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Supplementary Data

Visible-light-driven activation of persulfate over cyano and hydroxyl groups co-

modified mesoporous g-C₃N₄ for boosting bisphenol A degradation

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EXPERIMENTAL SECTION

Catalyst Characterization.

The X-ray diffraction (XRD) patterns were performed on a Scintag-XDS-2000 diffractometer (PANalytical B.V., Netherlands) with Cu K α radiation (λ = 0.15418 nm). Surface electronic states and chemical compositions were analyzed through Xray photoelectron spectroscopy (XPS) on a VG Scientific ESCALAB Mark II spectrometer (V.G. Scientific Ltd., UK). Elemental analyses (EA) of different samples were measured on a Vario EL III element analyzer. Fourier transform infrared (FTIR) spectra on KBr pellets of catalysts were carried out on a Bruker Tensor 27 FTIR spectrophotometer (Bruker, Germany) with a resolution of 4 cm⁻¹. The field emission-scanning electron microscopy (FE-SEM) images were obtained by using a SU8010 microscope (Hitachi, Japan) and high resolution transmission electron microscopy (HRTEM) images were recorded on a Hitachi-7650 microscope (Hitachi, Japan) at 200 kV. N₂ adsorption and desorption isotherms were obtianed on a ASAP 2020 HD88 analyzer and the corresponding specific surface areas were examined by the Brunauer-Emmett-Teller (BET) equation. UV-vis diffuse reflectance spectra (UV-vis DRS) were recorded at a Cary 7000 spectrophotometer (Agilent, USA) with an integrating sphere using BaSO₄ as the reference. The electron paramagnetic resonance (EPR) of solid powder samples and DMPO-SO4-, DMPO-'OH, DMPO-'O₂⁻ in solution were measured on a Bruker A300-10/12 spectrometer (Bruker, Germany). Photoluminescence (PL) emission spectra were implemented via a CARY ECLIPSE fluorescence spectrophotometer at the excitation wavelength of 350 nm. The photoelectrical properties were measured on a PGSTAT302N electrochemical workstation (Metrohm, Switzerland) with a standard three-electrode cell system containing a saturated calomel electrode (SCE) as the reference electrode, DMCN/FTO ($2.0 \times 1.0 \text{ cm}^2$) as the working electrode and Pt foil as the counter electrode. Na₂SO₄ solution ($0.1 \text{ mol} \cdot \text{L}^{-1}$) was used as the electrolyte. The transient photocurrent response was evaluated under pulsed light irradiation (light on or off cycles: 60 s). Electrochemical impedance spectroscopy (EIS) was conducted with the frequency range from 100000 - 0.01 Hz. The Mott-Schottky (M-S) plots were obtained in the dark at 1000 Hz.



Fig. S1. Degradation of BPA under visible light irradiation (420 nm $\leq \lambda \leq$ 780 nm) over DMCN-3.5 synthesized with different stirring time. Reaction conditions: [BPA]₀= 20 mg·L⁻¹, [catalyst]₀= 0.5 g·L⁻¹, [PS]₀= 1 g·L⁻¹, initial pH 6.2.



Fig. S2. (A) XRD patterns and (B) XPS survey spectra of different catalysts.



Fig. S3. (A) Chemical kinetic curves for BPA degradation under various conditions; (B) TOC removal rates in different catalytic systems: (a) DMCN-3.5/Vis, (b) bulk g- $C_3N_4/PS/Vis$, (c) DMCN-1/PS/Vis, (d) DMCN-2/PS/Vis, (e) DMCN-3.5/PS/Vis and (f) DMCN-5/PS/Vis; And (C) Chemical kinetic curves for BPA degradation over different catalysts by PS activation. Reaction conditions: [BPA]₀ = 20 mg·L⁻¹, [Catalyst] = 0.5 g·L⁻¹, [PS] = 1.0 g·L⁻¹, initial pH 6.2, visible light irradiation.



Fig. S4. Mott-Schotty plots of bulk $g-C_3N_4$ and DMCN-3.5 measured at a frequence of 100 Hz in the dark.



Fig. S5. DMPO spin-trapping EPR spectra in methanol dispersion for DMPO- O_2^{-} in the DMCN-3.5/PS/Vis system.

Samples	$S_{BET}(m^2 \cdot g^{-1})$	Pore volume ($cm^3 \cdot g^{-1}$)	Pore diameter (nm)
Bulk g-C ₃ N ₄	5.23	0.03	20.43
DMCN-1	82.43	0.25	12.13
DMCN-2	163.46	0.43	10.54
DMCN-3.5	212.32	0.57	10.73
DMCN-5	298.30	0.71	9.48

Table S1 BET surface areas and pore volumes of bulk $g-C_3N_4$ and DMCN catalysts.

Samplas	Contents (%) of different oxygen species			
Samples	H ₂ O (O ₁ , 532.3 eV)	(N) ₂ C–OH (O ₂ , 531.4 eV)		
Bulk g-C ₃ N ₄	100	0		
DMCN-1	70.33	29.67		
DMCN-2	66.53	33.47		
DMCN-3.5	64.12	35.88		
DMCN-5	61.12	38.88		

Table S2 Summarized O 1s XPS data for bulk $g-C_3N_4$ and DMCN catalysts.

	Contents (%) of	different carbon species			
Samples	C–C, C=C	$C-NH_x$	N-C=N	С–О	
	(C ₁ , 284.8 eV)	(C ₂ , 286.3 eV)	(C ₃ , 288.3 eV)	(C ₄ , 289.0 eV)	
Bulk g-C ₃ N ₄	10.02	5.37	84.61	0	
DMCN-1	6.44	2.36	84.97	6.23	
DMCN-2	4.21	2.73	85.12	7.94	
DMCN-3.5	4.56	3.19	84.01	8.24	
DMCN-5	3.44	3.76	83.26	9.54	

Table S3 Summarized C 1s XPS data for bulk g-C $_3N_4$ and DMCN catalysts.

	Contents (%) of different nitrogen species				
Samples	C-N=C	$N-(C)_3$	C–N–H	Charging	
1	(N ₁ , 398.8 eV)	$(N_2, 400.3 \text{ eV})$	(N ₃ , 401.3 eV)	effect	
				(N ₄ , 404.5 eV)	
Bulk g-C ₃ N ₄	67.74	16.35	11.84	4.07	
DMCN-1	67.58	16.75	11.01	4.66	
DMCN-2	68.04	16.82	10.47	4.67	
DMCN-3.5	69.52	17.16	8.64	4.73	
DMCN-5	69.69	17.82	7.70	4.79	

Table S4 Summarized N 1s XPS data for bulk $g-C_3N_4$ and DMCN catalysts.

Sample	N_2/N_1	N_3/N_1	C_2/C_3	C_{4}/C_{3}	O_2/O_1
Bulk g-C ₃ N ₄	0.241	0.175	0.063	0	0
DMCN-1	0.247	0.163	0.028	0.073	0.422
DMCN-2	0.247	0.154	0.032	0.093	0.503
DMCN-3.5	0.247	0.124	0.038	0.098	0.560
DMCN-5	0.256	0.110	0.045	0.115	0.636

Table S5 Distribution state for O, C and N elements derived from the detail XPS investigation.

Scavengers	RSs Quenched	$k (\min^{-1})$
No scavenger	/	0.317
EDTA-2Na	h^+	0.027
$K_2Cr_2O_7$	e⁻	0.058
MeOH	SO_4 -+ ·OH	0.074
TBA	•ОН	0.256
N ₂	O ₂ •-	0.296
MeOH+ N ₂ + EDTA-2Na	SO_4 ·-+ ·OH+ O_2 ·-+ h^+	0.004

Table S6 Scavengers used, RSs quenched and the corresponding values of k_{app} for BPA degradation in the DMCN-3.5/PS/Vis system.