

## Supplementary Information

*Achieving significantly enhanced visible-light photocatalytic efficiency using polyelectrolyte: the composites of exfoliated titania nanosheet, graphene, and poly(diallyl-dimethyl-ammonium chloride)*

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## **Experimental Section**

### ***Materials***

The PDDA and GO were purchased from Sigma-Aldrich and XFNANO Materials Tech Co. Ltd., respectively and TiO<sub>2</sub> was provided by Aladdin Reagent Company. K<sub>2</sub>CO<sub>3</sub>, LiCO<sub>3</sub>, HCl, MoO<sub>3</sub>, and methylene blue were purchased from Beijing Chemical Reagent Plant and (C<sub>4</sub>H<sub>9</sub>)<sub>4</sub>NOH was obtained from Sinopharm Chemical Reagent Co., Ltd.

### ***Preparation of the titania nanosheets***

The titania nanosheets were prepared as reported previously<sup>1</sup>. Briefly, The titanate crystals, K<sub>0.8</sub>[Ti<sub>1.73</sub>Li<sub>0.27</sub>]O<sub>4</sub>, was synthesized by a conventional solid-state reaction. Subsequently, the solid was treated with 1 M HCl to obtain the protonated derivative, H<sub>1.07</sub>Ti<sub>1.75</sub>O<sub>4</sub>H<sub>2</sub>O. Exfoliation of the layered titanate was conducted by intercalation of the tetrabutylammonium hydroxide solution ((C<sub>4</sub>H<sub>9</sub>)<sub>4</sub>NOH) into the layers of protonated titanate under gentle agitation.

### ***Characterization***

The morphology and microstructure of the samples were characterized on an S-4800 field emission scanning electron microscope (FE-SEM, Hitachi Ltd., Tokyo, Japan). The elemental distribution was determined by SEM (SU1510, Hitachi Ltd., Tokyo, Japan) equipped with an energy-dispersive spectrometry (EDS). Transmission electron micrographs (TEM) were acquired on the using FEI TECNAI G2 20 transmission electron microscopy. The thickness of the titania nanosheets was determined by atomic force microscopy (AFM, Dimension 3100, Veeco, USA). The X-ray diffraction (XRD) patterns of the powders were obtained by Cu K<sub>α</sub> radiation on a Rigaku diffractometer and Fourier-transform infrared (FTIR) spectra in the frequency range of 400-4000 cm<sup>-1</sup> were acquired on the 100 FTIR spectrometer (Peikin Elmer., USA). The UV-vis absorption spectra were collected in the diffused reflectance mode from powders on a PerkinElmer Lambda 35 UV-vis spectrophotometer. The spectra were collected at 200-2000 nm referenced to BaSO<sub>4</sub>. The photoelectrochemical experiments were performed on a CHI660C electrochemical workstation in a three-electrode system with the as-prepared sample as the working electrode, Pt wire as the counter electrode, and platinum-saturated calomel electrode (SCE) as the reference electrode. The visible light was generated by a 300 W Xenon lamp and 0.1 M Na<sub>2</sub>SO<sub>4</sub> aqueous solution was used as the electrolyte. The photo response of the photocatalysts was assessed by at 0.0 V. TGA was conducted from room temperature to 700 °C on a Perkin-Elmer TGA 7 in air at an increasing temperature rate of 10 °C/min. The UV-vis absorbance was monitored on the MAPADA UV-6100 UV-Vis spectrophotometer. The concentration of the titania nanosheets was determined by inductively-coupled plasms mass spectrometry (ICPMS, Perkin Elmer Optima 8300 Series). The total organic carbon (TOC) value was obtained on the total organic carbon analyzer (Shimadzu, TOC-L CPH).

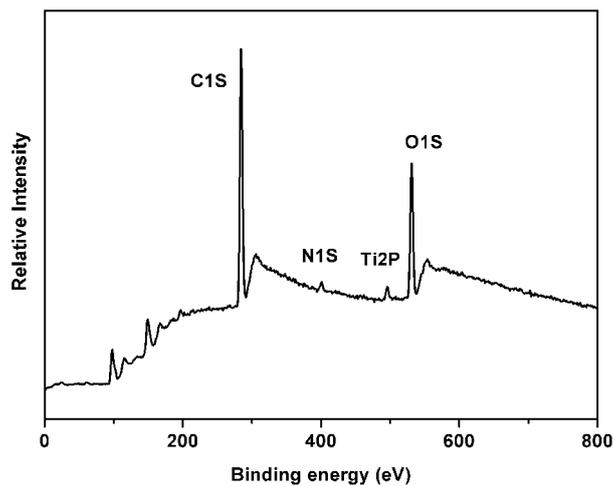


Figure S1. Survey scan of titania nanosheet.

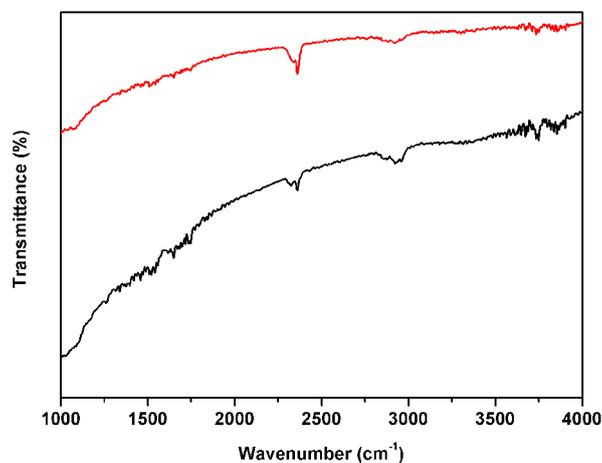


Figure S2. FTIR spectra of titania nanosheet (red line) and titania nanosheet annealing at 300°C for 1 hour (black line).

The existence of  $(C_4H_9)_4NOH$  in the titania nanosheet before and after calcination is confirmed by the doublet of the  $-CH_2-$  asymmetric and symmetric stretching vibration at  $2924\text{ cm}^{-1}$  and  $2873\text{ cm}^{-1}$ ,  $-OH$  stretching vibration at  $3722\text{ cm}^{-1}$  and C-N stretching vibration at  $1076\text{ cm}^{-1}$ .

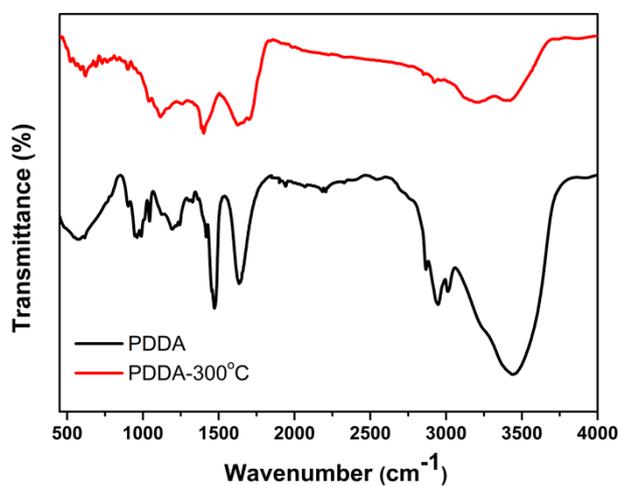


Figure S3 FTIR spectra of PDDA and PDDA calcination at 300°C for 1h

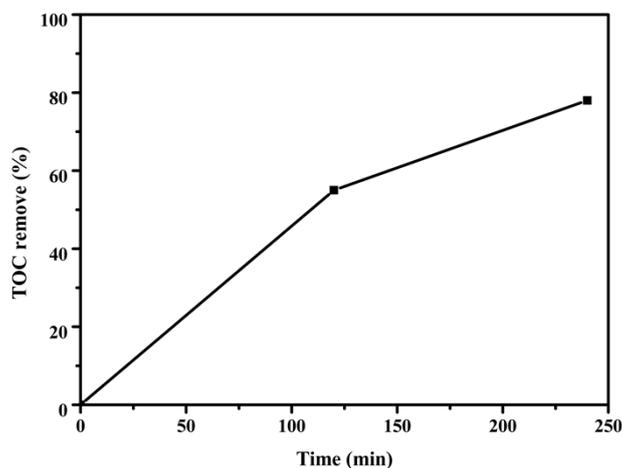


Figure S4. TOC removal rate in degradation of MB ( $3 \times 10^{-5}$  M)

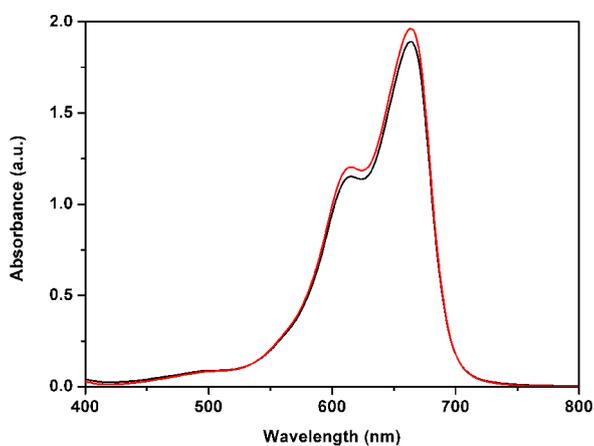


Figure S5. UV-vis absorption spectra of MB aqueous solution under visible light irradiation for 0 min (red line) and 240 min (black line) in the presence of composite before annealing.

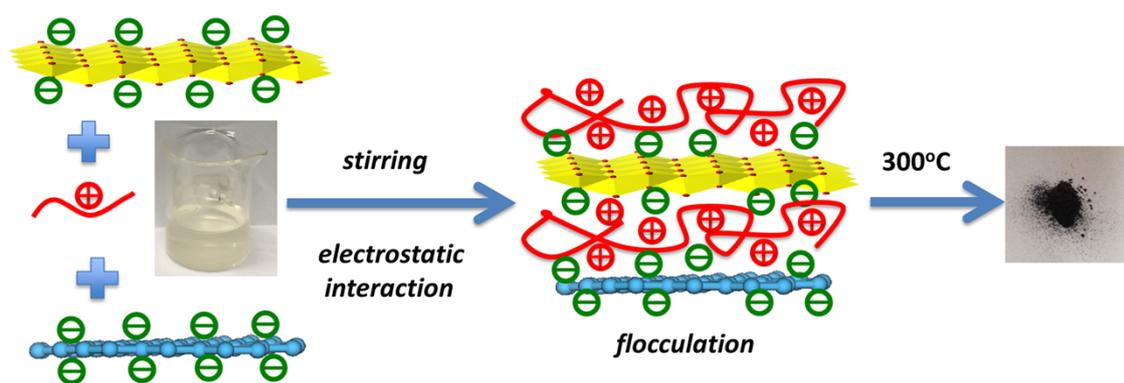


Figure S6. Transfer process of titania nanosheet from liquid to solid phase.

First, the titania nanosheet, graphene oxide and PDDA was assembled by electrostatic flocculation and collected as precipitate. Afterwards the liquid proportion in the complex catalyst was further removed by high temperature calcination.

Table S1. Elemental composition of the surface of the composite catalyst.

Element	Wt. %	Atom %
C	61.82	68.54
O	23.41	19.48
N	11.70	11.12
Ti	3.07	0.85

1 T. Tanaka, Y. Ebina, K. Takada, K. Kurashima, T. Sasaki. Chem. Mater. **2003**, 15, 3564-3568.