



Fabrication of stable, efficient and recyclable p-CuO/n-ZnO thin film heterojunction for visible light driven photocatalytic degradation of organic dyes

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ARTICLE INFO

Article history:

Received 14 October 2015

Received in revised form

28 October 2015

Accepted 29 October 2015

Available online 31 October 2015

Keywords:

Semiconductors

Interfaces

Thin films

photocatalytic property

ABSTRACT

We report the photocatalytic activity of p-CuO/n-ZnO thin film heterojunction fabricated by modifying our previously reported simple galvanic technique. The fabricated heterojunction exhibits excellent photocatalytic activity and re-usability towards degradation of three different dyes under visible light source. Various samples have been prepared with different compositions to interpret the degradation mechanism.

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1. Introduction

Dyestuff released from textile industries comprises one of the largest groups of organic compounds that cause colouration of water and cause great loss of aquatic life. Removal of these colours is a priority for ensuring a clean and safe aquatic environment. Visible light is a major component (50%) of sunlight as compared to UV light, which is only about 4% of total solar radiation. Therefore, the development of visible-light driven photocatalysts has recently become a very popular topic of research. Several efforts have been given for developing natural light driven coupled semiconductor system to avoid recombination of photo-generated electrons and holes [1–4]. Most of the visible light driven photocatalysts that have been studied previously were in powder-form. There are only a few reports on thin film based photocatalysts that can utilise natural solar light from a practical standpoint [5,6]. Sungmook Jung and Kijung Yong deposited CuO over ZnO to form a heterostructure on a steel mesh. They studied the degradation of the dye AO7 for nearly 8 h. to get around 90% degradation, under UV irradiation [7]. In our case, we could reach complete degradation of MB within 2hrs 30 mins with galvanically deposited CuO/ZnO thin films, with only visible light irradiation. Usually, powder photocatalysts have disadvantages related to catalyst

recovery when used in liquid–solid heterogeneous catalysis processes. On the other hand, when the catalyst particles are nanoscale, they often face the problem of serious agglomeration due to high surface energy. These issues give rise to difficulties in catalyst recycling, therefore, hindering its applications. The film based photocatalyst can overcome these limitations. In addition, as a film on a substrate, it can be easily separated from the solutions. Furthermore, it can be recycled just by simple treatments, such as washing with deionized water and acetone, respectively, and then drying. In this communication, we report the fabrication of an efficient, stable, non-toxic and low-cost thin film of p-CuO/n-ZnO heterostructure based visible-light driven photocatalyst.

2. Experimental

CuO thin films were synthesized by modifying our previously reported procedure on ITO and ZnO coated ITO substrates [8,9]. A resistance of 500 Ω was introduced in the external circuit between ITO and Zn rod (Fig. S1, ESI). Introduction of external resistance ensures slow and uniform deposition of CuO thin film with good adherence property. To demonstrate the potential application of the prepared heterojunction catalyst in the degradation of organic contaminants, we have studied the photocatalytic degradation of MB, RB and MO dyes with the assistance of H_2O_2 under visible light source and neutral pH condition.

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3. Results and discussions

To demonstrate the potential application of p-CuO and p-CuO/n-ZnO heterojunction with nanoporous structures in the degradation of organic contaminants, we have investigated their photocatalytic activities by choosing the photocatalytic degradation of three model pollutant dyes such as MB, RB and MO in the presence of H_2O_2 . Total surface area of the catalyst films was kept 40 sq cm for each degradation study. To interpret the degradation mechanism, various samples have been prepared with different compositions and designated the heterostructure samples with surface areas 10, 20, 30 and 40 sq cm CuO coverage over 40 sq cm ZnO films as ZC1, ZC2, ZC3 and ZC4, respectively. All samples (40 sq. cm) were immersed in 60 mL of dye solution (1.7×10^{-5} M) and kept for 30 min in dark to equilibrate. The light irradiation was carried out using a 200 W tungsten lamp which was placed vertically over the reaction vessel at a distance of 10 cm which gives a total optical irradiance of 70 mW cm^{-2} at the surface of dye solution. At given time intervals, UV–Vis absorption spectra of dye solution were recorded using a UV–Vis spectrophotometer. Fig. 1A, B and C demonstrate that the catalytic activity of ZC3 is better than that of the other compositions for all the pollutants and the observed rate constant values have been summarized in Table 1. We can find that under visible light irradiation, the pure ZnO films exhibit very weak ability for decolorization of all the dyes. The poor degradation ability of the pure ZnO films can be ascribed to the wide band gap, as the visible light can not excite electrons in the valance band, to jump to the conduction band. The

Table 1

Rate constant values of samples with different compositions.

Pollutant	ZnO	CuO	ZC1	ZC2	ZC3	ZC4
MB	0.0017	0.017	0.0063	0.011	0.028	0.012
RB	0.0016	0.01	0.004	0.0061	0.014	0.0067
MO	0.0005	0.0079	0.0033	0.0047	0.011	0.0056

CuO films exhibit an effective ability for decolorization of all the dyes, but the photocatalytic activity is lower than the CuO/ZnO heterojunction (ZC3) prepared by CuO electrodeposition, with surface area 30 sq cm. That is to say, the hetero-junction ZC3 exhibits a higher photocatalytic activity in the degradation of all the dyes than pure CuO films. Furthermore, the electrodeposited CuO area affects the photocatalytic activity of the CuO/ZnO heterojunction films. When the deposited CuO area is less than 30 sq cm, the photocatalytic activities of the CuO/ZnO heterojunction films decrease with the lowering of CuO electrodeposition area; again when the CuO area is greater than 30 sq cm, the photocatalytic activities of the CuO/ZnO heterojunction films decrease with the increase of CuO electrodeposition area. More the deposited CuO area less will be the available bare surface of ZnO film. This indicates that an appropriate CuO area is playing an important role for the degradation mechanism. The highest photocatalytic activity in the case of ZC3 is probably attributed to optimum surface area ratio of CuO: bare ZnO (3:1). Greater surface area of CuO improves optical absorption resulting in more number of electron and hole pairs and the p–n heterojunction favours the separation

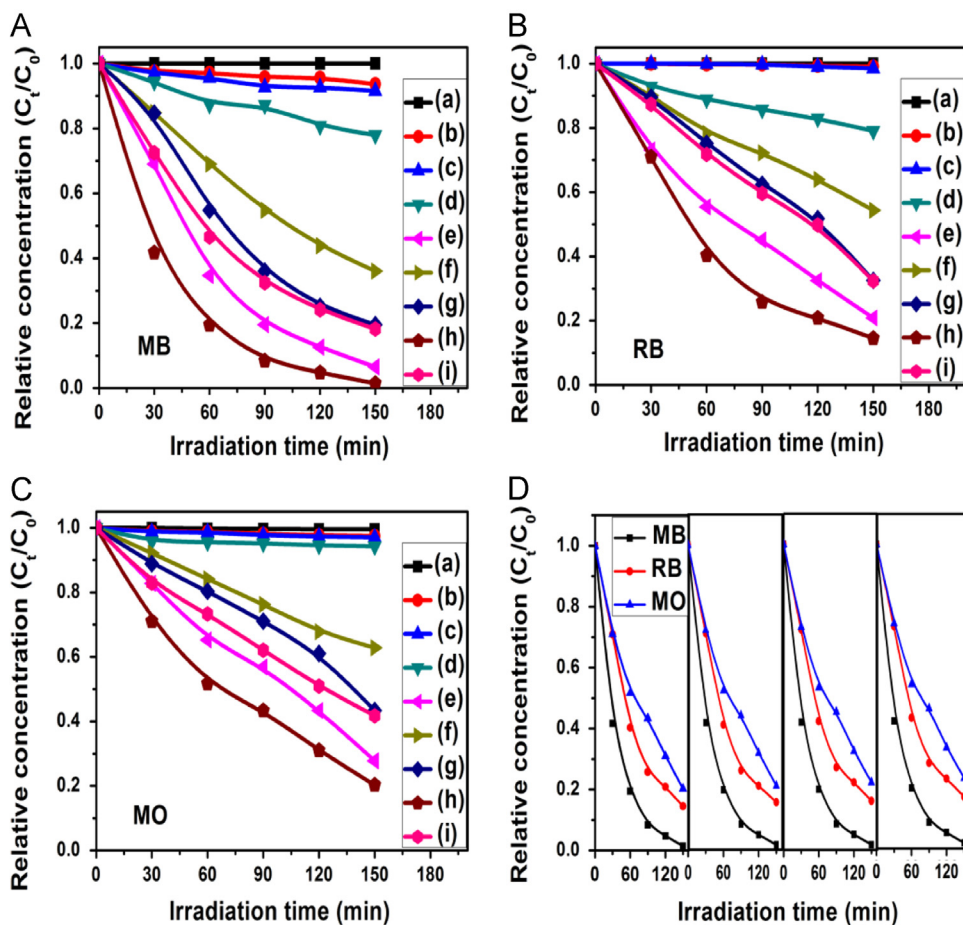


Fig. 1. Reaction profile of photocatalytic degradation of (A) MB (B) RB (C) MO dyes in (a) dark, (b) light, (c) light with H_2O_2 and in presence of H_2O_2 with catalyst films (d) ZnO (e) CuO (f) ZC1 (g) ZC2 (h) ZC3 (i) ZC4 under visible light irradiation. (D) Relative dye concentration versus light exposure time for four consecutive cycles of operation for ZC3.

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