



Enhanced photoinduced stability and photocatalytic activity of AgBr photocatalyst by surface modification of Fe(III) cocatalyst



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ABSTRACT

Recently, AgBr material was demonstrated to be a new and efficient visible-light photocatalyst for the decomposition of various organic compounds. Owing to its excellent photosensitive properties, however, AgBr phase is unavoidably decomposed into metallic Ag under visible-light irradiation, resulting in an obvious destroy of its surface structure. In this study, Fe(III) cocatalyst was grafted on the surface of AgBr particles to form Fe(III)/AgBr photocatalysts by an impregnation method and their photocatalytic performance was evaluated by the photocatalytic decolorization of methyl orange solution under visible-light irradiation. It was found that the Fe(III) cluster could act as a new and effective cocatalyst not only to improve the photocatalytic activity of AgBr photocatalyst, but also remarkably enhance the photoinduced stability of photosensitive AgBr. After surface coating by Fe(III) cocatalyst (8.2 at.%), the photocatalytic activity of AgBr photocatalyst can be greatly improved by a factor of 73% even after five cycles of photocatalytic reactions. Simultaneously, the decomposed amount of AgBr can be significantly deduced from 8.8 at.% to 2.9 at.% by the surface loading of Fe(III) cocatalyst. On the basis of the experimental results, a possible mechanism for the enhanced photocatalytic activity and photoinduced stability of AgBr by Fe(III) cocatalyst was proposed. Compared with the well-known noble metal cocatalysts (e.g., Pt, Au, Ag), the present abundant and cheap Fe(III) cocatalyst can be regarded as one of the ideal cocatalyst for the smart design and development of high-performance photocatalytic materials in various potential applications.

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1. Introduction

Developing stable and effective photocatalytic materials for the elimination of organic pollutants has been demonstrated to be one of the important investigation objectives in photocatalysis [1–3]. TiO₂, a traditional photocatalyst, was widely investigated because of its non-toxicity and long-term stability [4–6]. However, the TiO₂ photocatalyst still cannot be widely used in practical applications due to its limited visible-light absorption and low photocatalytic performance. Thus, it is highly required to develop new and highly efficient visible-light-responded photocatalytic materials. Recently, various Ag-based compounds such as AgCl [7–9], AgBr [10–12], AgI [13,14], and Ag₃PO₄ [15,16] were demonstrated to be a new family of highly efficient visible-light photocatalytic materials.

In our previous studies, we have also reported the highly efficient Ag-based photocatalytic materials such as Ag₂O [17], Ag₂CO₃ [18], Ag₃PO₄ [19,20], AgCl [21–23], and AgI [24]. Compared with the typical N-TiO₂ visible-light photocatalyst, it is found that the Ag-based photocatalysts usually show a significantly higher photocatalytic performance for the decomposition of various organic substances in aqueous solution [21]. However, it is very interesting and highly required to further improve their photocatalytic performance from the view point of potential applications. In addition, compared with the well-known photocatalytic materials such as TiO₂, one of the obvious disadvantages is that the photoinduced stability of bare Ag-based photocatalysts is quite poor owing to its strong photosensitivity, resulting in an obvious destroy of its surface structure under light irradiation. Unfortunately, less investigation about the enhanced photoinduced stability of photosensitive Ag-based materials has been concerned.

Silver halides (AgX, X=Cl, Br and I), a family of excellent photosensitive Ag-based material, have been extensively used in photographic films for the formation of latent image via their photoinduced decomposition mechanism [25]. Therefore, the

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photosensitive silver halides are unavoidably decomposed into metallic Ag under visible-light irradiation. By absorbing incident light, the photoinduced electrons and holes are generated in the conduction band (CB) and valence band (VB) of silver halides, respectively. The photogenerated electrons in the CB can be captured by surface lattice Ag^+ ions to form metallic Ag_n clusters, whereas the photogenerated holes oxidize lattice X^- to release X_2 , resulting in the photodecomposition of silver halides and the formation of metallic Ag. As a consequence, it can be expected that if the photogenerated electrons in the CB of AgX can be quickly captured by other electron acceptors before reducing the lattice Ag^+ , it is possible for us to improve the photoinduced stability of photosensitive AgX phase. Considering the fact that the photogenerated electrons in the CB of semiconductors can be rapidly transferred via the surface modification of cocatalysts (such as Au, Pt, Ag, and graphene) [26–29], it is quite possible that the photoinduced stability of photosensitive AgX can be greatly enhanced by loading effective cocatalysts on the surface of AgX.

In our previous studies, the Fe(III) cluster was demonstrated to be one of the new and efficient cocatalysts to greatly improve the photocatalytic performance of TiO_2 [30]. Compared with the expensive noble-metal nanoparticles, the Fe element is nontoxic and abundant in natural resources. In this study, Fe(III) cocatalyst was grafted on the surface of AgBr particles to form Fe(III)/AgBr photocatalysts by an impregnation method and their photocatalytic performance was evaluated by the photocatalytic decolorization of methyl orange solution under visible-light irradiation. The effect of Fe(III) cocatalyst on the photocatalytic activity and photoinduced stability of AgBr particles was investigated. It was found that the Fe(III) cluster could act as a new and effective cocatalyst not only to improve the photocatalytic activity of AgBr photocatalyst, but also remarkably enhance the photoinduced stability of photosensitive AgBr. To the best of our knowledge, this is the first report about the enhanced photocatalytic activity and photoinduced stability of AgBr photocatalyst by Fe(III) cocatalyst. Compared with the well-known noble metal cocatalysts (e.g., Pt, Au, Ag), the present abundant and cheap Fe(III) cocatalyst can be regarded as one of the ideal cocatalysts for the smart design and development of high-performance photocatalytic materials. In addition, considering the effective inhibition effect for the rapid decomposition of photosensitive AgBr material by surface cocatalyst, the work may provide a new insight for the potential applications of various photosensitive Ag-based materials.

2. Experimental

2.1. Synthesis of AgBr photocatalyst

The starting aqueous solutions of AgNO_3 solution (0.1 mol L^{-1}) and NaBr solution (0.1 mol L^{-1}) were first prepared. The synthesis of AgBr powder was prepared by a simple participation reaction between Ag^+ and Br^- ions in the solution. In a typical synthesis, 35 mL AgNO_3 solution was poured into 35 mL of NaBr solution under stirring. After stirring for 5 min, the resultant yellow suspension was maintained at 60°C for 2 h. The resultant powder was recovered by filtration, rinsed with distilled water, and dried at room temperature.

2.2. Modification of AgBr photocatalyst by Fe(III) cocatalyst

The Fe(III)/AgBr photocatalyst was prepared by an impregnation technique. In a typical preparation, 0.5 g of AgBr powder was dispersed into 75 mL $\text{Fe}(\text{NO}_3)_3$ solution under stirring. After stirring for 15 min, the suspension solution was maintained at 60°C for 2 h. The resultant powder was recovered by filtration, rinsed with distilled

water, and dried at 60°C to obtain modified Fe(III)/AgBr photocatalysts. To investigate the effect of $\text{Fe}(\text{NO}_3)_3$ concentration on the microstructures and photocatalytic performance of AgBr photocatalyst, the $\text{Fe}(\text{NO}_3)_3$ concentration was controlled to be 0.001, 0.005, 0.01, and 0.05 mol L^{-1} , and the resultant Fe(III)/AgBr photocatalyst can be referred to as Fe(III)/AgBr (0.001 M), Fe(III)/AgBr (0.005 M), Fe(III)/AgBr (0.01 M), and Fe(III)/AgBr (0.05 M), respectively.

For comparison, the bare AgBr were also treated in distilled water under an identical experimental condition.

2.3. Characterization

X-ray diffraction (XRD) patterns were obtained on a D/MAX-RBX-ray diffractometer (Rigaku, Japan). X-ray photoelectron spectroscopy (XPS) measurements were done on a KRATOA XSAM800 XPS system with Mg $\text{K}\alpha$ source. All the binding energies were referenced to the C 1s peak at 284.8 eV for the surface adventitious carbon. Morphological analysis was performed with an S-4800 field emission scanning electron microscope (FESEM) (Hitachi, Japan) with an acceleration voltage of 10 kV. UV–vis absorption spectra were obtained using a UV–visible spectrophotometer (UV-2550, SHI-MADZU, Japan).

2.4. Photocatalytic activity

MO is a simple azo dye that has been widely used as a model system to probe the photocatalytic performance of various photocatalysts [21]. It is generally considered to be very stable to light and difficult to decompose, though MO can absorb visible light at ca. 464 nm. The photocatalytic activity of the prepared samples was performed at ambient temperature. Experimental details were shown as follows: 0.1 g of the sample was dispersed into 10 mL of MO solution (20 mg/L) in a disk with a diameter of ca. 5 cm. The solution was allowed to reach an adsorption–desorption equilibrium among the photocatalyst, MO, and water before visible-light irradiation. A 350 W xenon lamp equipped with a UV-cutoff filter (providing visible light with $\lambda \geq 400 \text{ nm}$) was used as a visible-light source. The average light intensity striking the surface of the reaction solution was about 40 mW cm^{-2} . The concentration of MO was determined by an UV–visible spectrophotometer (UV-1240, SHI-MADZU, Japan). After visible-light irradiation for some time, the reaction solution was centrifuged to measure the concentration of MO. As for the methyl orange aqueous solution with low concentration, its photocatalytic decolorization is a pseudo-first-order reaction and its kinetics may be expressed as $\ln(c_0/c) = kt$, where k is the apparent rate constant, and c_0 and c are the methyl orange concentrations at initial state and after irradiation for t min, respectively [21,27].

3. Results and discussion

3.1. Morphology and microstructures

Fig. 1A and B shows the FESEM images of the AgBr particles before and after surface modification by Fe(III) cocatalyst. It is clear that the direct participation reaction method results in a wide particle-size distribution (0.3–1.5 μm) of the AgBr particles (Fig. 1A). The corresponding XRD pattern (Fig. 1C) suggests the formation of AgBr phase (JCPDS no. 06-0438). As for the Fe(III)/AgBr photocatalyst, it is clear that there is no obvious change for the morphology of the resultant Fe(III)/AgBr (Fig. 1B) owing to a low-temperature modification process (60°C). Moreover, in view of a low-concentration $\text{Fe}(\text{NO}_3)_3$ solution, no related diffraction peaks of Fe(III) compounds can be detected (Fig. 1C).

As the surface modification process of AgBr by Fe(III) cocatalyst is performed at a low temperature (60°C), it is believed that these

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