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Invited paper

Silver nanoparticles/graphitic carbon nitride nanosheets for improved visible-light-driven photocatalytic performance



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ABSTRACT

The binary nanocomposite of silver nanoparticles/graphitic carbon nitride nanosheets (AgNPs/g-C₃N₄ nanosheets) has been prepared through polymerization-photodeposition method. The UV-vis absorption spectra show that the composite material has a significant absorption in the visible region, and indirectly demonstrates the surface plasmon resonance (SPR) of Ag nanoparticles in composites. The as-prepared AgNPs/g-C₃N₄ nanosheets photocatalysts exhibit distinctly enhanced photocatalytic activities toward the degradation of Methyl orange (more than 95% after 60 min) under visible light irradiation, which is about 20 and 3 times higher than that of bulk g-C₃N₄ and g-C₃N₄ nanosheets. The cyclic stability test proves that hybrid photocatalyst has a strong stability in the degradation of organic dyes. The improved visible-light-driven photocatalytic activities of AgNPs/g-C₃N₄ nanosheets can be attributed to the SPR of Ag nanoparticles as well as generation, separation and transportation of the photogenerated carriers, and the higher specific surface area for improved visible-light utilization efficiency.

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1. Introduction

Semiconductor photocatalysis was widely deemed as a very promising strategy for restoring energy and addressing environmental issues because semiconductor catalysts and solar energy are the most economical and environment-friendly [1,2]. Seeking efficient, stable, inexpensive, and facile semiconductor photocatalysts is the final objective. Despite the immense amount of research on developing highly active semiconductor materials, there remains the general problem that most metal oxides only react under UV light driven, which accounts for ~5% of natural solar irradiation [3]. So far, many impressive photocatalysts have been discovered and have shown excited photoreactivity by exploring new materials, improving technologies and so on [4,5].

Recently, graphitic carbon nitride (g-C₃N₄), a polymer with a graphite-like structure, is receiving increasing attention because of its potential for photocatalytic applications [6]. Graphitic carbon nitride possesses a large bandgap ~2.7 eV. Its lowest unoccupied molecular orbital and highest occupied molecular orbital are

located respectively at ~-1.3 V and +1.4 V versus normal hydrogen electrode [7]. Additionally, g-C₃N₄ can be easily obtained by thermal condensation reaction of organic precursors such as melamine [8], urea [9], and dicyandiamide [10]. However, g-C₃N₄ suffers from rapid recombination rate of photoinduced charge carriers and poor electrical conductivity which leads to low photocatalytic activities [11]. Strategies that may increase the photocatalytic efficiency of g-C₃N₄ include doping with non-metal element [12–15], loading metal [16–19], improving surface morphology [20–23], and constructing heterojunction composites [24–26]. Kozuka et al. carried out pioneering work in introducing Au and Ag on the TiO₂ layer in photoelectrochemical evaluation [27]. Since then photocatalysts modified with plasmonic nanostructures of noble metals (such as Au, Ag, etc.) offered a new opportunity for developing efficient visible light-responsive composite materials. These noble metal nanoparticles characteristic of the surface plasmon resonance can act as actives for light trapping and form a locally enhanced electromagnetic field nearby the interface of the noble metal nanoparticles/semiconductor. In 2012, Liu et al. reported that graphitic carbon nitride (g-C₃N₄) with nanosheets morphology possessed of much superior photocatalytic activities compared with bulk g-C₃N₄ due to weak Van der Waals forces between layers [22].

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In this study, we improved the photocatalytic performance of $g\text{-C}_3\text{N}_4$, especially visible-light-driven photocatalytic performance, by introducing two modifications: (i) $g\text{-C}_3\text{N}_4$ nanosheets is prepared by direct thermal oxidation “etching” process of bulk $g\text{-C}_3\text{N}_4$ in air, which enhancing a high specific surface area for improved visible-light utilization efficiency. The BET surface area of $g\text{-C}_3\text{N}_4$ nanosheets is $162\text{ m}^2/\text{g}$ using the Barrett-Joyner-Halenda (BJH) method. After Ag nanoparticles deposited on the surface of $g\text{-C}_3\text{N}_4$ nanosheets, The BET surface area of composites is $67\text{ m}^2/\text{g}$. (ii) Silver nanoparticles are deposited on the surface of $g\text{-C}_3\text{N}_4$ nanosheets by simple photochemical deposition. The diameter of Ag nanoparticles is about 15 nm calculated from transmission electron microscopy (TEM) images. The deposited Ag nanoparticles can facilitate generation, separation and transportation of photo-generated electron-hole pairs, and the SPR is occurring on Ag nanoparticles. The visible-light photocatalytic activity is evaluated by the degradation of methyl orange (MO), methyl blue (MB), and rhodamine B (RhB) under visible light irradiation ($>420\text{ nm}$). The other phase and chemical structure, optical absorption properties as well as morphologies of the samples are well characterized.

2. Experimental section

2.1. Materials and reagents

All the reagents were purchased from Sinopharm Chemical Reagent Co. Ltd, which are of analytical-grade, and used as received without any further purification. Water used in all experiments was deionized.

2.2. Synthesis of samples

2.2.1. Preparation of bulk $g\text{-C}_3\text{N}_4$

The bulk $g\text{-C}_3\text{N}_4$ was prepared by the thermal polymerization of dicyandiamide at $550\text{ }^\circ\text{C}$ for 4 h with a ramping rate of $2\text{ }^\circ\text{C}/\text{min}$ in a

muffle furnace at air condition. The naturally cooled product was ground in an agate mortar to obtain a yellow powder sample.

2.2.2. Preparation of $g\text{-C}_3\text{N}_4$ nanosheets

$g\text{-C}_3\text{N}_4$ nanosheets was synthesized via a further thermal polymerization of the above bulk $g\text{-C}_3\text{N}_4$. Above all, the bulk $g\text{-C}_3\text{N}_4$ was put into a ceramic ark under ambient pressure in air, and then the ceramic ark was heated in a muffle furnace at $500\text{ }^\circ\text{C}$ for 3 h with a ramp rate of $5\text{ }^\circ\text{C}/\text{min}$. The yield of $g\text{-C}_3\text{N}_4$ nanosheets with a light yellow colour was about 5%.

2.2.3. Preparation of AgNPs/ $g\text{-C}_3\text{N}_4$ nanosheets

The silver nanoparticles/ $g\text{-C}_3\text{N}_4$ nanosheets was prepared as follows: firstly, 500 mg $g\text{-C}_3\text{N}_4$ nanosheets was dispersed in 500 mL deionized water followed by sonication for 60 min. 1 mM AgNO_3 and 1 mM trisodium citrate were added into this mixed solution under agitation. After 40 min of stirring, the solution was transferred into a sealed UV light box irradiated with UV light for 60 min (wavelength 365 nm, power 56 W and working distance 0.3 m). Finally, the sample was collected by centrifugation using excess deionized water, and dried. Then, named as AgNPs/ $g\text{-C}_3\text{N}_4$ nanosheets.

2.3. Characterization

The powder X-ray diffraction (XRD) measurements were measured by an X-ray diffractometer (XRD, D8 Advance) using $\text{Cu K}\alpha$ radiation ($\lambda = 1.5418\text{ \AA}$). Transmission electron microscopy (TEM) experiments were conducted on a Hitachi ht7700 microscope operated at 100 kV. Before the TEM measurements, the samples were dispersed in ethanol, and then was supported by a holey carbon film on a Cu grid. Fourier-transform infrared spectroscopy (FT-IR) spectra were recorded with a Fouriertransform infrared spectrometer (FT-IR, PerkinElmer Spectrum Two) using the KBr method at room temperature. The Brunauer-Emmett-Teller (BET, Micromeritics Tristar II3020) specific surface

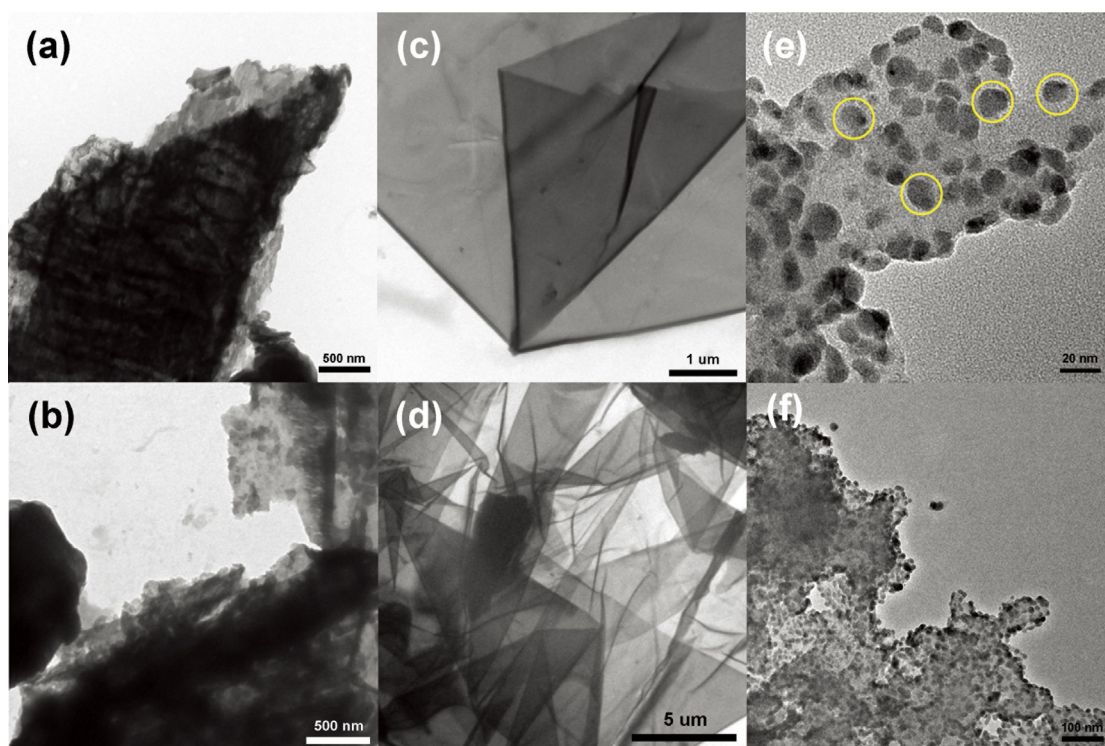


Fig. 1. TEM images of (a, b) Bulk $g\text{-C}_3\text{N}_4$, (c, d) $g\text{-C}_3\text{N}_4$ nanosheets, and (e, f) AgNPs/ $g\text{-C}_3\text{N}_4$ nanosheets. Within the circle is Ag Nanoparticles.

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