Advances and Challenges of Graphitic Carbon Nitride as a Visible-Light-Responsive Photocatalyst for Sustainable Water Purification

Supplementary Information

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Table S1 Representative Summary of the Photocatalytic Activity of g-C_3N_4-Based Photocatalysts for Degrading/Inactivating Waterborne Contaminants/Microorganisms

Photocatalyst	Contaminant	Reactivity (Reaction Rate Constant)	Light Source	Reactive Species	Ref (Year)
Bulk g-C_3N_4	Phenol	92.5% of degradation in 3 h	500 W Xe lamp ($\lambda > 420$ nm)	N/A	[1] (2011)
Bulk g-C_3N_4	2,4,6-Trichlorophenol	(1.79 h^{-1})	300 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}/HO_2^{\bullet}$	[2] (2013)
Bulk g-C_3N_4	○ Tetracycline ○ Salicylic acid ○ Ciprofloxacin ○ Ibuprofen	○ 86% of degradation in 4 h ○ 30% of degradation in 4 h ○ 60% of degradation in 4 h ○ 20% of degradation in 4 h	35 W Xe lamp	•OH, H_2O_2, h^+	[3] (2016)
Bulk g-C_3N_4	Virus MS2	> 7 log of inactivation in 5 h	300 W Xe lamp ($\lambda > 400$ nm)	$O_2^{\bullet-}$, e^-	[4] (2016)
Mesoporous g-C_3N_4	○ 4-Chlorophenol ○ Phenol	○ ~100% of degradation in 1 h (0.0526 min^{-1}) ○ ~96% of degradation in 1.5 h (0.0342 min^{-1})	300 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, •OH	[5] (2012)
Nanoporous g-C_3N_4	Phenol	(0.00846 h^{-1})	500 W Xe lamp ($\lambda > 420$ nm)	N/A	[6] (2013)
Porous g-C_3N_4	Phenol	(0.039 h^{-1})	500 W Xe lamp ($\lambda > 420$ nm)	N/A	[7] (2013)
Mesoporous g-C_3N_4	Bacterium *E. coli*	100% of inactivation in 4 h	300 W Xe lamp ($\lambda > 400$ nm)	h^+	[8] (2014)
Porous g-C_3N_4	Phenol	~35% of degradation in 3 h	300 W Xe lamp ($\lambda > 400$ nm)	N/A	[9] (2015)
Porous g-C_3N_4	Phenol	55% of degradation in 3 h	300 W Xe lamp ($\lambda > 400$ nm)	N/A	[10] (2015)

g-C_3N_4 nanosheets	Phenol	(0.2589 h^{-1})	500 W Xe lamp ($\lambda > 420$ nm)	N/A	[11] (2013)
Porous C-g-C_3N_4	○ Phenol ○ Atrazine ○ Sulfamethoxazole ○ Carbamazepine	○ (0.76 ± 0.08 m^2 (mol of photons)$^{-1}$) ○ (1.33 ± 0.18 m^2 (mol of photons)$^{-1}$) ○ (0.78 ± 0.02 m^2 (mol of photons)$^{-1}$) ○ (0.16 ± 0.003 m^2 (mol of photons)$^{-1}$)	1000 W Xe lamp ($\lambda > 400$ nm)	$^1O_2$, h^+	[12] (2016)
C-g-C_3N_4	Tetracycline	>95% of degradation in 1.5 h	Sunlight (78,000-80,000 lux)	$O_2^{\bullet-}$, h^+	[13] (2017)
Phosphate modified g-C_3N_4	Phenol	~80% of degradation in 1 h (0.0174 min^{-1})	150 W Xe lamp	N/A	[14] (2014)
P-porous g-C_3N_4 nanosheets	2,4-Diclorophenol	100% of degradation in 80 min	300 W Xe lamp ($\lambda > 400$ nm)	h^+	[15] (2017)
Oxidized g-C_3N_4	Sodium pentachlorophenol	(0.0613 h^{-1})	300 W Xe lamp ($\lambda > 420$ nm)	N/A	[16] (2014)
O-porous g-C_3N_4	Bisphenol A (BPA)	100% of degradation in 3 h (0.098 mmol g_{cat}^{-1} h^{-1})	500 W Xe lamp (visible light)	•OH	[17] (2017)
Reduced graphene oxide@ g-C_3N_4@α-S_8	Bacterium *E. coli*	> 6 log of inactivation in 4 h	300 W Xe lamp ($\lambda > 400$ nm)	•OH, H_2O_2 (aerobic); e^- (anaerobic)	[18] (2013)
g-C_3N_4/$Bi_5Nb_3O_{15}$	4-Chlorophenol	~100% of degradation in 1 h	300 W Xe lamp (680 nm > λ > 400 nm)	$O_2^{\bullet-}$, h^+	[19] (2013)
Ag/g-C_3N_4	4-Nitrophenol	98% of degradation in 2 h	300 W Xe lamp (680 nm > λ > 400 nm)	$O_2^{\bullet-}$, •OH, h^+	[20] (2013)
AgX/g-C_3N_4 (X = Br, I)	4-Chlorophenol	30% and 53% of degradation in 6 h	300 W Xe lamp ($\lambda > 400$ nm)	N/A	[21] (2013)

CeO_2/g-C_3N_4	4-Chlorophenol	45% of degradation in 5 h	300 W Xe lamp ($\lambda > 400$ nm)	N/A	[22] (2013)
Co_3O_4/mesoporous g-C_3N_4	Bisphenol A (BPA)	93.6% of degradation in 3 h ($0.014\ min^{-1}$)	500 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, •OH	[23] (2013)
$H_3PW_{12}O_{40}$/g-C_3N_4 nanotubes	Diethyl phthalate	100% of degradation in 24 h	300 W Xe lamp ($\lambda > 420$ nm)	N/A	[24] (2013)
Polyoxometalates/g-C_3N_4	Phenol	($3.99 \times 10^{-3}\ min^{-1}$)	575 W metal halide lamp	N/A	[25] (2015)
g-C_3N_4/TiO_2	Isoniazid	($8.33 \times 10^{-3}\ min^{-1}$)	N/A	$O_2^{\bullet-}$, •OH	[26] (2015)
WO_3/g-C_3N_4	Methyl tertiary butyl ether (MTBE)	96.7 % of degradation	500 W Xe lamp equipped with a visible light filter	N/A	[27] (2015)
g-C_3N_4/TiO_2	Ciprofloxacin	~95% of degradation in 1 h	150 W tungsten lamp	•OH, h^+, e^-	[28] (2015)
Pt/g-C_3N_4	4-Chlorophenol	100% of degradation in 6 h	300 W Xe lamp ($\lambda > 420$ nm)	N/A	[29] (2015)
g-C_3N_4/ZnO	Phenol	(0.02 and $0.015\ min^{-1}$)	60 W tungsten lamp	$O_2^{\bullet-}$, •OH	[30] (2015)
Au/porous g-C_3N_4/graphene	Ciprofloxacin	($\sim 0.014\ min^{-1}$)	500 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, •OH	[31] (2015)
$ZnIn_2S_4$/g-C_3N_4	Phenol	72.3% of degradation in 4 h	300 W Xe lamp ($\lambda > 420$ nm)	•OH, h^+	[32] (2015)
$Cd_{0.2}Zn_{0.8}S$/g-C_3N_4 nanosheet	Phenol	>76.1% of degradation in 3 h	500 W Xe lamp ($\lambda > 420$ nm)	N/A	[33] (2015)
MoS_2 nanosheets/TiO_2/g-C_3N_4	Atrazine	86.5% of degradation in 3 h	500 W Xe lamp	$O_2^{\bullet-}$, •OH, h^+	[34] (2016)
V_2O_5/g-C_3N_4	Tetracycline	75.7% of degradation in 2 h	250 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, h^+	[35] (2016)

$Co_{0.5}Zn_{0.5}Fe_2O_4$/g-C_3N_4	Chloromycetin	96% of degradation in 4 h	300 W Xe lamp with a UV cutoff filter	N/A	[36] (2016)
g-C_3N_4/mullite	Tetracycline	92% of degradation in 4 h ($0.00602\ min^{-1}$)	500 W Xe lamp ($\lambda > 400$ nm)	$O_2^{\bullet-}$, h^+	[37] (2016)
g-C_3N_4/activated carbon	Phenol	100% of degradation in 160 min ($0.630\ h^{-1}$)	350 W Xe lamp ($\lambda > 400$ nm)	N/A	[38] (2016)
N-graphene quantum dots-$BiVO_4$/g-C_3N_4	Tetracycline	91.5% of degradation in 30 min ($0.0815\ min^{-1}$)	250 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, •OH	[39] (2016)
C/X-TiO_2@C_3N_4 nanotubes (X = N, F, Cl)	4-Chlorophenol	~100% of degradation in 6 h	300 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, h^+	[40] (2016)
Nb_2O_5/g-C_3N_4	Tetracycline	○ 76.2% of degradation in 150 min ($0.0096\ min^{-1}$) ○ 90.1% of degradation in 1 h ($0.038\ min^{-1}$)	○ 250 W Xe lamp ($\lambda > 420$ nm) ○ 250 W Xe lamp	$O_2^{\bullet-}$, h^+	[41] (2016)
Polyaniline/g-C_3N_4 nanosheets	Phenol	16.3% of degradation in 4 h	300 W Xe lamp ($\lambda > 420$ nm)	N/A	[42] (2016)
g-C_3N_4/SiO_2 hydrogel	Phenol	~20% of degradation in 10 h	500 W Xe lamp ($\lambda > 420$ nm)	N/A	[43] (2016)
g-C_3N_4/zinc phthalocyanine nanofibers	Carbamazepine	~98% of degradation in 10 h	Sunlight (82,961 lux)	$O_2^{\bullet-}$, h^+	[44] (2016)
g-C_3N_4/TiO_2	Ciprofloxacin	88.1% of degradation in 3 h	300 W Xe lamp ($\lambda > 400$ nm)	h^+	[45] (2016)
g-C_3N_4-Ag/ZnO	Tetracycline	80% of degradation in 1 h	Sunlight	$O_2^{\bullet-}$, h^+, e^-	[46] (2016)

Carbon quantum dots/g-C_3N_4	Tetracycline	(0.00642 min^{-1})	250 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, h^+	[47] (2016)
Polypyrrole@Ag/g-C_3N_4	Danofloxacin mesylate, tetracycline, ciprofloxacin, gatifloxacin, enrofloxacin	~90% of degradation in 1 h	300 W Xe lamp (visible light)	$O_2^{\bullet-}$, h^+	[48] (2016)
Amorphous AgSiO/g-C_3N_4 nanosheets	Tetracycline	○ ~97.5 of degradation in 140 min (0.0116 min^{-1}) ○ ~90.5% of degradation in 40 min (0.0798 min^{-1})	○ 500 W Xe lamp ($\lambda > 400$ nm) ○ 500 W Xe lamp (UV)	•OH, h^+	[49] (2016)
$BiVO_4$/g-C_3N_4	4-Nitrophenol	100% of degradation in 2 h	300 W Xe lamp	$O_2^{\bullet-}$, •OH, h^+	[50] (2016)
SnO_2/B-P-g-C_3N_4	Phenol	~75% of degradation in 1 h	150 W Xe lamp ($\lambda > 420$ nm)	N/A	[51] (2017)
$CdIn_2S_4$/g-C_3N_4	Tetracycline	~80% of degradation in 1.5 h	500 W tungsten light lamp	$O_2^{\bullet-}$, h^+	[52] (2017)
ZrO_2/Fe-hollow g-C_3N_4	Metsulfuron methyl	~100% of degradation in 1 h	500 W Xe lamp (720 nm> λ > 420 nm)	N/A	[53] (2017)
Fe_2O_3/g-C_3N_4	4-Nitrophenol	~96% of degradation in 80 min, with the addition of H_2O_2 (0.0743 min^{-1})	300 W Xe lamp ($\lambda > 400$ nm)	$O_2^{\bullet-}$, •OH, h^+	[54] (2017)
Ag@g-C_3N_4 nanosheets@$BiVO_4$	Tetracycline	90.76%, ~80%, and ~12% of degradation in 1 h	300 W Xe lamp (λ > 350, 420, and 760 nm)	$O_2^{\bullet-}$, •OH, h^+	[55] (2017)
Carbon nanospheres/g-C_3N_4	Sulfachloropyridazine	100% of degradation in 2 h (0.0381 min^{-1})	Metal halide lamp (2.31 μW cm^{-2})	$O_2^{\bullet-}$	[56] (2017)

g-C_3N_4/$BiPO_4$	Ciprofloxacin	96.6% of degradation in 2 h	250 W high pressure Hg lamp (UV)	$O_2^{\bullet-}$, h^+	[57] (2017)
Graphene oxide/Ag_2CrO_4/g-C_3N_4	○ Phenol ○ Oxytetracycline	○ 94.21% of degradation in 1.5 h ○ 81.34% of degradation in 1.5 h	300 W Xe lamp ($\lambda > 420$ nm)	$O_2^{\bullet-}$, •OH, h^+	[58] (2017)
OH-graphene quantum dots/mesoporous g-C_3N_4	Tetracycline	70% of degradation in 2 h	300 W Xe lamp ($\lambda > 400$ nm)	$O_2^{\bullet-}$	[59] (2017)

Table S2 Representative Summary of Photoluminescence Quantum Yield of g-C_3N_4 in Steady-State Photoluminescence Measurements

Photocatalyst	Synthesis Method	Particle Size	Photoluminescence Quantum Yield	Reference (Year)
Bulk g-C_3N_4	Thermal polycondensation of melamine	N/A	4.8%	[60] (2013)
Nanosheet g-C_3N_4	Liquid exfoliation of bulk g-C_3N_4, which was prepared via thermal polycondensation of melamine	Diameter of 70-160 nm, thickness of 2.5 nm	19.6%	[60] (2013)
Nanosheet g-C_3N_4	Liquid exfoliation of acid treated bulk g-C_3N_4, which was prepared via thermal polycondensation of melamine	Diameter of 70 nm, thickness of 2.13 nm	5.5%	[61] (2015)
Nanoparticulate g-C_3N_4 (QDs)[a]	Microwave mediated synthesis of formamide	Diameter of 2-15 nm, thickness of 0.3-2.1 nm	29%	[62] (2012)
Nanoparticulate g-C_3N_4 (QDs)	Microwave-assisted solvothermal method of citric acid, urea, and oleic acid	Diameter of 2.8 nm	27.1%	[63] (2015)
Nanoparticulate g-C_3N_4 (QDs)	Microwave-assisted solvothermal method of guanidine hydrochloride and EDTA	Diameter of 3.2-6.5 nm, thickness of 1.7 nm	35%	[64] (2014)
Nanoparticulate g-C_3N_4 (QDs)	Hydrothermal reaction of phosphonitrilic chloride trimer and g-C_3N_4 QDs, which was obtained via liquid exfoliation of melamine derived bulk g-C_3N_4	Diameter of 4-12 nm, thickness of 0.35-1.21 nm	91-96%	[65] (2016)
Nanoparticulate g-C_3N_4 (QDs)	Thermal reaction of citric acid and thiourea	Diameter of 2.75 nm	14.5%	[66] (2015)
Nanoparticulate g-C_3N_4 (QDs)	Thermal reaction of urea and sodium citrate	Diameter of 2.6-5.5 nm, thickness of 1.5-2.5 nm	42%	[67] (2013)

[a] QDs represents quantum dots.

Table S3 Representative Summary of Fluorescence Lifetime of Photocatalysts in Time-Resolved Photoluminescence Measurements

Photocatalyst	Synthesis Method	Excitation/Emission Wavelength (nm)	Average Fluorescence Lifetime (ns)	Reference (Year)
TiO_2 Anatase	Commercially available from Merck	355/450	450	[68] (1997)
TiO_2 Anatase	Hydrothermal treatment of a $TiOCl_2$-NH_3 solution, and subsequent calcination for the resultant solids	355/450	320-460	[68] (1997)
TiO_2 Rutile	Commercially available from Janssen	355/450	200	[68] (1997)
TiO_2 Rutile	A diluted $TiOCl_2$ solution was boiled (pH < 1.1), and resultant solids were calcinated	355/450	180-400	[68] (1997)
TiO_2 Degussa P25	Commercially available	337/N/A	2.14	[69] (2006)
TiO_2	Sol-gel	337/N/A	2.55	[69] (2006)
TiO_2	Polyol method	260/410	0.81	[70] (2012)
N, S-doped TiO_2	Polyol method	260/410	3.57	[70] (2012)
WO_3	Commercially available from Sigma-Aldrich	N/A	5.78	[71] (2014)
Cu^{2+} modified WO_3	Commercially available from Showa Titanium Inc. Japan	470/780	0.188	[72] (2012)
$BiVO_4$	Hydrothermal treatment of $Bi(NO_3)_3 \cdot 5H_2O$ and NH_4VO_3	N/A	3.87	[73] (2015)
Bulk g-C_3N_4	Thermal polycondensation of dicyandiamide	405/475	5.2	[74] (2013)
Bulk g-C_3N_4	Thermal polycondensation of dicyandiamide	340/450	9.86	[75] (2015)
Bulk g-C_3N_4	Thermal polycondensation of dicyandiamide	330/468	6.98[a]	[76]

				(2012)
Bulk g-C_3N_4	Thermal polycondensation of melamine	405/500	6.22	[77] (2014)
Bulk g-C_3N_4	Thermal polycondensation of melamine	365/460	2.885	[78] (2015)
Bulk g-C_3N_4	Thermal polycondensation of melamine	N/A	3.88[a]	[79] (2016)
Bulk g-C_3N_4	Thermal polycondensation of melamine	N/A	9.05[a]	[80] (2016)
Bulk g-C_3N_4	Thermal polycondensation of melamine	377/460	6.4	[81] (2015)
Bulk g-C_3N_4	Thermal polycondensation of urea	380/440 380/563	2.42 4.37	[82] (2015)
Porous g-C_3N_4	Thermal polycondensation of melamine and cyanuric acid	405/475	3.6-5.6	[74] (2013)
Porous g-C_3N_4	Thermal polycondensation of melamine and cyanuric acid	405/500	4.7	[77] (2014)
Barbituric acid-doped, porous g-C_3N_4	Thermal polycondensation of melamine, cyanuric acid, and barbituric acid	405/500	1.51-3.3	[77] (2014)
g-C_3N_4 with hydrogenated defects	Hydrogen thermal treatment of bulk g-C_3N_4	800/450-470 800/500-520 800/560-580 (Two photon absorption)	0.74-1.20 0.71-1.41 0.58-1.30	[83] (2015)
Quinoline-doped g-C_3N_4	Thermal polycondensation of urea and 2,4-dibromoquinoline	380/436 380/510	1.47 3.20	[82] (2015)
P-doped, porous g-C_3N_4	Thermal exfoliation of P-doped, bulk g-C_3N_4, which was synthesized from thermal polycondensation of 2-aminoethylphosphonic acid and melamine	365/460	3.927	[78] (2015)
Protonated g-C_3N_4	Hydrochloric acid treated bulk g-C_3N_4, which was synthesized from thermal polycondensation of dicyandiamide	340/450	18.4	[75] (2015)

Openly-structured g-C_3N_4 microspheres	Bulk g-C_3N_4 was first prepared via thermal polycondensation of melamine, and it was dissolved in acidic aqueous solution. The precipitated white solids was separated, dried, and thermally treated.	N/A	4.91[a]	[79] (2016)
K-doped g-C_3N_4	Thermal polycondensation of melamine and KCl	N/A	2.73[a]	[80] (2016)
K and OH-doped g-C_3N_4	Thermal polycondensation of melamine, KCl, and NaOH	N/A	2.03[a]	[80] (2016)
Fe^{3+} modified, K and OH-doped g-C_3N_4	Thermal polycondensation of melamine, KCl, and NaOH, and subsequent modification with Fe^{3+}	N/A	2.52[a]	[80] (2016)
Nanosheet g-C_3N_4	Thermal exfoliation of bulk g-C_3N_4 synthesized from thermal polycondensation of dicyandiamide	330/448	7.98[a]	[76] (2012)
Nanosheet g-C_3N_4	Liquid exfoliation of bulk g-C_3N_4 synthesized from thermal polycondensation of melamine	377/460	7.3	[81] (2015)

[a] Calculated from short, medium, and long lifetimes and their relative intensity.

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