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Ag₂MoO₄ nanoparticles encapsulated in g-C₃N₄ for sunlight photodegradation of pollutants

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ABSTRACT

Graphitic carbon nitride (g-C₃N₄), a promising metal-free photocatalyst, shows a high thermal stability, excellent chemical stability, and great optical absorption of solar light. However, a short life of activated electron-hole pairs limits g-C₃N₄ in practical applications. In this work, Ag₂MoO₄ nanoparticles encapsulated in g-C₃N₄ (Ag₂MoO₄/g-C₃N₄) was synthesized with a facile *in-situ* precipitation method. A variety of characterization techniques were applied for analyzing the compositions, morphologies and optical properties of Ag₂MoO₄/g-C₃N₄. The band structure of Ag₂MoO₄ produces a synergistic effect with g-C₃N₄, which can efficiently increase solar light absorption and reduce the recombination rate of the photo-induced electron-hole pairs. Therefore, this hybrid catalyst presents a much higher photocatalytic activity for the degradation of various organic pollutants (bisphenol A, acyclovir, and methyl orange) and strong stability under both artificial and real sunlight, which is promising for practical application.

1. Introduction

Since the 21st century, energy crisis and environmental pollution become more and more serious with the significant growth of population. Water pollution has attracted more attention as resources of drinking water are remarkably reducing. Organic wastes from industries and our daily living are toxic and harmful, which can contaminate water and eventually threaten human health [1]. Some of organic wastes like dyes [2], phenol [3], phenolic compounds [4,5], and pharmaceuticals etc. [6,7] cannot be directly bio-degraded [8,9]. Therefore, treatments of these organic wastes in water have been a hot research topic all over the world. Many methods have been explored, including photocatalysis [8–11], adsorption [12], electrochemical decomposition [13], microbial degradation [14], advanced oxidation [15] and others [16,17]. Photocatalysis is considered as a promising and environmentally friendly method for using clean solar energy and no emission of harmful substances (CO₂ and H₂O). Graphitic carbon nitride (g-C₃N₄) was discovered for water-splitting in 2009, which attracted great attention afterwards from all over the world [18–20].

Meanwhile, the excellent properties of g-C₃N₄, such as great thermal and chemical stability, good optical absorption for sunlight, make it one of the most popular photocatalysts in environmental and energy applications [21–23]. However, pristine g-C₃N₄ has not yet reached the requirements for practical applications due to the short time of the recombination of photogenerated electron-hole pairs [24,25].

Introduction of Ag or its compounds is considered as an effective strategy to enhance the photocatalytic activity of g-C₃N₄ by hindering the electron-hole recombination. For example, Ag [26–28], Ag₂O [29–32], Ag₂CO₃ [33], Ag₂MoO₇ [34], Ag₃VO₄ [35], Ag₃PO₄ [36,37], Ag/AgWO₃ [38], and Ag/AgBr [39–43] have been attempted for enhancing photocatalytic activity of pristine g-C₃N₄. Another Ag-based compound, Ag₂MoO₄, was also investigated because of its excellent properties like great electrical conductivity, high antimicrobial activity, nontoxicity and great photocatalytic activity [44–48] for degradation of organic dyes. However, it can only be excited under ultraviolet light irradiation due to a wide bandgap of 3.05 eV [49–51]. Recently, Ag₂MoO₄/g-C₃N₄ was reported for degradation of a dye under visible light irradiation [52], but more research on its synthesis, catalytic

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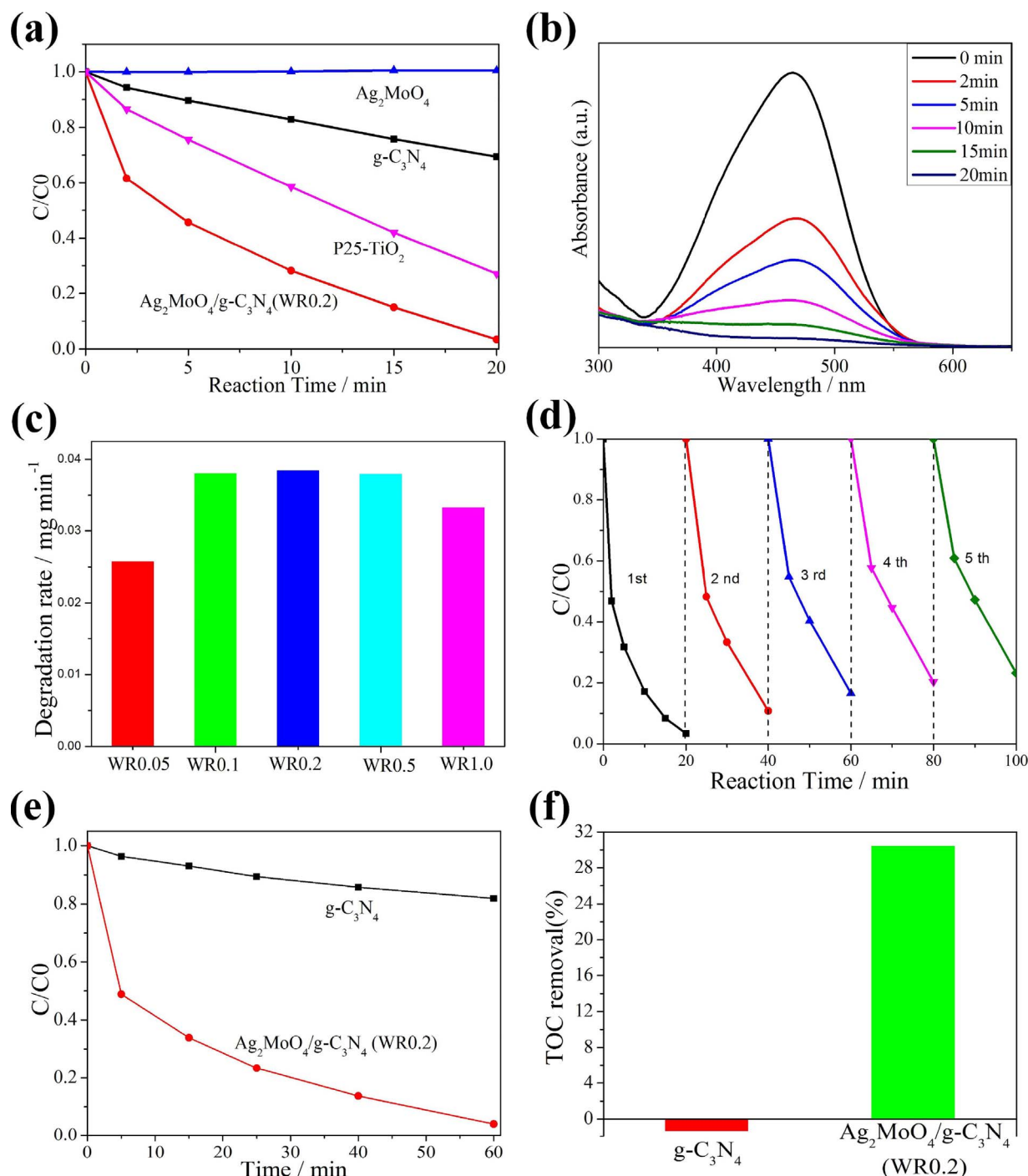


Fig. 1. (a) Photodegradation of MO with Ag_2MoO_4 , $\text{g-C}_3\text{N}_4$, $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4(\text{WR0.2})$ and P25-TiO_2 . (b) The given time UV/Vis spectrum of MO degradation solution by $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4(\text{WR0.2})$. (c) Photodegradation rate of MO with different $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4$ samples under 20 min reaction. (d) Recycle tests of $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4(\text{WR0.2})$ for photodegradation of MO. The photocatalytic reactions of (a-d) were conducted with simulated sunlight (Xe-light). (e) Photodegradation of MO with $\text{g-C}_3\text{N}_4$ and $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4(\text{WR0.2})$ under natural sunlight irradiation. (f) TOC removal of MO degraded by $\text{g-C}_3\text{N}_4$ and $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4(\text{WR0.2})$ after photocatalytic reaction under natural sunlight irradiation.

mechanism and wide application for different organic pollutants is still required.

In this paper, we report a simple synthesis of $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4$ hybrids using a facile *in-situ* precipitation method. The $\text{Ag}_2\text{MoO}_4/\text{g-C}_3\text{N}_4$ samples showed much higher photocatalytic activity than pristine $\text{g-C}_3\text{N}_4$ in degradation of organic pollutants including bisphenol A, acyclovir, and methyl orange (MO) under sunlight irradiation. The photocatalytic reaction mechanism and enhancement mechanism of this hybrid catalyst under sunlight were also investigated.

2. Experiments

2.1. Chemicals

Urea ($\text{CO}(\text{NH}_2)_2$, Aladdin Chemistry Reagent Co., Ltd, $\geq 99.5\%$), sodium molybdate ($\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$, Tianjin Chemical Reagent Co., Ltd, $\geq 99.0\%$), silver nitrate (AgNO_3 , Guangzhou Jinhua Chemical Reagent Co., Ltd, $\geq 99.8\%$) were used with no further purification. Ultrapure water with a resistance of $18.2 \text{ M}\Omega \cdot \text{cm}$ was obtained from an instrument Unique-R20.

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