Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Experiment

Preparation of g-C_3N_4 nanosheets. The bulk g-C_3N_4 was purchased from Carbodeon Ltd (Finland). The g-C_3N_4 nanosheets were prepared by thermal treatment of purchased bulk g-C_3N_4 under H_2 atmosphere. In detail, 30 mg of the bulk g-C_3N_4 was placed in the center of a quartz circular tube and was heated at 400 °C for 4h with a ramp rate of 3°C/min under a H_2 flow of 40 mL min^{-1}. The g-C_3N_4 nanosheets were obtained as light yellow powder.

Preparation of N-doped TiO_2. The N-doped TiO_2 was synthesized according to literature [1]. In detail, 3.6 g of TiO_2 powder was grounded with $(NH_4)_2CO_3$ (0.4 g), followed by heating at 200 °C with heating rate of 200 °C/h in air.

Characterization. Powder X-ray diffraction (XRD) patterns were obtained from a Bruker D8 Advanced diffraction-meter with Cu Kα radiation with the scanning angle ranging from 10° to 60°. Transmission electron microscopy (TEM) images were acquired using JEOL JEM2100 microscopy at an acceleration voltage of 200kV. Atomic force microscopy (AFM) was performed on Thermo microscope auto probe CP research system. X-ray photoelectron sprctra (XPS) were obtained on a RBD upgraded PHI-5000C ESCA system (Perkin Elmer) with Mg Kα radiation (hν=1253.6 eV). The Fourier transform infrared (FT-IR) experiment was carried out on a Nexus 870 spectrometer. ^{13}C and ^{1}H cross-polarisation magic angle spinning nuclear magnetic resonance spectra were recorded by a Bruker Avance 400 spectrometer at room temperature with a 4 mm triple resonance Bruker MAS probe spinning at 12 kHz, and the number of scan is 5000 and 32, recycle time is 8 and 0.5 s respectively. UV-vis diffuse reflectance spectroscopy was taken on a Hitachi U-3010 UV-vis spectrometer. Photoluminescence spectra (PL) were measured on a jobinYvon SPEX Fluorolog-3-P spectroscope. Photocurrent was acquired on a CHI 660B electrochemical workstation in a standard three-electrode system under illumination using a 300 W Xe lamp. Time-resolved fluorescence decay spectra were performed by an Edinburgh FLSP920 spectrophotometer with the excitation wavelength at 340 nm and the emission wavelength at 460 nm.

Photocatalytic activity measurement. The photocatalytic activities of bulk g-C_3N_4

and g-C_3N_4 nanosheets were tested by the degradation of methylene blue (MB) under visible light irradiation. In detail, 0.08 g catalyst was added into 200 mL of 10 mg L^{-1} MB solution in a quartz tube reactor at 25 °C. The mixture was kept stirring in dark for 60 min to reach adsorption-desorption equilibrium before a 500 W Xe lamp was turned on. At given time intervals of irradiation, 2 mL of suspension was withdrawn, and then filtered to determine the residual concentration of MB solution spectrophotometrically at 664 nm.

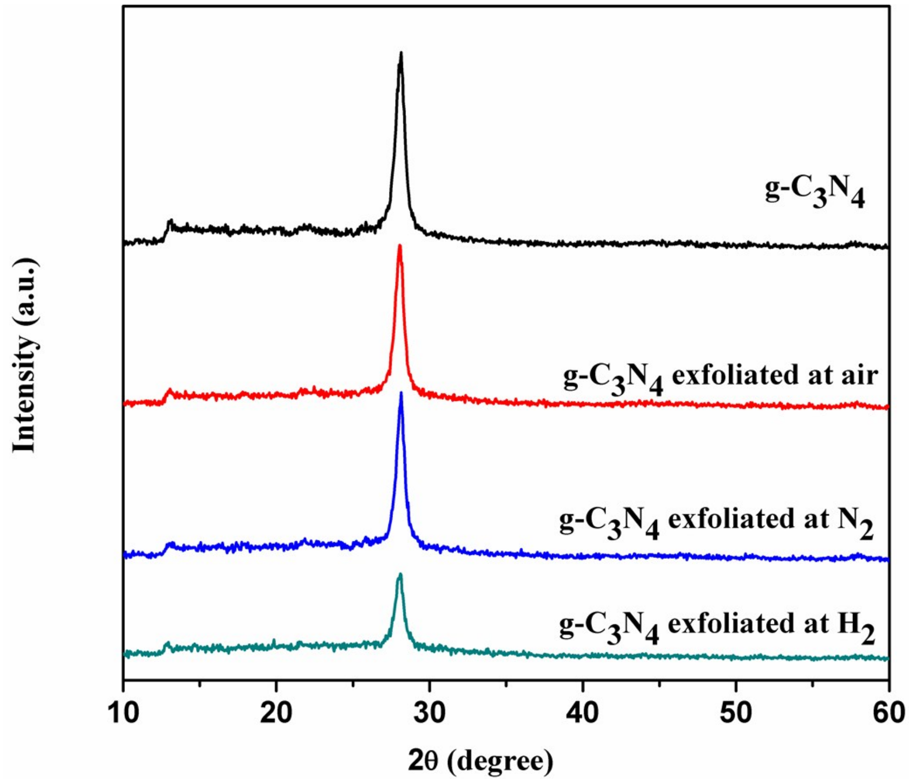

Fig S1 XRD patterns of bulk g-C_3N_4 and g-C_3N_4 exfoliated under air, N_2 and H_2 atmosphere.

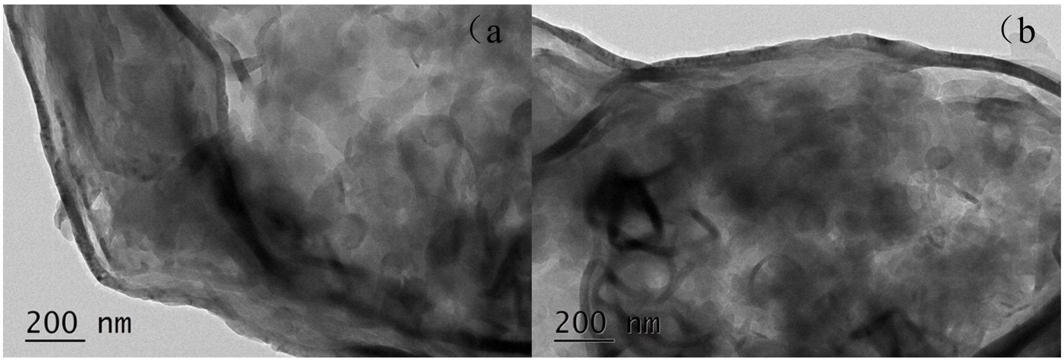

Fig S2 TEM images of g-C_3N_4 exfoliated under (a) air and (b) N_2 atmosphere.

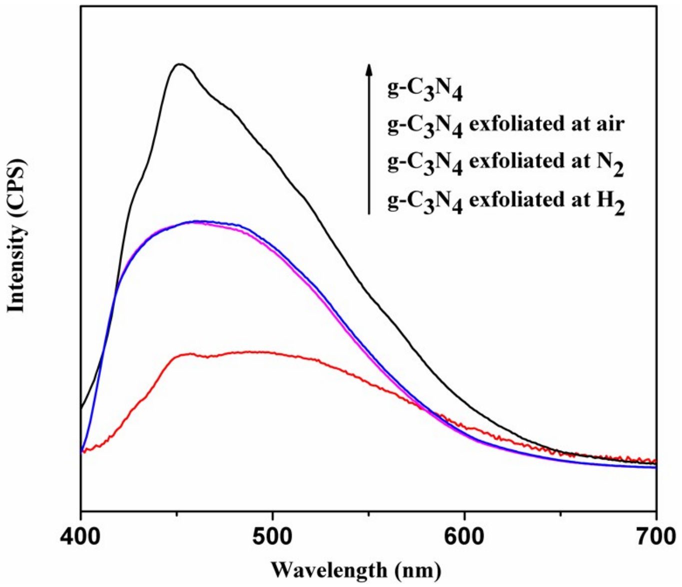

Fig S3 Photoluminescence (PL) spectra of bulk g-C_3N_4 and g-C_3N_4 exfoliated under air, N_2 and H_2 atmosphere.

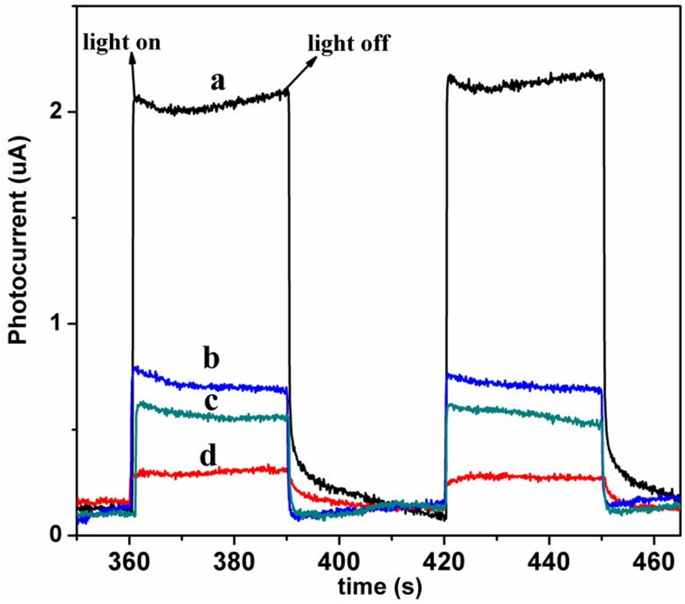

Fig S4 Transient photocurrent response of bulk g-C_3N_4 and g-C_3N_4 exfoliated under different atmosphere (a: g-C_3N_4 exfoliated under H_2, b: g-C_3N_4 exfoliated under N_2, c: g-C_3N_4 exfoliated under air and d: bulk g-C_3N_4).

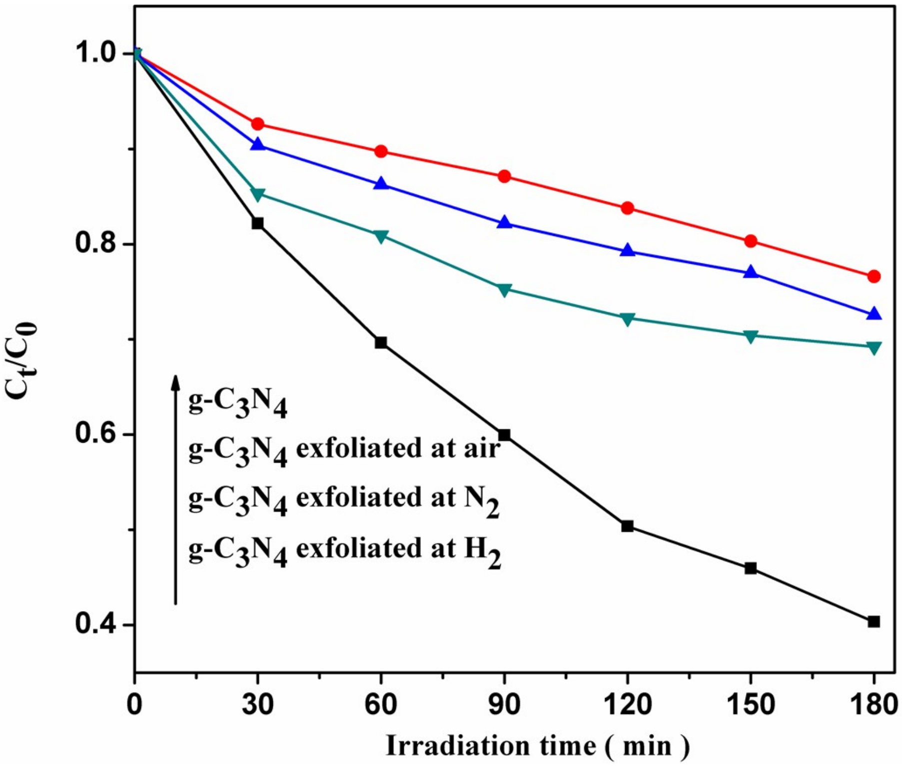

Fig S5 Photocatalytic degradation of MB over bulk g-C_3N_4 and g-C_3N_4 exfoliated under air, N_2 and H_2 atmosphere.

Table S1. C species content (mol %) of the samples based on XPS results.

Photocatalyst	C=C	C-NH_2/C-NH	C=N
bulk g-C_3N_4	17.93	23.94	58.13
g-C_3N_4 nanosheets	7.56	14.83	77.61

Table S2. C, N and H content (wt.%) of the samples according to elemental analysis.

Photocatalyst	C wt.%	N wt.%	H wt.%	C/N (molar ratio)	C/H (molar ratio)
bulk g-C_3N_4	35.37	62.28	1.82	0.66	1.63
g-C_3N_4 nanosheets	34.97	61.05	1.89	0.68	1.54

Reference:

[1] I. C. Kang, Q. Zhang, S. Yin, T. Sato and F. Saito, *Environ. Sci. Technol.*, 2008, **42**, 3622.