

## Supporting Information

### Enhancement of Mineralization Ability of $C_3N_4$ via lower Valence Position by Tetracyanoquinodimethane Organic Semiconductor

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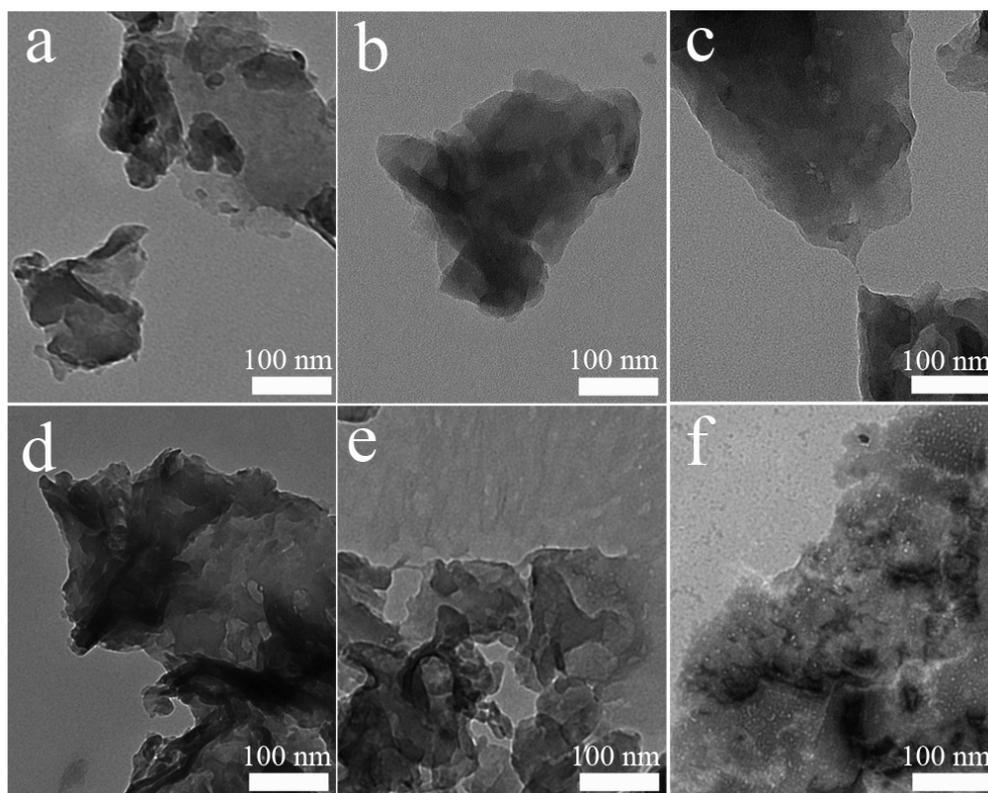


Figure S1 TEM images of pure g- $C_3N_4$  (a) TCNQ- $C_3N_4$  with TCNQ mass fraction as 1 %, 5 %, 10 %, 20% (b-e) and pure TCNQ (f)

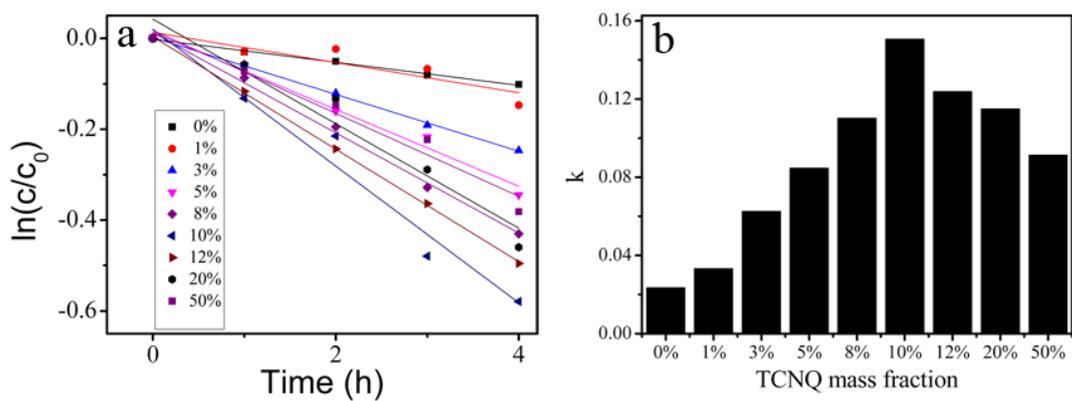


Figure S2 (a) Photocatalytic degradation of phenol and (b) the apparent rate constants over pure  $g\text{-C}_3\text{N}_4$  (0%), pure TCNQ (100%) and TCNQ- $\text{C}_3\text{N}_4$  with different TCNQ mass fraction (1% ~ 50%) under simulated sunlight irradiation.

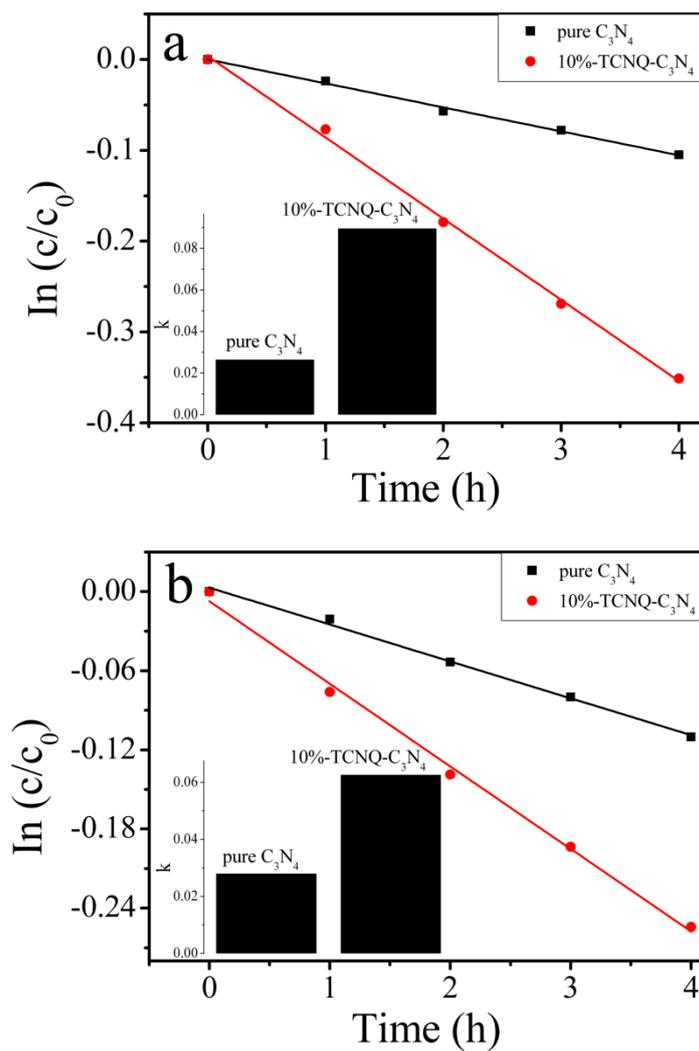


Figure S3 Photocatalytic degradation of 2,4-dichlorophenol (a) and bisphenol A (b), (inset) the apparent rate constants over pure g- $C_3N_4$  (0%) and 10%-TCNQ- $C_3N_4$  under visible light irradiation ( $\lambda > 420$  nm).

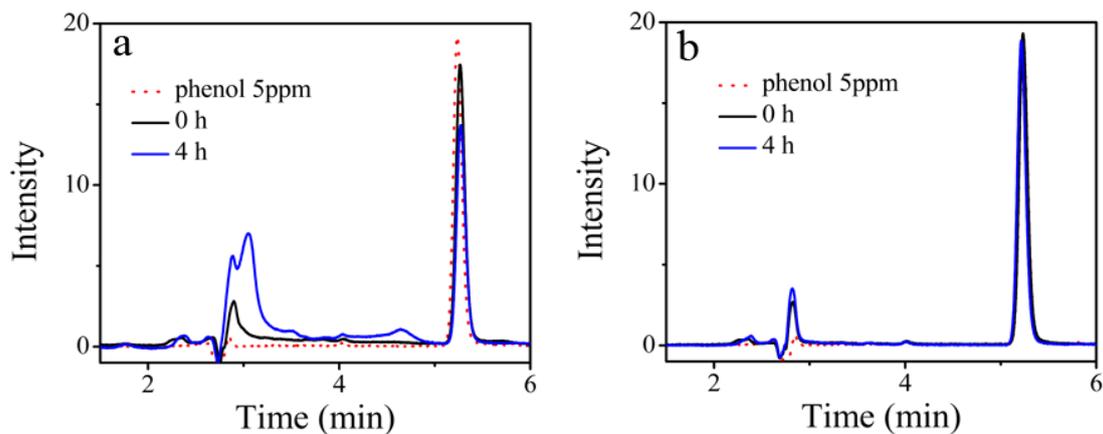


Figure S4 HPLC chromatograms of phenol and after photocatalytic degradation by TCNQ-C<sub>3</sub>N<sub>4</sub> (a) and pure C<sub>3</sub>N<sub>4</sub> for 4 h monitored at 275 nm ([phenol] = 5 ppm, catalyst = 25 mg/50 mL)

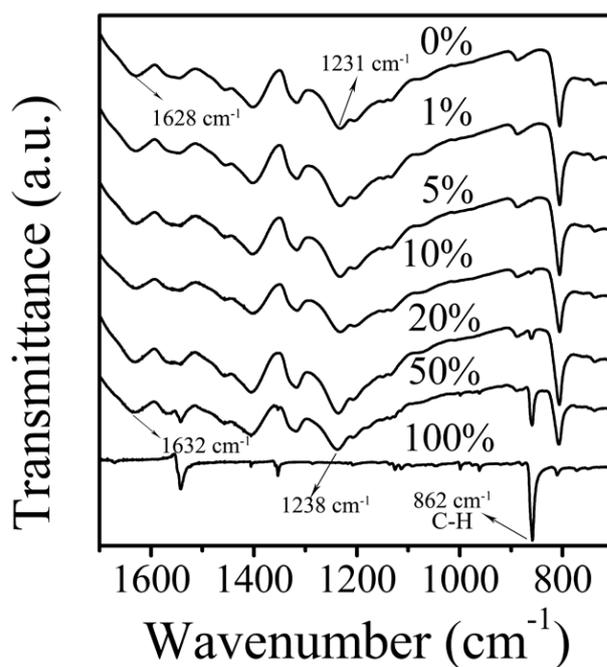


Figure S5 IR spectra of g-C<sub>3</sub>N<sub>4</sub>, pure TCNQ and TCNQ-C<sub>3</sub>N<sub>4</sub> materials.

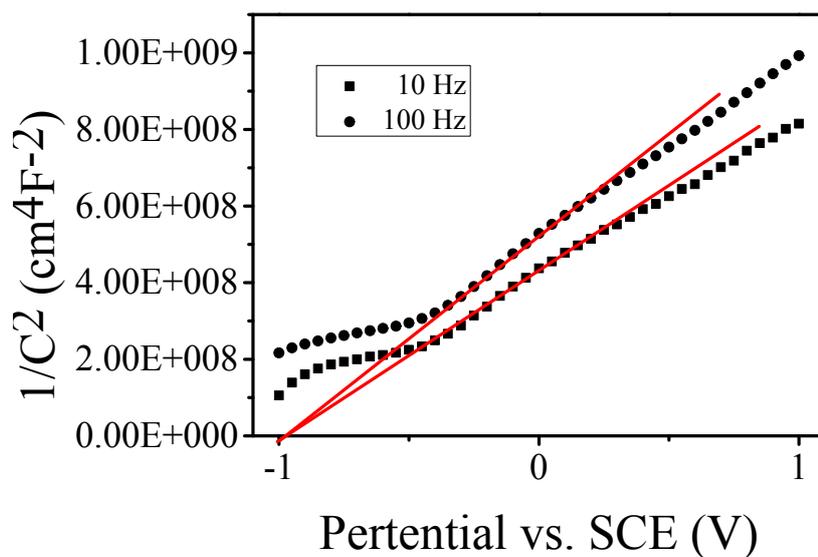


Figure S6 Mott-Schottky (MS) plots of pure  $C_3N_4$  film electrodes at a frequency of 10 Hz and 100 Hz in an aqueous solution of  $Na_2SO_4$  (0.1 M).

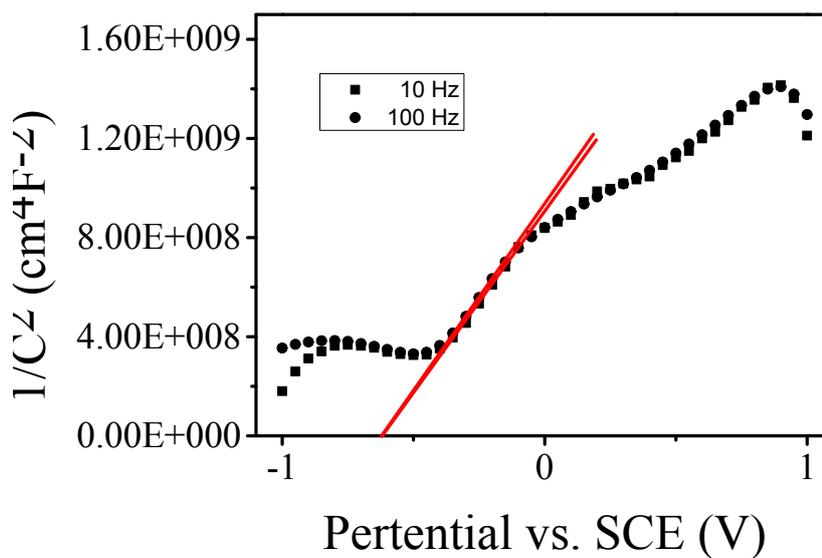


Figure S7 Mott-Schottky (MS) plots of pure TCNQ film electrodes at a frequency of 10 Hz and 100 Hz in an aqueous solution of  $Na_2SO_4$  (0.1 M).

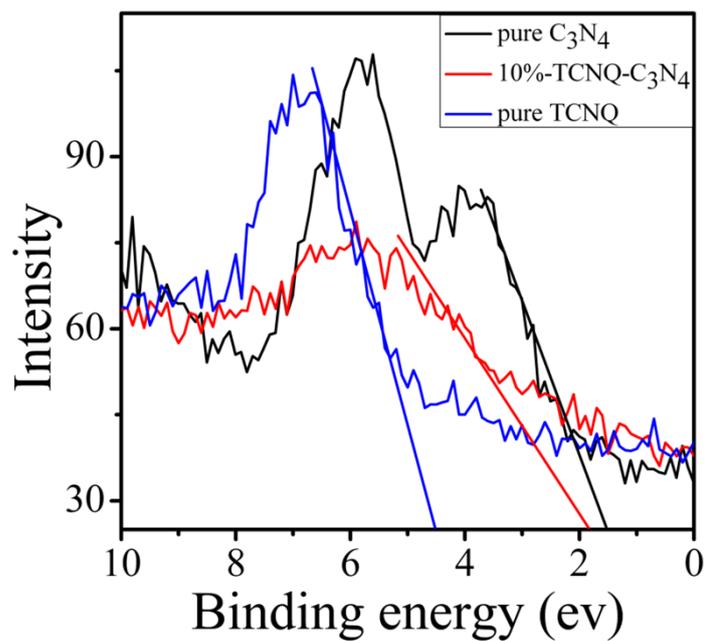


Figure S8 The valence band spectra of X-ray photoelectron spectroscopy for pure C<sub>3</sub>N<sub>4</sub>, 10%-TCNQ-C<sub>3</sub>N<sub>4</sub> and pure TCNQ.

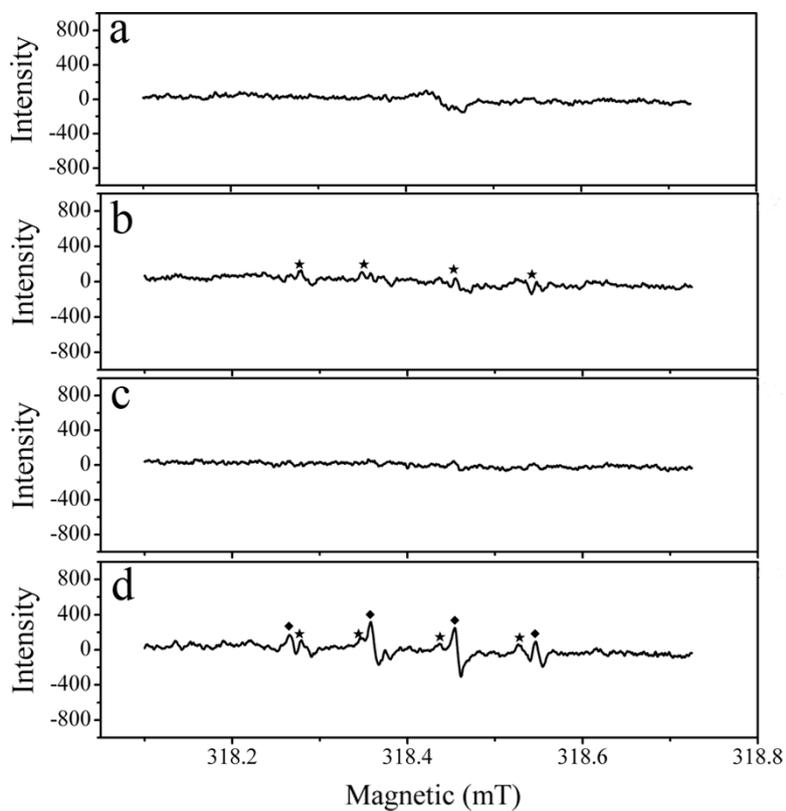


Figure S9 ESR spectra of 10%-TCNQ-C<sub>3</sub>N<sub>4</sub> in dark (a) 10%-TCNQ-C<sub>3</sub>N<sub>4</sub> under visible light irradiation ( $\lambda > 420$  nm) in water (b) pure C<sub>3</sub>N<sub>4</sub> in dark (c) and pure C<sub>3</sub>N<sub>4</sub> under visible light irradiation ( $\lambda > 420$  nm) in water (d). ★ label as superoxide radicals  
◆ label as hydroxy radical.

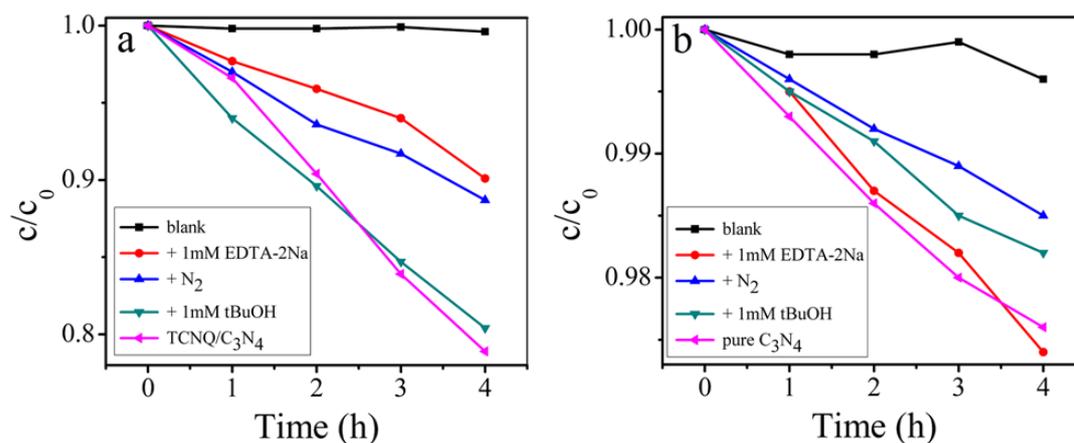


Figure S10 The plots of photogenerated carriers trapping in the system of photodegradation of phenol by 10%-TCNQ-C<sub>3</sub>N<sub>4</sub> and pure C<sub>3</sub>N<sub>4</sub> under visible light irradiation ( $\lambda > 420$  nm)

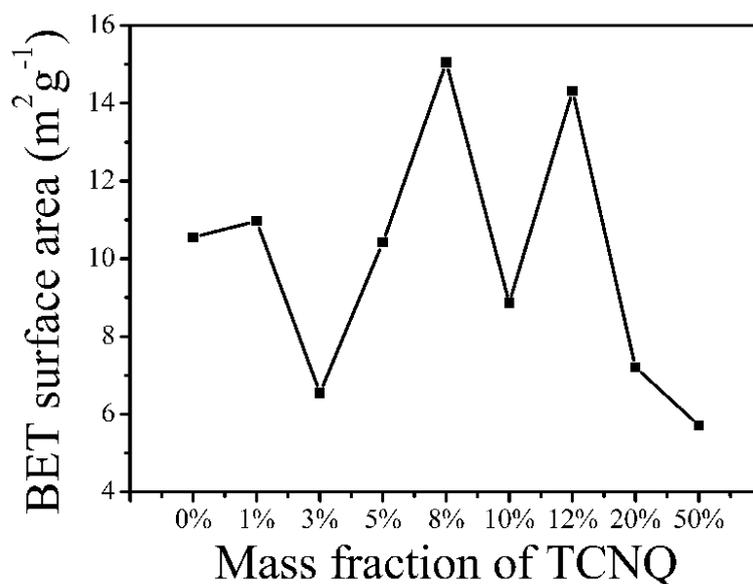


Figure S11 BET specific surface areas of pure g-C<sub>3</sub>N<sub>4</sub>, pure TCNQ and TCNQ-C<sub>3</sub>N<sub>4</sub> with different TCNQ mass fraction.