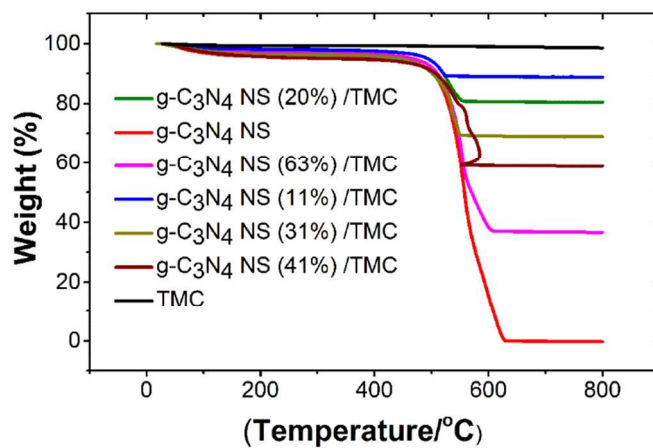


# g-C<sub>3</sub>N<sub>4</sub>/TiO<sub>2</sub> Mesocrystals Composite for H<sub>2</sub> Evolution under Visible Light Irradiation and Its Charge Carriers Dynamics

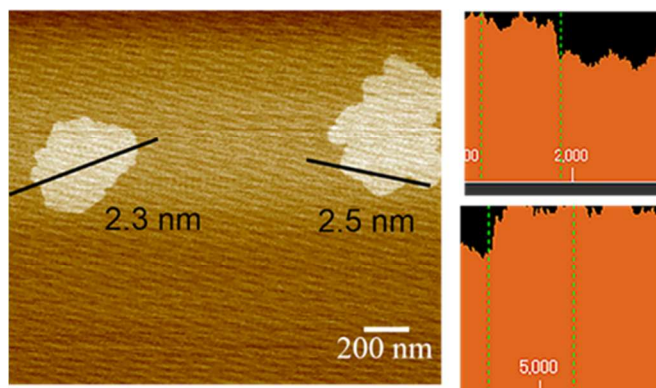
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8-1, Ibaraki, Osaka 567-0047, Japan.

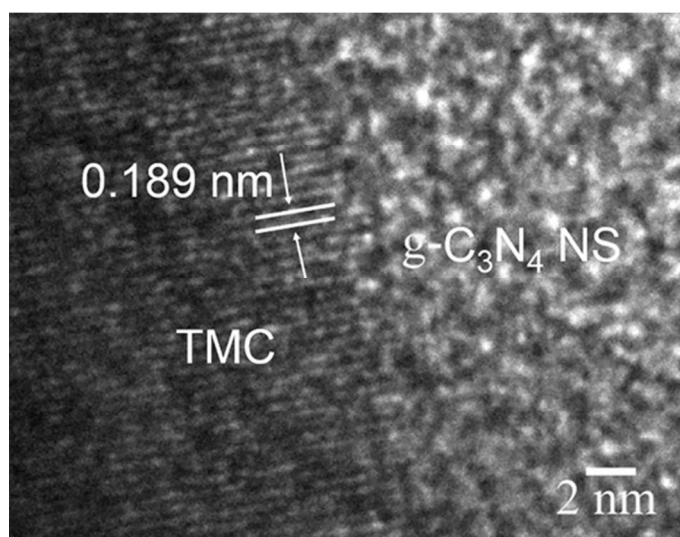
<sup>\*</sup>Author to whom correspondence should be addressed. E-MAIL: majima@sanken.osaka-  
u.ac.jp (T.M.)



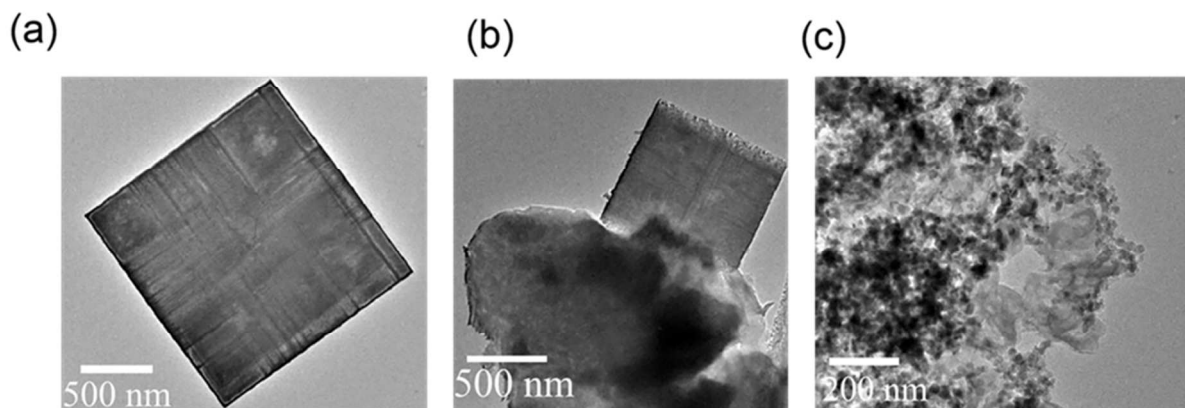
**Figure S1.** TGA curves of pure g-C<sub>3</sub>N<sub>4</sub> NS and g-C<sub>3</sub>N<sub>4</sub> NS/TMC composite with different wt%.



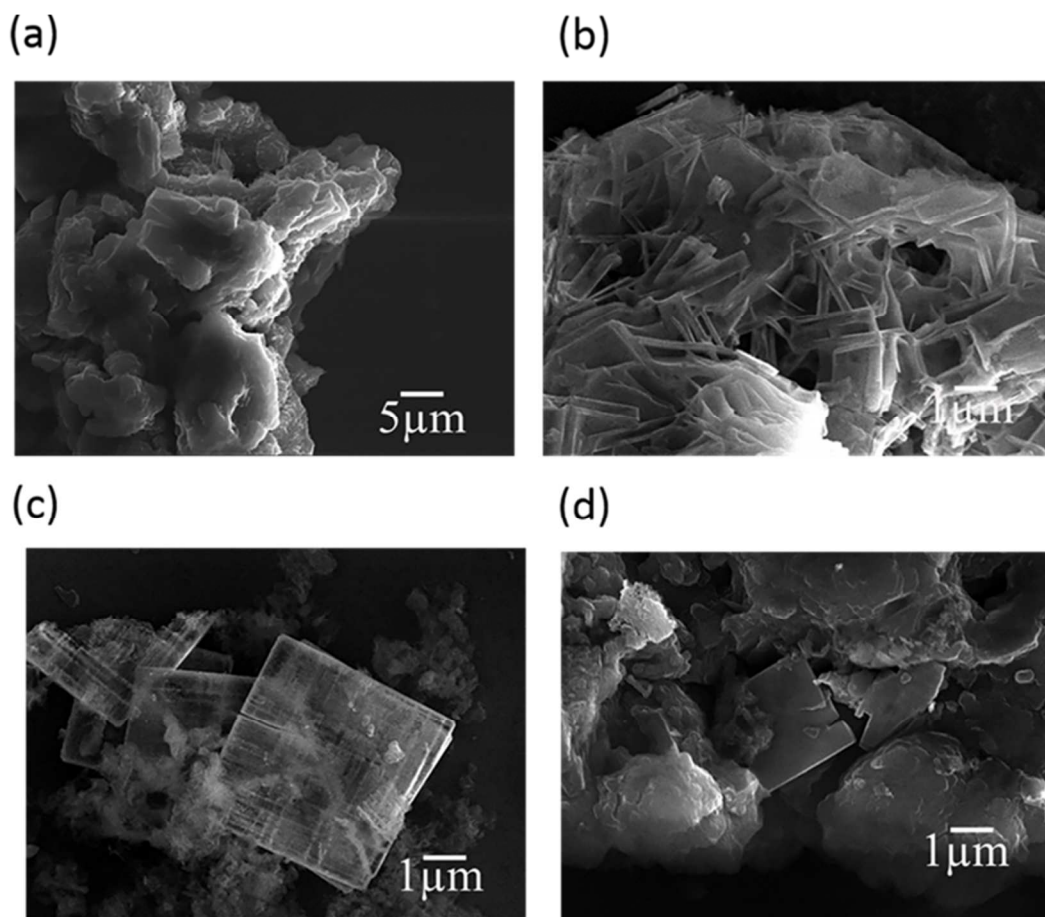
**Figure S2.** AFM image of g-C<sub>3</sub>N<sub>4</sub> NS.



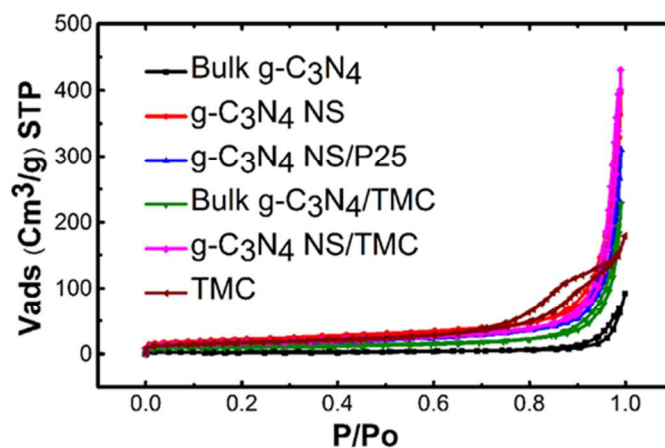
**Figure S3.** HRTEM image of g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/TMC.



**Figure S4.** TEM images of TMC (a), bulk g-C<sub>3</sub>N<sub>4</sub> (31 wt%)/TMC (b), and g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%) /P25 (c).



**Figure S5.** SEM images of bulk g-C<sub>3</sub>N<sub>4</sub> (a), g-C<sub>3</sub>N<sub>4</sub> NS (b), g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/TMC (c), and bulk g-C<sub>3</sub>N<sub>4</sub> (31 wt%)/TMC (d).

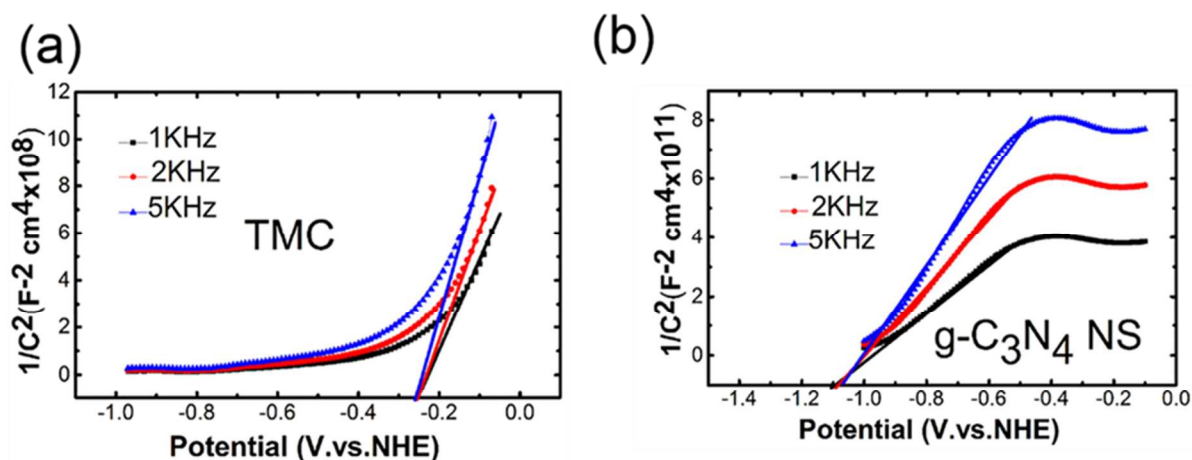


**Figure S6.** N<sub>2</sub> adsorption-desorption isotherm of the prepared samples.

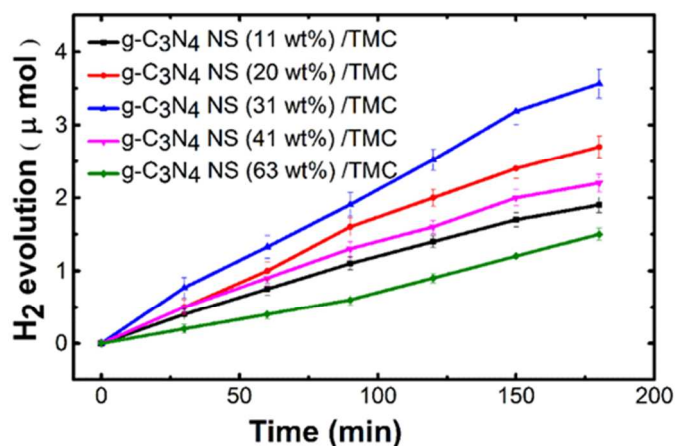
**Mott-Schottky (MS) measurement.** The MS plots were measured with a frequency of 200 Hz and amplitude of 10 mV. The electrodes were immersed in 0.1 M Na<sub>2</sub>SO<sub>4</sub> aqueous solution.

$$1/C_{sc}^2 = [2/e\epsilon_0\epsilon N_d A^2] [(E-E_{fb}) - (K_b T/e)]$$

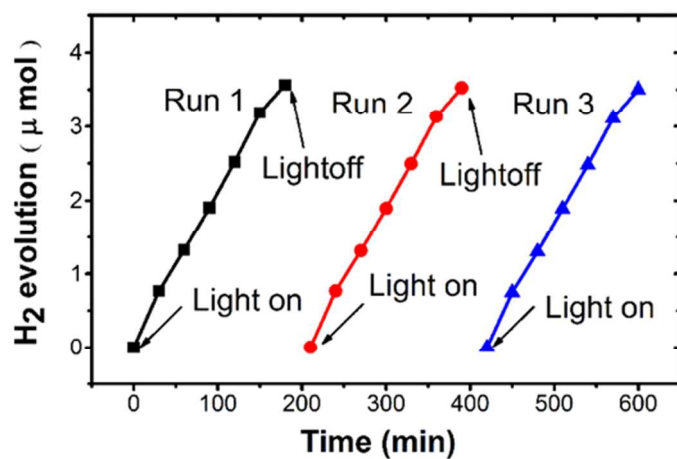
where  $C_{sc}$  is the space charge capacitance (F cm<sup>-2</sup>),  $A$  is the area,  $N_d$  is the donor density,  $e$  is the elementary charge ( $1.62 \times 10^{-19}$  C),  $\epsilon$  is the relative dielectric constant of the semiconductor,  $\epsilon_0$  is the permittivity of vacuum ( $8.85 \times 10^{-14}$  F cm<sup>-1</sup>),  $E$  is the applied potential (V),  $K_b$  is the Boltzmann constant ( $1.38 \times 10^{-23}$  J K<sup>-1</sup>) and  $T$  is the temperature. According to the equation, three curves can be drawn using  $E$  against  $1/C_{sc}^2$ , and  $E_{fb}$  of the corresponding electrode is ascertained by the x-intercept



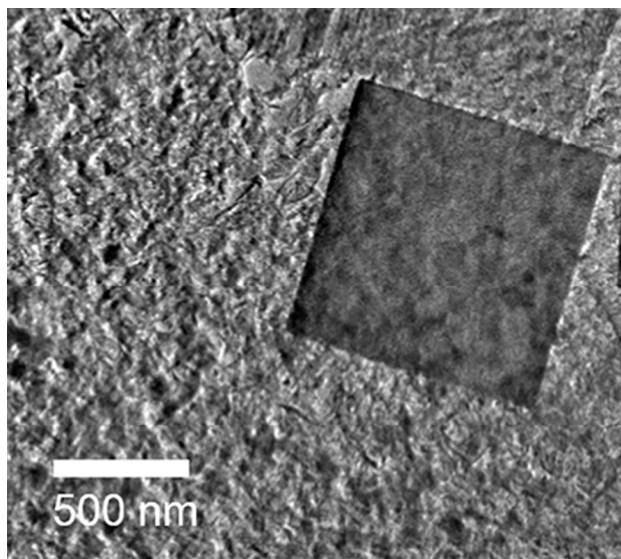
**Figure S7.** Mott-Schottky plot of TMC (a) and g-C<sub>3</sub>N<sub>4</sub> NS (b).



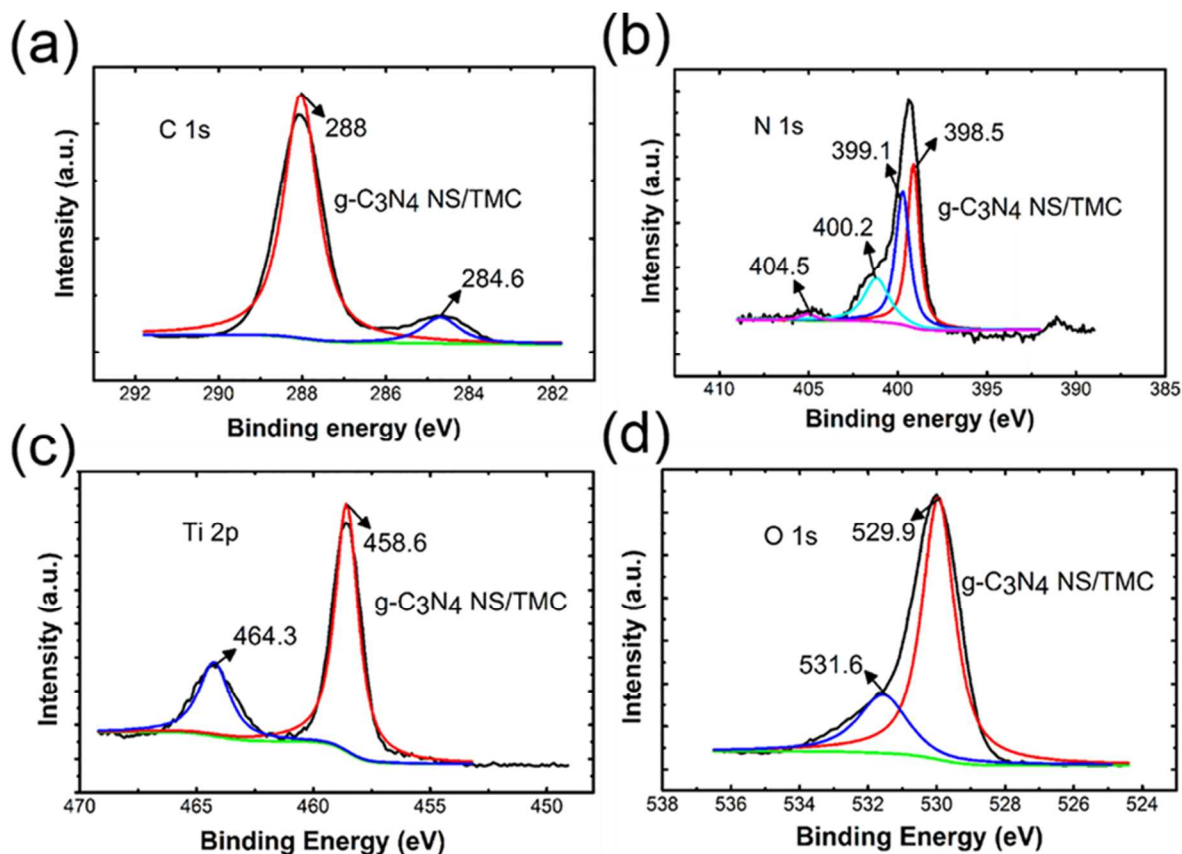
**Figure S8.** Visible-light photocatalytic activities for H<sub>2</sub> evolution ( $\lambda > 420$  nm) of g-C<sub>3</sub>N<sub>4</sub> NS /TMC with different wt% of g-C<sub>3</sub>N<sub>4</sub> NS.



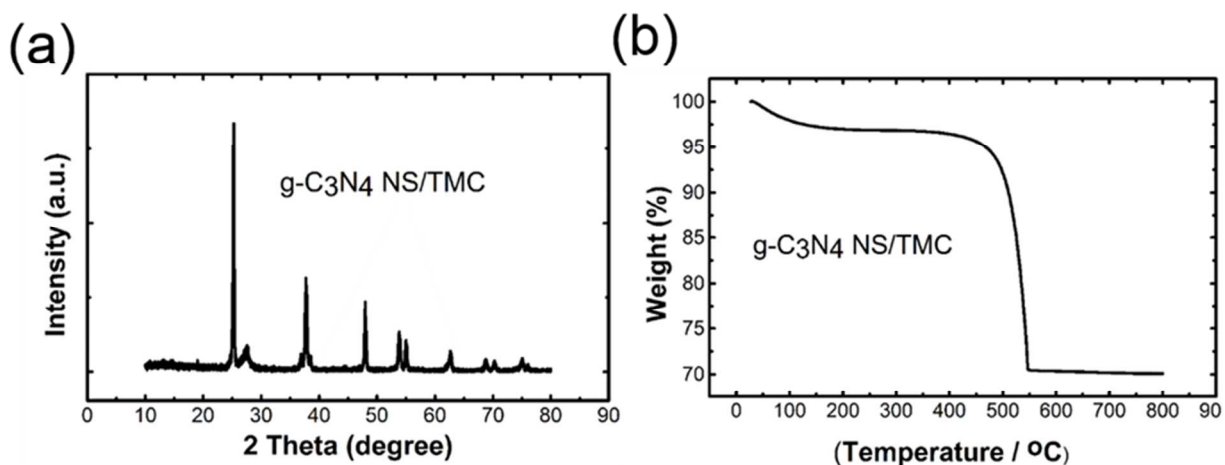
**Figure S9.** Stability test of g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/TMC under visible light irradiation ( $\lambda > 420$  nm).



**Figure S10.** TEM image of g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/TMC after photocatalytic H<sub>2</sub> evolution under visible light irradiation ( $\lambda > 420$  nm).

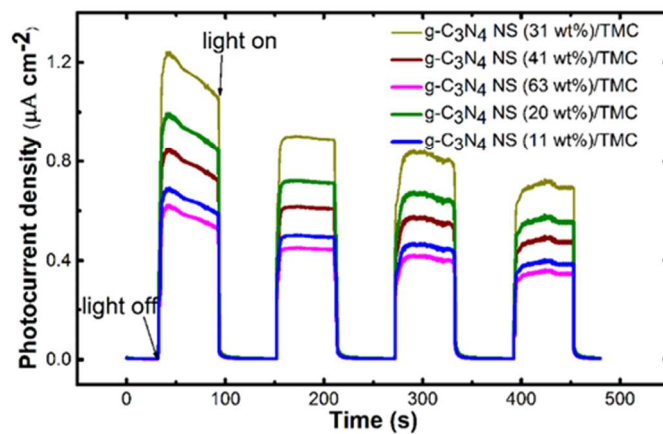


**Figure S11.** High resolution XPS spectra of C 1s (a), N 1s (b), Ti 2p (c), and O 1s (d) of g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/TMC after photocatalytic H<sub>2</sub> evolution under visible light irradiation ( $\lambda > 420$  nm).

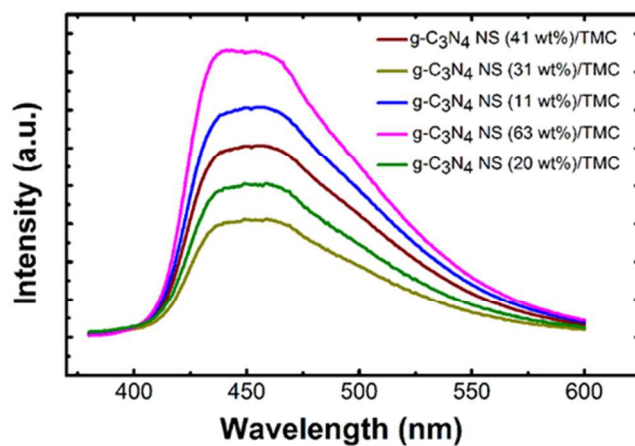


**Figure S12.** (a) XRD and (b) TGA curve of g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/TMC after photocatalytic H<sub>2</sub> evolution under visible light irradiation ( $\lambda > 420$  nm).



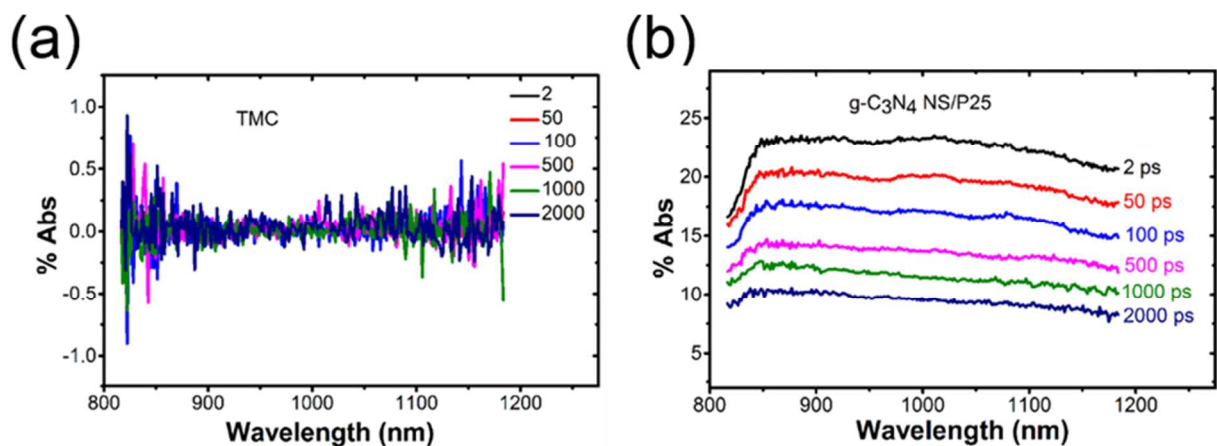


**Figure S13.** Photocurrent response of g-C<sub>3</sub>N<sub>4</sub> NS/TMC with different wt% of g-C<sub>3</sub>N<sub>4</sub> NS.

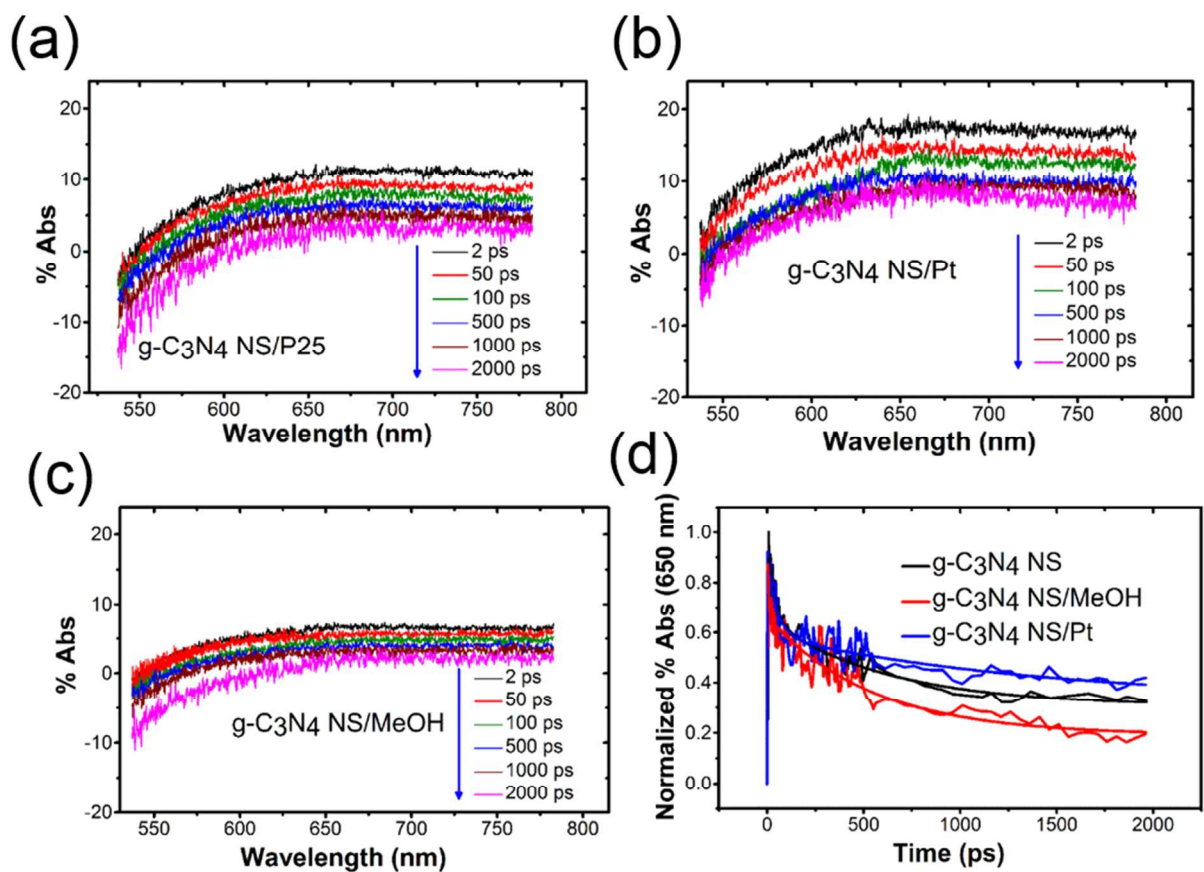


**Figure S14.** Room-temperature PL emission spectra of g-C<sub>3</sub>N<sub>4</sub> NS/TMC with different wt% of g-C<sub>3</sub>N<sub>4</sub> NS.





**Figure S15.** Time-resolved diffuse reflectance spectra of TMC (a) and g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/P25



(b).

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**Figure S16.** Time-resolved diffuse reflectance spectra of g-C<sub>3</sub>N<sub>4</sub> NS (31 wt%)/P25 (a), g-C<sub>3</sub>N<sub>4</sub> NS/Pt (b), g-C<sub>3</sub>N<sub>4</sub> NS/MeOH (c), and time decay profile observed at 650 nm for g-C<sub>3</sub>N<sub>4</sub> NS, g-C<sub>3</sub>N<sub>4</sub> NS/MeOH, and g-C<sub>3</sub>N<sub>4</sub> NS/Pt (d).

**Table S1.** Elemental analysis of g-C<sub>3</sub>N<sub>4</sub> NS and g-C<sub>3</sub>N<sub>4</sub> NS /TMC with different wt%.

Sample	C%	N%	C/N molar ratio
g-C <sub>3</sub> N <sub>4</sub> NS	35.6	62.1	0.67
g-C <sub>3</sub> N <sub>4</sub> NS (11 wt%)/TMC	3.9	6.9	0.67
g-C <sub>3</sub> N <sub>4</sub> NS (21 wt%)/TMC	7.2	12.7	0.66
g-C <sub>3</sub> N <sub>4</sub> NS (31 wt%)/TMC	11.3	19.5	0.67
g-C <sub>3</sub> N <sub>4</sub> NS (41 wt%)/TMC	15	26.3	0.67
g-C <sub>3</sub> N <sub>4</sub> NS (63 wt%)/TMC	22.8	39.7	0.67

**Table S2.** The structural characteristics of TMC, g-C<sub>3</sub>N<sub>4</sub>, and g-C<sub>3</sub>N<sub>4</sub>/TMC composite.

Sample	Surface area (m <sup>2</sup> g <sup>-1</sup> )	Pore volume (cm <sup>3</sup> g <sup>-1</sup> )	Pore size (nm)
bulk g-C <sub>3</sub> N <sub>4</sub>	8.4	0.09	24.3
g-C <sub>3</sub> N <sub>4</sub> NS	72.2	0.55	14
g-C <sub>3</sub> N <sub>4</sub> NS (31 wt%)/P25	52.3	0.45	16.5
bulk g-C <sub>3</sub> N <sub>4</sub> (31 wt%)/TMC	34.2	0.33	19
g-C <sub>3</sub> N <sub>4</sub> NS (31 wt%)/TMC	57.4	0.66	15.5

**Table S3.** The fitted parameters of fluorescence lifetime for different samples.

Samples	A <sub>1</sub>	τ <sub>1</sub> (ns)	A <sub>2</sub>	τ <sub>2</sub> (ns)
bulk g-C <sub>3</sub> N <sub>4</sub>	0.46	0.5	0.54	2.2
g-C <sub>3</sub> N <sub>4</sub> NS	0.43	0.74	0.57	3.0
g-C <sub>3</sub> N <sub>4</sub> NS (31 wt%)/P25	0.41	0.9	0.59	3.3
bulk g-C <sub>3</sub> N <sub>4</sub> (31 wt%)/TMC	0.40	0.9	0.60	3.5
g-C <sub>3</sub> N <sub>4</sub> NS (31 wt%)/TMC	0.35	1.1	0.65	4.8