



The BiOBr/regenerated cellulose composite film as a green catalyst for light degradation of phenol



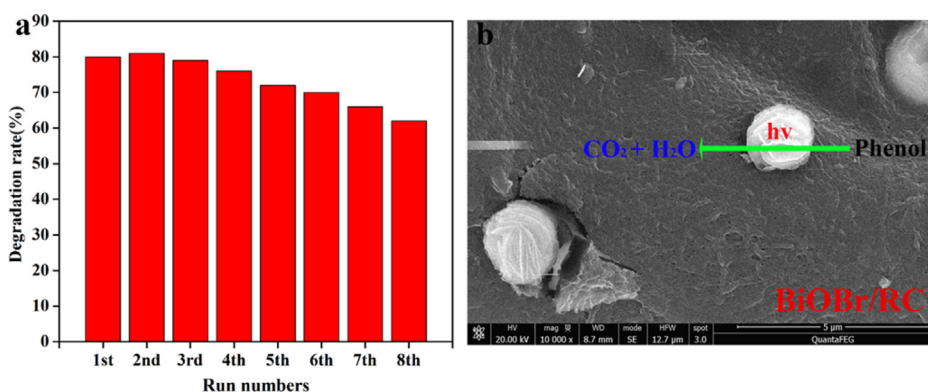
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HIGHLIGHTS

- The BiOBr/RC has been prepared via in-situ synthesis method.
- The influence of the porous cellulose film to the BiOBr photodegradation of phenol was investigated.
- The BiOBr/RC exhibited good photocatalytic activity for the photodegradation of phenol.
- The BiOBr/RC is a portable catalyst.

GRAPHICAL ABSTRACT



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ABSTRACT

A portable catalyst, BiOBr/regenerated cellulose composite films (BiOBr/RC), was prepared for the application of photocatalytic irradiation under visible light. It exhibited good photocatalytic activity for the photodegradation of phenol which means it has potential for the application in photodegradation of organic pollutant. Cellulosic biomass is a kind of green material, and the composite film is easy to recycle and has good reproducibility. So it's a "green" processing for the catalyst can be removed easily from pollution solution after being used and to be used for the next run.

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1. Introduction

Currently, organic-inorganic hybrid material plays an increasingly important role due to its advantages like electrical, magnetic and optical properties as well as the combined properties of organic and inorganic materials [1]. Especially, biodegradable composites

derived from renewable resources, which could be employed as organic moieties, has attracted the attention of researchers from diverse areas [2,3]. The obtained organic-inorganic hybrid biomaterials not only possess the improved structural and functional properties, but also show remarkable advantage of exhibiting biocompatibility, biodegradability and some other novel properties [4].

Photocatalytic degrading organic pollutants via semiconductors is a promising treatment strategy nowadays [5]. The oxide semiconductors, such as TiO₂ [6,7], ZnO [8,9], Fe₂O₃ [10],

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oxyhalides [11] (BiOX, X = F, Cl, Br, I), WO_3 [12] and so forth, have been widely employed as photocatalyst, owing to the distinct size-dependent physicochemical and opto-electronic properties. After the photocatalytic activity under UV- or visible-light illumination had been unveiled, bismuth oxyhalides and their potential application in decomposing organic pollutants have been largely explored [13,14]. Among them, semiconductor BiOBr has attracted enormous attention due to its suitable band gap, excellent photocatalytic activity and stability under UV and visible light irradiation [15–18]. The intrinsic indirect band gap of BiOBr endows this lamellar-structured p-type semiconductor [18] with excellent mobility and a prolonged transfer path for photogenerated electrons [19]. Limited by their repeatability, BiOBr photocatalysts still cannot meet the requirements of practical application. Therefore, it is essential to develop an efficient, green, low-cost and simply prepared catalyst with excellent repeatability for removal of organic pollutant. Organic-inorganic hybrid material with BiOBr particles as inorganic moieties is a good selection to solve the problem.

Cellulose, one of the most abundant naturally-biodegradable polymers, featuring renewability, good biodegradability, and excellent biocompatibility, is usually considered as good candidates to be host material because it can improve the stability, retain the special morphology and control the growth of nanoparticles [20,21]. Based on the above-mentioned properties, cellulose is chosen as organic moieties of organic-inorganic hybrid material to host the BiOBr particles.

Hence, we used a simple method to prepare BiOBr/RC in this paper. A novel solvent of 5.0 wt% LiOH/12.0 wt% urea aqueous solutions made it possible to dissolve cellulose quickly with being precooled to -12°C [22,23]. The regenerated cellulose films (RC) at the wet state possesses surface porous structure. So the porous cellulose fibers were employed as an effective nanoreactor as well as particle stabilizer of metal particles in situ synthesis from metal precursor solution [24,25]. The metal-salt in solution was adsorbed onto the surface of RC and subsequently gave the nanoparticles on the thin films via hydrolysis, forming the BiOBr/RC. In our present work, the BiOBr/RC was employed as portable photocatalyst under visible light irradiation. We hope that this work can provide some meaningful information to develop a simple method to prepare BiOBr/RC in aqueous solvent system. The BiOBr/RC were characterized by different technique (SEM, TGA, FT-IR and UV-vis diffuse spectroscopy) and used for the photocatalytic degradation of phenol under visible light irradiation. Moreover, this is a “green” processing for the photocatalyst can be easily removed from pollutant solution and regenerated by washing with deionized water, which meets the requirements of sustainable development and resource-conserving economy.

2. Experimental

2.1. Materials

The cellulose sample used in this study was α -cellulose (particle size: 25 μm , Chengdu Chemical Fiber Co., Ltd., China). All other chemical reagents were analytical grade and used without further purification. Deionized water was used for all experiments.

2.2. Preparation of cellulose composite film

A 5.0 wt% LiOH/12.0 wt% urea aqueous solution was precooled to -12°C in a refrigerator. The desired amount of cellulose was immediately dispersed into the solvent under vigorous stirring for 5 min at ambient temperature to obtain a transparent cellulose solution with 4.5% concentration. In order to eliminate air bubbles, the resultant cellulose solution was subjected to centrifugation at 8000 rpm

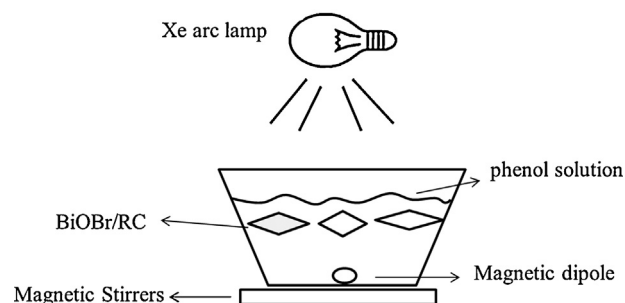


Fig. 1. Schematic diagram of the experimental set-up of photocatalytic degradation of phenol.

for 10 min at 10°C . The viscous bubble-free solution was cast on a glass plate to give about 0.25 mm-thick layer, and then immersed into 9.0 wt% Na_2SO_4 /7.0 wt% H_2SO_4 aqueous solution for 5 min to obtain RC. The resultant RC was washed with excess deionized water to remove the residual chemical reagents, and preserved in deionized water.

2.3. Preparation of BiOBr/cellulose composite films

To prepare BiOBr/RC, 1 g $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ was dissolved into 50 mL 1 M HNO_3 solution under stirring at room temperature until the solution was clarified and transparent solution. The RC was immersed into $\text{Bi}(\text{NO}_3)_3$ solutions for 12 h in a round culture dish, then, were immersed into the deionized water for 2 min, subsequently, were immersed into 1 M NaBr solution for 10 min. Finally, the composite films were washed by deionized water three times, and then some composite films were pasted on culture dish and dried with Vacuum freeze drying for characterization. Other composite films were immersed into the deionized water for photocatalytic degradation test before used. The original cellulose film was coded as RC. The BiOBr/cellulose composite films were coded as BiOBr/RC composite films.

2.4. Preparation of BiOBr particles

To prepare BiOBr, 0.01 mol $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ were dissolved into 100 mL 1 M HNO_3 solution under stirring at room temperature, then the $\text{Bi}(\text{NO}_3)_3$ solution was added to the 0.1 mol/L NaBr solution drop by drop under stirring for 1 h at room temperature. The resulting product was collected by filtration, washed several times with anhydrous ethanol and deionized water, and then dried at 60°C for 12 h.

2.5. Photocatalytic degradation of phenol

The photocatalytic performances of the BiOBr/RC were evaluated by the degradation of phenol under visible light irradiation at room temperature. Three pieces of the wet BiOBr/RC (1 cm \times 2 cm) were immersed into 20 mL 7 mg/L aqueous phenol solution. The light source was a 500 W Xe arc lamp. The distance between the liquid surface of the suspension and the light source was set about 10 cm. Schematic diagram of the experimental set-up of photocatalytic degradation of phenol is shown in Fig. 1. Prior to irradiation, the solution with the BiOBr/RC was stirred in the dark for 1 h to insure that the phenol was adsorbed to saturation on the surface of catalyst. The whole degradation process of magnetic stirring speed was about 800 rpm. Reaction solution was collected for analysis in visible light irradiation every 0.5 h times. The solution of collection was 0.5 mL and removal of impurities with centrifugal. The concentration of phenol was obtained by the 4-aminoantipyrene

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