

## Supporting Information

### *Construction and preparation of a novel 2D metal-free few-layer BN modified graphene-like g-C<sub>3</sub>N<sub>4</sub> with enhanced photocatalytic performance*

Jiemin Gu<sup>a</sup>, Jia Yan<sup>a</sup>, Zhigang Chen<sup>a</sup>, Haiyan Ji<sup>a</sup>, Yanhua Song<sup>b</sup>, Yamin Fan<sup>a</sup>, Hui Xu<sup>a,\*</sup>, Huaming Li<sup>a,\*</sup>

<sup>a</sup> School of the Environment and Safety Engineering, Institute for Energy Research, Jiangsu University, Zhenjiang 212013, P. R. China. E-mail: xh@ujs.edu.cn. lihm@ujs.edu.cn

<sup>b</sup> School of Environmental and Chemical Engineering, Jiangsu University of Science and Technology, Zhenjiang 212003, P. R. China

#### Characterization of the photocatalysts

X-ray diffraction (XRD) analyses were carried out to detect the crystal structure of the 2D BN/g-C<sub>3</sub>N<sub>4</sub> samples. And it was radiated with Cu K $\alpha$  ( $\lambda=1.5418$  Å) in the scope of  $2\theta=10-80^\circ$  using Bruker D8 diffractometer. X-ray photoelectron spectroscopy (XPS) analysis was performed on an ESCALab MKII X-ray photoelectron spectrometer radiated by Mg K $\alpha$ . The transmission electron microscopy (TEM) images were picked up at 200 kV with a JEOL-JEM-2010 (JEOL, Japan). The energy-dispersive X-ray spectroscopy (EDS) and elemental mapping measurements were carried out to detect the elemental composition and distribution using a 10 kV acceleration voltage. Ultraviolet visible (UV-Vis) diffuse reflection spectra (DRS) were measured on an UV-Vis spectrophotometer (Shimadzu UV-2450, Japan) ranging from 200 to 800 nm using BaSO<sub>4</sub> as reflectance standard substance. The Fourier transform infrared spectra (FT-IR) of the composites were implemented using Nicolet Nexus 470 spectrometer. Photocurrents and Electrochemical impedance spectroscopy (EIS) were detected on an electro-chemical workstation (CHI 660B, Chen Hua Instrument Company, Shanghai, China). Electron spin resonance (ESR) was operated on JES FA200 (JEOL) using a Xenon lamp with a 420 nm cutoff filter as source of light, and the 5,5-dimethyl-1-pyrroline-N-oxide (DMPO) was adopted to capture the spin-trapping signals.

#### Photoelectrochemical measurements

The 2D BN/g-C<sub>3</sub>N<sub>4</sub> electrodes were prepared to investigate the separation efficiency of photo-generated holes and electrons. The photocurrent was conducted in a standard three-electrode system, where the saturated Ag/AgCl electrode was employed as an reference electrode, the platinum wire was used as a counter electrode and one indium-tin oxide glass (ITO) was utilized as a working electrode. 5.0 mg samples were spread out ultrasonically into 1.0 mL of ethanol. To prepare the sample

modified ITO electrode (named as ITO/2D BN/g-C<sub>3</sub>N<sub>4</sub>), 20  $\mu$ L of the obtained mixture (5 g L<sup>-1</sup>) was dropped on the ITO slice (0.5 $\times$ 1 cm<sup>2</sup>) and then dried. The photocurrent analyses were all measured with the fixed 0 V potential (vs. saturated Ag/AgCl). The phosphate buffered saline (pH=7.0, 0.1 mol L<sup>-1</sup>) was adopted as electrolyte solution during photocurrent measurements. And the light source came from a 500W Xe arc lamp. The Nyquist plots were measured ranging from the frequency of 100000~100 kHz. And they were implemented in the 0.1 M KCl solution containing 5 mM Fe(CN)<sub>6</sub><sup>3-</sup>/Fe(CN)<sub>6</sub><sup>4-</sup>. All electrochemical measurements aforementioned were performed at sunless conditions.

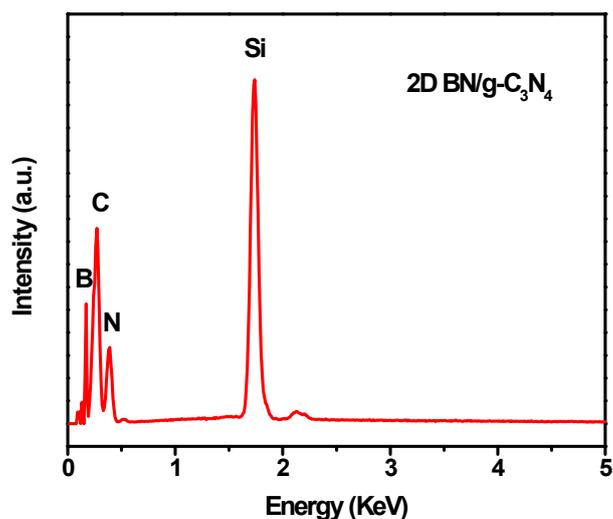


Figure S1 EDS of 2D BN/g-C<sub>3</sub>N<sub>4</sub> sample.

### FT-IR analysis

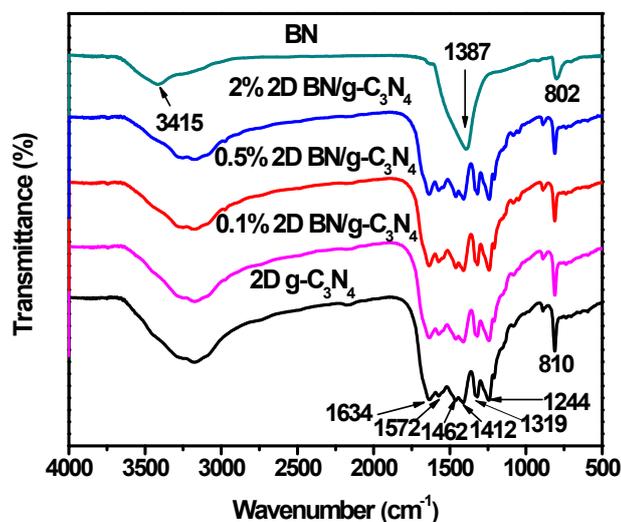


Fig. S2 FT-IR spectra of BN, 2D g-C<sub>3</sub>N<sub>4</sub> and 2D BN/g-C<sub>3</sub>N<sub>4</sub> composites.

The FT-IR spectra were explored to explore whether the introduction of BN will influence the structure and the surface groups of 2D g-C<sub>3</sub>N<sub>4</sub>. And the corresponding FT-IR spectra of the pristine 2D g-C<sub>3</sub>N<sub>4</sub>, BN and 2D BN/g-C<sub>3</sub>N<sub>4</sub> catalysts with different BN contents are exhibited in Fig. S2. The broad absorption band located near 3150 cm<sup>-1</sup> is associated to the -OH stretching vibrations from the adsorbed H<sub>2</sub>O molecules and terminal NH or NH<sub>2</sub> at the defect sites exist in the aromatic ring.<sup>1,2</sup> For bare 2D g-C<sub>3</sub>N<sub>4</sub>, the band at 810 cm<sup>-1</sup> is associated with out-of-plane C-N bending modes of triazine units. The two peaks located at 1634 and 1572 cm<sup>-1</sup> are ascribed to C=N stretching vibration, while those peaks at 1244, 1319 and 1412 cm<sup>-1</sup> belong to the aromatic C-N stretching.<sup>3,4</sup> As for bare BN, the peak at 1387 cm<sup>-1</sup> is assigned to the in plane mode of B-N stretching vibration. And it corresponds to the sp<sup>2</sup> replacement in BN.<sup>5,6</sup> Besides, the absorption band at 802 cm<sup>-1</sup> is associated with the out-of-plane B-N-B bending vibrations.<sup>7</sup> In addition, the absorption band at around 3415 cm<sup>-1</sup> is related to the stretching vibration of terminated -OH groups on its (002) facet edges of the BN, which are generated by the O-containing boric acid and urea during the synthesis process.<sup>8,9</sup> For 2D BN/g-C<sub>3</sub>N<sub>4</sub> catalysts, the characteristic peaks of BN cannot be observed, which is possibly ascribed to the small contents of BN. And all 2D BN/g-C<sub>3</sub>N<sub>4</sub> composites present similar characteristic peaks to that of 2D g-C<sub>3</sub>N<sub>4</sub>, suggesting that introducing BN did not change the structure of 2D g-C<sub>3</sub>N<sub>4</sub>.

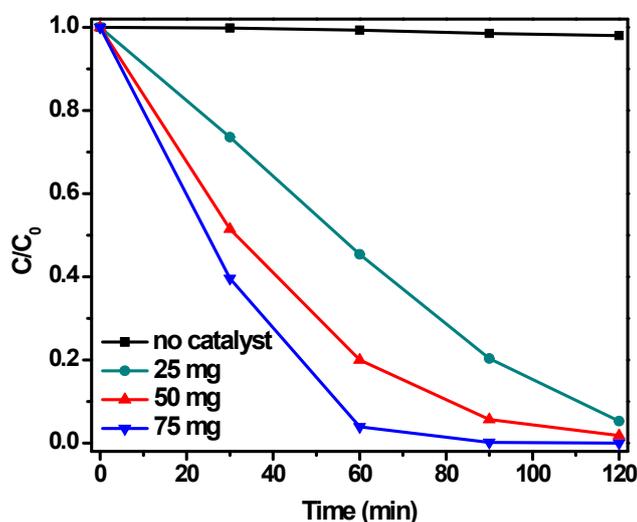


Figure S3 The photocatalytic activities towards RhB degradation with different consumptions of 0.5% 2D BN/g-C<sub>3</sub>N<sub>4</sub> samples.

The photocatalytic degradation performances towards RhB with different photocatalyst concentrations were assessed, and the corresponding results are shown in Figure S3. In the photocatalytic experiments, 0, 25, 50 and 75 mg of the as-prepared 0.5% 2D BN/g-C<sub>3</sub>N<sub>4</sub> samples were added into 50 mL 10 mg L<sup>-1</sup> of RhB

solution, respectively. The reaction conditions were all same except for the dosage of catalyst. Obviously, after irradiated for 120 min, RhB could not be degraded in the absence of catalyst, suggesting that the self-degradation of RhB could be neglected. In addition, with the increasing of the samples used in the reaction, the degradation efficiencies are improved gradually, indicating that the dosage of catalyst would greatly impact the photocatalytic performance in the degradation reaction.

## Reference

- [1] J. H. Liu, T. K. Zhang, Z. C. Wang, G. Dawson and W. Chen, *J Mater Chem.*, 2011, 21, 14398.
- [2] S. B. Yang, Y. J. Gong, J. S. Zhang, L. Zhan, L. L. Ma, Z. Y. Fang, R. Vajtai, X. C. Wang and P. M. Ajayan, *Adv. Mater.*, 2013, 25, 2452.
- [3] X. F. Li, J. Zhang, L. H. Shen, Y. M. Ma, W. W. Lei, Q. L. Cui and G. T. Zou, *Appl. Phys. A: Mater. Sci. Process.*, 2009, 94, 387.
- [4] Y. J. Wang, X. J. Bai, C. S. Pan, J. He and Y. F. Zhu, *J Mater Chem.*, 2012, 22, 11568.
- [5] J. Di, J. X. Xia, M. X. Ji, B. Wang, S. Yin, Q. Zhang, Z. G. Chen and H. M. Li, *Appl. Catal., B*, 2016, 183, 254.
- [6] C. J. Huang, C. Chen, M. W. Zhang, L. H. Lin, X. X. Ye, S. Lin, M. Antonietti and X. C. Wang, *Nat. Commun.*, 2015, 6.
- [7] D. Liu, Z. F. Jiang, C. Z. Zhu, K. Qian, Z. Y. Wu and J. M. Xie, *Dalton Trans.*, 2016, 45, 2505.
- [8] X. M. Lv, J. X. Wang, Z. X. Yan, D. L. Jiang and J. Liu, *J. Mol. Catal. A: Chem.*, 2016, 418, 146.
- [9] Q. H. Weng, B. J. Wang, X. B. Wang, N. Hanagata, X. Li, D. Q. Liu, X. Wang, X. F. Jiang, Y. Bando and D. Golberg, *ACS Nano*, 2014, 8, 6123.