Supplementary Material

# Noble-metal-free TiO<sub>2</sub> coated carbon nitride layers for enhanced visible-light-driven photocatalysis

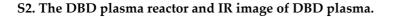
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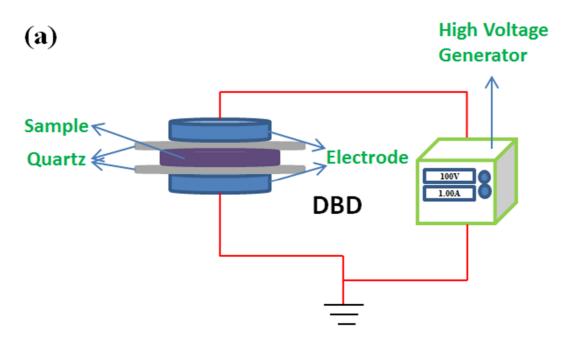
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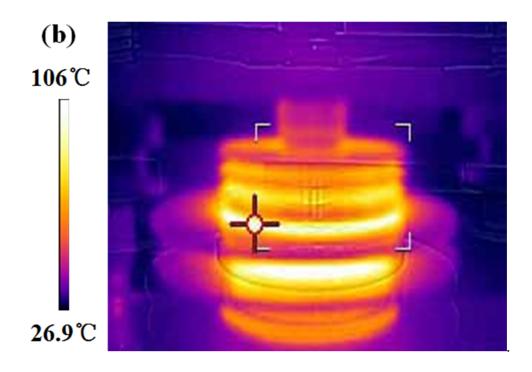
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#### S1. Synthesis of g-C<sub>3</sub>N<sub>4</sub>

g-C<sub>3</sub>N<sub>4</sub> powder was synthesized according to a classic procedure of Ma et al [1]. The pure g-C<sub>3</sub>N<sub>4</sub> powder was synthesized by heating melamine powder directly. Typically, 10 g melamine was putted into a closed alumina crucible, then heated at 550°C for 4 h with the rate of 5°C/min in the muffle furnace. The as-prepared yellow product was grounded for further use.







**Figure S1.** (a) The DBD plasma reactor and (b) IR image of DBD plasma reactor during samples preparation.

### S3. The schematic illustration for the preparation of TCN composites photocatalysts.

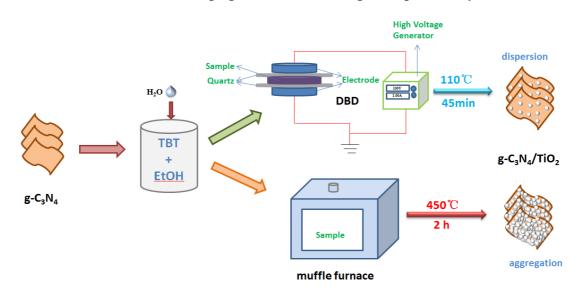


Figure S2. The schematic illustration for the preparation of TCN composites photocatalysts

# S4. Photocurrent tests of TCN50-D at different potentials.

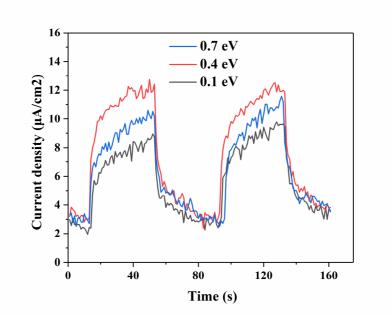
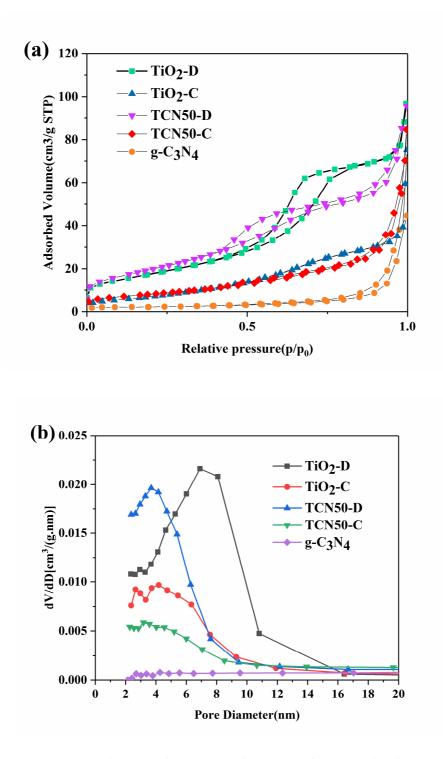


Figure S3. Photocurrent tests of TCN50-D at different potential.

# S5. Nitrogen adsorption-desorption isotherms.

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Sample	Sbet (m <sup>2</sup> /g)	Pore volume(cm³/g)	Average pore radius(nm)	Crystallite size (nm)
TiO <sub>2</sub> -D	64.6±6.1	$0.16 \pm 0.02$	2.99±0.3	14.3
TiO <sub>2</sub> -C	28.2±2.4	$0.10 \pm 0.01$	6.79±0.6	17.8
TCN10-D	$117.2 \pm 10.3$	0.17±0.02	2.29±0.2	13.7
TCN30-D	94.9±8.8	$0.16 \pm 0.02$	$2.80 \pm 0.3$	13.4
TCN50-D	72.8±6.3	$0.14 \pm 0.01$	$2.86 \pm 0.3$	12.3
TCN50-C	29.3±2.7	$0.11 \pm 0.01$	6.02±0.6	14.4
TCN70-D	43.4±4.0	$0.12 \pm 0.01$	$4.04 \pm 0.4$	11.4
TCN90-D	35.6±3.1	$0.11 \pm 0.01$	$5.38 \pm 0.5$	10.8
<b>g-</b> C <sub>3</sub> N <sub>4</sub>	7.1±0.9	$0.06 \pm 0.01$	$11.34 \pm 1.0$	/

Table S1. Surface and structural characterization of TiO<sub>2</sub>, g-C<sub>3</sub>N<sub>4</sub> and TCNX composites.



**Figure S4.** Nitrogen adsorption–desorption isotherms (a) and pore-size distribution curves (b) of  $g-C_3N_4$ , TiO<sub>2</sub> and TCN50 samples.

S6. Thermogravimetric Analysis curves

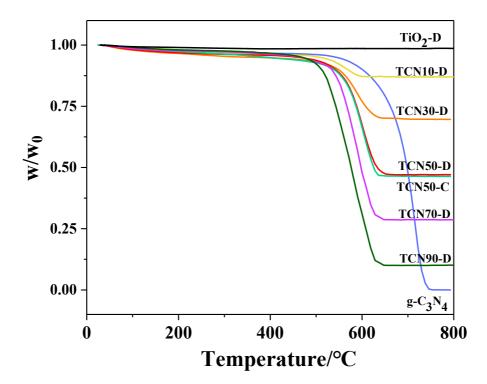
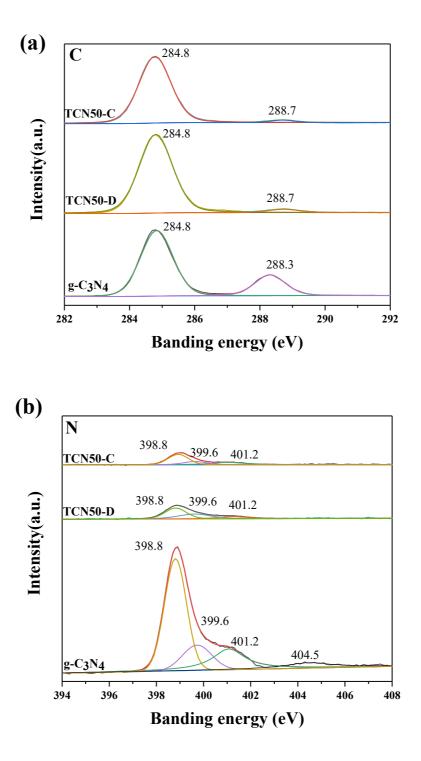
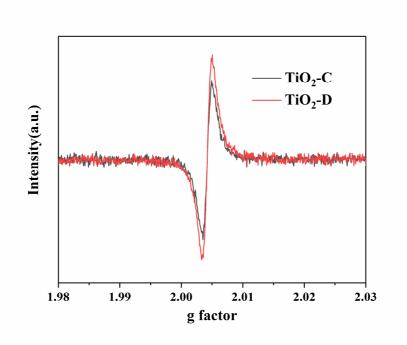


Figure S5. TGA curves of the g-C<sub>3</sub>N<sub>4</sub>, TiO<sub>2</sub>-D and TCNX composite photocatalysts.



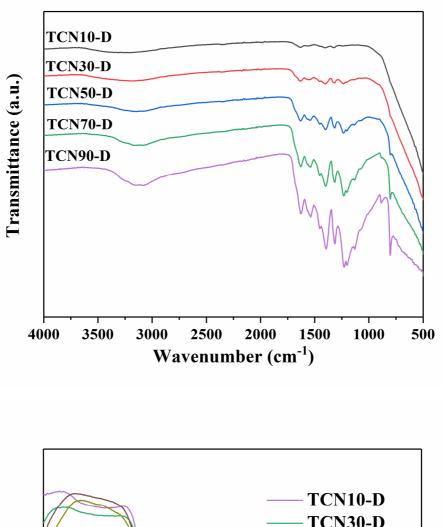
**Figure S6.** (a) The high-resolution XPS spectra C 1s, (b) N 1s of g-C<sub>3</sub>N<sub>4</sub>, TCN50-C and TCN50-D samples.

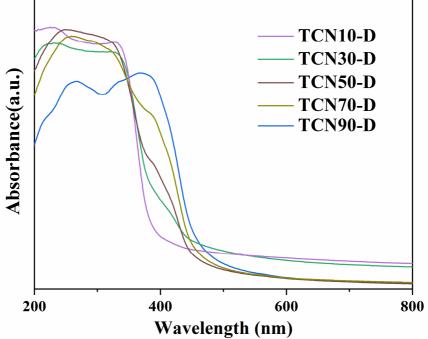
# S8. The EPR spectra of TiO<sub>2</sub>-C and D samples



**Figure S7.** The EPR spectra of TiO<sub>2</sub>-C and D samples.

S9. FTIR spectra, UV-vis DRS and XPS VB spectra of TCNX.





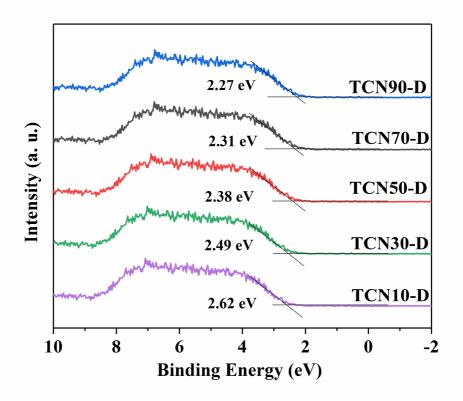
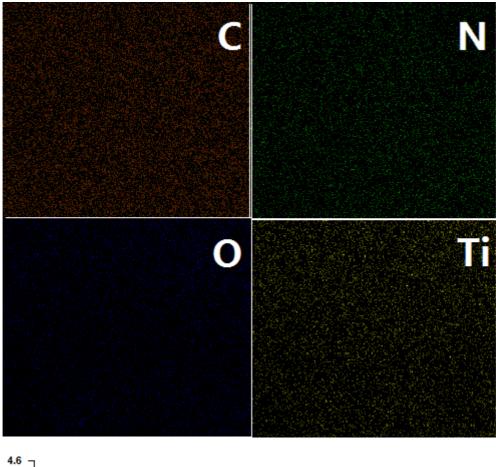


Figure S8. FTIR spectra, UV-vis DRS and XPS VB spectra of TCNX.

### S10. EDS patterns and element mapping of TCN50-D sample



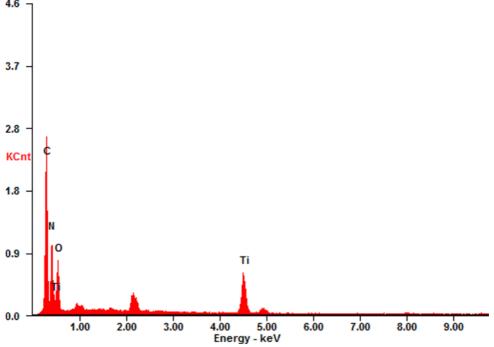
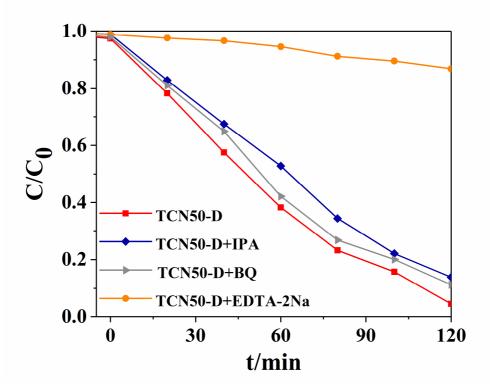


Figure S9. EDS patterns and element mapping of TCN50-D sample.



**Figure S10.** Trapping experimental of photogenerated radicals and holes in TCN50-D sample for the RhB degradation.

The photogenerated active substance trapping experiments was to determine the active substance. Before the start of the experiment of RhB degradation, a certain amount of benzoquinone, 2-propanol and ethylenediaminetetraacetic acid disodium salt were added to the mixture, corresponding to the scavenger of superoxide radicals, hydroxyl radicals and photogenerated holes, respectively.

### S12. Valence band of XPS

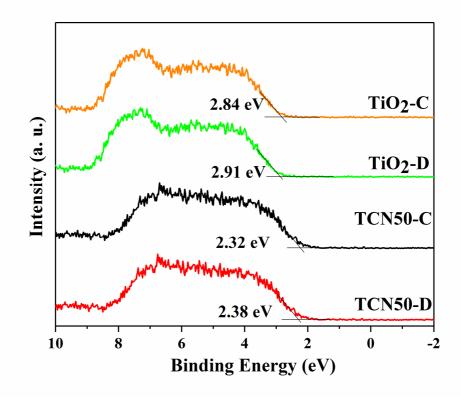


Figure S11. VB XPS of TiO<sub>2</sub>-C, TiO<sub>2</sub>-D, TCN50-C and TCN50-D.





### Reference

[1] Ma, J.; Tan, X.; Yu, T.; Li, X. Fabrication of  $g-C_3N_4/TiO_2$  hierarchical spheres with reactive {001} TiO2 crystal facets and its visible-light photocatalytic activity. *Int. J. Hydrogen Energ.* 2016, *41*, 3877-3887.

By measuring the intersection of the slope of the XPS valence band curve and the X axis, the valence band (VB) of the material can be determined.