

Soft-template Synthesis of Mesoporous Carbon Nitride with Enhanced Photocatalytic H₂ Evolution under Visible Light

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Supporting Information

Experimental Section

1. Materials

Melamine, sulfuric acid (H₂SO₄) and triethanolamine were purchased from Chengdu Kelong Chemical Reagent Factory. Pluronic P123 was purchased from Aldrich. All reagents were used as received without further treatment.

2. Synthesis of mesoporous graphitic carbon nitride

The following is a typical procedure for synthesis of worm-like mesoporous graphitic carbon nitride.

The melamine (5.0 g) and Pluronic P123 (1.0 g) were dispersed in distilled water (100 mL) by heating at 100 °C for 1 h under reflux. White precipitate was produced by adding sulfuric acid solution (3 mL, H₂SO₄:H₂O = 1:1 in volume) to the solution. After cooling down to room temperature, the precipitate was collected by filtration, and then dried in an oven at 80 °C for over night.

The precipitate (3.0 g) was pressed into a quartz boat and then placed in the middle region of a quartz tube with 30 mm in inner diameter and 1000 mm in length. The quartz tube was heated to 380 °C in 5 minutes and then heated to 600 °C in a muffle furnace for 4 h at a heating rate of 1 °C min⁻¹ in the flow of Ar gas. After the reaction, the quartz tube was cooled down to room temperature in the flow of Ar gas. Finally, the product was then calcined at 500 °C for 2 h in air.

3. Characterization

Elemental analyses (C, H, N, S) were performed on a Euro EA3000 elemental analyser (Euro Vector S.P.A., Italy). XRD patterns were recorded on a X-Pert Pro diffractionmeter with CuKα radiation ($\lambda = 1.5406 \text{ \AA}$) at a scanning speed of 4° min⁻¹. UV-vis diffuse reflection spectra were measured using a UV-vis spectrophotometer (UV2100, Shimadzu) and converted from reflection to

absorbance by the Kubelka-Munk method. The BET specific surface areas were measured by N₂ adsorption at 77 K on an Quantachrome Instruments (Autosorb 1-C). The Fourier transform infrared spectra (FTIR) of the samples were performed using a Fourier transform infrared (FTIR) spectrometer (Nicolet 670) only in the mid infrared range of the instrument (400 ~ 4000 cm⁻¹) for samples dispersed in KBr pellets in 1:99 ratio. The morphology of the composite materials was characterized using Tecnai G² F20 S-TWIN microscope.

4. Photocatalytic reactions

The photocatalytic H₂-production experiments were performed in a 300 mL Pyrex cell connected to a closed gas circulation and evacuation system. A 300 W Xe arc lamp (Cermax® Xenon arc lamp, PE300BF) through an optical cutoff filter (HOYA, L42) to eliminate ultraviolet light was used as the light source. The loading of Pt (0.5 wt. %) co-catalyst on the g-C₃N₄ was performed by an in-situ photodeposition method. In a typical photocatalytic experiment, the prepared g-C₃N₄ (0.3 g) was suspended in 200 mL of aqueous solution containing equivalent molar amount of H₂PtCl₆ and 20 mL of triethanolamine as a sacrificing agent. The suspension was then thoroughly degassed and irradiated by Xe lamp (300 W). The temperature of the reactant solution was maintained at 283 K by a flow of cooling water during the reaction. The amount of H₂ produced was analyzed with an on-line TCD gas chromatograph (SPSIC, GC-102AT, argon carrier). The activities of different catalysts were compared by the average rate of H₂ evolution in the first 4 h.

The apparent quantum efficiency (QE) was estimated by the method described previously^{1,2} with irradiation light at 420 ± 10 nm by using combined band-pass and cut-off filters, and the QE was calculated according to the following equation:

$$QE(\%) = \frac{2 \times H}{I} \times 100$$

where *H* and *I* represent the numbers of evolved H₂ molecules and incident photons, respectively. The number of incident photons was 1.3 × 10²¹ photons h⁻¹ which was measured by using a calibrated Si photodiode.

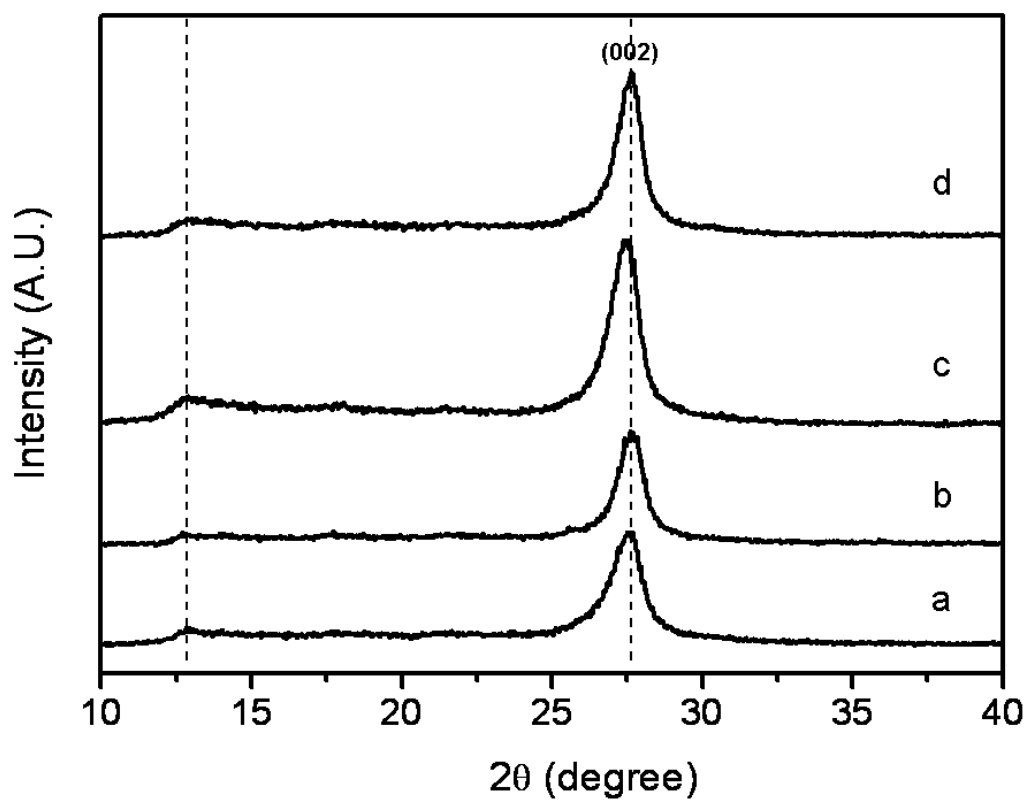


Figure S1 XRD patterns of g-C₃N₄ prepared without (a, b) and with (c, d) using P123 surfactant. b and d denote the samples calcined at 500 °C for 2 h in air.

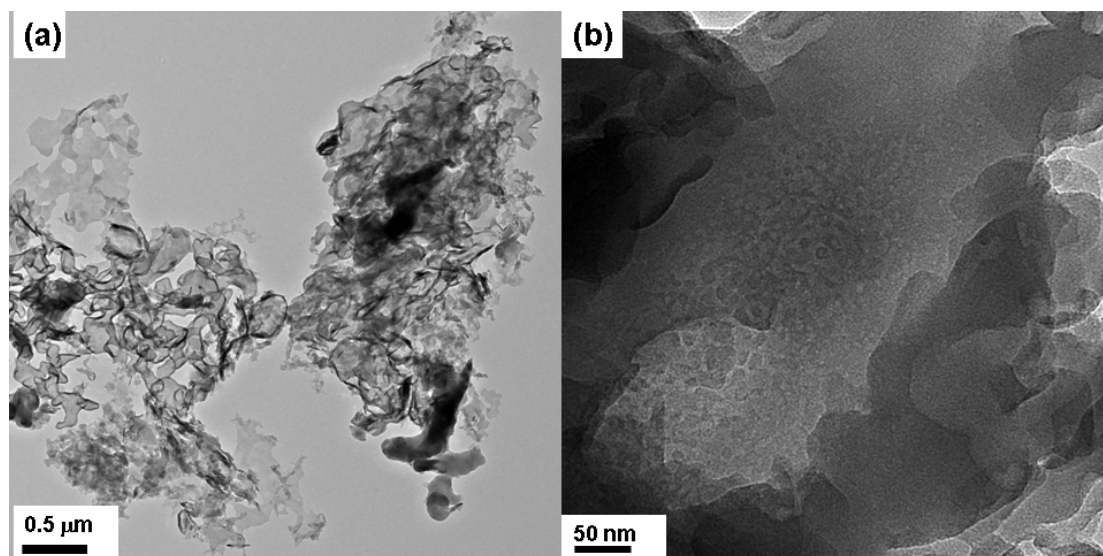


Figure S2 The TEM image of g-C₃N₄ prepared with using P123 surfactant as soft template before calcination in air. (a) low magnification; (b) high magnification.

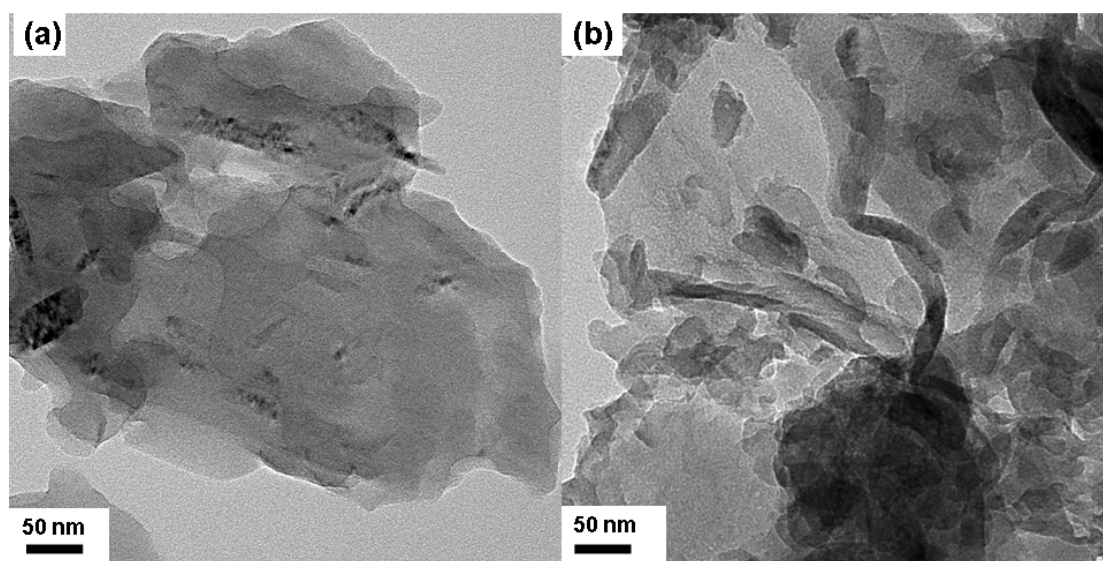


Figure S3 The TEM image of g-C₃N₄ prepared without using P123 surfactant. (a) without calcined in air; (b) calcined at 500 °C for 2 h in air.

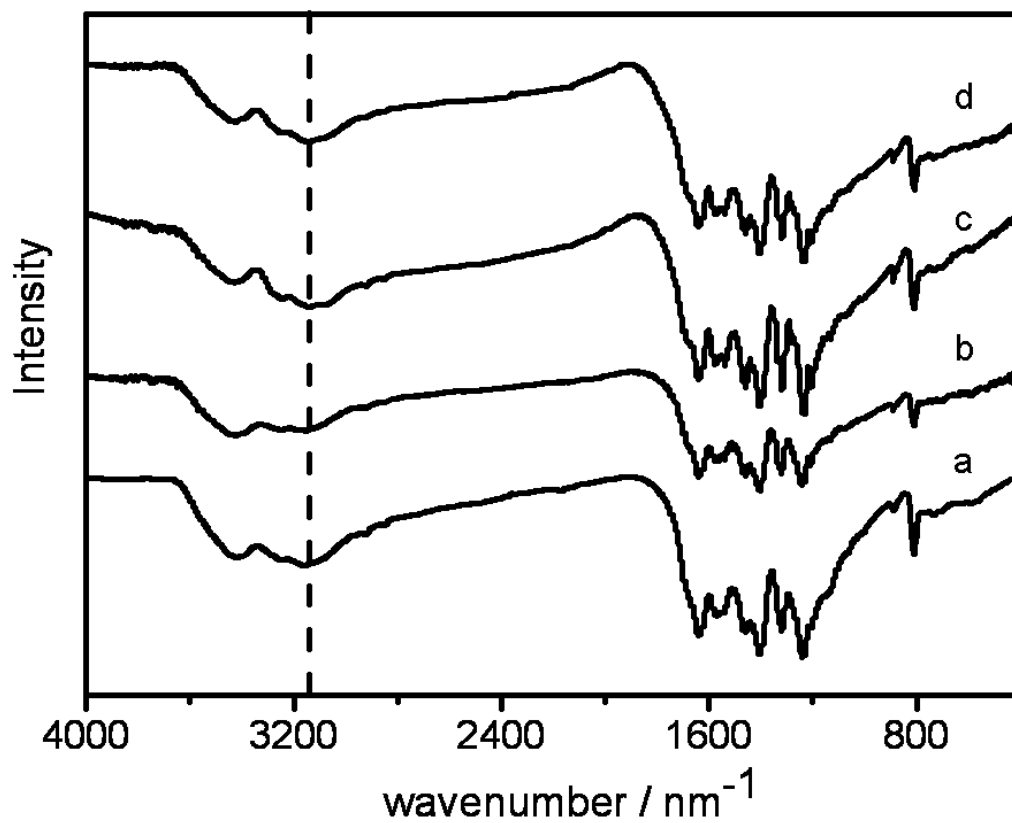


Figure S4 FT-IR spectra of $\text{g-C}_3\text{N}_4$ prepared without (a, b) and with (c, d) using P123 surfactant. b and d denote the samples calcined at 500 °C for 2 h in air.

Supporting References

- (1) Yan, H.; Yang, J.; Ma, G.; Wu, G.; Zong, X.; Lei, Z.; Shi, J.; Li, C. *J. Catal.* **2009**, 266, 165-168.
- (2) Wang, X.; Maeda, K.; Thomas, A.; Takanabe, K.; Xin, G.; Carlsson, J. M.; Domen, K.; Antonietti, M. *Nat. Mater.* **2009**, 8, 76-80.