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Supporting Information

A gradual carbon doping graphitic carbon nitride towards the metal-free visible light photocatalytic hydrogen evolution

Zhou Chen†, Tingting Fan†, Xiang Yu†, Qiuling Wu†, Qiuhui Zhu‡, Lizhong Zhang‡, Jianhui Li†, Weiping Fang†, Xiaodong Yi†*

† National Engineering Laboratory for Green Chemical Productions of Alcohols-ethers-esters, College of Chemistry and Chemical Engineering, Xiamen University, Fujian 361005, P. R. China

‡ Department of Chemistry and Applied Chemistry, Changji University, Changji 831100, P. R. China.

*Corresponding author: xdyi@xmu.edu.cn.

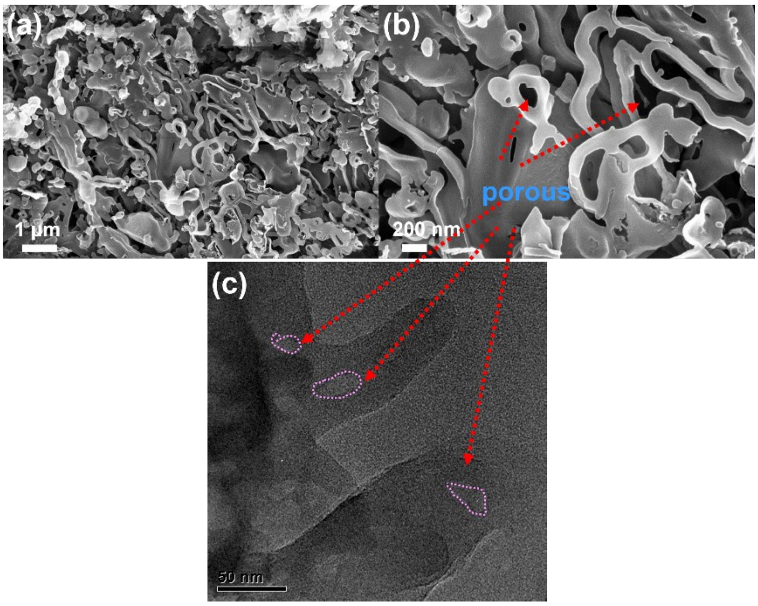

Figure S1. SEM (a, b) and TEM (c) images of C-rich g-C_3N_4.

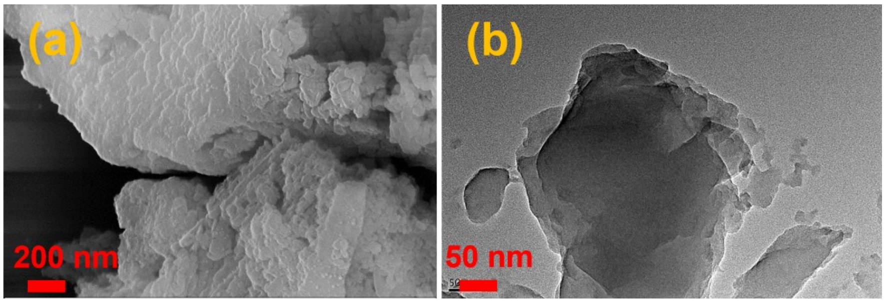

Figure S2. (a) SEM and (b) TEM images of bulk-C_3N_4.

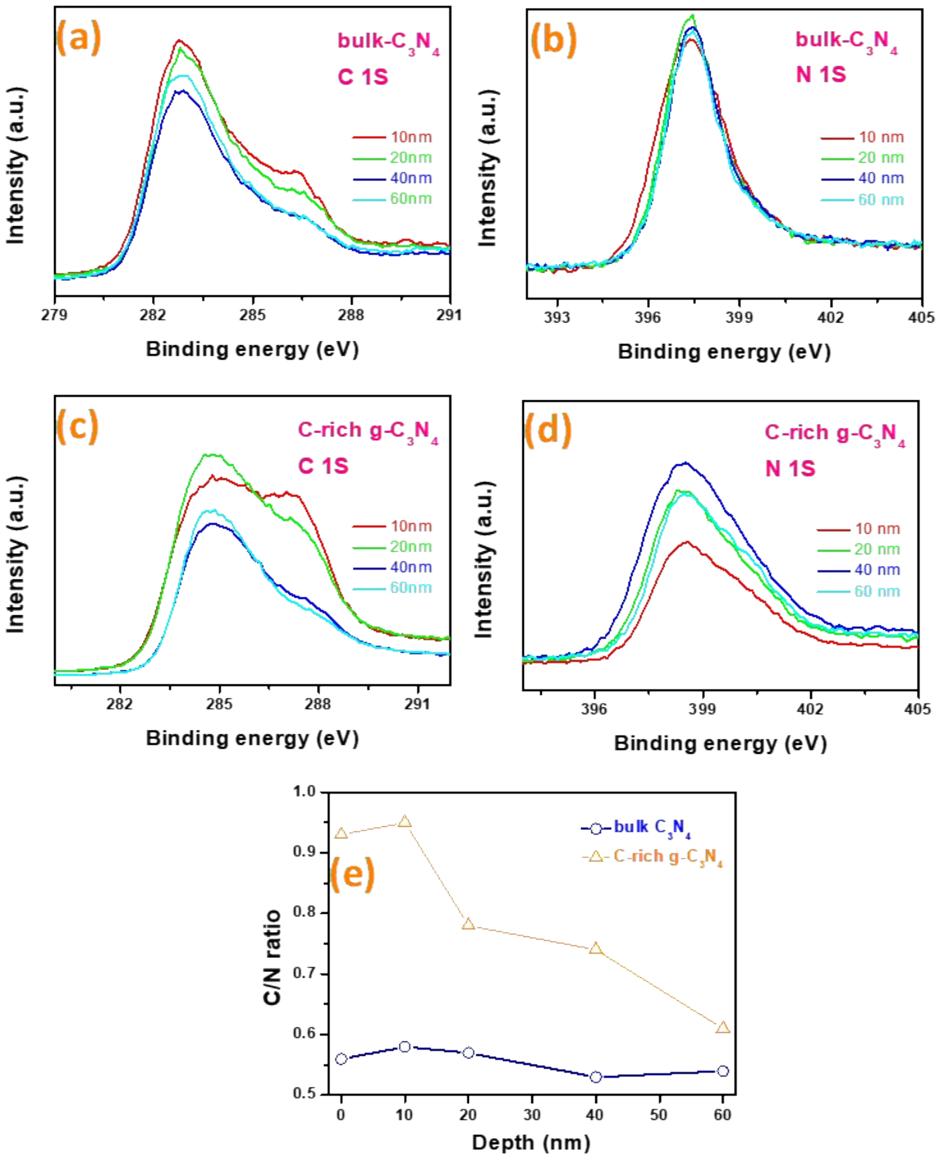

Figure S3. XPS signals with different depth of C 1s (a), N 1s (b) for bulk-C_3N_4, C 1s (c), N 1s (d) for C-rich g-C_3N_4 and Diagram of the variation with depth of the C/N ratio for both samples.

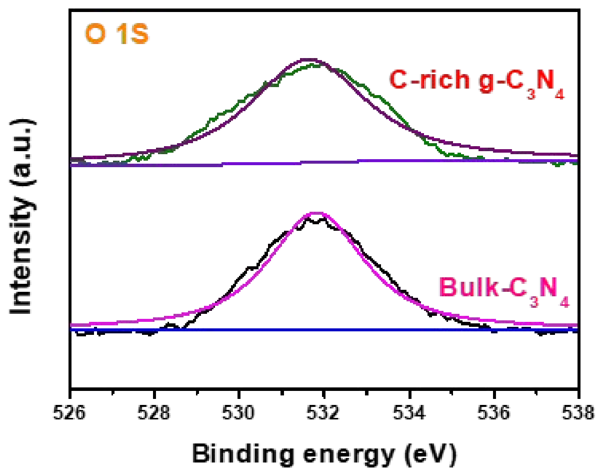

Figure S4. XPS spectra of O 1s for bulk-C_3N_4 and C-rich g-C_3N_4 photocatalysts.

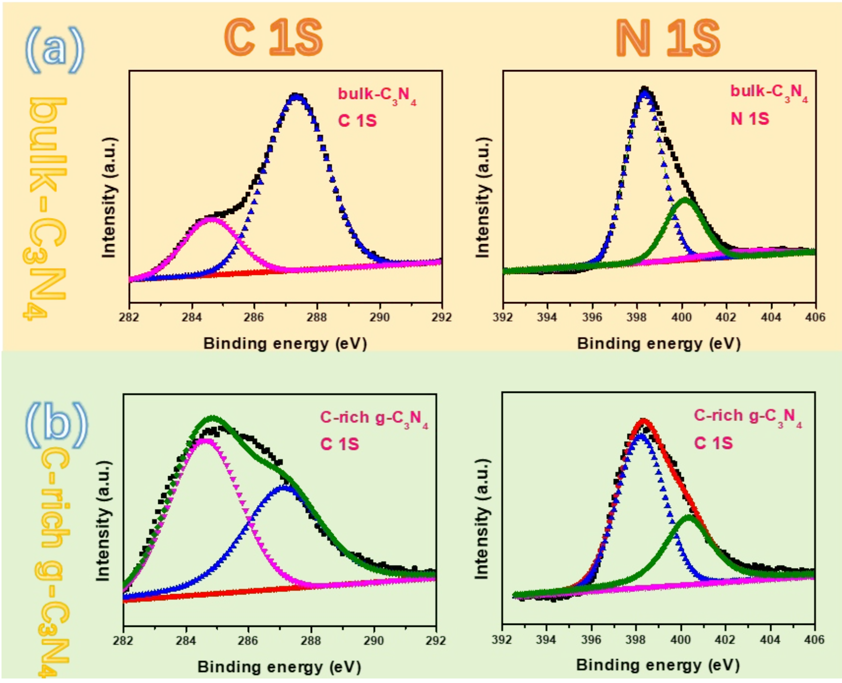

Figure S5. XPS spectra of C 1s and N 1s of (a) bulk-C_3N_4 and (b) C-rich g-C_3N_4 photocatalysts.

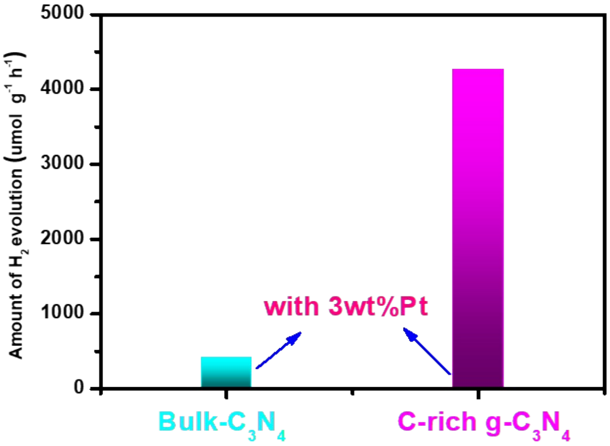

Figure S6. H_2 generation rate over bulk-C_3N_4 and C-rich g-C_3N_4 with 3wt% Pt as co-catalyst (300-W Xe lamp, 35 ml aqueous triethanolamine solution (10 vol %).

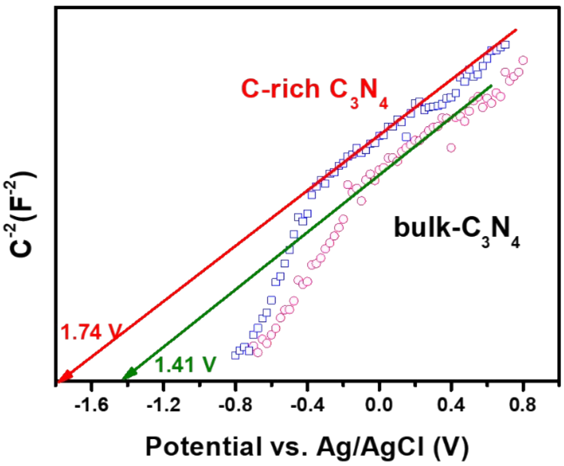

Figure S7. Mott-Schottky plots of bulk-C_3N_4 and C-rich g-C_3N_4 in 0.2 M Na_2SO_4 aqueous solution.

Table S1. Recently published g-C_3N_4 based materials towards noble metal free photocatalytic hydrogen generation.

catalysts	Light source	Conditions	Hydrogen generate rate ($\mu mol\ h^{-1}\ g^{-1}$)	Reference
g-C_3N_4–10% Cu_3P	300W xenon lamp（λ > 420 nm）	17 vol% triethanolamine	159.41	[1]
g-C_3N_4–2% acetylene -2%CuS	350W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	348	[2]
g-C_3N_4–2% Co_2P	300W xenon lamp（λ > 420 nm）	17 vol% triethanolamine	128.4	[3]
g-C_3N_4–5% H-MoSe	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	136.8	[4]
g-C_3N_4–3% Co_2P	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	53.3	[5]
g-C_3N_4–3.5% Ni_2P	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	474.7	[6]
g-C_3N_4–5% $Ni_{12}P_5$	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	535.7	[7]
g-C_3N_4–0.01% WS_2	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	101	[8]
g-C_3N_4–3.5% Ni_3P	300W xenon lamp（λ > 400 nm）	20 vol% triethanolamine	120	[9]
g-C_3N_4–2% Ni_2P	300W xenon lamp（λ > 400 nm）	10 vol% triethanolamine	82.5	[10]
g-C_3N_4–2% $Ni_{12}P_5$	350W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	126.61	[11]
C-rich g-C_3N_4	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine	125.1	This work

Table S2. Recently published g-C_3N_4 based materials towards photocatalytic hydrogen generation.

Materials	Elements	Light source	Conditions	Hydrogen generate rate (μmol h^{-1} g^{-1})	Apparent quantum efficiency	Reference
Amorphous C_3N_4	C, N	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine, 1 wt% Pt	2035	420 nm 6.1 %	[12]
CQD/g-C_3N_4	C, N	300W xenon lamp（λ > 420 nm）	33.3 vol% methanol, 3 wt% Pt	3538.3	420 nm 10.94 %	[13]
GD-C3N4	C, N	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine, 3 wt% Pt	23060	420 nm 31.07 %	[14]
CMB–HCl–C_3N_4	C, N, Cl	A white LED array	10 vol% triethanolamine, 3 wt% Pt	380	420 nm 1.6 %	[15]
Carbon-rich g-C_3N_4 Nanosheets	C, N	300W xenon lamp（λ > 400 nm）	12 vol% triethanolamine, 3 wt% Pt	3960	420 nm 4.52 %	[16]
CCN_{550}	C, N	300W xenon lamp（λ > 420 nm）	10 vol% methanol, 3 wt% Pt	600	420 nm 6.8 %	[17]
CN-Br-3	C, N, Br	300W xenon lamp（λ > 420 nm）	10 ml lactic acid, 1 wt% Pt	1354	420 nm 14.3 %	[18]
3D porous C_3N_4 monolith	C, N	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine, 3 wt% Pt	2900	Not mentioned	[19]
Holey Ultrathin g-C_3N_4	C, N	300W xenon lamp（λ > 400 nm）	12 vol% triethanolamine, 3 wt% Pt	2860	420 nm 4.03 %	[20]

High-crystalline g-C_3N_4	C, N	300W xenon lamp（λ > 400 nm）	10 vol% triethanolamine, 3 wt% Pt	808.5	420 nm 6.17 %	[21]
O-CN2	C, N	300W xenon lamp（λ > 420 nm）	10 vol% lactic acid, 1 wt% Pt.	1062.4	420 nm 13.2 %	[22]
"seaweed" g-C_3N_4	C, N	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine, 3 wt% Pt	9900	420 nm 7.8 %	[23]
g-C_3N_4 nanomesh	C, N	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine, 3 wt% Pt	8510	420 nm 5.1 %	[24]
C-rich g-C_3N_4	C, N	300W xenon lamp（λ > 420 nm）	10 vol% triethanolamine, 3 wt% Pt	4268	420 nm 6.8 %	This work

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