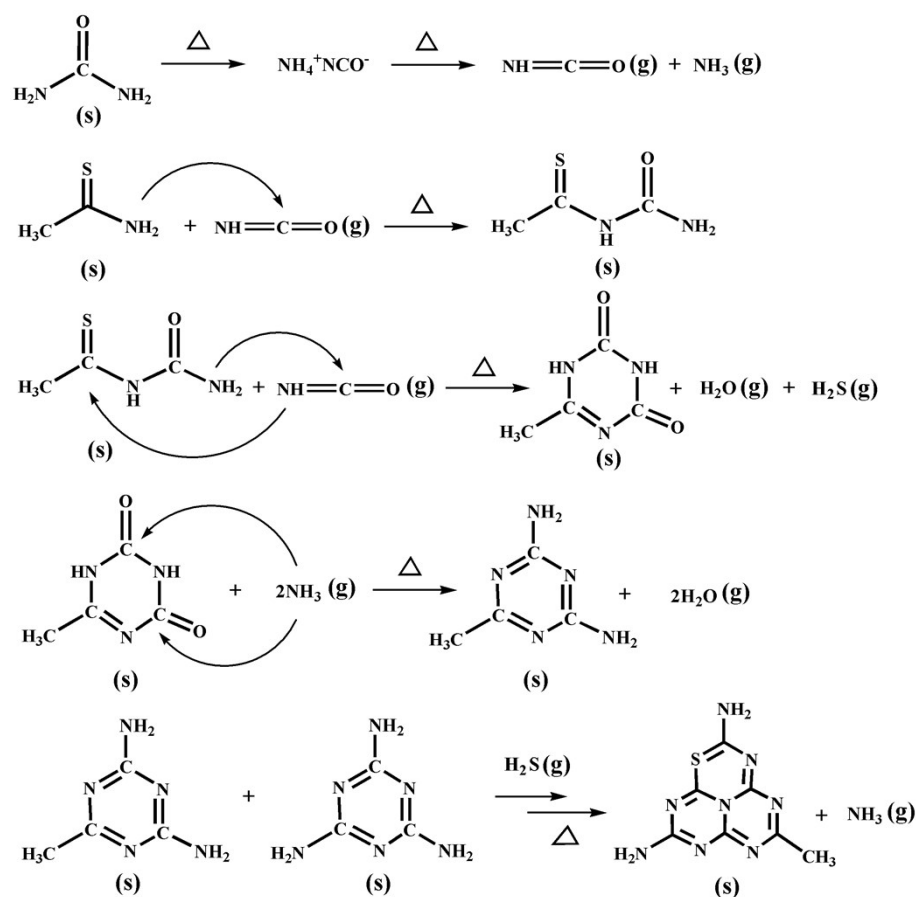


## Preparation and enhanced photocatalytic performance of sulfur doping terminal-methylated g-C<sub>3</sub>N<sub>4</sub> nanosheets with extended visible-light response

Yunfeng Li<sup>a,\*</sup>, Shuai Wang<sup>a</sup>, Wei Chang<sup>a</sup>, Luohong Zhang<sup>a</sup>, Zhansheng Wu<sup>a</sup>, Shuyan Song<sup>b,\*</sup> and Yan Xing<sup>c,\*</sup>



**Figure S1.** The proposed reaction mechanism of polymerization steps for urea and TAA.

**Table S1.** The DFT calculated overall energy ( $E_o$ ) and formation energy ( $E_f$ ) of substituting potential N atoms by S atom in melon and methylated melon.

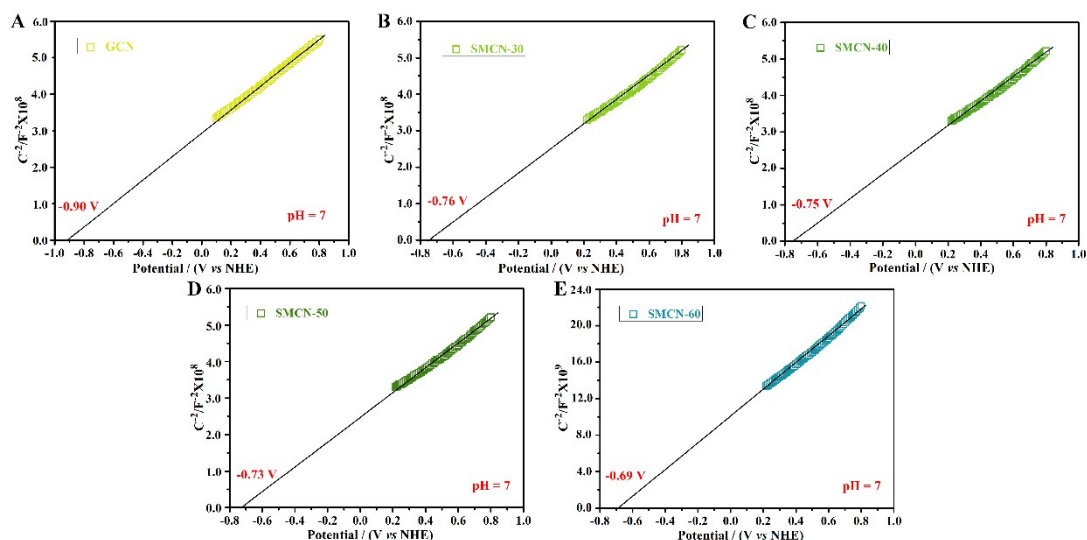
System <sup>a</sup>	M,S <sub>1</sub> -g-C <sub>3</sub> N <sub>4</sub>	M,S <sub>2</sub> -g-C <sub>3</sub> N <sub>4</sub>	M,S <sub>3</sub> -g-C <sub>3</sub> N <sub>4</sub>	M,S <sub>4</sub> -g-C <sub>3</sub> N <sub>4</sub>
$E_o$ (eV)	-22139.67	-22139.82	-22139.80	-22140.36
$E_f$ (eV)	-1.51	-1.66	-1.64	-2.20

<sup>a</sup>The four possible systems with different doping positions are shown in Figure 1.

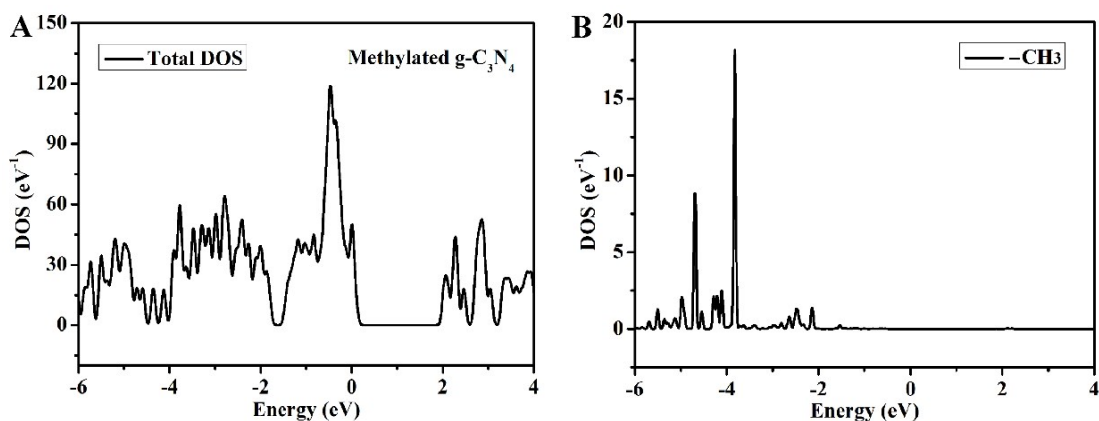
**Table S2.** The summary of relevant parameters for the as-prepared photocatalysts.

	GCN	SMCN-30	SMCN-40	SMCN-50	SMCN-60
Specific Surface Area (m <sup>2</sup> g <sup>-1</sup> ) <sup>a</sup>	42.56	52.63	67.62	90.49	67.23
Pore Volume (cm <sup>3</sup> g <sup>-1</sup> ) <sup>b</sup>	0.18	0.26	0.34	0.44	0.33
Bandgaps (eV) <sup>c</sup>	2.62	2.21	1.94	1.85	1.83
CB Potential (eV) <sup>d</sup>	-0.90	-0.76	-0.75	-0.73	-0.69
VB Potential (eV)	1.72	1.45	1.19	1.12	1.14
Kinetic Constants (min <sup>-1</sup> ) <sup>e</sup>	0.024	0.044	0.052	0.072	0.046
H <sub>2</sub> Evolution Rate (mmol g <sup>-1</sup> h <sup>-1</sup> )	1.67	—	—	6.34	—

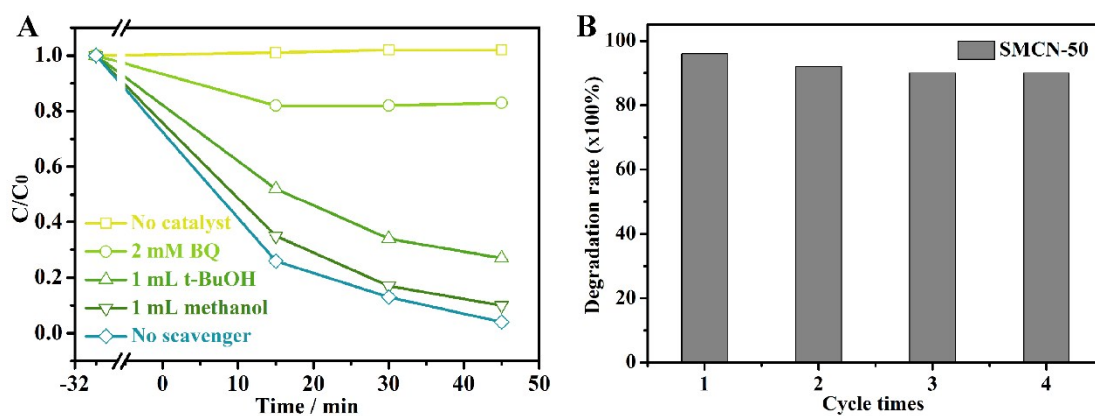
<sup>a</sup>BET surface area ( $P/P_0 = 0.2-0.9$ ); <sup>b</sup>Pore volume based on BJH method; <sup>c</sup>Based on the Kubelka-Munk function  $(F(R)hv)^n$  ( $n = 1/2$ ); <sup>d</sup>Obtained from the Mott-Schottky plots; <sup>e</sup>Approximated as a pseudo-first-order process by linear transforms  $\ln(C_0/C_t) = kt$ .



**Figure S2.** Mott-Schottky plots of (A) GCN and (B-E) SMCN samples.

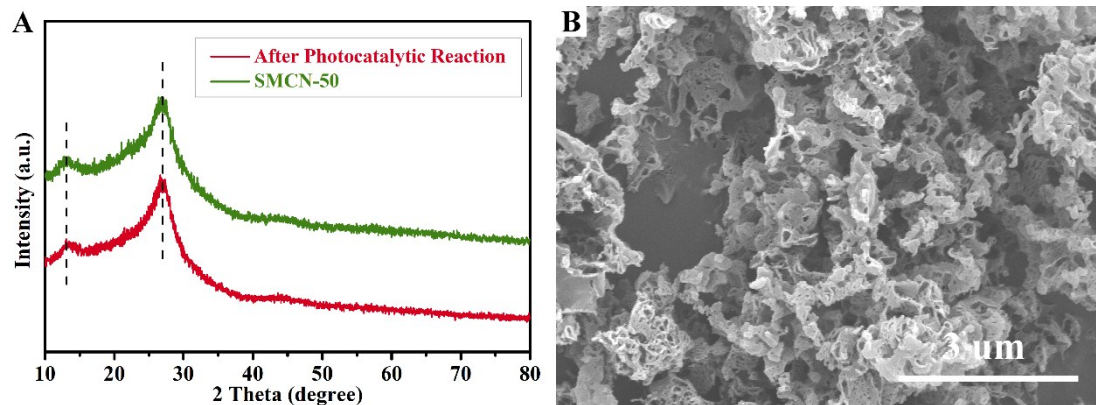


**Figure S3.** (A) Total DOS of methylated  $g\text{-C}_3\text{N}_4$  without S doping, (B) The DOS of methyl group in SMCN sample.



**Figure S4.** (A) Plots of photo-generated carriers trapped during the photocatalytic degradation of Procion Red MX-5B, (B) Stability of SMCN-50 sample during four

consecutive photocatalytic degradation cycles.



**Figure S5.** (A) XRD patterns and (B) SEM image of the reacted SMCN-50 sample.