

High crystallinity and conjugation promote polarization degree in O-doped g-C₃N₄ for removing organic pollutants

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Section S1. Degradation of methylene blue (MB) solution by g-C₃N₄ treated with different concentration of HNO₃

The degradation of MB was recorded on a multi-position photochemical reactor under a 1000 W xenon lamp simulated solar ($\lambda \geq 420$ nm) irradiation. In experiment process, 25 mg O-doped g-C₃N_{4-x} ($x = 0.1, 0.2, 0.5, 1, \text{ and } 2$ mol L⁻¹ HNO₃) were devoted in a quartz tube with 50 mL MB (1×10^{-5} mol L⁻¹), then dispersing under magnetic stirring. Then the lamp was turned on to start the photocatalysis reaction after 30 min of adsorption-desorption equilibrium. Sampling 2 ml every 30 min and kept for 3 h, then the concentration of supernatant after centrifugation was measured by using an ultraviolet visible spectrophotometer ($\lambda_{\text{max}} = 663$ nm).

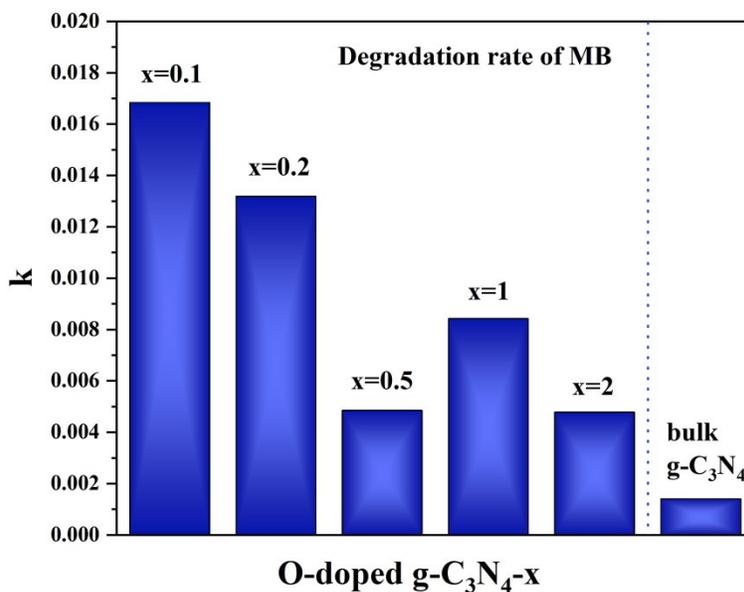


Figure S1. The degradation rate of MB solution by O-doped g-C₃N_{4-x} (x = 0.1, 0.2, 0.5, 1, and 2 mol L⁻¹ HNO₃).

Section S2. Photocatalytic performance test

Photocatalytic experiments were conducted in a multi-position photochemical reactor under a 1000 W xenon lamp simulated solar ($\lambda \geq 420$ nm). 25 mg sample was dispersed in 50 mL solution (phenol, [pHOH] = 5 ppm; bisphenol A, [BPA] = 5 ppm; diclofenac sodium, [DCF] = 5 ppm) in a quartz tube with a rotor, and mixed in an ultrasonic cleaner for 30 min to form a homogeneous composite solution. Then the mixture was transferred to a multi-position photochemical reaction apparatus and stirred in dark for 30 minutes to reach adsorption equilibrium, subsequently, the lamp ($\lambda \geq 420$ nm) was turned on to start the photocatalysis reaction. For the detection of degradation products of pHOH, BPA, and DCF, 2 ml of the supernatant were removed every 1.5 h ($t_{\text{total}} = 7.5$ h) and centrifuged (with the 0.45 μm PTFE filter) to remove the photocatalyst for determining the concentration of residual pollutants by HPLC (LC-20AT, Japan) with a tunable UV absorption detector at 270, 270 and 275 nm and a Roc-C18 column (150 mm \times 4.6 mm, 5 μm).

Besides, we conducted pHOH for the radical trapping experiments under xenon lamp irradiation. The scavenger for each reactive species was added to the reaction solution like that described for the photocatalytic experiment. The concentrations of tert-Butyl Alcohol (*t*BuOH), p-benzoquinone (PBQ) and EDTA-2Na as scavengers are 5×10^{-4} mol L⁻¹. The pseudo-first-order rate constant for contaminant photocatalytic degradation was obtained by performing a linear regression of the natural log of contaminant concentrations vs time.

Section S3. Electrochemical experiment details

The photocurrent measurement, Mott-Schottky (MS) plots, and Electrochemical impedance spectroscopy (EIS) were conducted on a CHI 660B electrochemical system (Shanghai, China) using a standard three-electrode cell with a working electrode, a platinum wire counter electrode, and a standard calomel electrode (SCE) reference electrode. Na₂SO₄ (0.5 M) was used as the electrolyte solution. The working electrode was prepared according to the following process. Twenty milligrams of the as-prepared sample was suspended in 0.5 mL of DMF, which was then dip-coated on a 10 mm × 20 mm indium-tin-oxide (ITO) glass electrode. The electrode was then annealed at 350 °C for 1 h at a heating rate of 6 °C min⁻¹. Visible light irradiation was provided by MVL-210 with a UV cutoff filter. Electrochemical impedance spectroscopy (EIS) was applied to the electrode over the frequency range 0.05 to 1×10^5 Hz with a sinusoidal ac perturbation of 5 mV.

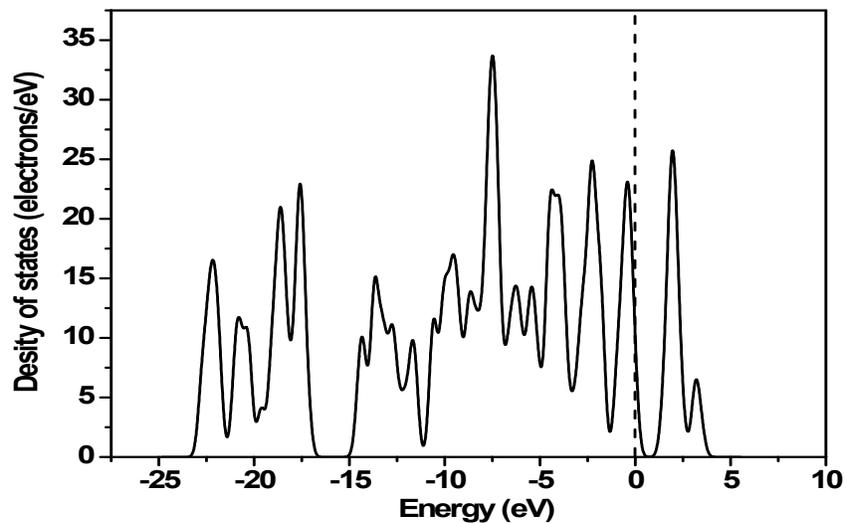


Figure S2. The first-principles calculations of energy (DOS) of bulk $g\text{-C}_3\text{N}_4$.

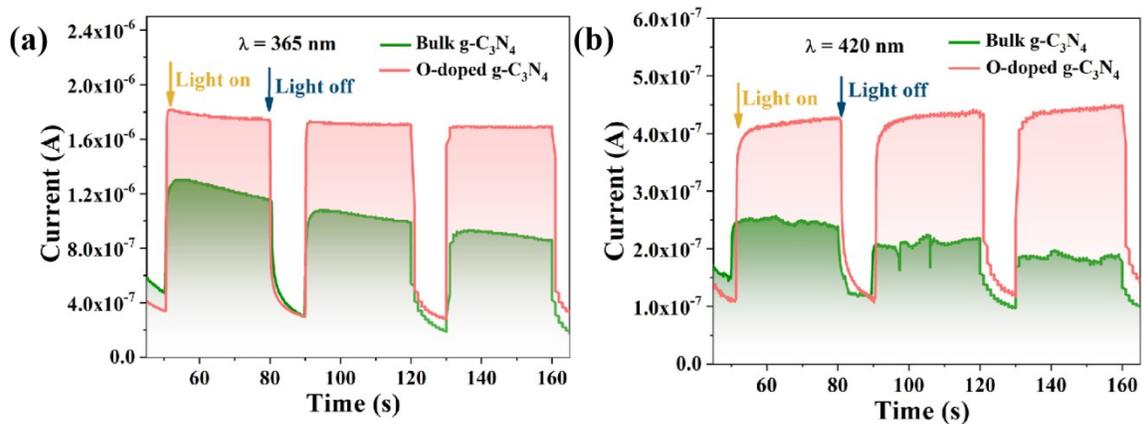


Figure S3. The photocurrent at (a) 365 nm and (b) 420 nm for three samples.

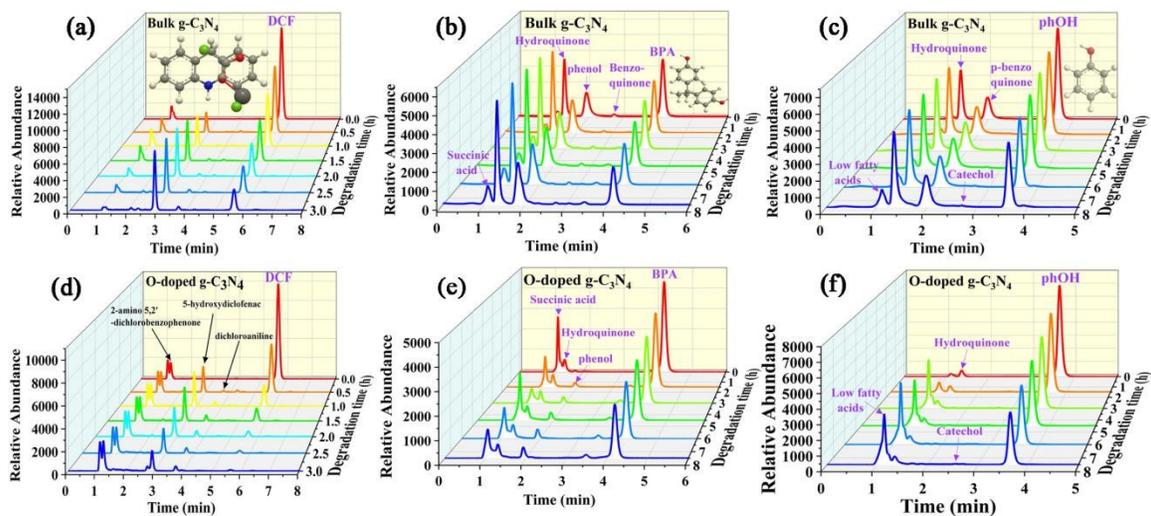


Figure S4. HPLC diagram of (a, d) DCF, (b, e) BPA, and (c, f) phOH by bulk $g\text{-C}_3\text{N}_4$ and O-doped $g\text{-C}_3\text{N}_4$.