Supporting information

1. Experimental section

1.1 Materials

Natural graphite powder (325 mesh) was commercially obtained from Alfa-Aesar, TiO_2 (P25, 20% rutile and 80% anatase) was purchased from Degussa, melamine were purchased from Aldrich. Concentrated sulfuric acid (98%, H_2SO_4) and potassium permanganate ($KMnO_4$) were purchased from Shanghai Chemical Reagent Co., Ltd (China). Sodium nitrate ($NaNO_3$), polyethylene glycol (PEG), potassium persulfate ($K_2S_2O_8$), phosphorus pentoxide (P_2O_5), hydrogen peroxide (30%, H_2O_2) and MB were purchased from Sinopharm Chemical Reagent Corp (China). Double distilled water was used during the experimental process. All reactants were of analytical purity and used as received without further purification.

1.2 Preparation of GO

GO was synthesized by the modified Hummers' method.[1] In detail, 3 g of graphite was put into a mixture of 2.5 g of $K_2S_2O_8$, 12 mL of concentrated H_2SO_4 and 2.5 g of P_2O_5. The solution was heated to 80 °C and kept stirring for 4.5 h in oil bath. Then the mixture was diluted with 500 mL of water, and the product was obtained by filtering using 0.2 μm Nylon film and dried under ambient condition. Thereafter, 15 g $KMnO_4$ was added gradually with stirring, to prevent the temperature of the mixture from exceeding 10 ^{o}C. The ice bath was then removed and the mixture was stirred at 35 ^{o}C for 2 h. The reaction was terminated by adding 700 mL of distilled water and 20 mL of 30 % H_2O_2 solution. Finally, the GO was recovered by filtration and drying.

1.3 Synthesis of g-C_3N_4 nanosheet

Pure g-C_3N_4 nanosheet was synthesized by directly heating melamine. 5 g melamine powder was put into an alumina crucible and heated to 500 ^{o}C for 2 h with a heating rate of 2 ^{o}C/min. After cooling to room temperature, g-C_3N_4 was obtained in a powder form.

1.4 Preparation of GO/g-C_3N_4 hybird

GO/g-C_3N_4 hybrid photocatalyst was prepared via sonochemical approach. Typically, 0.1 g GO were initially dissolved in 100 mL water. And then, a certain amount of g-C_3N_4 powder was added into the above solution. The suspension was ultrasonicated for 1 h and then dried at 60℃ for 6 h. With this method, GO/g-C_3N_4 composite photocatalysts was obtained. To investigate the effect of GO content on the photocatalytic performance rates of GO/g-C_3N_4 composites, the weight percentages of GO to g-C_3N_4 were varied from 0 to 7 (0, 1, 3, 5 and 7 wt%) by varying the weight of g-C_3N_4, and the samples were presented as x%GO/g-C_3N_4, where x is the weight content of GO. The physically mixed sample of 95% g-C_3N_4 nanosheets and 5% GO (presented as 5%GO+C_3N_4) was prepared by hand gridding GO and g-C_3N_4 powder.

1.5 Analytical and testing instruments

Scanning electron microscopy (SEM) images were recorded by Quanta 400FEG at an accelerating voltage of 30 kV. High resolution transmission electron microscopy (HRTEM) of samples was performed using Tecnai G2 F20 S-Twin with an accelerating voltage of 200 kV. Atomic force microscopy (AFM) was conducted using a Bruker Multimode8, the frequency is 0.801 Hz, the samples were prepared by dropcasting dilute mixtures of samples in ethanol onto heated, freshly cleavedmica substrates. An etched silicon tip was used as a probe to image the samples. The X-ray photoelectron spectra (XPS) of GO/g-C_3N_4 hybrid were measured using a Kratos AXIS Ultra DLD X-ray photoelectron spectrometer. X-ray diffraction (XRD) data for samples were collected using a Rigaku D/MAX 24000 diffractometer with Cu Kα radiation. The infrared absorption spectra were recorded over the frequency range from 400 to 4000 cm^{-1} using a Nicolet 6700 FT-IR spectrophotometer. UV-vis diffuse reflectance spectroscopy (DRS) measurements were carried out using a Hitachi UV-3600 UV-vis spectrophotometer equipped with an integrating sphere attachment. The analysis range was from 250 to 800 nm, and $BaSO_4$ was used as reflectance standard. The photoluminescence (PL) spectra of the photocatalyst were measured by FLS920 combined fluorescene lifetime and steady state spectrometer with excitation wavelength of 325 nm. Wavelength of LED light was measured by PMS-50, light power was performed by UV-2000Z, voltage and current were performed by GPS-2303. The Brunauer-Emmett-Teller (BET) specific surface area values were determined by using nitrogen adsorption data at 77 K obtained by a Micromeritics ASAP 2010 system with multipoint BET method. The photoelectrochemical measurements were measured on CHI-660D electrochemical system, using a conventional three-electrode cell. The counter and the reference electrodes were platinum wire and saturated calomel electrode (SCE), respectively. The electrolyte solution was 1 M Na_2SO_4. The working electrodes were prepared as follows: the 0.1 g ground sample was mixed with 0.02 g PEG and 0.5 ml distilled water was added to make slurry. The slurry was then injected onto a 1.0 cm ×1.0 cm ITO glass electrode and these electrolytes were dried at 60 °C for 2 h and then calcined at 250 °C for 4 h.

1.6 Measurement of photocatalytic activity

A 5×20 LED array was welded on copper clad laminate, and used as light source. The photocatalytic experiments were carried out in a reactor containing the 30 mL 10 mg/L aqueous solution of Methylene blue (MB) and 0.03 g photocatalysts. The distance between the LED light source and the reactor was 1 cm. Before irradiation, the suspension was magnetically stirred in the dark for 1 h to establish adsorption equilibrium under ambient conditions. Then, the mixture was exposed to the LED light irradiation. Then, the mixture was exposed to the LED light irradiation. At the given irradiation time, the concentration of MB was quantified by the absorbance, and the absorbance wavelength used in the spectrophotometer analysis to determine MB concentration was about 664 nm. The degradation efficiency was calculated as follows:

$$\eta = \frac{\mathrm{C}_0 - C}{\mathrm{C}_0} \times 100\% \qquad \text{(S1)}$$

Where C_0 is the absorbance of original MB solution and C is the absorbance of the MB solution after UV light irradiation.

The decomposition behavior of MB by photocatalytic oxidative process can be described by Langmuir-Hinshelwood rate equation, equation S2 can be seen as follows:

$$\frac{1}{r} = \frac{1}{k} + \frac{1}{kKC} \qquad \text{(S2)}$$

Where C is the the absorbance of the MB solution after LED light irradiation at time t; k, and K are the rate constant, and the equilibrium adsorption constant, respectively. Integrating equation S3 gives

$$\ln\frac{C_0}{C} = k_{app}t \qquad (S3)$$

Where C_0 is the initial dye concentration in solution, k_{app} is the apparent pseudo-first-order rate constant, and t is the reaction time.

References

1. W. S. Hummers and R. E. Offeman, *J. Am. Chem. Soc.*, 1958, **80**, 1339; G. Williams and P.V. Kamat, *Langmuir*, 2009, **25**, 13869.

As indicated in Fig.S1, 5×20 LED arrays were used as the light source of the photocatalytic degradation device, the direct current of LED light is 0.4 A and the direct voltage is about 17.0 V. Fig.S1b shows the wavelength spectrum of the single LED at the condition of 20 mA and 25 ^{o}C. The result showed that the emission center wavelength of LED is 410 nm with very narrow wavelength range.

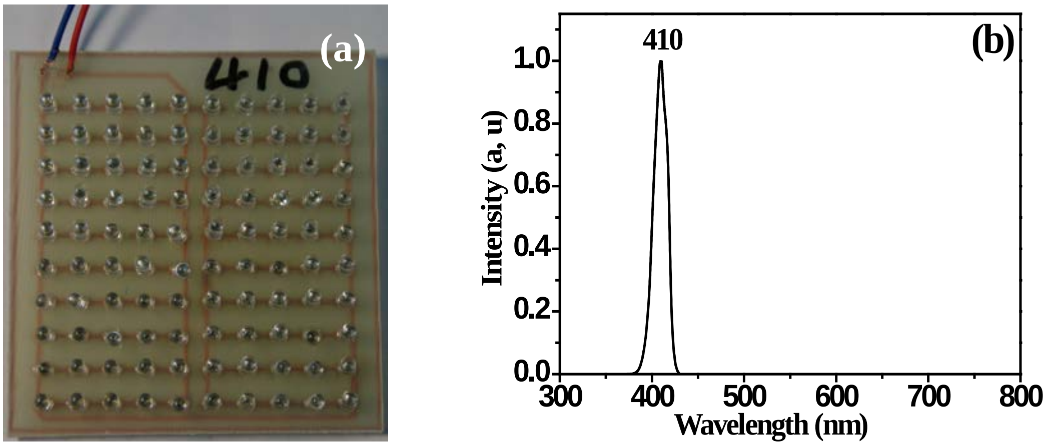

Fig.S1 (a) photograph of LED arrays and (b) the wavelength spectrum of LED lamp

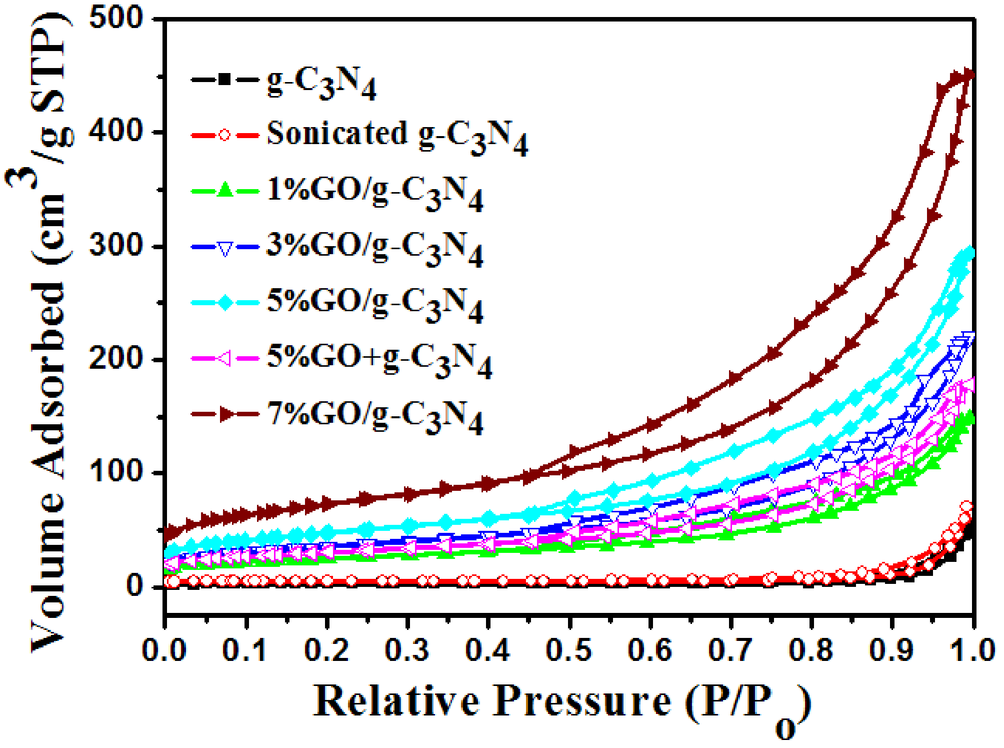

Fig.S2 Isotherms for N_2 adsorption-desorption

Table.S1 BET surface area of the as-prepared samples

Samples	BET surface area (m^2/g)
g-C_3N_4	14.3
Sonicated g-C_3N_4	16.4
1%GO/g-C_3N_4	48.6
3%GO/g-C_3N_4	112.4
5%GO/g-C_3N_4	137.3
7%GO/g-C_3N_4	161.1
5%GO+g-C_3N_4	62.4