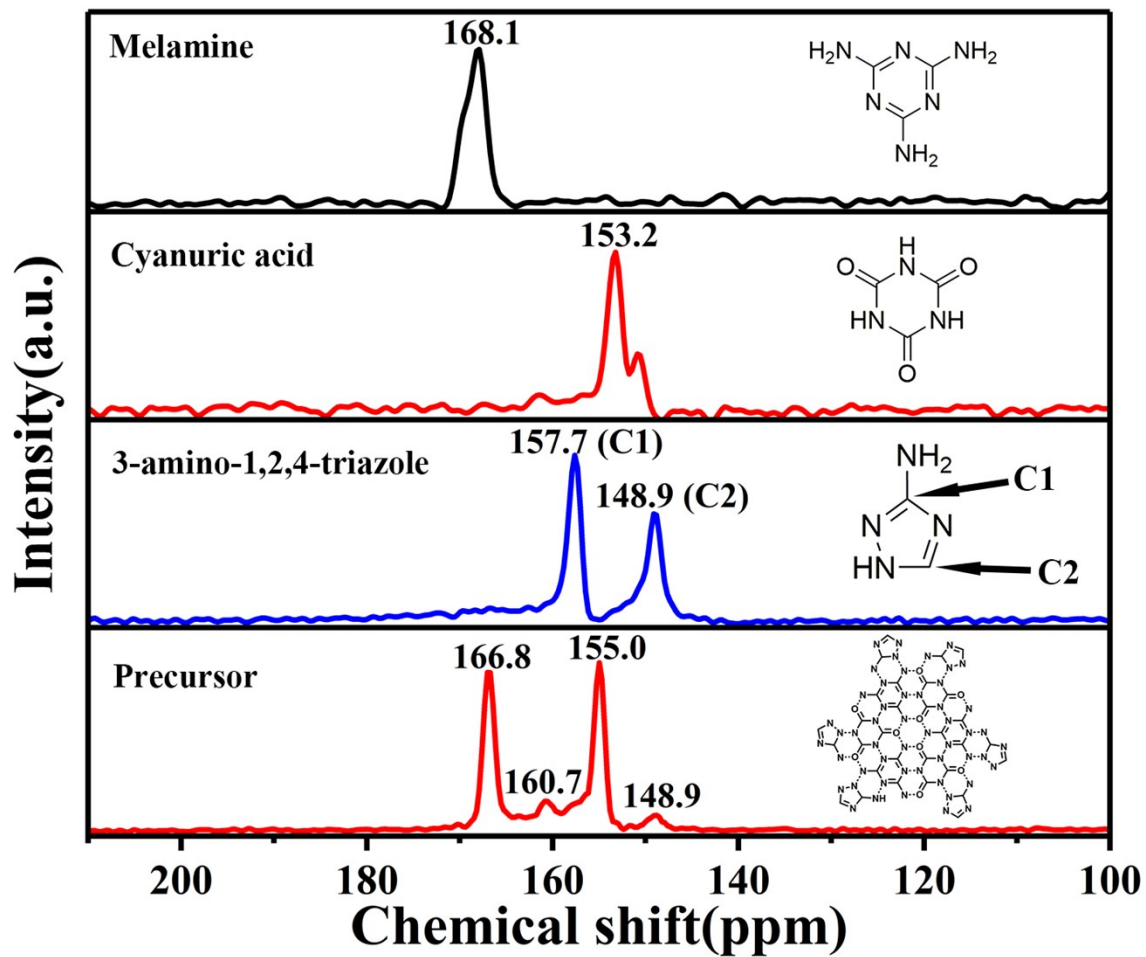
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**Figure S1.** XRD pattern of precursor (red line), commercial cyanuric acid (black line), commercial melamine (green line) and commercial 3-amino-1,2,4-triazole (blue line).



**Figure S2.** The FT-IR spectra of commercial melamine (black line), the precursor (red line), commercial cyanuric acid (green line) and commercial 3-amino-1,2,4-triazole (AT, blue line).



**Figure S3.** Solid-state  $^{13}\text{C}$  MAS NMR spectra of melamine, cyanuric acid 3-amino-1, 2, 4-triazole and precursor.

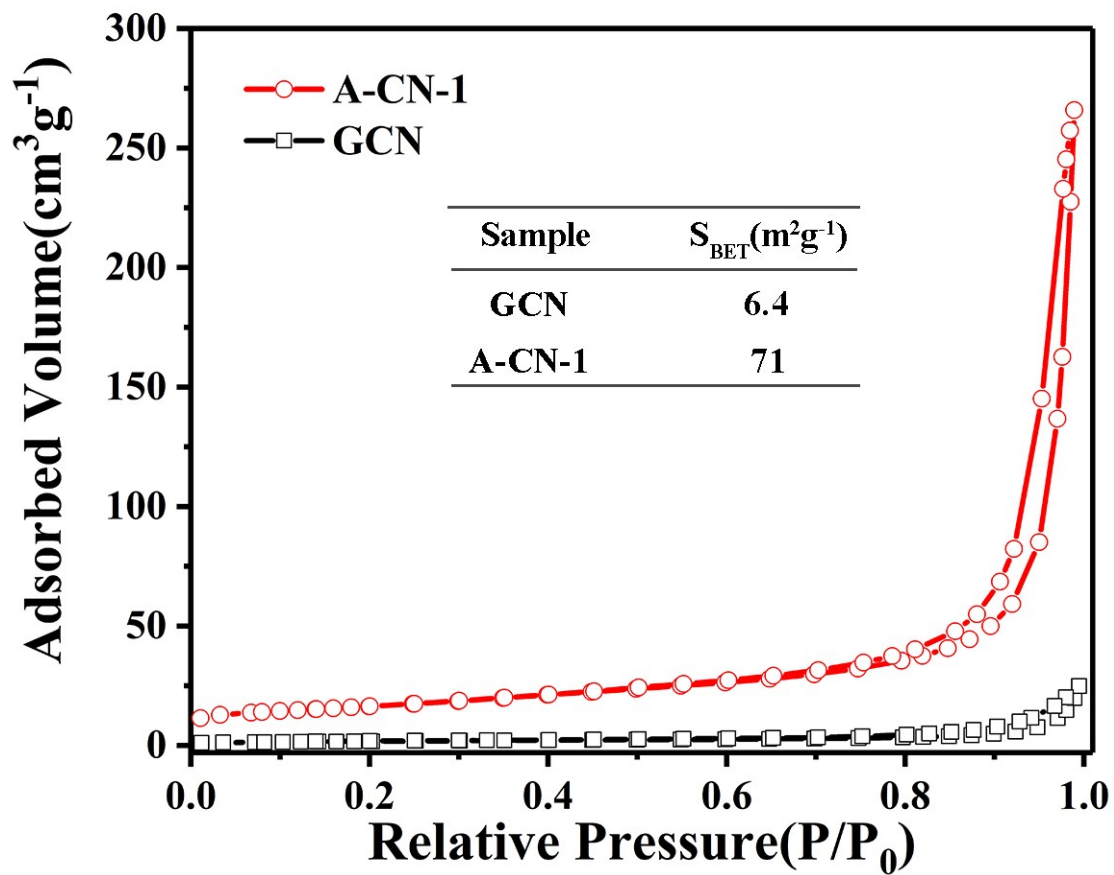


Figure S4. N<sub>2</sub>-sorption isotherms of GCN and A-CN-1.

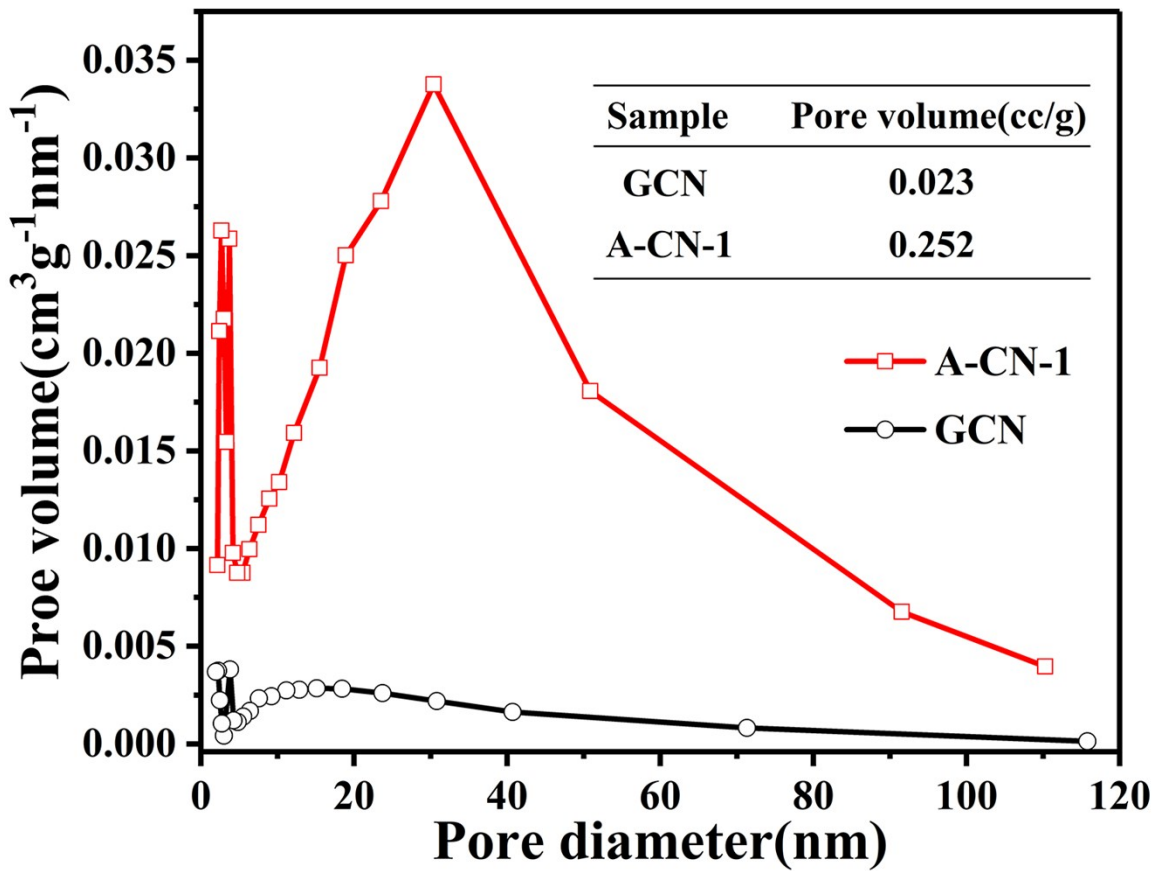
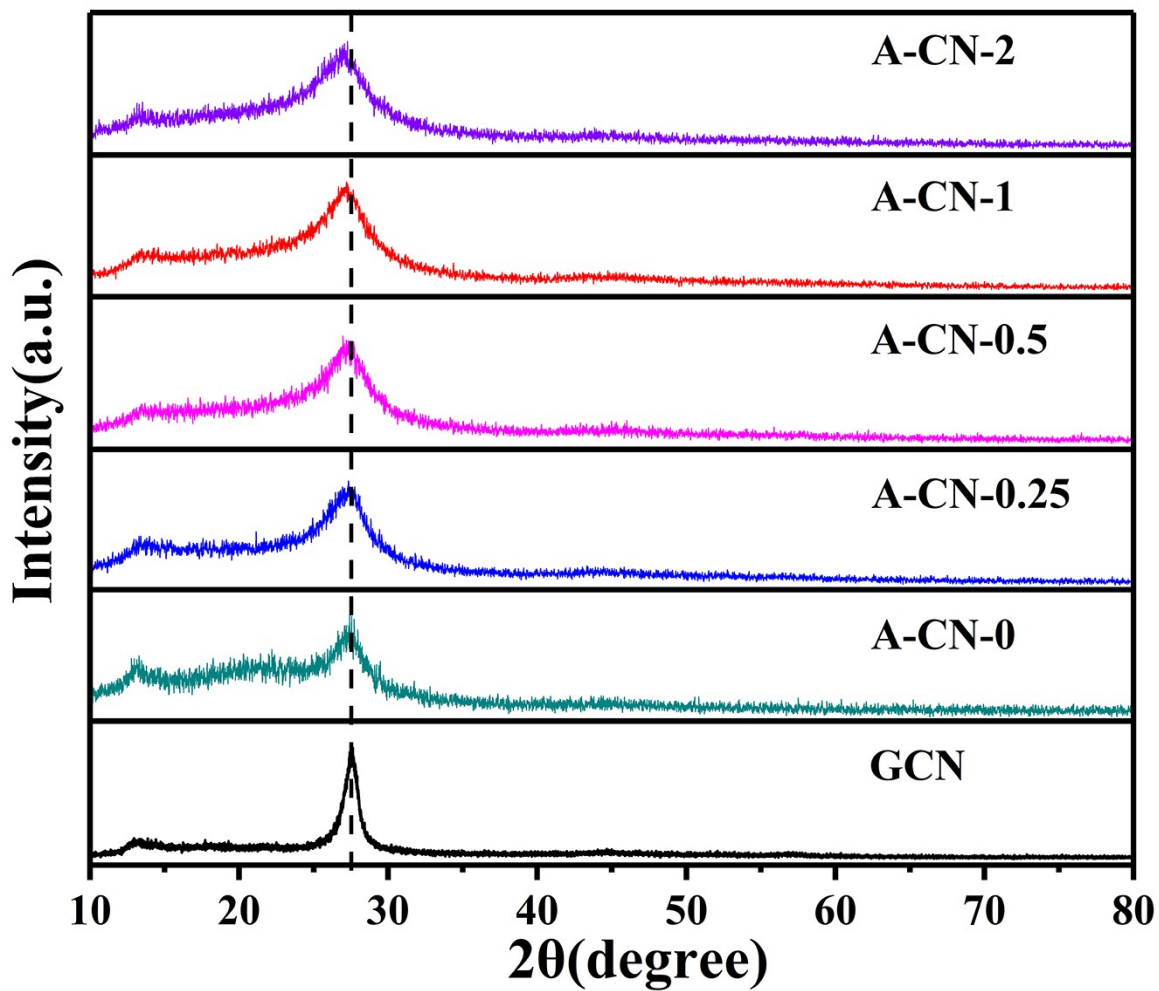


Figure S5. The pore size distribution of GCN and A-CN-1.



**Figure S6.** XRD pattern of serials of products with different proportions 3-amino-1, 2, 4-triazole and reference sample GCN.

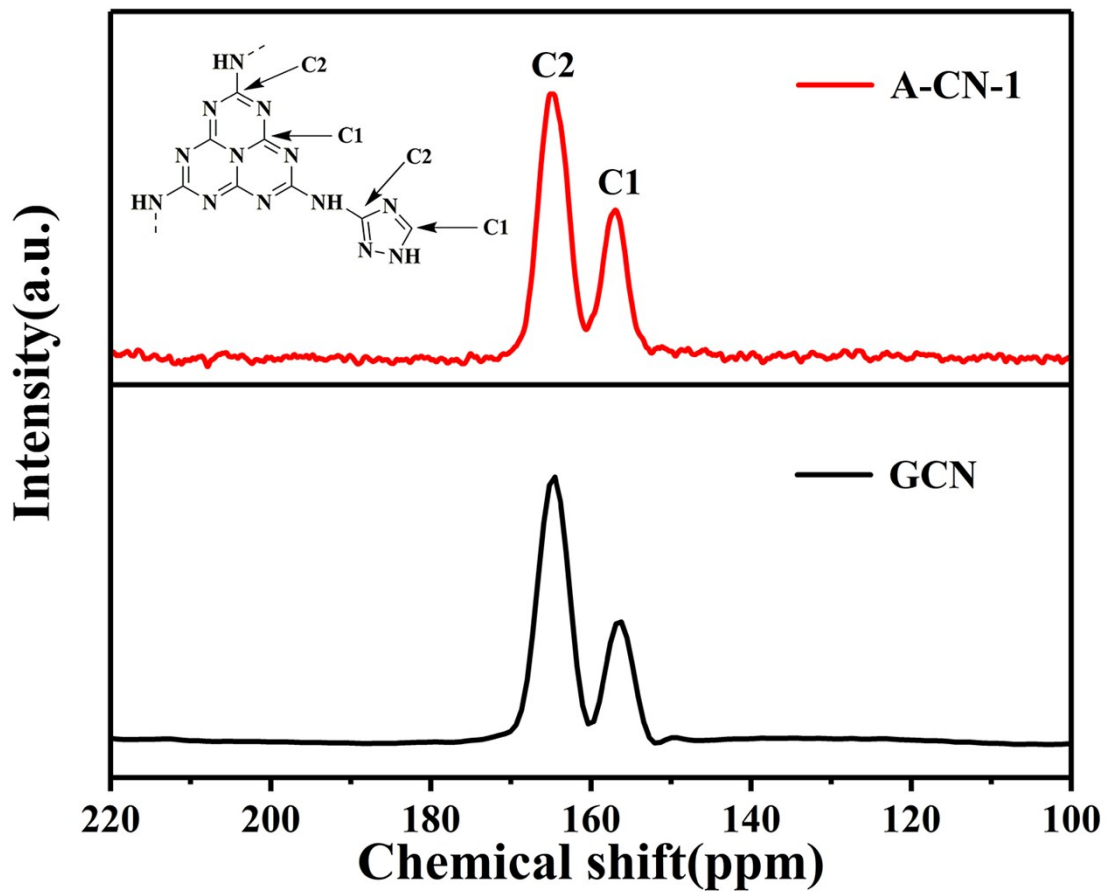


Figure S7. Solid-state  $^{13}\text{C}$  MAS NMR spectra of GCN and A-CN-1.



**Table S1.** XPS analysis of GCN and A-CN-1

<b>Sample</b>	<b>C (Atom conc%)</b>	<b>N (Atom conc%)</b>	<b>P (Atom conc%)</b>	<b>N/C</b>
GCN	41.12	55.46	0	1.35
A-CN-1	40.45	58.88	0	1.46

**Table S2.** Organic elemental analysis of GCN and A-CN-1.

<b>Sample</b>	<b>C (wt%)</b>	<b>N (wt%)</b>	<b>N/C</b>
GCN	35.12	56.34	1.38
A-CN-1	34.11	58.98	1.48

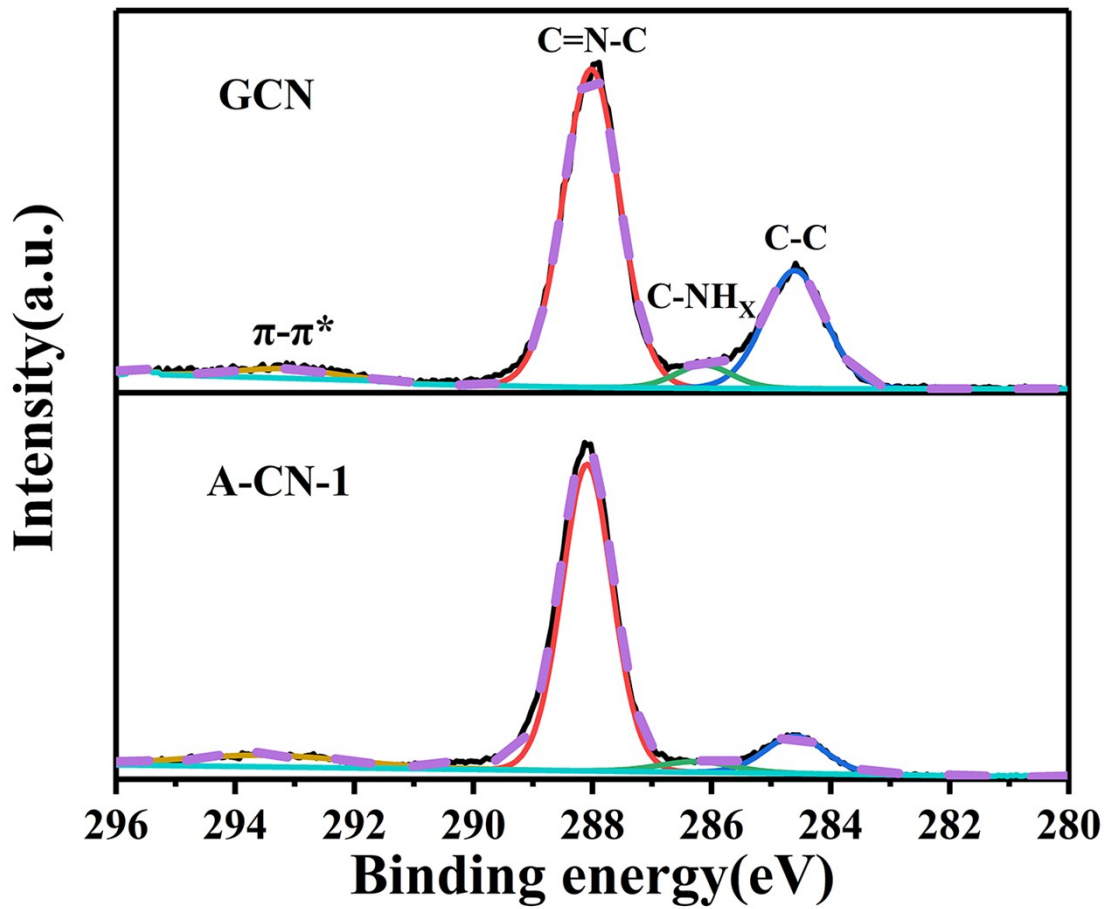


Figure S8. C 1s XPS spectra of GCN and A-CN-1.

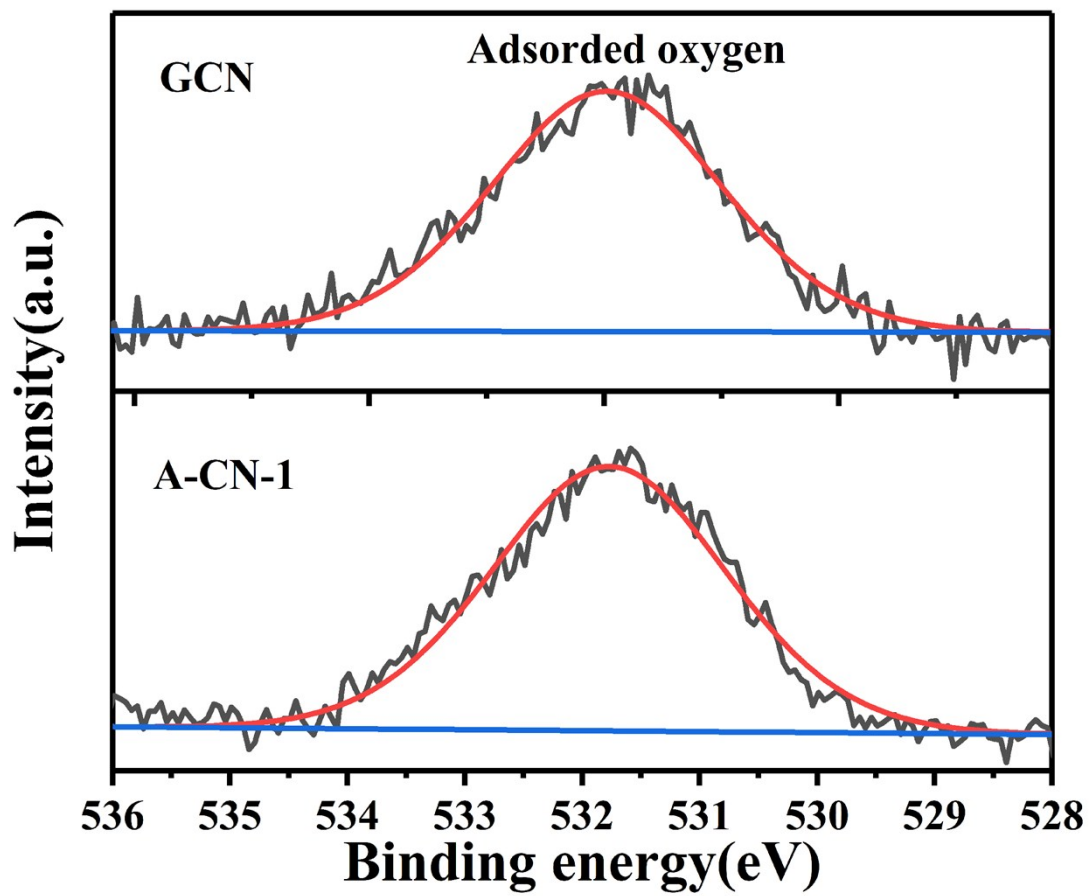


Figure S9. O 1s XPS spectra of GCN and A-CN-1.

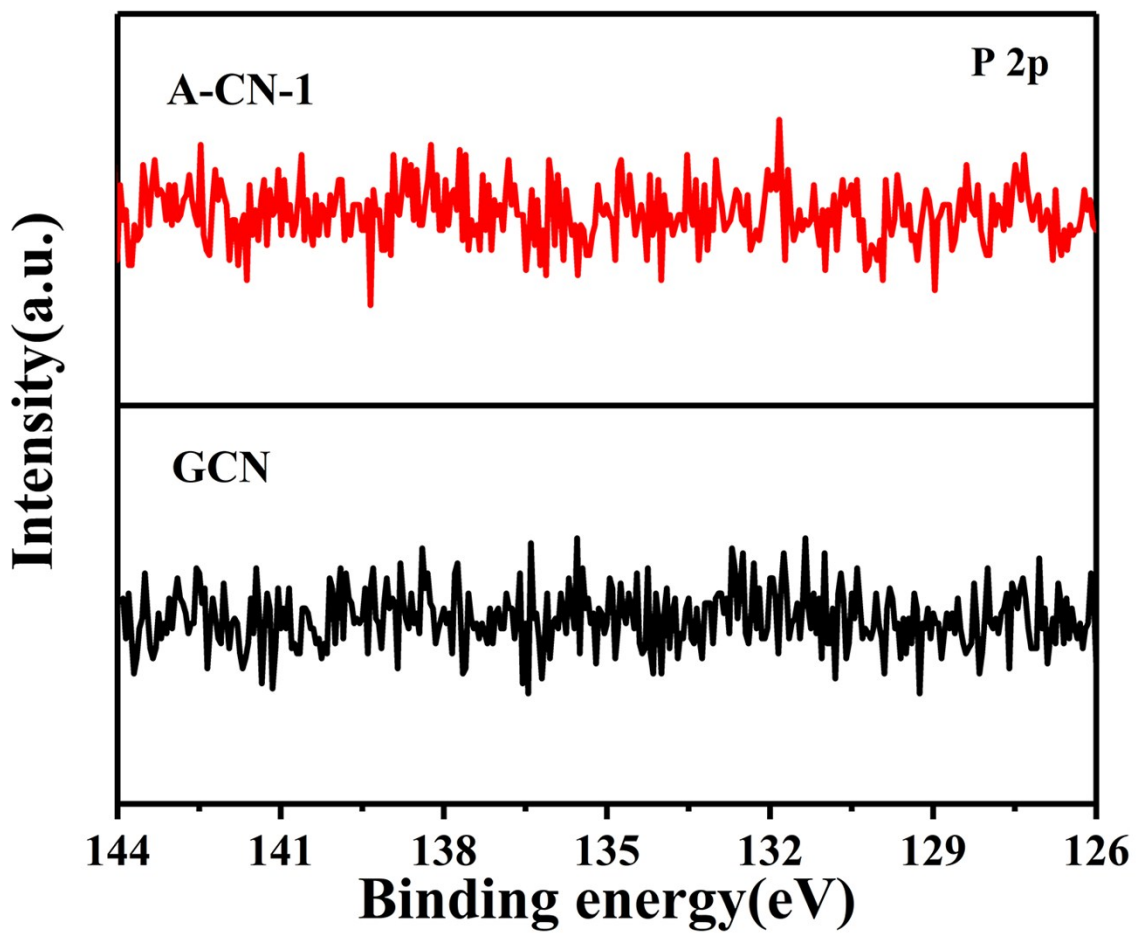


Figure S10. P 2p XPS spectra of GCN and A-CN-1.

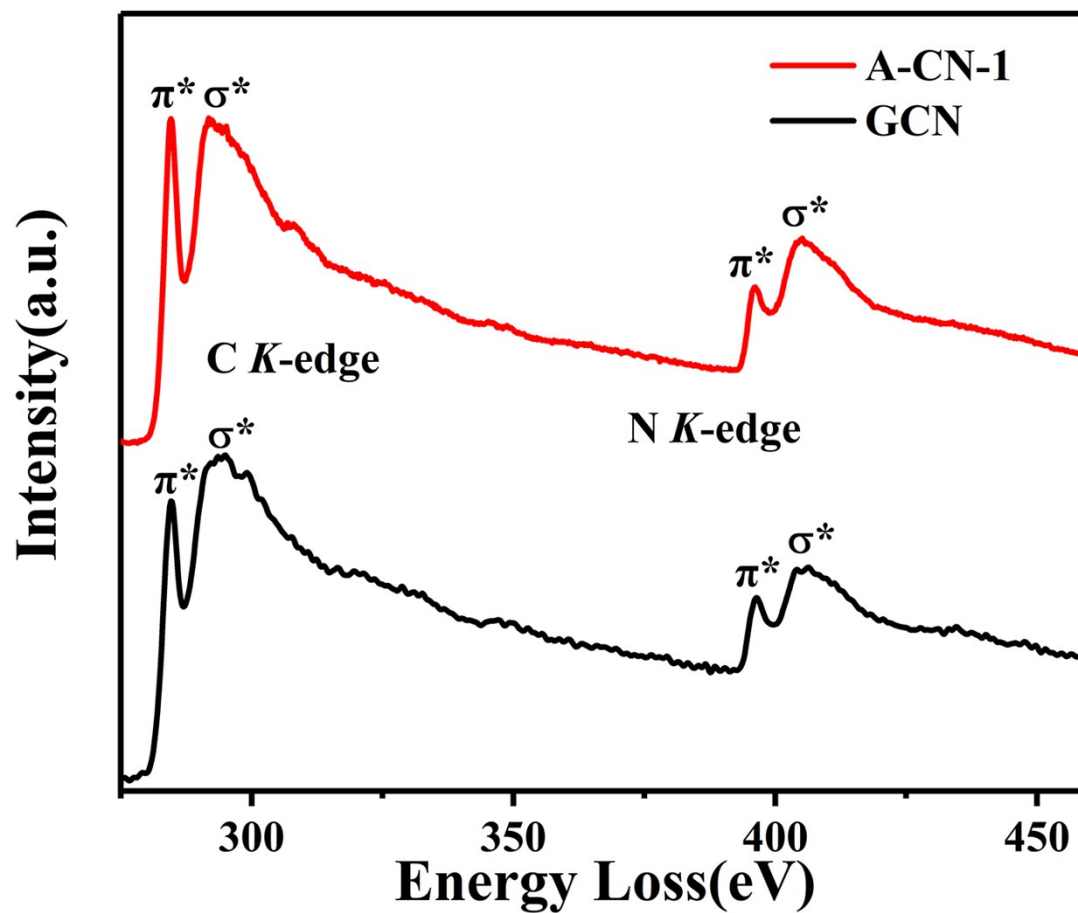
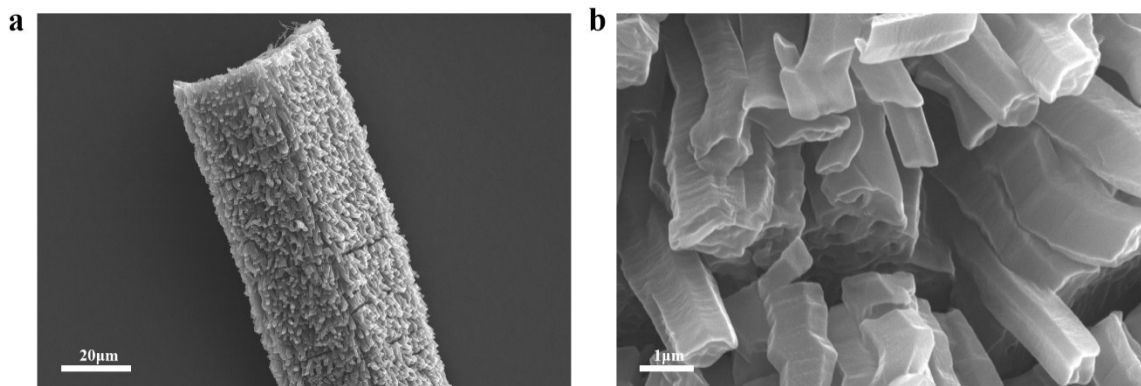


Figure S11. EEL spectrum of GCN and A-CN-1.



**Figure S12.** SEM images (a, b) of A-CN-1 after photocatalytic hydrogen production process.

**Table S3.** Kinetic analysis of emission decay for GCN and A-CN-1.

<b>Sample</b>	<b><math>\tau_1</math>(ns)</b>	<b>Rel.%</b>	<b><math>\tau_2</math>(ns)</b>	<b>Rel.%</b>	<b><math>\tau_3</math>(ns)</b>	<b>Rel.%</b>
GCN	1.47	31.62	5.76	47.68	32.75	20.70
A-CN-1	1.10	29.99	5.01	46.17	27.88	23.84



**Table S4.** Summary of C<sub>3</sub>N<sub>4</sub>-base nanostructures photocatalysts reported for hydrogen evolution rate.

Catalyst	Hydrogen evolution rate (μmol/h)	Hydrogen evolution rate (μmol/h/g)	Experimental conditions	Light source filter (nm)	BET(m <sup>2</sup> g <sup>-1</sup> )	Reference
A-CN-1	71 (10mg)	7100	1wt% Pt 20% methanol	AM 1.5	71	This work
g-C <sub>3</sub> N <sub>4</sub> /KNbO <sub>3</sub> (1:1)	101.9 (100mg)	1019.38	2 wt% Pt 20%TEOA	AM 1.5	--	1
UCN	47.2 (80mg)	590	3 wt% Pt 10% TEOA	AM 1.5	69.6	2
KSCN-treated Melon	24.7 (20mg)	1235	8 wt% Pt 20% methanol	AM 1.5	55	3
Urea-CN <sub>x</sub>	56.2 (20mg)	2810	2 wt% Pt 10% methanol	AM 1.5	64.6	4
C <sub>3</sub> N <sub>4</sub> -Ni-Tu-TETN	51 (100mg)	510	0.05 mmol Ni <sup>2+</sup> 3 mmol Tu 22% TEOA	AM 1.5	--	5
CNHS	57.4 (20mg)	2860	3 wt% Pt 10% TEOA	>400	277.89	6
PCN-S	79.8 (50mg)	1596	1 wt% Pt 20% TEOA	>400	122.6	7
P-TCN	67 (100mg)	670	1wt% Pt 20% methanol	>420	22.9	8
R-CN-350	19 (10mg)	1900	3wt% Pt 10% TEOA	>420	29.9	9
CN-NaK	278 (50mg)	5560	3wt% Pt 10% TEOA	>420	74	10

**Reference:**

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