Supplementary Information For:

Simple Pyrolysis of Urea into Graphitic Carbon Nitride with Recyclable Adsorption and Photocatalytic Activity

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1. Preparation of g-C_3N_4:

Graphitic carbon nitride (g-C_3N_4) was synthesized by thermal treatment of urea (10g, AR, Sinopharm Chemical Reagent Co.,Ltd) in a crucible with a cover under ambient pressure in air. After dried at 80 °C, the urea was put in a Muffle Furnace (Isotemp Programmable Muffle Furnace 650-750 Series, Fisher Scientific) and heated to 550 °C for 3 hours to complete the reaction. The yellow-colored product was washed with nitric acid (0.1 mol L^{-1}) and distilled water to remove any residual alkaline species (e.g. ammonia) adsorbed on the sample surface, and then dried at 80 °C for 24 hours. The weight of the yellow colored powders was 0.4 g.

2. Characterization:

EDX were taken on Quanta 400FEG (FEI). TEM images were obtained on Tecnai G2 F20 S-Twin (FEI, 200 kV). UV-Vis absorption spectra were obtained on a UV-2550 (Shimadzu). XRD patterns were identified by X'Pert-Pro MPD (Cu Kα, $\lambda = 1.5406$ Å). UV-Vis diffuse-reflectance spectra (UV-Vis DRS) were recorded on Cary 5000 with Integrating Sphere Attachment (incidence angle to reflecting sample: 12.5°). FTIR transmission spectra were performed on Nicolet 6700 IR spectrometer by using KBr pellets. XPS spectra were measured on an ESCALAB 250 x-ray photoelectron spectrometer using Al Kα x-ray as an excitation source.

3. Photocatalytic decomposition of methylene blue (MB):

The g-C_3N_4 photocatalyst (100 mg) was dispersed in 100 mL MB solution with initial concentration 10 mg L^{-1} by sonication for 10 min. Then it was kept in dark for 1 hour under stirring to reach saturation adsorption. After that, the photocatalytic decomposition of methylene blue (MB) started under visible light irradiation (>400 nm). The light source was a Xe lamp (PLS-SXE 300, 4.1 mW cm^{-2}) with filter cut-off 400 nm. The recycle experiments were performed. After filtering the sedimentation of photocatalyst by a vacuum filter, the obtained photocatalyst was re-dispersed in 100 mL MB solution (initial concentration 10 mg L^{-1}) by sonication for 10 min. The adsorption and photocatalytic process were the same with the above mentioned one.

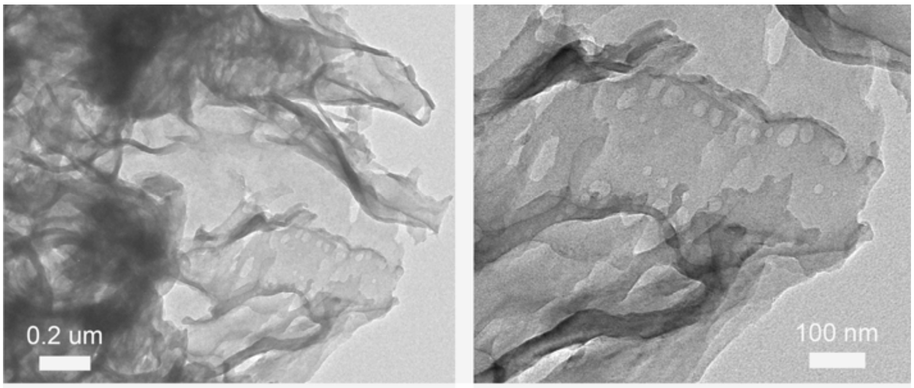
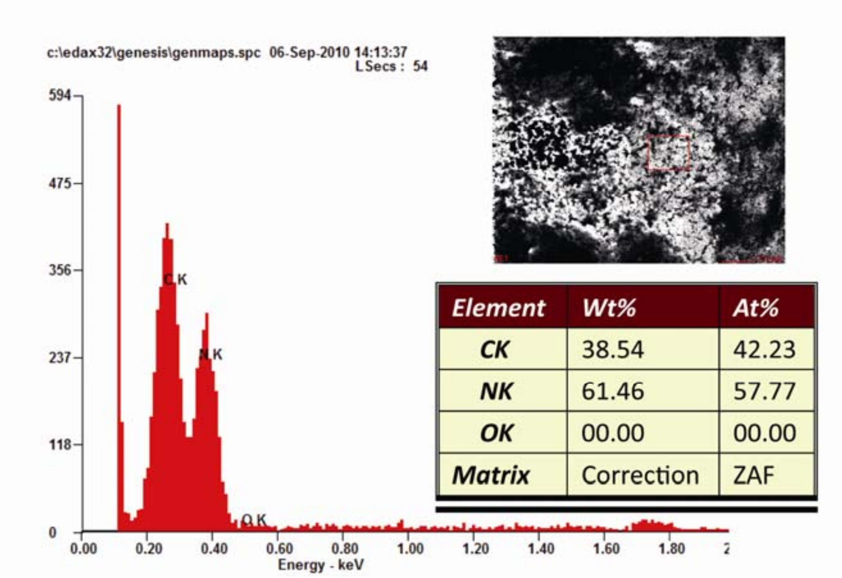


Figure S1. TEM and magnified TEM images for graphitic carbon nitride (g-C_3N_4).



Element	*Wt%*	*At%*
CK	38.54	42.23
NK	61.46	57.77
OK	00.00	00.00
Matrix	Correction	ZAF

Figure S2. The EDX spectrum for elemental compositions of the yellow colored g-C_3N_4.

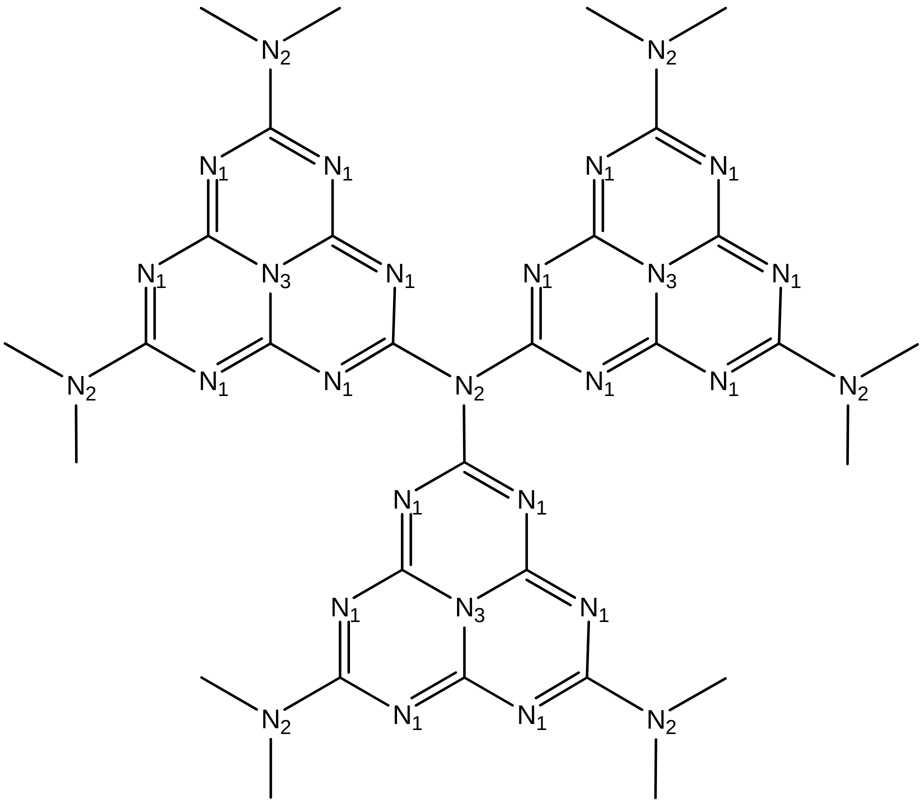


Figure S3. The molecular structure of heptazine-derived repeating units of g-C_3N_4. N_1, N_2 and N_3 represent three types of deconvoluted N1s signal in XPS spectra.

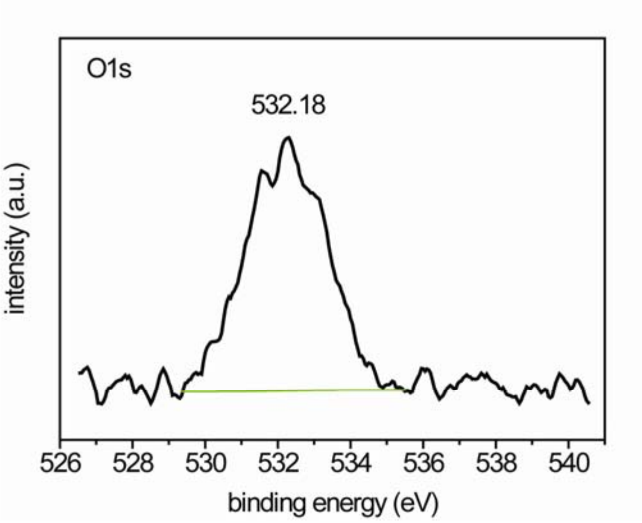


Figure S4. The O1s XPS spectrum for g-C_3N_4. From the XPS peak table, the O At% was 2.01%.

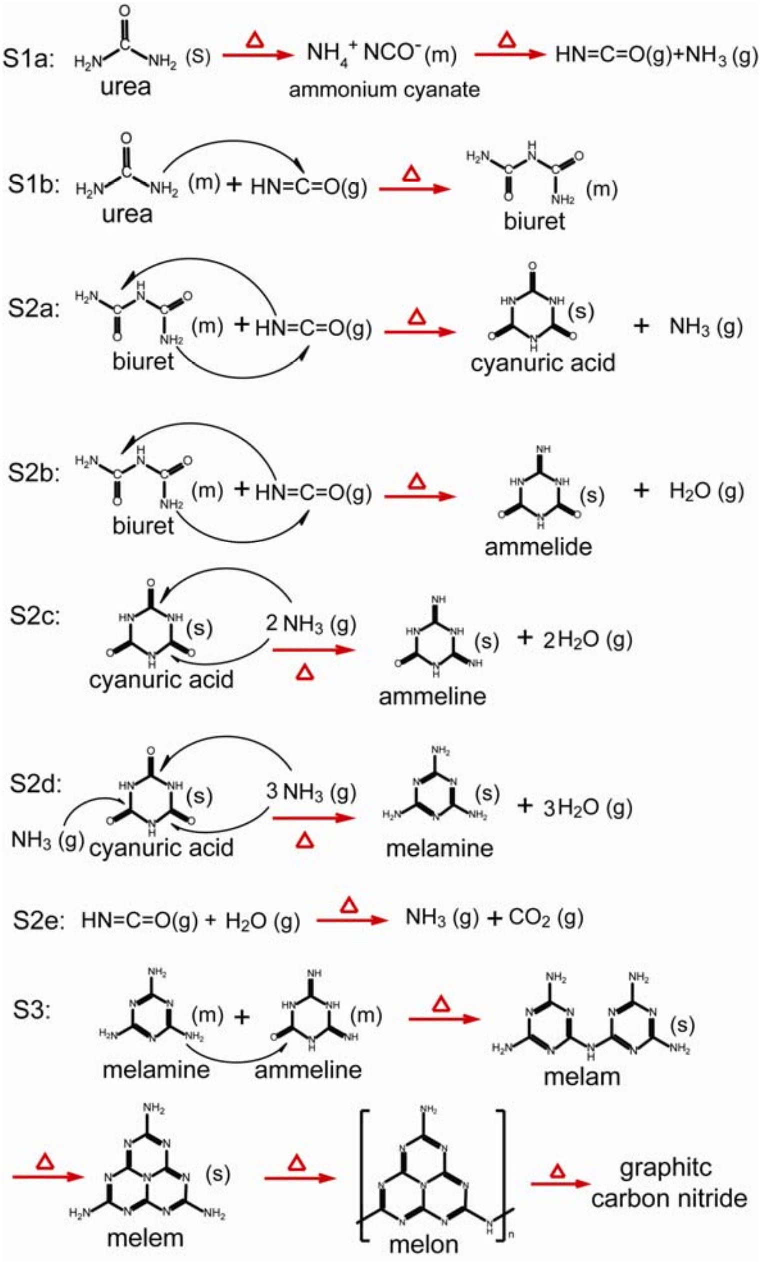


Figure S5. The proposed reaction mechanism for the pyrolysis of urea into g-C_3N_4. S: the reaction steps. The oxygen (O) could be eliminated from the final product at S2b as H_2O (g) and S2e CO_2 (g).

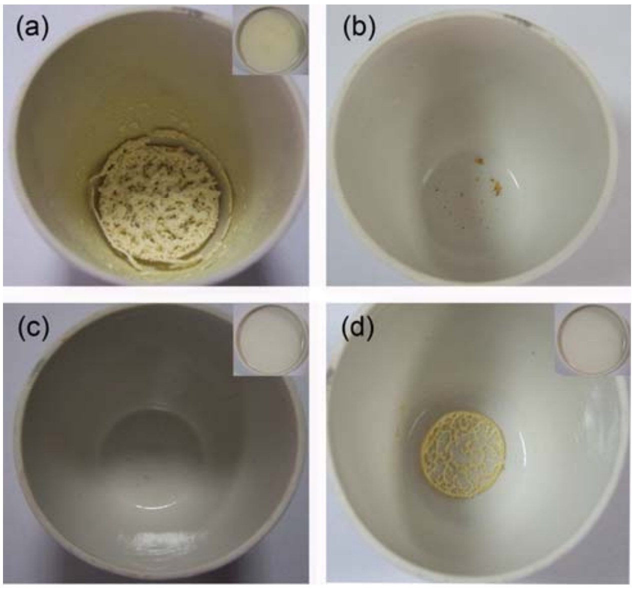


Figure S6. The photos show the retainability and delayed-release effects of pyrolysis-generated self-supporting atmosphere, reaction temperature and amount of starting urea on producing g-C_3N_4 solid. (a) Solid was produced in a covered crucible with 5 g urea at 550 °C. (b) No solid was produced in an open crucible with 10 g urea at 550 °C. (c) No solid was produced in a covered crucible with 5 g urea at 600 °C. (d) Solid produced in a covered crucible with 3 g urea at 550 °C. Inset: Cover of the crucible for (a), (c) and (d), and no cover for (b).

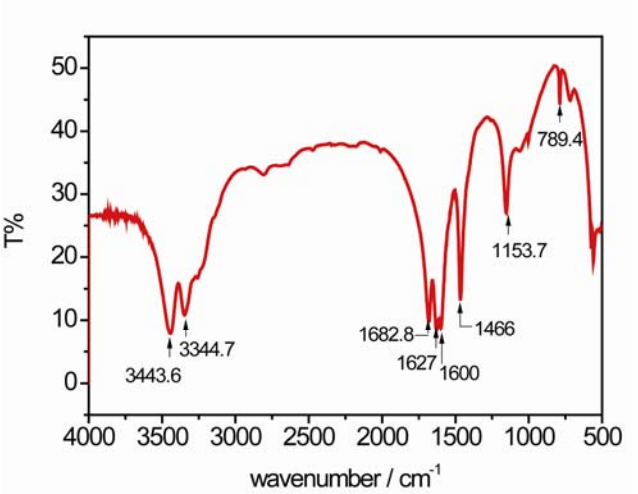


Figure S7a. The FTIR spectra of starting urea. The FTIR bands at 3443.6 and 3344.7 cm^{-1} are N-H stretching vibration. The 1682.8 cm^{-1}represents the C=O stretching vibration. The 1627 and 1600 cm^{-1} are assigned to N-H bending vibration. The 1466 cm^{-1} are identified to C-N stretching vibration.

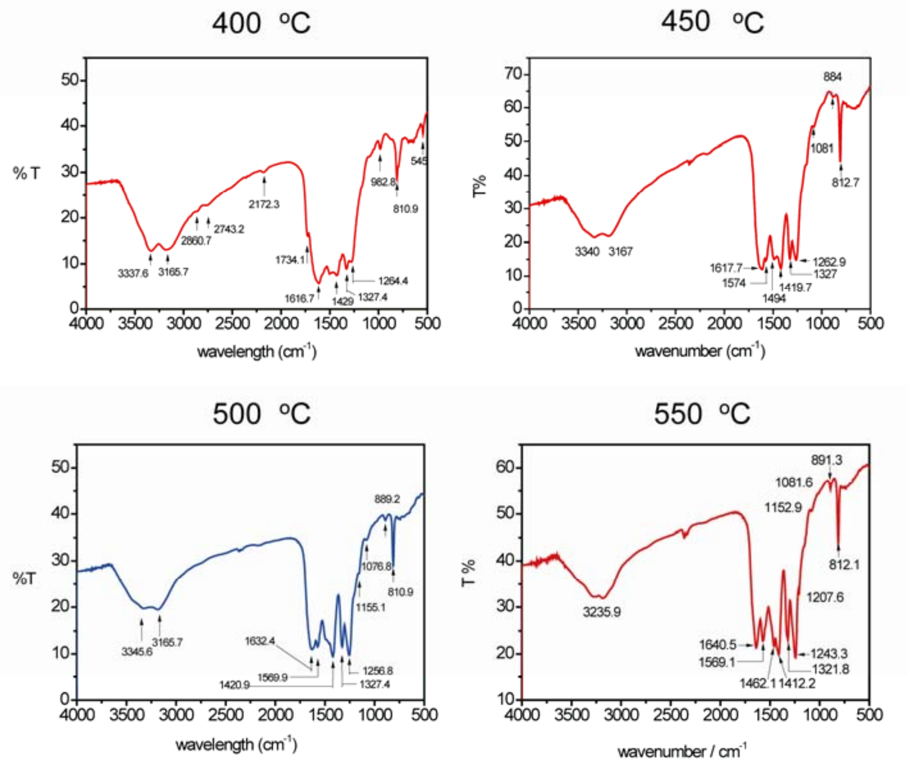


Figure S7b. The FTIR spectra for the evolution of the molecular structure of the graphitic carbon nitride obtained at different temperature.

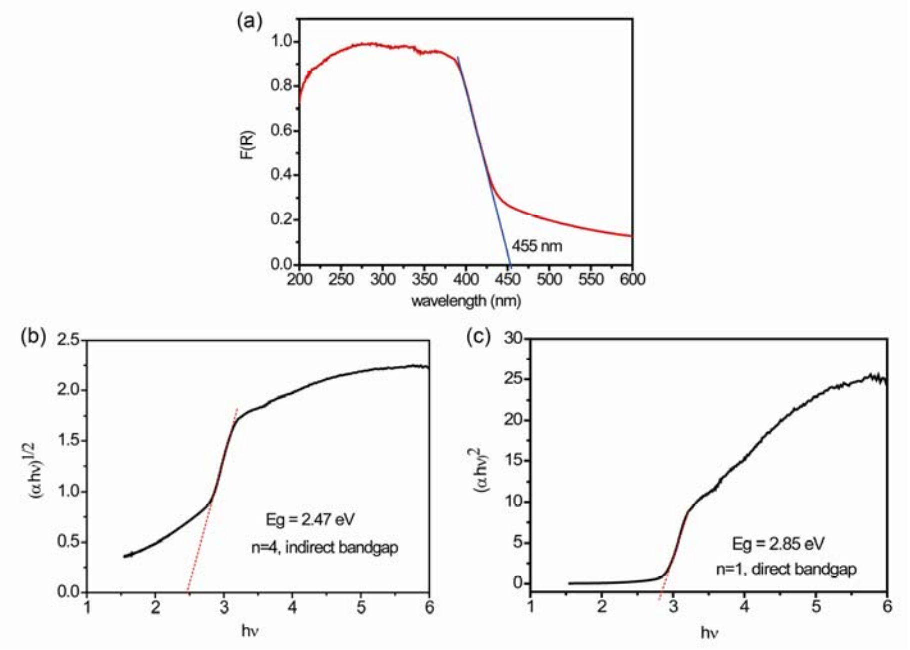


Figure S8. The estimated optical absorption bandgap of g-C_3N_4. It follows the equation: $\alpha h\upsilon = A(h\upsilon - Eg)^{n/2}$, in which α, υ, A, and Eg are the absorption coefficient, light frequency, proporitionality constant and bandgap, respectively. n=1, direct absorption and n=4, indirect absorption [1].

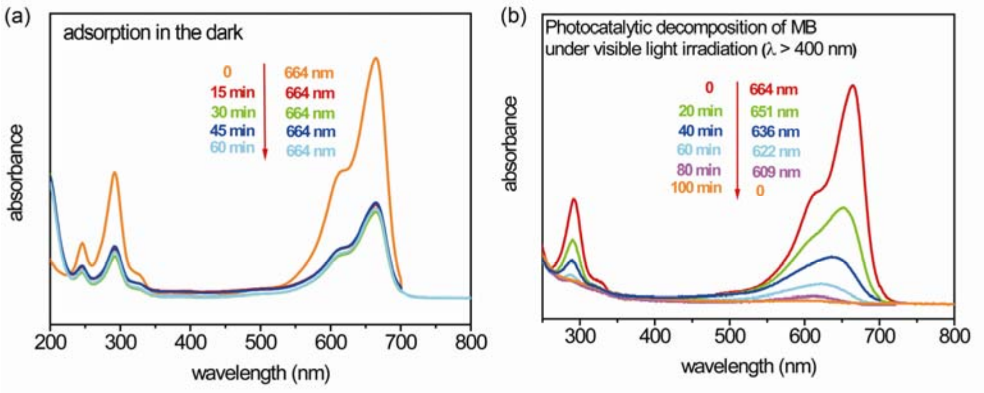


Figure S9. The change of optical absorption of MB solution during the adsorption in the dark (a) and photocatalytic degradation process under visible light irradiation (> 400 nm) (b).

References:

1. (a) Z. G. Yi, J. H. Ye, N. Kikugawa, T. Kako, S. X. Ouyang, H. Stuar-Williams, H. Yang, J. Y. Cao, W. J. Luo, Z. S. Li, Y. Liu, R. L. Withers, *Nat. Mater.* 2010, **9**, 559-564. (b) M. A. Butler, *J. Appl. Phys.* 1977, **48**, 1914-1920.