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# Highly efficient composite visible light-driven Ag–AgBr/g-C<sub>3</sub>N<sub>4</sub> plasmonic photocatalyst for degrading organic pollutants

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## ABSTRACT

As a potential visible-light photocatalyst, the efficiency of graphite-like carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) has been limited by its wide band gap. In this paper, Ag/AgBr- functionalized g-C<sub>3</sub>N<sub>4</sub> nanostructured composites have been fabricated through in situ photoreduction of AgBr/g-C<sub>3</sub>N<sub>4</sub> prepared by an oil/water microemulsion method, which effectively inhibits the agglomeration of AgBr on the surface of g-C<sub>3</sub>N<sub>4</sub>. The as-prepared Ag/AgBr/g-C<sub>3</sub>N<sub>4</sub> is used as a stable plasmonic photocatalyst for photodegradation of methyl orange (MO) pollutant under visible light. Compared with single-phase g-C<sub>3</sub>N<sub>4</sub>, the catalytic activities of Ag/AgBr/g-C<sub>3</sub>N<sub>4</sub> nanocomposites are greatly enhanced mainly due to fast separation of photo-generated electron–hole pairs and improved absorption of visible light. The material shows good stability of its photocatalytic activity upon reusability test, which could lead to potential applications in pollution treatment.

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## 1. Introduction

For decades, the creation of efficient photocatalysts working with visible light has been considered as one of the promising strategies to solve the future energy shortages by converting sustainable solar energy into electric and chemical energies [1,2]. Among all photocatalysts, graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>), a novel metal-free photocatalyst, has attracted much attention due to its abundance, non-toxicity and amenability to chemical modification [3,4], making it suitable for a range of applications [5–7]. As an analogue to graphite, g-C<sub>3</sub>N<sub>4</sub> possesses sp<sup>2</sup> hybridization of carbon and nitrogen forming the delocalized conjugated  $\pi$  structures. It has been reported that this unique structure can efficiently arouse a rapid photo-induced charge separation and a relatively slow charge recombination in electron-transfer processes [8,9]. However, the g-C<sub>3</sub>N<sub>4</sub> only responses in a limited region of visible light and utilizes a very small part of solar energy due to its large band gap of 2.7 eV, which greatly hinders its practical applications [10,11].

Recently, Ag/AgX has been proved to be a promising plasmonic photocatalyst [12]. The metallic Ag<sup>0</sup> species on the surface could dramatically amplify the absorption of visible light and suppress the photocorrosion of AgX [13,14]. Thus, various endeavors have been made to improve the catalytic activity of large-band gap

semiconductors by coupling with Ag/AgBr [15,16]. However, to the best of our knowledge, only a few reports have been related to the synthesis of Ag/AgBr nanoparticles coupled with g-C<sub>3</sub>N<sub>4</sub> [17,18]. Herein, we develop a facile oil/water microemulsion method followed by subsequent light-driven fabrication of g-C<sub>3</sub>N<sub>4</sub> with Ag/AgBr, which makes the Ag/AgBr nanoparticles highly dispersed on the sheets of g-C<sub>3</sub>N<sub>4</sub>.

## 2. Experimental section

The g-C<sub>3</sub>N<sub>4</sub> was synthesized by directly heating urea according to a reported procedure [19]. The Ag/AgBr/g-C<sub>3</sub>N<sub>4</sub> photocatalysts were fabricated as follows: a certain amount of the prepared g-C<sub>3</sub>N<sub>4</sub> was dispersed in 10 mL of deionized water and sonicated for 1 h. Then 0.009 g of AgNO<sub>3</sub> was added into the above suspension and stirred for 30 min in the dark. After that 500  $\mu$ L of chloroform containing 0.03 g hexadecyltrimethylammonium bromide (CTAB) was added into the above Ag<sup>+</sup>-g-C<sub>3</sub>N<sub>4</sub> suspension drop by drop. After stirring for another 30 min, the dispersion with a pale yellow color was irradiated using a 300 W Xe lamp (Feilai Bo Technology Co., Ltd., China) equipped with a 400 nm cut-off filter. The product was obtained by centrifugation, washed with ethanol and deionized water, and dried at 333 K for 12 h. The Ag/AgBr/g-C<sub>3</sub>N<sub>4</sub> composites with different mass ratios were synthesized using the same method through changing the amount of g-C<sub>3</sub>N<sub>4</sub>. The samples were denoted as Ag/AgBr/g-C<sub>3</sub>N<sub>4</sub>-X (X=10, 15, 25, 30,

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35, 45) when the mass ratio of  $\text{AgNO}_3$  to  $\text{g-C}_3\text{N}_4$  were 10%, 15%, 25%, 30%, 35%, 45%, respectively.

Powder X-ray diffraction (XRD) measurements were determined by Siemens D5005 Diffractometer with  $\text{Cu K}\alpha$  radiation ( $\lambda=1.5418 \text{ \AA}$ ). The surface characteristics and microstructures of the as-prepared samples were examined through a JEM-2100F transmission electron microscope (TEM) with an accelerating voltage of 200 kV. Thermo ESCALAB 250 X-ray photoelectron spectroscopy (XPS) was employed for investigation of the surface composition of the catalysts. Photoluminescence (PL) spectra were obtained with FLSP920 Edinburgh Fluorescence Spectrometer. UV-vis diffuse reflectance spectroscopy (DRS) were collected through a Cary 500 spectrometer.

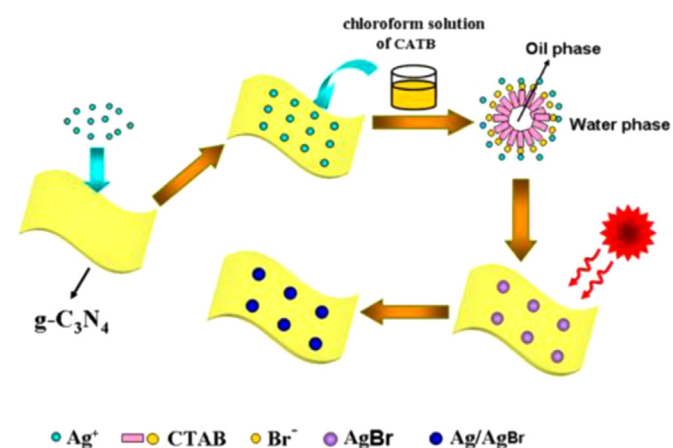
The photocatalytic activities of the samples were evaluated through photodegradation of MO under visible light irradiation

supplied by a 300 W Xe lamp with a 400 nm cut-off filter. The suspension including the photocatalysts (50 mg) and MO solution (100 mL,  $10 \text{ mg L}^{-1}$ ) were laid in a beaker. Before irradiation, the reaction suspension was ultrasonicated for 10 min and stirred in the dark for 40 min to reach adsorption-desorption equilibrium. Then the suspension was exposed to visible-light irradiation with magnetic stirring. At given time intervals, 3 mL of suspension was obtained and with the photocatalyst removed by centrifuged. The degradation results were monitored by UV-vis-NIR (Purkinje General, TU-1900) spectrophotometer.

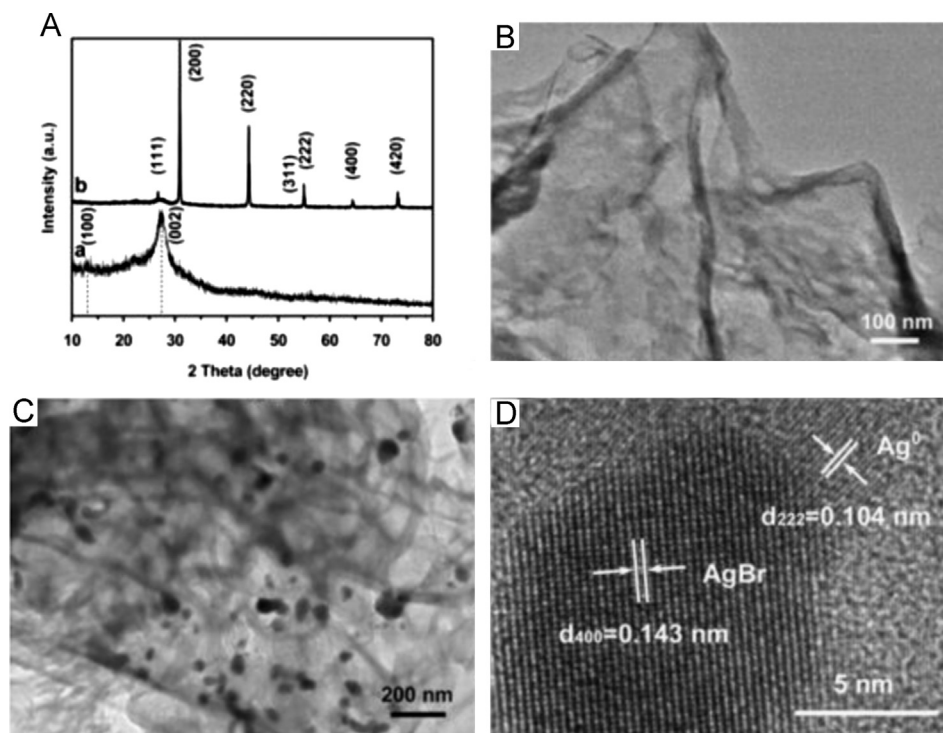
### 3. Results and discussion

In this work,  $\text{Ag/AgBr/g-C}_3\text{N}_4$  photocatalysts are fabricated by the in situ photoreduction of  $\text{AgBr/g-C}_3\text{N}_4$  hybrids prepared by a microemulsion method at room temperature (Scheme 1). First, the as-prepared  $\text{g-C}_3\text{N}_4$  is dispersed into deionized water by sonication. When  $\text{AgNO}_3$  solution is added, amino groups on the sheet surface of  $\text{g-C}_3\text{N}_4$  can coordinate to the  $\text{Ag}^+$  ions tightly. Second, with the addition of chloroform containing CTAB, an oil/water microemulsion is formed. The  $\text{Ag}^+$  attached to the surface of  $\text{g-C}_3\text{N}_4$  reacts with  $\text{Br}^-$  from CTAB to generate  $\text{AgBr}$  nanoparticles. Microemulsion system facilitates the well-dispersion of the as-prepared  $\text{AgBr}$  particles on  $\text{g-C}_3\text{N}_4$  sheets. Finally, the resulting  $\text{AgBr/g-C}_3\text{N}_4$  is illuminated under visible light and  $\text{Ag/AgBr/g-C}_3\text{N}_4$  nanocomposites are obtained.

The phase structures of the samples were examined by XRD. As shown in Fig. 1A, the dominant diffraction peak at  $27.7^\circ$  corresponds to the (0 0 2) diffraction planes of  $\text{g-C}_3\text{N}_4$  and a small diffraction peak at approximately  $13^\circ$  can be indexed to in-plane of tris-triazine units [6,20]. After decoration with  $\text{Ag/AgBr}$  nanoparticles, the composite shows seven new diffraction peaks, which all can be indexed to cubic structure of  $\text{AgBr}$  (JCPDS 06-0438). No clear peaks of  $\text{Ag}^0$  are observed, possibly due to the low content of



**Scheme 1.** Schematic diagram of the fabrication of  $\text{Ag/AgBr/g-C}_3\text{N}_4$ -30 nanocomposites.



**Fig. 1.** (A) XRD patterns of pure  $\text{g-C}_3\text{N}_4$  (a) and  $\text{Ag/AgBr/g-C}_3\text{N}_4$ -30 composite (b), (B) TEM image of pure  $\text{g-C}_3\text{N}_4$ , TEM (C) and HRTEM images (D) of  $\text{Ag/AgBr/g-C}_3\text{N}_4$ -30 composite.

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