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## **Supplementary Information**

## Physical vapor deposition (PVD): a method to fabricate modified g-C<sub>3</sub>N<sub>4</sub> sheets

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Fig. S1 Thermal gravimetric analysis (TGA) of g-C<sub>3</sub>N<sub>4</sub>



Fig. S2 Py-GC/MS of bulk g-C<sub>3</sub>N<sub>4</sub> at 550 °C.

Table S1 The area ratio of gaseous derivatives and corresponding Mr from Fig. S2.

Peak	1	2	3	4	5	6	7
Area%	4.90	6.68	4.02	7.72	6.65	4.21	9.95
Mr	256	264	281	341	370	572	662

Only the Area% of derivatives molecules which exceeds 4% was listed.



**Fig. S3** Total ion chromatogram of Py-GC/MS of bulk  $g-C_3N_4$  at 700 °C (a) and FTIR of liquid water (top) and gaseous derivatives (bottom) derived from bulk  $g-C_3N_4$  at 700 °C (b)

Iable	52	Area	ratios	and	possible	relative	molecular	mass	(Mr)	01	gaseous
derivat	ives	derive	d from	g-C <sub>3</sub> ]	N <sub>4</sub> corresp	onding to	otal ion chro	omatog	ram in	Fig	. S2a

	1	2	3	4	5	6	7	8
Area% (>2%)	20.5	23.1	2.2	2.5	2.6	3.8	2.0	4.3
Mr	68	66	156	196	223	222	243	368

The pyrolysis gas was firstly investigated by pyrolyzer coupled with gas chromatography and mass spectrometry (Py-GC/MS). The m/z obtained by Py-GC/MS ranges from 45 to 750 (full scan) which also indicates the MS section can not measure more large m/z than 750. The obvious peaks from C=N stretching vibration appear at about 2260 nm<sup>-1</sup> by TG-IR, while there not exists tri-s-triazine-based structure in the spectrum. It can be considered that the low temperature (180 °C) of pipeline between TG and IR result into the deposition of the pyrolysis gas on its wall.

	g-C	3N4	CNS	8
	binding energy	area	binding energy	area
N-H	401.1	17339	401.1	9185
N–(C)3	399.9	54576	400.2	19063
C-N=C	398.6	193086	398.8	95221

Table S3 Binding energies and corresponding areas of XPS N1s of samples

**Table S4** Relative ratios of N–H, N–(C)3 and C–N=C of g-C<sub>3</sub>N<sub>4</sub> and CNS by N1s spectral analysis

sample	N-H/C-N=C	N-(C)3/C-N=C
g-C <sub>3</sub> N <sub>4</sub>	0.043	0.28
CNS	0.096	0.20

**Table S5** Surface and bulk atomic ratio (C/Nr, C/N) of the samples by XPS spectral analysis and elemental analysis

sample	C/Nr atm%	C/N atm%
g-C <sub>3</sub> N <sub>4</sub>	0.686	0.670
CNS	0.694	0.679



Fig. S4 ESR spectra of DMPO-spin adduct measured in the aqueous solution of CNS exposed to visible light ( $\lambda$ > 400 nm). Note that aqueous solution of CNS was purged with Ar to remove the dissolved O<sub>2</sub>



Fig. S5 Mechanism diagram of pristine g-C<sub>3</sub>N<sub>4</sub> modified by PVD.