

Supporting Information

Surface defect engineering and morphology control of graphitic carbon nitride synergistically improved photocatalytic performance

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2. Experimental

2.1 Characterization

The X-ray diffraction (XRD) patterns of the prepared samples are collected on a powder X-ray diffractometer (Cu K α radiation source, D8, Bruker). Fourier-transform infrared (FT-IR) spectra are registered on Thermo-Smart-iTR at room temperature. The morphology is characterized by transmission electron microscopy (TEM) using JEM-1400. The specific surface area and porosity are measured by an ASAP 2020 HD88 Surface Area and Porosity Analyzer. X-ray photoelectron spectroscopy (XPS) measurements are conducted via a Thermo Scientific K-Alpha spectrometer with a monochromatic Al K α source at 1486.6 eV to analyze the surface electronic states. Electron paramagnetic resonance (EPR) spectra are obtained using A300-10/12, Specord 2,450 spectrometer at 77 K. UV-vis diffuse reflectance (DRS) spectra are registered by using UV-vis spectrophotometer (Hitachi UV-3010) with BaSO₄ as standard. Photoluminescence (PL) measurements are conducted on a HITACHI F-7100 fluorescence spectrophotometer at room temperature with excitation wavelength at 350 nm. The active species could be detected by Electron spin resonance (ESR) with (MiniScope MS4000) using 5, 5-dimethyl-1-pyrroline N-oxide (DMPO) as free radical trapping agent. Total organic carbons are measured on a multi N/C 3100 (AnalytikJena, Germany) TOC analyzer. Atomic force microscopy (AFM) morphologies are collected on Dimesion Edge.

2.2 Photocatalytic activity tests

The photocatalytic performance of samples are estimated by degradation of the RhB solution under visible light. A 250 W Xe lamp with a filter ($\lambda > 420$ nm) is served as visible light source. In a typical degradation experiment, 0.05 g of catalyst powder is uniformly dispersed in RhB aqueous solution (50 ml, 10 mg/L) by ultrasonic dispersion and stirred in the dark for 30 min to establish the adsorption-desorption equilibrium. Then turn on the Xe lamp with continuous stirring and use cold flowing water to maintain 25 °C of the photoreactor. In every irradiation interval, 3 mL suspension is sampled and centrifuged. Finally, the concentration of RhB

solution is analyzed at 554 nm absorbance value by the UV-vis spectrophotometer.

2.3 Electrochemical measurements

Photoelectrochemical measurements are implemented by an electrochemical working station (CHI760E) with a standard three-electrode system using the Ag/AgCl as reference electrode, the Pt foil as counter electrode, and the samples coated at FTO glass as working electrode, respectively. The working electrode is prepared by suspension drop method. First, the sample is uniformly dissolved in a mixed solution of Nafion and ethanol to obtain a slurry, and then the slurry is suspended in the middle of the FTO glass. The transient photocurrent response and Electrochemical Impedance Spectroscopy (EIS) test of the catalysts are carried out in 0.5 M Na₂SO₄ aqueous solution.

2.4 Active species trapping experiments

To explore the reactive species involved in the degradation process over CNDNS, active species trapping experiments are carried out. The ethylenediamine tetraacetic acid disodium salt (EDTA-2Na, 1 mM), isopropanol (IPA, 1 mM), and 1,4-benzoquinone (BQ, 1 mM) are usually selected as the scavengers for trapping holes (h⁺), hydroxyl (\cdot OH), and superoxide free radicals (\cdot O₂⁻), respectively. The experiment process is based on the above photocatalytic activity.

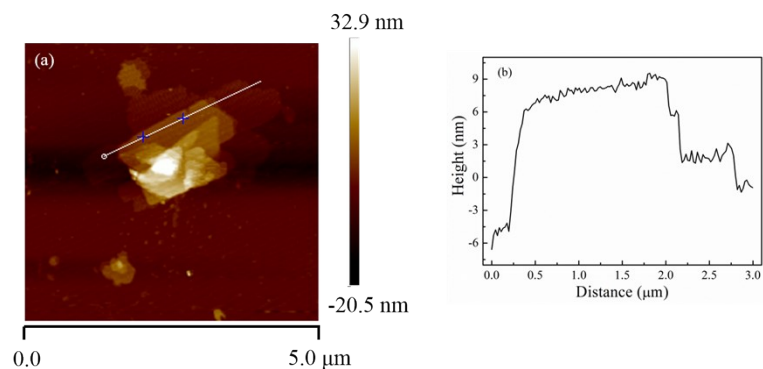


Figure S1. AFM (a) and corresponding height images (b) of CNDNS-2.

Table S1. The detailed information of BET specific surface area and BJH pore volume.

Sample	Surface area (m ² /g)	Pore volume (cm ³ /g)
CN	5.26	0.04
CNDNS-1	35.59	0.21
CNDNS-1	106.32	0.42

Table S2. The result of N 1s XPS spectra of CN and CNDNS-2.

	CN			CNDNS-2		
	C-N=C	N-(C ₃)	C-N-H	C-N=C	N-(C ₃)	C-N-H
FWHM						
Area	1.285	1.699	0.932	1.217	1.740	1.076
Molar ratio	219561.5	54861	12514.89	209647.7	66275.2	18088.8
(C-N=C/N-(C ₃))	4.0			3.16		

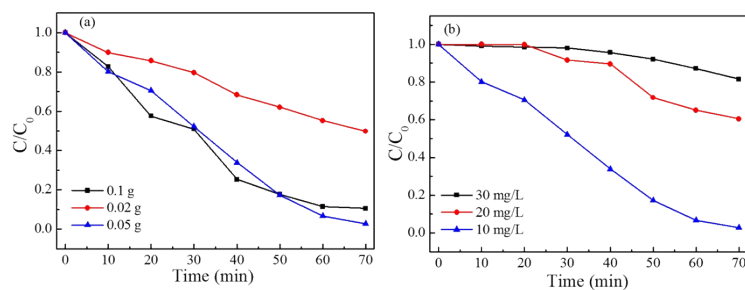


Figure S2. (a) Diagram of degradation efficiency by adjusting the amount of catalyst. (b) Diagram of degradation efficiency by adjusting the concentration of pollutants.

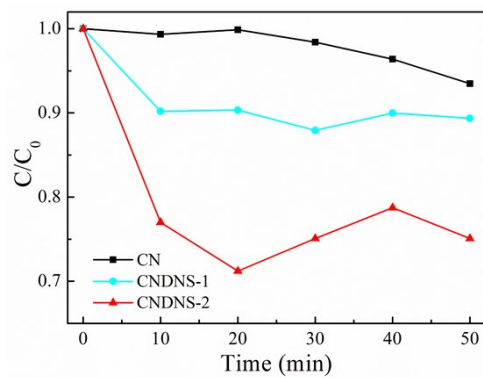


Figure S3. Absorption properties of RhB in the dark.

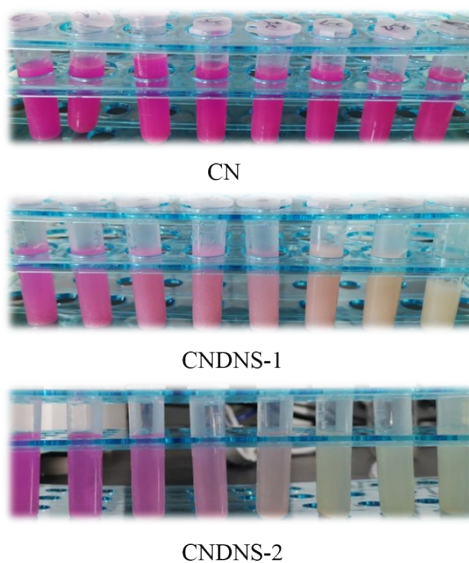


Figure S4. Effect pictures of CN, CNDNS-1, and CNDNS-2 degradation of RhB over time.

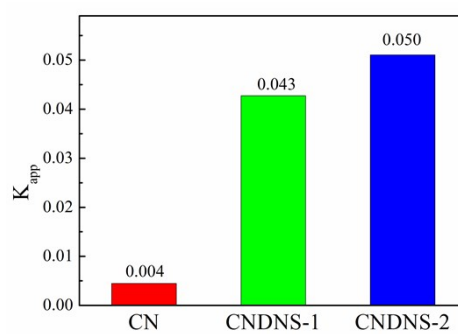


Figure S5. Values of reaction rate constants over the prepared samples.

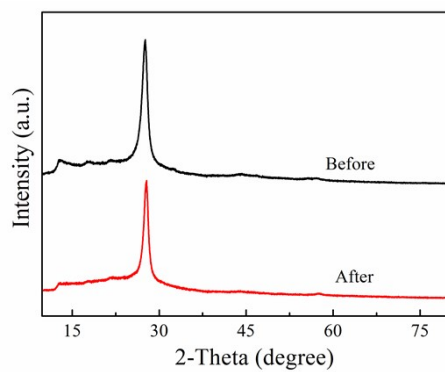


Figure S6. XRD patterns of CNDNS-2 before and after the photocatalytic reaction.

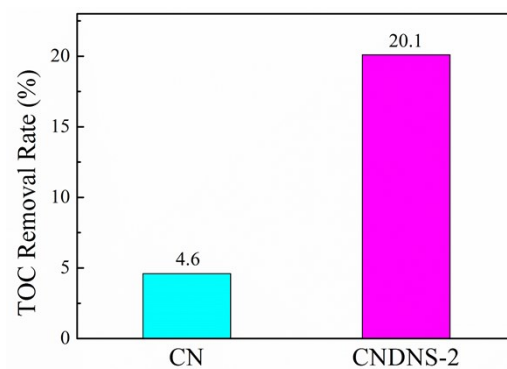


Figure S7. TOC removal ratio of RhB ($C_0 = 10 \text{ mg L}^{-1}$) over CN and CNDNS-2 under visible light irradiation.