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## Facile solvothermally method assisted g-C<sub>3</sub>N<sub>4</sub> post-grafting with aromatic amine dyes for highly photocatalytic hydrogen evolution

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Fig. S1 The TEM images of samples (a) g-C<sub>3</sub>N<sub>4</sub>, (b) g-C<sub>3</sub>N<sub>4</sub>/TPA-CNCHO, (c) g-C<sub>3</sub>N<sub>4</sub>/PTZ-CNCHO, (d) g-C<sub>3</sub>N<sub>4</sub>/CZ-CNCHO.



Fig. S2 The element map of g-C<sub>3</sub>N<sub>4</sub>/TPA-CNCHO (C (green), N (purple), S (yellow)).



Fig. S3 The N<sub>2</sub> adsorption desorption isotherms and pore size distribution of samples (a) g-C<sub>3</sub>N<sub>4</sub>, (b) g-C<sub>3</sub>N<sub>4</sub>/TPA-CNCHO, (c) g-C<sub>3</sub>N<sub>4</sub>/PTZ-CNCHO and (d) g-C<sub>3</sub>N<sub>4</sub>/CZ-CNCHO.

Table S1. The surface area and pore diameter of g-C<sub>3</sub>N<sub>4</sub>, g-C<sub>3</sub>N<sub>4</sub>/TPA-CNCHO, g-C<sub>3</sub>N<sub>4</sub>-PTZ-CNCHO and g-C<sub>3</sub>N<sub>4</sub>-CZ-CNCHO.

Sample	$S_{BET}(m^2/g)$	Pore diameter <sub>-max</sub> (nm)
g-C <sub>3</sub> N <sub>4</sub>	95.0	34.9
g-C <sub>3</sub> N <sub>4</sub> -TPA-CNCHO	82.5	35.4
g-C <sub>3</sub> N <sub>4</sub> -PTZ-CNCHO	86.6	36.1
g-C <sub>3</sub> N <sub>4</sub> -CZ-CNCHO	88.1	35.9



Fig. S4 XPS spectra of C1s of g-C<sub>3</sub>N<sub>4</sub>(a) and g-C<sub>3</sub>N<sub>4</sub>/PTZ-CNCHO (b), N1s of g-C<sub>3</sub>N<sub>4</sub>(c) and N1s of g-C<sub>3</sub>N<sub>4</sub>/PTZ-CNCHO (d)



Fig. S5 Optical photos of relevant samples.



Fig. S6 LSV curves of g-C<sub>3</sub>N<sub>4</sub>, g-C<sub>3</sub>N<sub>4</sub>/TPA-CNCHO, g-C<sub>3</sub>N<sub>4</sub>/PTZ-CNCHO and g-C<sub>3</sub>N<sub>4</sub>/CZ-CNCHO.



Fig. S7 The WCA of g-C<sub>3</sub>N<sub>4</sub> (a), g-C<sub>3</sub>N<sub>4</sub>/TPA-CNCHO (b), g-C<sub>3</sub>N<sub>4</sub>/PTZ-CNCHO (c) and g-C<sub>3</sub>N<sub>4</sub>/CZ-CNCHO (d).



 $Fig. \ S8 \ The \ photocatalytic \ hydrogen \ evolution \ rate \ of \ g-C_3N_4, \ g-C_3N_4/TPA-CNCHO \ and \ g-C_3N_4-TPA-CNCHO.$ 

Photocatalyst	Modification method	Modification conditions	Photocatalytic Reaction conditions	H <sub>2</sub> production activity (n times of g-C <sub>3</sub> N <sub>4</sub> )	Other photocatalytic activity	Ref.
		insert gas				
melamine /		PPL lined	triethanolamine	58.1		
p-benzaldehyde	copolymerizati	autoclave	(1EOA) $\lambda > 420 \text{ nm}$	(n=2)	none	81
	on	250°C,6h				
Urea/		550°C, 2h,	TEOA	226 $\mu$ mol·h <sup>-1</sup>		
p-benzaldehyde		5°C/min	420 nm<λ<780nm	(n=2)	none	S2
	Schiff's base					
g-C <sub>3</sub> N <sub>4</sub> /	reaction	250°C, 5h,	TEOA	92.4 μmol·h <sup>-1</sup>		62
p-benzaldehyde	(solid	5°C/min	λ≥420 nm	(n=4.1)	none	53
	medium)					
g-C3N4/ Feqpy-BA	Schiff's base reaction (liquid medium)	blue LED light 24°C	TEOA λ=460 nm	none	The number of $CO_2$ conversion is 2554 and the selectivity is 95%.	S4
g-C <sub>3</sub> N <sub>4</sub> / TPA-CNCHO g-C <sub>3</sub> N <sub>4</sub> / PTZ-CNCHO g-C <sub>3</sub> N <sub>4</sub> / CZ-CNCHO	Schiff's base reaction (liquid medium)	insert gas, DDM 240°C,15h	AA λ≥400 nm	$\begin{array}{c} 16232.7\\ \mu mol \cdot h^{-1} \cdot g^{-1}\\ (n=40)\\ 13266.9\\ \mu mol \cdot h^{-1} \cdot g^{-1}\\ (n=33)\\ 11822.8\\ \mu mol \cdot h^{-1} \cdot g^{-1}\\ (n=29)\end{array}$	none	this work

Table S2. The comparison of other dye grafted photocatalyst via Schiff's base reaction

for photocatalytic H<sub>2</sub> production or other use.



Fig. S9 DFT calculated geometry structures and electron densities of LUMOs and HOMOs of (a) TPA-CNCHO, (b) PTZ-CNCHO and (c) CZ-CNCHO at B3LYP/6-31G(d) level.

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