Supporting Information

In-situ intercalation and exploitation of Co_3O_4 nanoparticles grown on carbon nitride nanosheets for highly efficient degradation of methylene blue

Yan Li,[a] Lei Wang,[a]* Yuting Xiao,[b] Guohui Tian,[a] Chungui Tian,[a] Honggang Fu[a]*

[a] *Key Laboratory of Functional Inorganic Material Chemistry, Ministry of Education of the People's Republic of China, Heilongjiang University, Harbin 150080, China. E-mail: wanglei0525@hlju.edu.cn, fuhg@vip.sina.com, fuhg@hlju.edu.cn*

[b] *Key Laboratory of Jiangxi Province for Persistent Pollutants Control and Resources Recycle, Nanchang Hangkong University, Nanchang, Jiangxi 330063, China.E-mail: yutingxiao9@sina.com*

Table S1. The denoted name and synthesized samples for various samples.

Samples	**concentration of $Co(NO_3)_2$**
1-Co_3O_4/S-C_3N_4	6 mM
Co_3O_4/S-C_3N_4	23mM
4-Co_3O_4/S-C_3N_4	46 mM

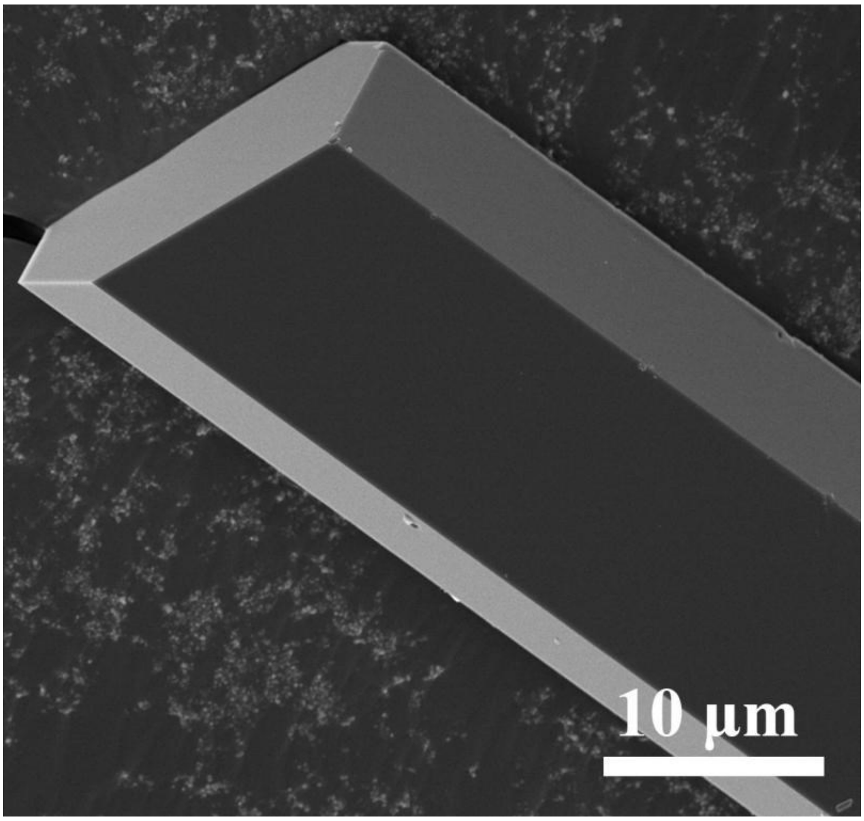


Fig. S1 SEM image of C_3N_4 precursor**.**

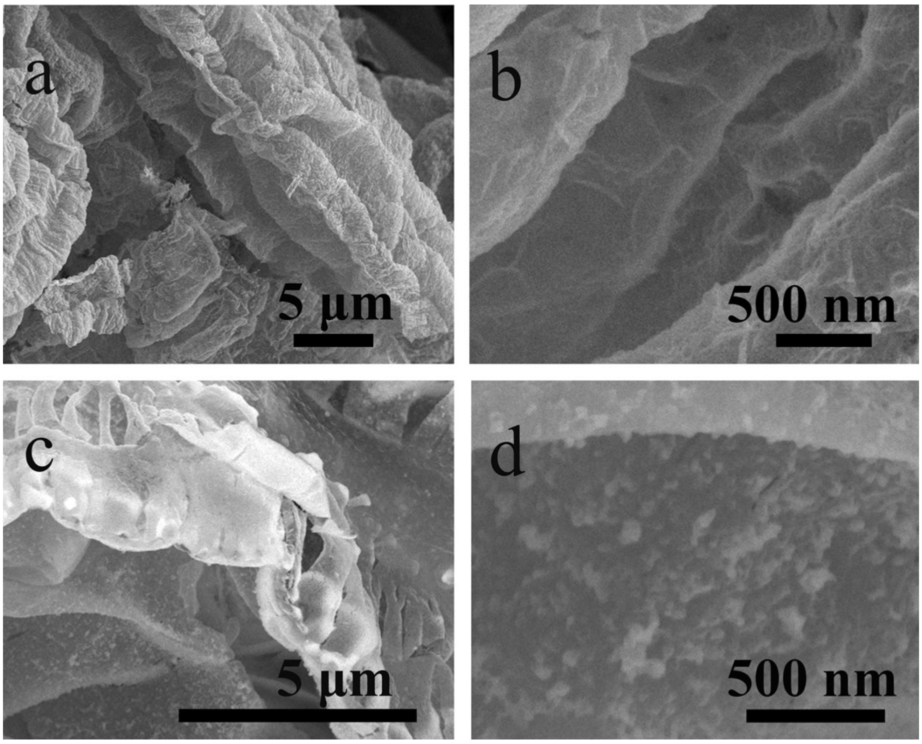


Fig. S2 SEM images of (a, b) 1-Co_3O_4/S-C_3N_4, (c, d) 4-Co_3O_4/S-C_3N_4.

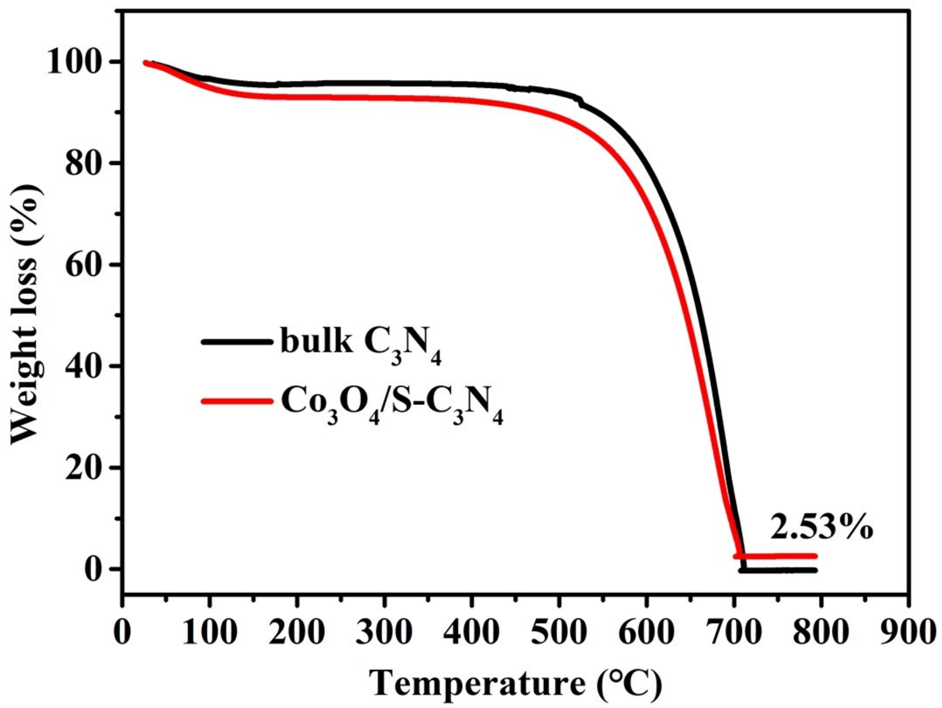


Fig. S3 TG curves of bulk C_3N_4 and Co_3O_4/S-C_3N_4 samples tested in air ambient with a heating rate of 10 ^oC min^{-1}.

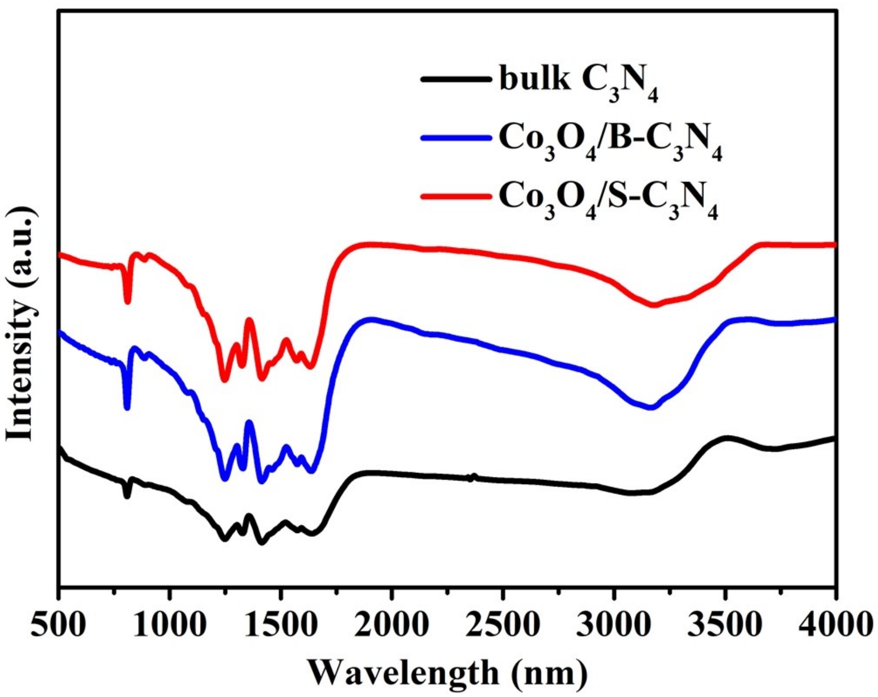


Fig. S4 FT-IR spectra of various samples.

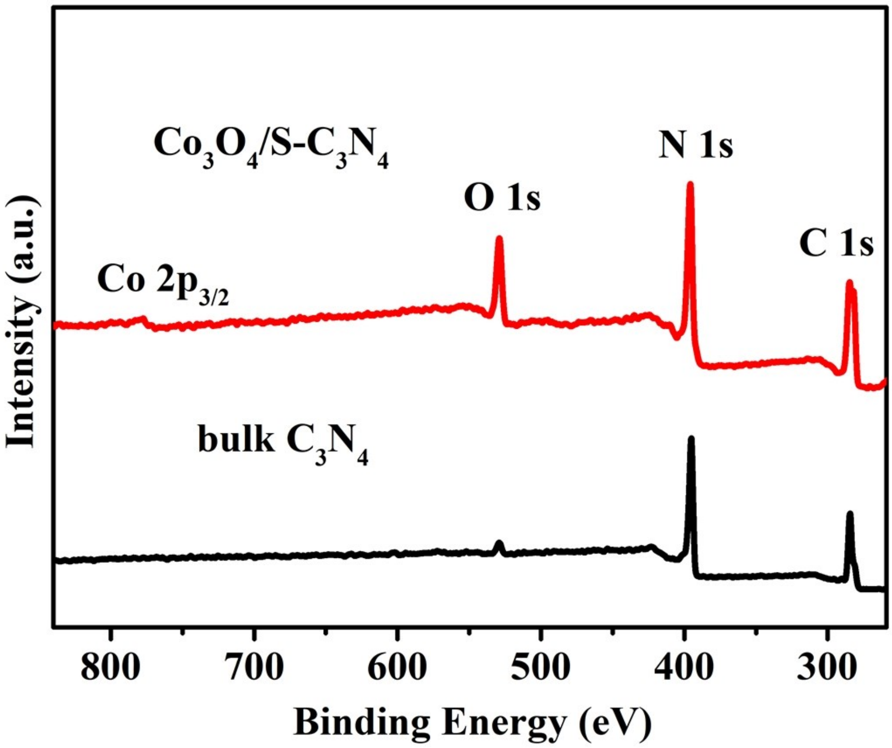


Fig. S5 Wide XPS spectrum of the bulk C_3N_4 and Co_3O_4/S-C_3N_4 samples.

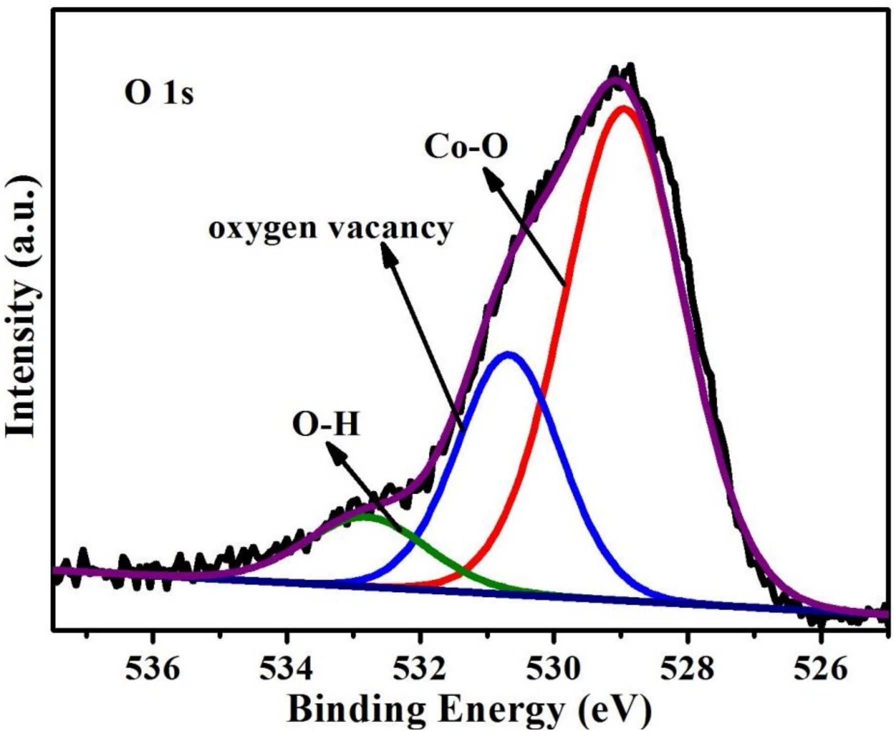


Fig. S6 XPS spectra of O 1s for Co_3O_4/S-C_3N_4 sample.

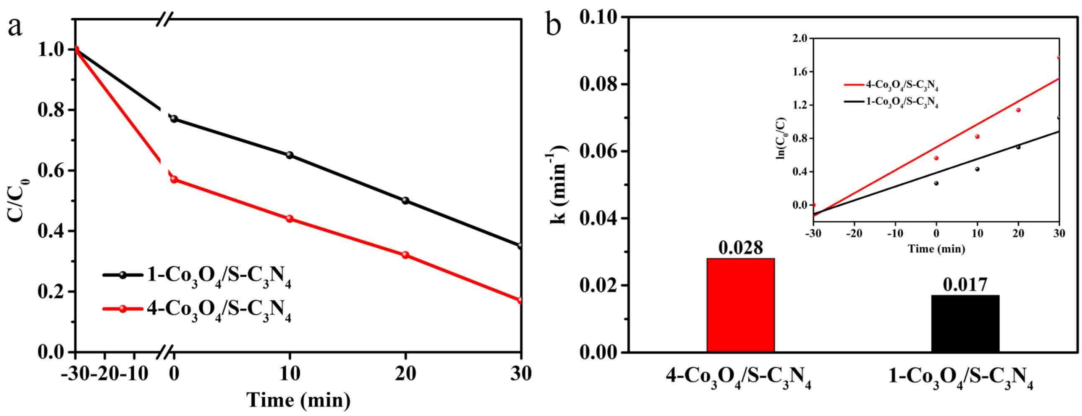


Fig. S7 Methylene blue degradation efficiency diagram of sample 1-Co_3O_4/S-C_3N_4 and 4-Co_3O_4/S-C_3N_4

Table S2. Comparison the photocatalytic degration rate of **Co_3O_4/S-C_3N_4** with the reported C_3N_4-based heterojunctions.

Catalyst	Catalyst usage (mg)	Methylene blue concentration (mg L^{-1})	Reaction time (min)	Degradation rate (%)	References
α-ZnTcPc/g-C_3N_4	50	10	50	94.49	S1
N-ZnO/g-C_3N_4	100	20	90	95	S2
g-C_3N_4/Ag_3PO_4/NCDs	30	10	40	99	S3
g-C_3N_4/TiO_2/Ag	6	7.5	60	100	S4
WO_3/g-C_3N_4	50	50	90	95	S5
TiO_2/Na-g-C_3N_4	100	20	120	100	S6
Ag_2O/g-C_3N_4	100	10	30	100	S7
TiO_{2-x}/Ag/g-C_3N_4	25	10	180	99	S8
g-C_3N_4-RGO-TiO_2	50	30	180	92	S9
g-C_3N_4/Ag_2CrO_4	10	10	120	99.1	S10
Ag-Fe3O4/g-C3N4	25	10	120	99	S11
$Zn_{0.25}Cd_{0.75}S$/g-C_3N_4	10	10	120	92.25	S12
g-C_3N_4/Fe@ZnO	100	10	90	95	S13
g-C_3N_4/MoS_2/Bi_2O_3	50	20	90	98.5	S14
$CoFe_2O_4$/g-C_3N_4	20	10	180	97.3	S15
$ZnFe_2O_4$/g-C_3N_4	30	10	120	98	S16
$CaFe_2O_4$ 30%/g-C_3N_4	100	10	120	90	S17
PANI/C_3N_4	25	11	120	~ 80	S18
Co_3O_4/S-C_3N_4	**10**	**10**	**30**	**99.5**	**This work**

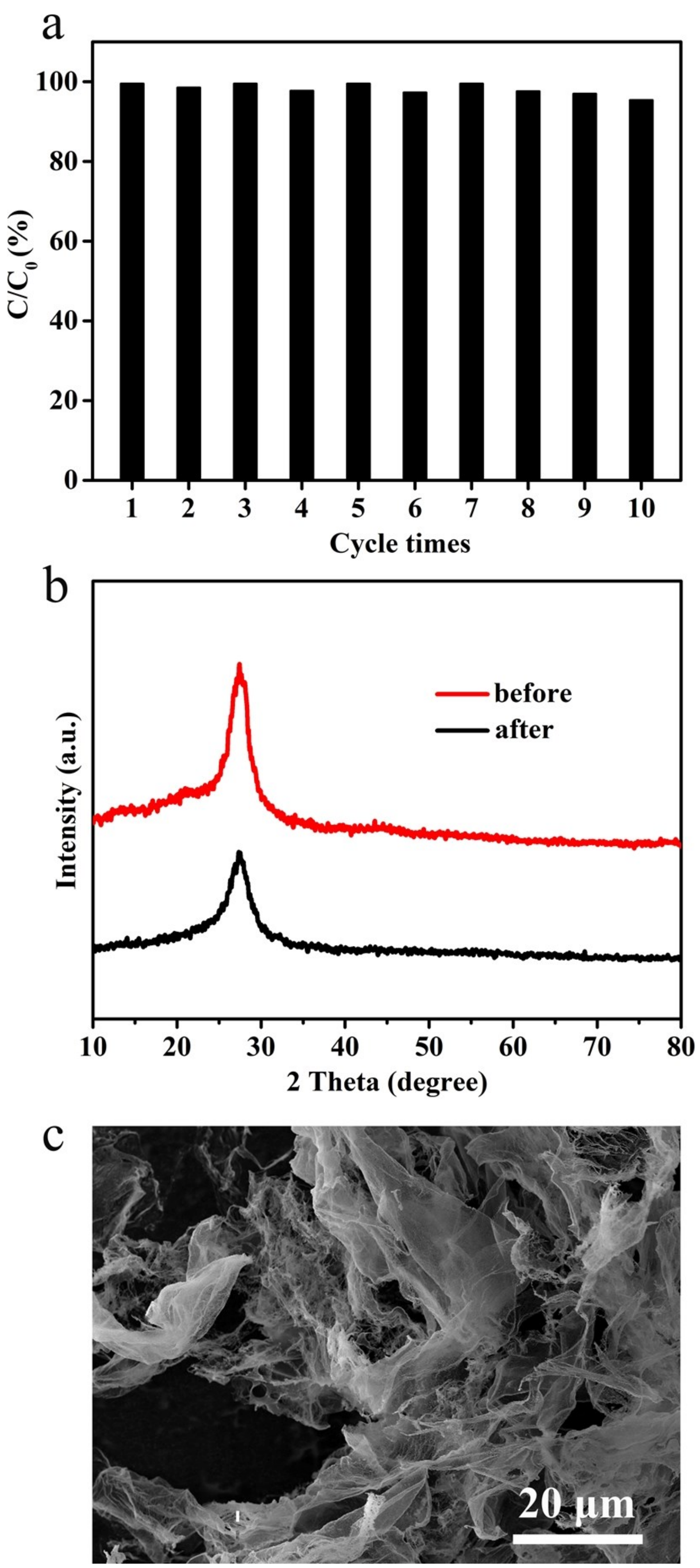


Fig. S8 Degradation ratios for 10 photocatalytic cyclic tests of Co_3O_4/S-C_3N_4 sample.

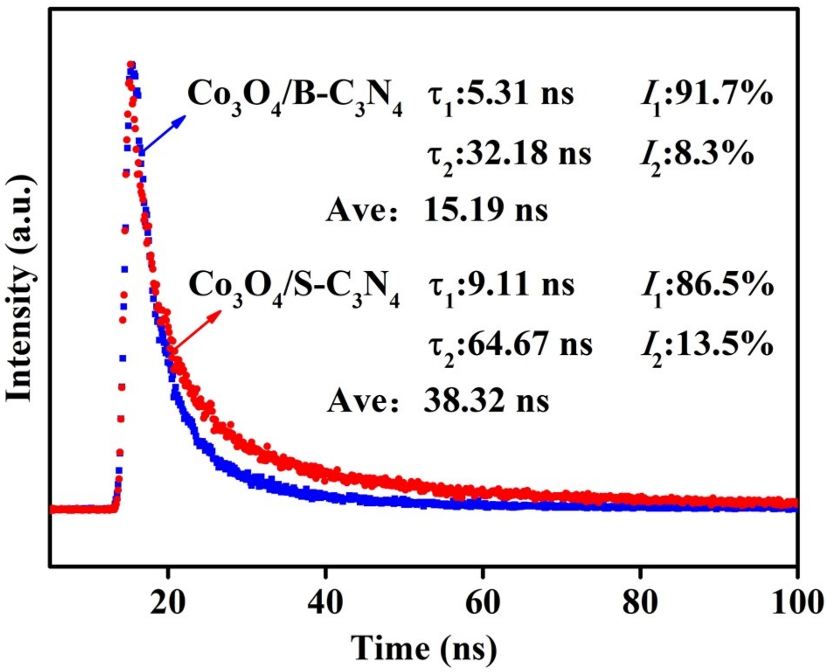


Fig. S9 Time-resolved PL decay spectra of Co_3O_4/B-C_3N_4 and Co_3O_4/S-C_3N_4 samples.

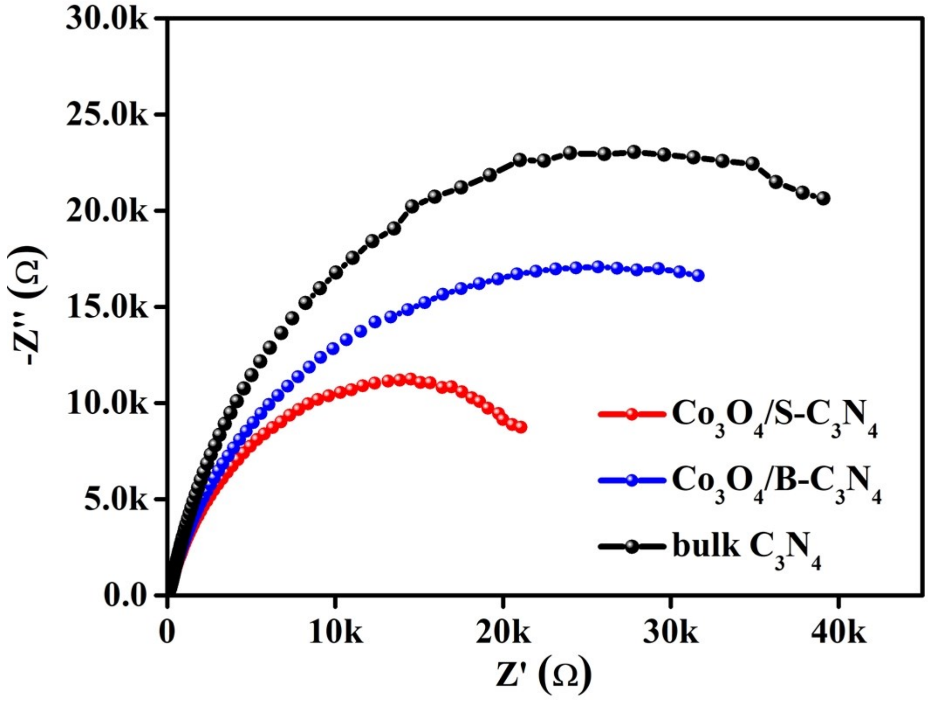


Fig. S10 EIS Nyquist plots of bulk C_3N_4, Co_3O_4/B-C_3N_4 and Co_3O_4/S-C_3N_4 electrodes with visible light irradiation.

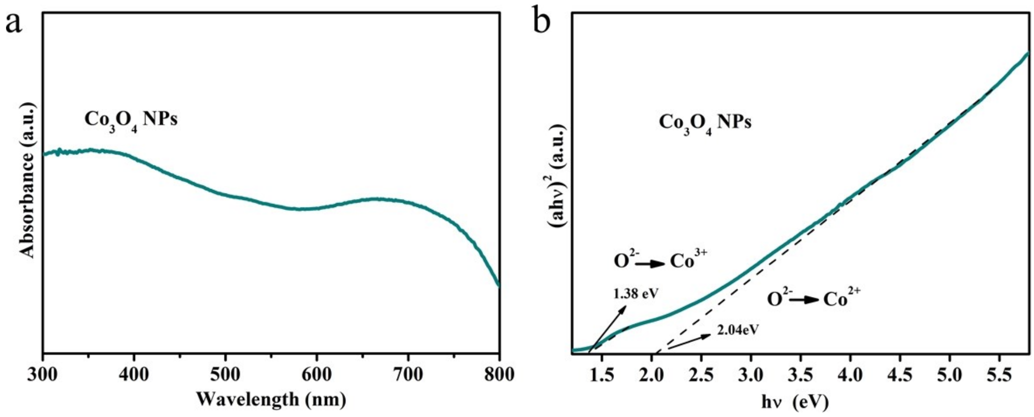
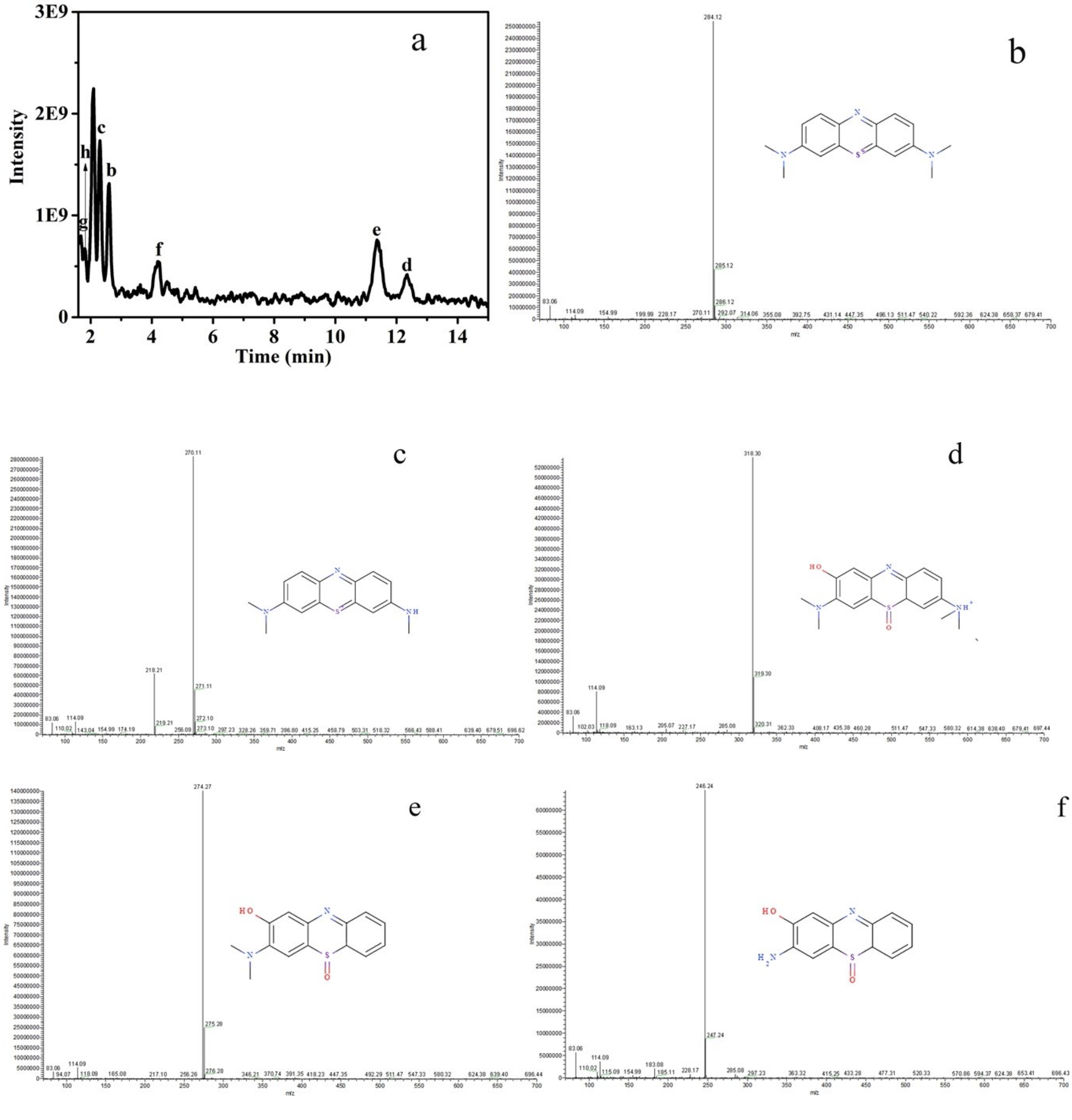


Fig. S11 (a) UV-vis absorption spectra of Co_3O_4 NPs, (b) Optical bandgap diagrams of Co_3O_4 NPs.



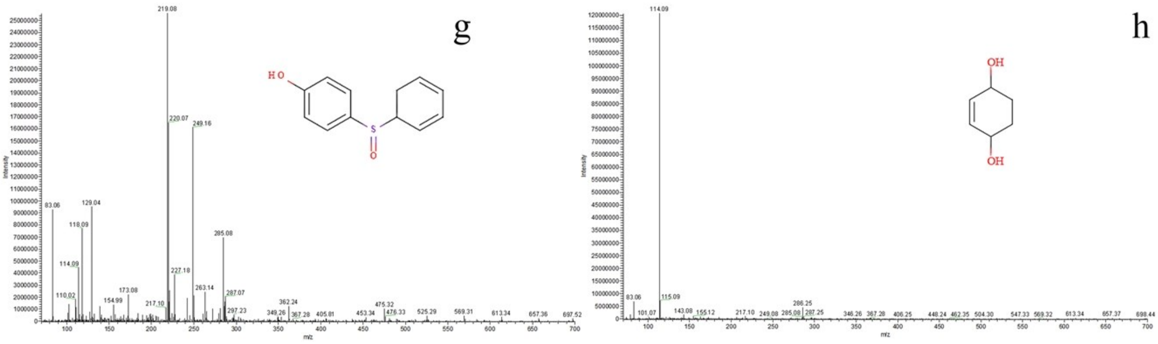


Fig. S12 (a) Total ion flow diagram, (b-h) the mass spectrum of the sample Co_3O_4/S-C_3N_4 obtained by the LC-MS during the photocatalytic degradation of methylene blue after 20 min photodegradation.

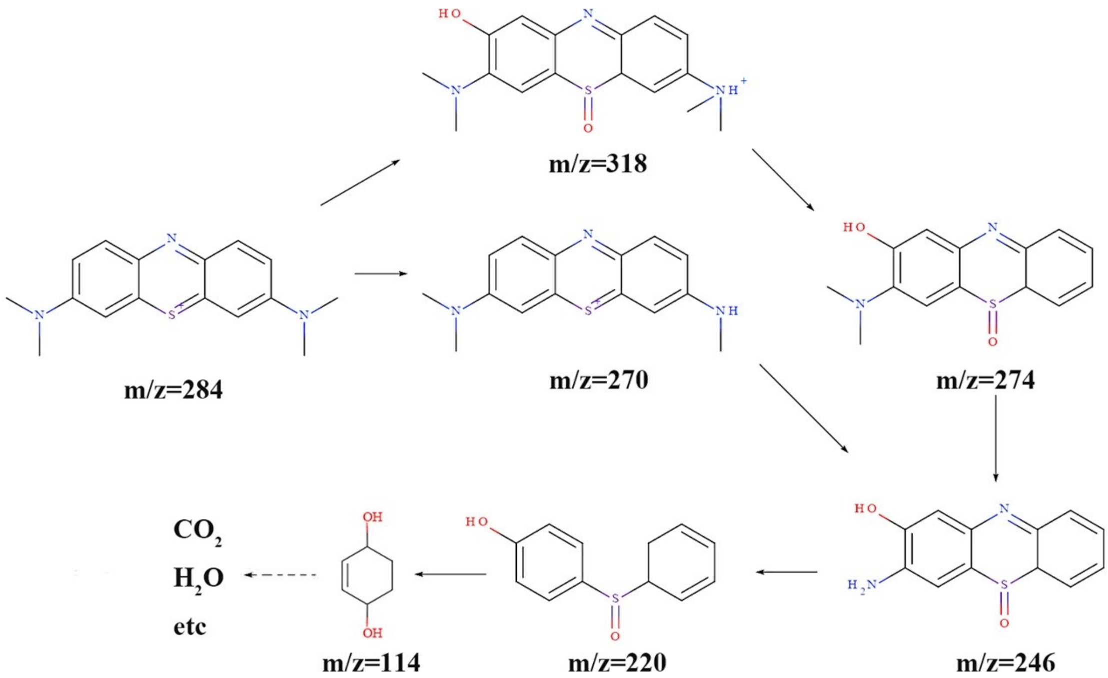


Fig. S13 Propose a way to degrade MB by Co_3O_4/S-C_3N_4 photocatalysis

The molecular ion peak at m/z=284 is attributed to MB (**Fig. S12b)**. The first step decomposition of MB molecules should be done followed the two means: On the one hand, it could be directly broken the -N-CH_3 bond to produce an intermediate with a m/z=270 (**Fig. S12c)**. On the other hand, the S atoms of the C-S^+=C group in MB molecules could be oxidized by •OH radicals,[19,20] at the same time, the active species •OH and •O_2^- attack the benzene ring in the methylene blue ion, resulting in the intermediate with m/z=318 was formed **(Fig. S12d)**. The low bond energy of N-CH_3 is easy to be attacked by the active molecule •OH and form an intermediate with m/z=274 (**Fig. S12e)**. Under the attack of •OH, an intermediate with m/z=114 is formed after the substitution reaction (m/z=246) and addition reaction (m/z=220). After a series of oxidation processes, methylene blue was degraded to form CO_2, H_2O and other small molecules.

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