Supporting Information

Green synthesis of ultrathin edge-activated foam-like carbon nitride nanosheets for enhanced photocatalytic performance under visible light irradiation

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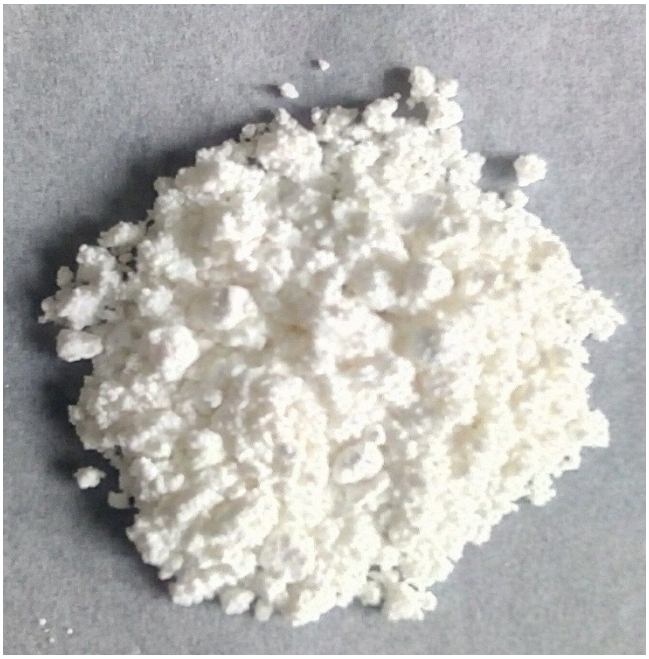

Fig. S1 Image of as-prepared PCN in wet atmosphere with loose and white appearance.

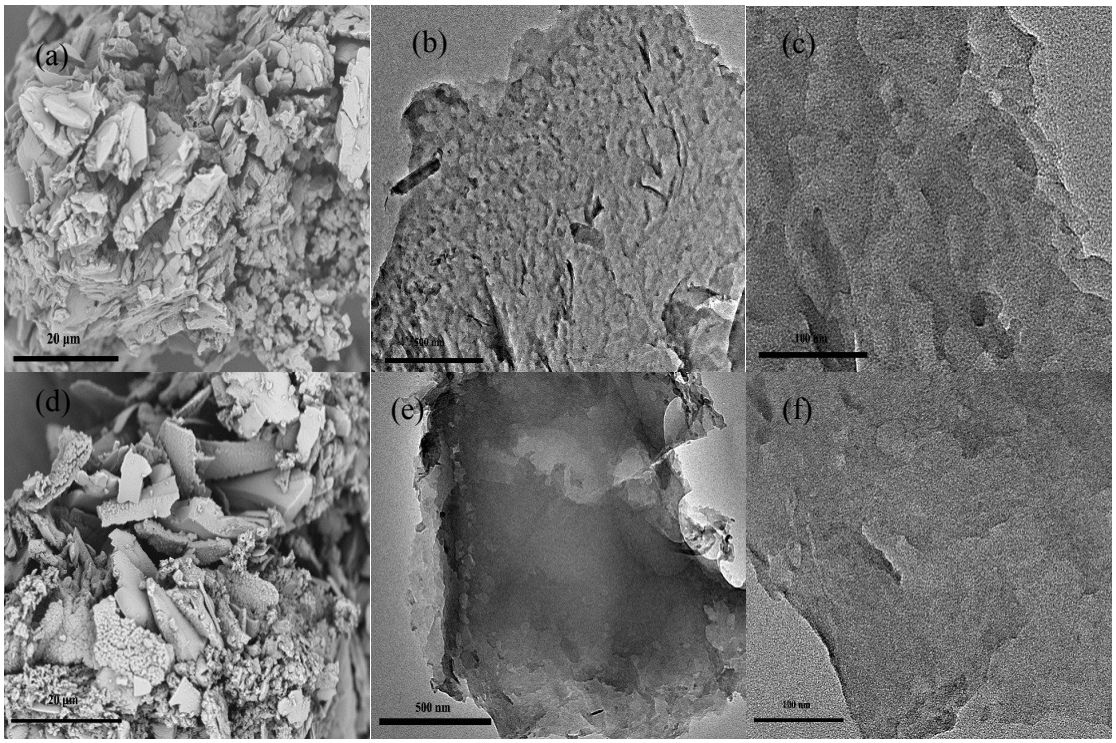

Fig. S2 (a) SEM image of MCN-air, (b) TEM image, and (c) High magnification TEM of MCN-air. (d) SEM image of MCN-N_2, (e) TEM image, and (f) High magnification TEM of MCN-N_2.

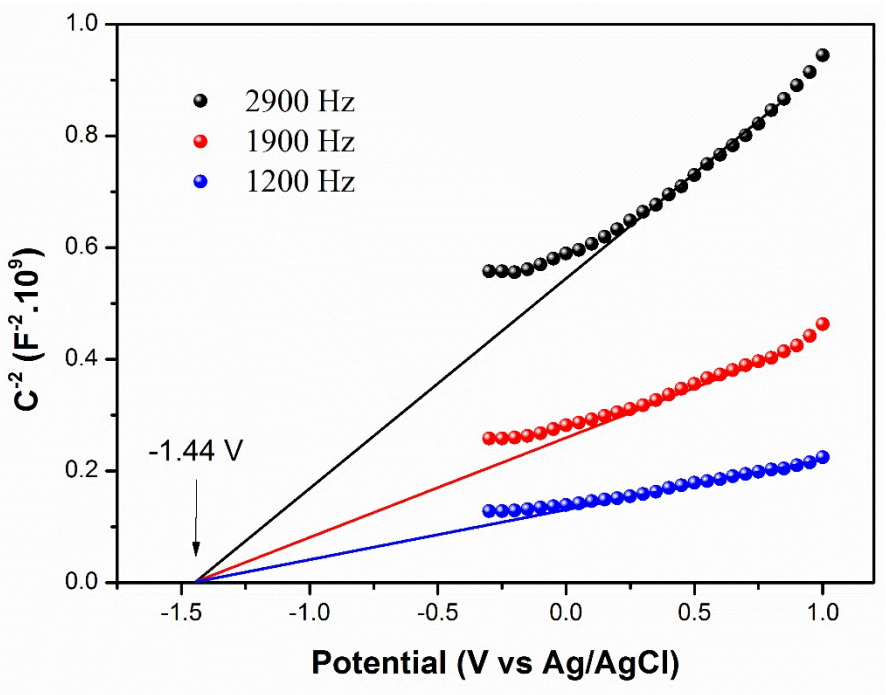

Fig. S3 Mott-Schottky plots of MFCN-wet nanosheets at various frequencies.

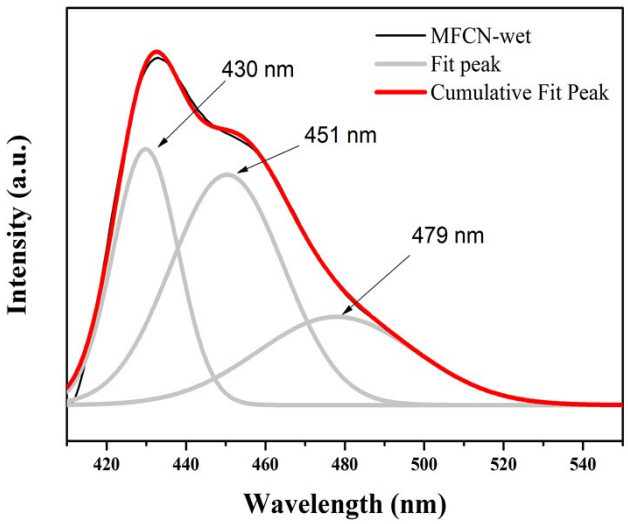

Fig. S4 Gaussian fitting of PL emission spectra of MFCN-wet.

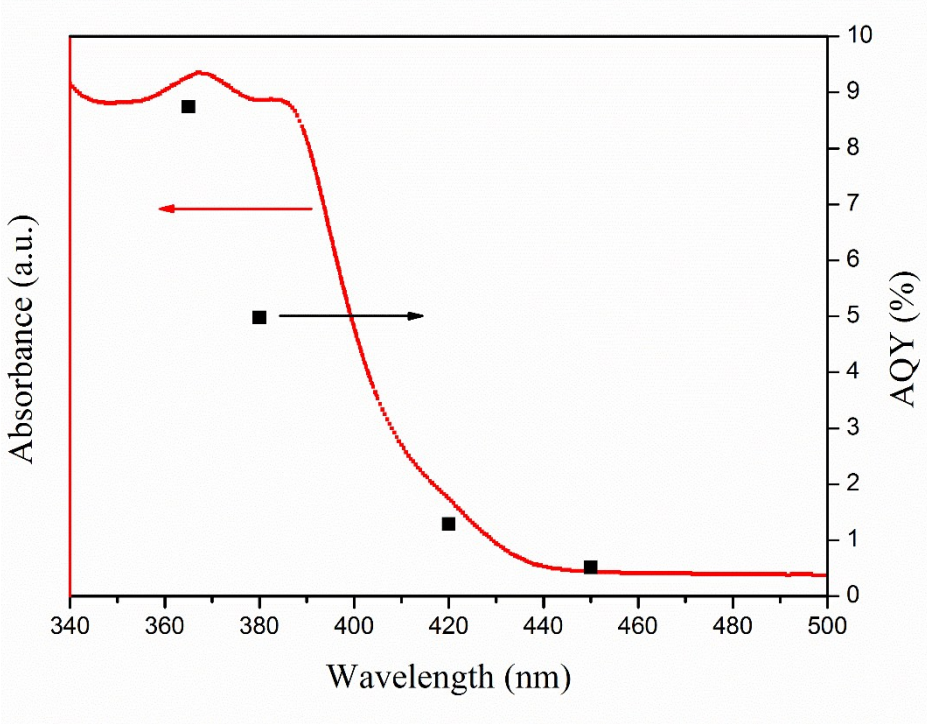

Fig. S5 Wavelength dependence of AQY on the H_2 evolution in the MFCN-wet nanosheets.

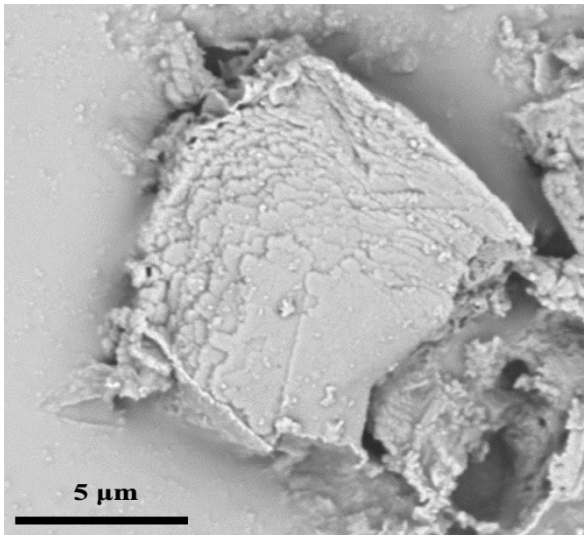

Fig. S6 SEM image of MFCN-wet nanosheets after 16 h visible light irradiation for H_2 evolution.

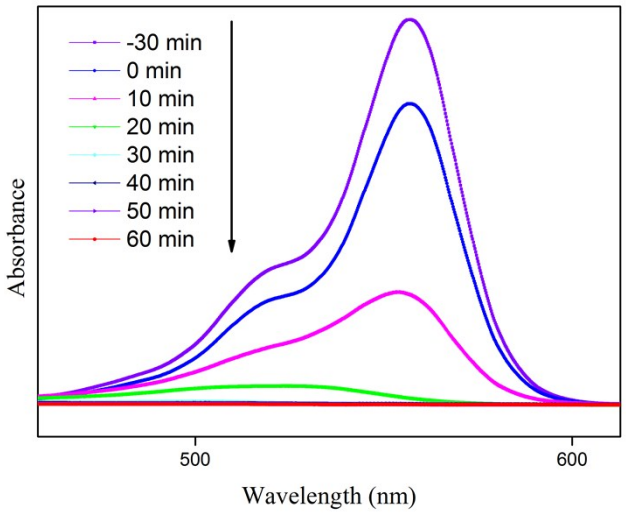

Fig. S7 UV-visible absorption spectra of RhB after different irradiation time under visible light (λ = 420 nm) in the presence of MFCN-wet.

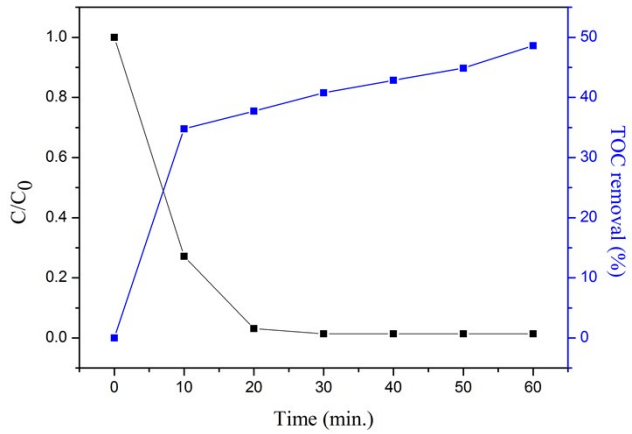

Fig. S8 The degradation (left) and mineralization (right) of RhB during MFCN-wet photocatalytic degradation process.

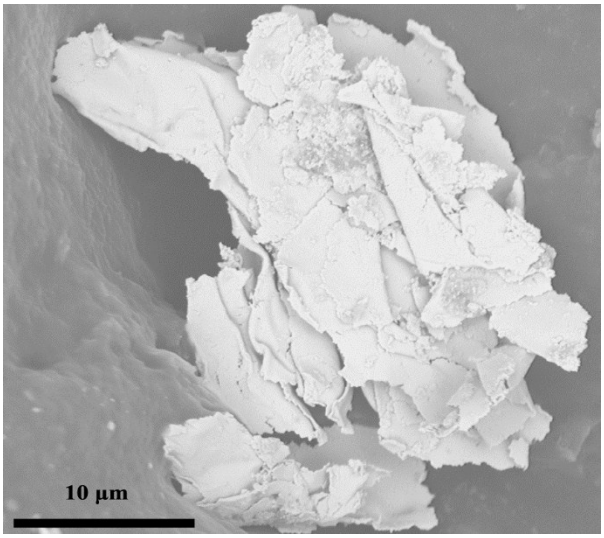

Fig. S9 SEM image of MFCN-wet nanosheets after 5 runs of RhB photodegradation under visible light irradiation.

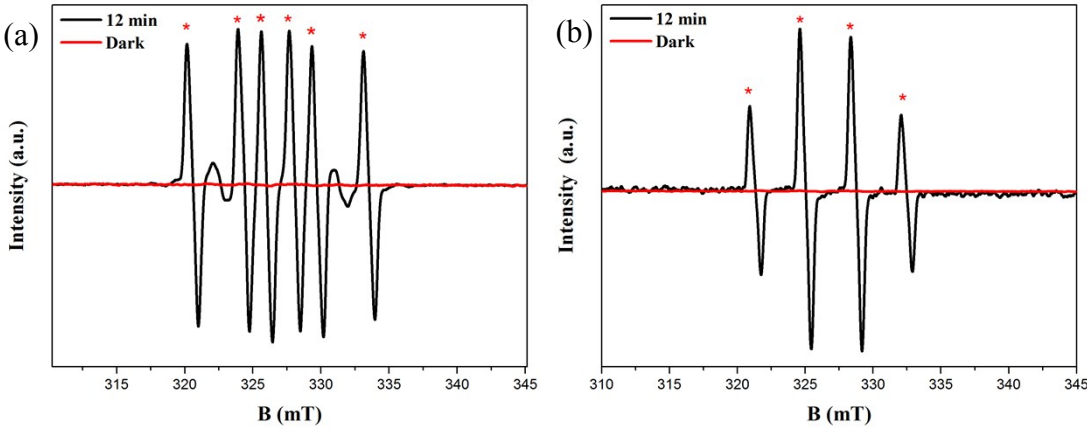

Fig. S10 Electron spin resonance spectroscopy (ESR) of MFCN-wet photocatalyst without and with visible light irradiation ($\lambda \geq 420$ nm); (a) Superoxide radical test in methanol solution with DMPO as radical trapper, and (b) Hydroxyl radical test in aqueous solution with DMPO as radical trapper.

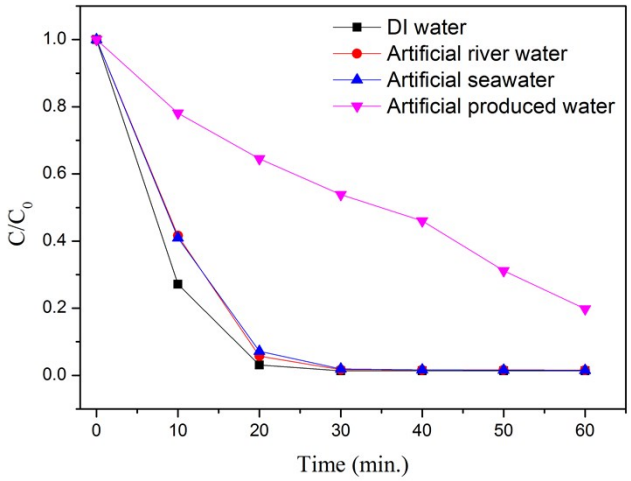

Fig. S11 Photocatalytic degradation of RhB in different artificial water environments.

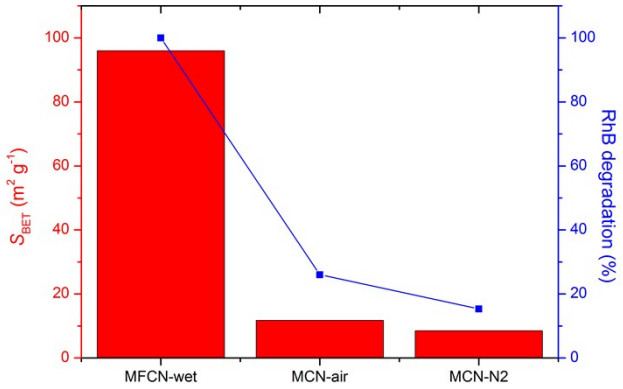

Fig. S12 Normalization of RhB degradation (%) after 30 min with the surface area of as-prepared materials.

Table S1 Comparison of typical metal-free synthesis process of nanostructured carbon nitride photocatalysts and their performance under visible light irradiation.

Material	Approach	Preparation method	Chemicals consumed	Synthesis condition (steps/time/cost)	Functional groups	Surface area ($m^2 g^{-1}$)	Layer thickness (nm)	Application	Efficiency	Industrial green criteria	Ref.
Distorted g-C_3N_4	Top-down	Hydrothermal	HF	3 steps/15 h/ medium	Structural distortion	53.2	20	Hydrogen evolution	1167.7 μmol g^{-1} h^{-1}, (3 wt% Pt co-catalyst), AQY = 1.7%	Harsh solvent, time-consuming	1
Single layer g-C_3N_4	Top-down	Exfoliation	Methanol	3 steps/ 7.5 h/ low	-NH_2	-	0.5	RhB degradation	k = 0.0326 min^{-1}	Chemical-consuming, less-efficiency	2
Holey ultrathin g-C_3N_4 nanosheets	Top-down	Hydrothermal exfoliation	NaClO	2 steps/ 9 h/ low	-NH_2	170.7	4	Hydrogen evolution	890 μmol g^{-1} h^{-1}	Chemical-consuming	3
O-doped CN aerogel	Top-down	Self-assembly combined with hydrothermal process	-	4 steps/ 13 h/ medium	-NH_2, -OH	19.38	-	Hydrogen evolution	66.28 μmol g^{-1} h^{-1}, (3 wt% Pt co-catalyst), AQY = 7.43%	Time- and energy-consuming Less-efficiency	4
Ultrathin g-C_3N_4 nanosheets	Top-down	Exfoliation	Isopropanol	1 step/10 h/ low	-NH_2, -C≡N	384	2	Hydrogen evolution	1860 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst)	Chemical-consuming	5
3D g-C_3N_4 nanobelt	Top-down	Hydrothermal	$H_2C_2O_4$, ethanol, acetone	3 steps/ more than 32 h/ high	-OH, -C=O, -NH_2	62.7	-	Hydrogen evolution	1360 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst), AQY = 12%	Harsh solvents, time- and energy-consuming	6
Ultrathin g-C_3N_4 nanosheets	Top-down	Post-thermal etching	-	2 steps/ 8 h/ low	-NH_2	131.2	-	Hydrogen evolution, RhB degradation	459.94 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst), k = 0.07346 min^{-1}	Less-efficiency	7
Crystalline gCN nanosheets	Top-down	Exfoliation	KCl, LiCl, isopropanol	3 steps/ 23 h/ medium	-NH_2	203	3.6	Hydrogen evolution	1060 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst), AQE = 8.57%	Harsh solvents, Time- and energy-consuming	8
Monolayer g-C_3N_4 nanosheet	Top-down	Exfoliation	Isopropano, ethanol, dimethyl formamide	2 steps/ 14 h/ medium	-NH_2	59.4	0.38	RhB degradation oxidation of benzyl alcohol	Degradation in 70 min, 19.4% conversion	Chemical-consuming, less-efficiency	9
Functionalized g-C_3N_4 nanosheets	Top-down	Chemical oxidation	H_2SO_4, $KMnO_4$	2 steps/4.5 h/ medium	-COOH, -C=O	-	-	E-coli disinfection	k = 0.068 min^{-1}	Harsh solvents, chemical-consuming	10

Onion-ring like g-C_3N_4	Bottom-up	SiO_2 template	SiO_2, NH_4HF_2	2 steps/ 17 h/ medium	-NH_2, -C=O	29.9	-	Hydrogen evolution	1900 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst)	Harsh solvents, Time-consuming	11
Crystalline CN	Bottom-up	Protonation	HCl	2 steps/ -/ low	-NH_2	17.73	-	RhB degradation	k = 0.01296 min^{-1}	Harsh solvent, less-efficiency	12
Triazine–heptazine copolymer	Bottom-up	Ionothermal synthesis	NaCl, KCl	2 steps/ 4 h/ low	-NH_2, -C≡N	74	-	Hydrogen evolution	5560 μmol g^{-1} h^{-1}, (3 wt% Pt co-catalyst), AQY = 32%	Chemical-consuming	13
Crystalline PCN	Bottom-up	Supramolecular aggregation + ionic melt polycondensation	2,4,6-triaminopyrimidine, ethanol, LiCl, KCl	2 steps/ more than 12 h/ high	-NH_2	93	-	Hydrogen evolution	8160 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst), AQY = 7%	Chemical-consuming	14
Porous g-C_3N_4	Bottom-up	Hard-template	HCl, HF, kaolinite	5 steps/ more than 6 h/ high	-NH_2, Defect pores	109	-	Hydrogen evolution	1917 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst)	Harsh solvents, chemical-consuming	15
Oxygen doped g-C_3N_4	Bottom-up	Co-pyrolysis with thermal polymerization	$(NH_4)_2S_2O_8$	1 step/ 4 h/ low	-C≡O, N-defected	36.9	-	Hydrogen evolution	395.96 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst), AQY = 0.79%	Chemical consuming, less-efficiency	16
Thin g-C_3N_4 nanosheets	Bottom-up	Hydrothermal and calcination	Thiourea, ethanol	2 steps/ 23 h / medium	-NH_2	44.2	4	Hydrogen evolution NO removal	991 μmol g^{-1} h^{-1}, (1 wt% Pt co-catalyst), 17.5% removal	Time- and chemical-consuming, less-efficiency	17
Porous g-C_3N_4 nanosheets	Bottom-up	Self-producing atmosphere	-	1 step/ 4 h/ low	-NH_2	38.51	-	RhB degradation	k = 0.0605 min^{-1}	Less-efficiency	18
Nanoporous α-C_3N_4	Bottom-up	Solution combustion	-	2 steps/ more than 3 h/ low	-NH_2	135.6	-	Hydrogen evolution, RhB degradation	70 μmol g^{-1} h^{-1}, (1.5 wt% Pt co-catalyst) Mostly degraded within 1 h	Less-efficiency	19
g-C_3N_4 microtube	Bottom-up	Hydrothermal-assisted thermal polymerization	-	2 steps/ 28 h/ medium	Tubular-like structure	12.8	-	Hydrogen evolution	1000 μmol g^{-1} h^{-1} (0.5 wt% Pt co-catalyst)	Time- and energy-consuming	20
g-C_3N_4 Nanofibers	Bottom-up	Poly addition + thermal polymerization	Ethanol, HNO_3	2 steps/ more than 2 h/ low	-NH_2	165	-	RhB degradation	k = 0.0412 min^{-1}	Harsh solvents, less-efficiency	21
Nanocage-like g-C_3N_4	Bottom-up	Hydrothermal + freeze drying + thermal polymerization	HCl	4 steps/ more than 8 h/ medium	-NH_2, N-vacancies	226.19	-	Hydrogen evolution	1851.4 μmol g^{-1} h^{-1} (3 wt% Pt co-catalyst), AQY = 3.01%	Harsh solvent, tedious multistep process	22
Ultrathin PCN	Bottom-up	Coupling of thermal	-	2 steps/ 6 h/ low	-NH_2, -OH,	96	3.8	Hydrogen evolution,	1880.2 μmol g^{-1} h^{-1}	Green, low-cost,	Present work

nanosheets	polymerization with layers etching in wet atmosphere	-NO, $-NO_2$	RhB degradation	(3 wt% Pt co-catalyst), AQY = 1.3% k = 0.15467 min^{-1}	template-free, high-efficiency, enrich active sites

Note: k is the degradation or disinfection rate constant of the pollutant. All hydrogen evolution tests were performed in DI water and all apparent quantum yields (AQY) were calculated at 420 nm.

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