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Visible light responsive Ag/TiO_2/MCM-41 nanocomposite films synthesized by a microwave assisted sol–gel technique

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ABSTRACT

A convenient and inexpensive method for the preparation of visible light responsive nanocomposite film was introduced in this study. Silver doped TiO_2 was incorporated into as-synthesized MCM-41, via a microwave assisted sol–gel technique. The nanocomposite film was formed by dip coating on a glass substrate. The characterization results displayed high adsorbability and photocatalytic properties of the Ag and MCM-41 enhanced TiO_2 photocatalyst. Performance of the nanocomposite film was tested by photocatalytic decolorization of MB dye, under UV and visible light irradiation. Ag/Ti/Si (0.1/1/2) exhibited the highest photocatalytic decolorization of methylene blue, with an efficiency of 81% under UV, and 30% under visible light irradiation. The kinetic rate constant of MB dye on the composite films followed pseudo first-order reaction law ($R^2 > 0.9$), arranged in the order of Ag/Ti/Si (0.1/1/2) > Ag/Ti/Si (0.1/1/1) > Ag/Ti/Si (0.1/1/0.5) > Ag/Ti/Si (0.1/1/0) > TiO_2.

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1. Introduction

Photocatalytic oxidation is one of the most promising pollution treatments, and is extensively applied to water and air purification [1–3]. Titanium dioxide (TiO_2) is known as a versatile photocatalyst, due to its low cost, non-toxicity and high photo-chemical stability [4]. Photocatalytic reaction occurs when photon energy ($h\nu$) irradiates onto the TiO_2 surface, greater than its band gap energy permits. The electrons can then migrate from the valence band (VB) to the conduction band (CB), creating an e^-–h^+ charged carrier. After jumping, the excited electrons then can move freely, and return to a ground state. Furthermore, these electrons can react with a molecule acceptor, such as O_2, to form an oxidizing agent or superoxide radical ($O_2^{-\cdot}$). In addition, a charge recombination does not occur whilst an adsorbed hydroxyl radical ($OH^{\cdot}$) at hole is created [5]. The superoxide radicals can breakdown the molecules of organic compounds, and thus destroy pathogens.

Generally, the photocatalytic process of TiO_2 requires that energy of light occurs under UV light ($\lambda < 387$ nm), leading to limitations of TiO_2 when applied under visible light ($\lambda > 400$ nm). Doping of TiO_2 with noble metals, such as Pt [6], Au [7] and Cu [8], has been researched to enhance photocatalytic activity under visible light. Among these metals, Silver (Ag) is an attractive doping agent which enhances the photocatalytic reactivity of TiO_2 under visible light, because silver particles can reduce the bang gap energy of TiO_2, and prevent e^-–h^+ recombination [9–11]. Yu et al. reported that Ag doped TiO_2 exhibited high adsorption rates in visible light. This is a result of the surface plasmon resonance photolysis of Ag nanoparticles, in which the electrons migrate to the conduction band of TiO_2 and are further trapped by O_2, producing oxidative species, resulting in the oxidation of organic pollutants [12]. It is reported that Ag doped TiO_2 coated onto a plastic substrate was applied to the treatment of BTEX gases, under visible light [13]. It was found that the maximum degradation efficiency of benzene was 79%, when doping 10 mol% Ag onto TiO_2 thin film. Even though Ag can improve the photocatalytic activity of TiO_2, loading of Ag onto TiO_2 causes a decrease in surface area, which may result in poor photocatalytic performance. Recently, TiO_2 thin film doped with a mesoporous silica (MCM-41) has been studied, in order to improve the adsorbability of TiO_2 thin film [14]. MCM-41 has a highly ordered hexagonal structure, with large surface area (>800 $m^2\,g^{-1}$), and this can enhance the specific surface area of TiO_2 thin film. Moreover, the functional groups of silanol (Si–OH) found in the MCM-41 structure can generate active hydroxyl radicals ($^{\cdot}OH$), which behave as an oxidizing agent and destroy organic pollutants during photocatalytic reactions [15–17]. However, a study of incorporating Ag and MCM-41 onto TiO_2 nanocomposite film, and an analysis of the synergistic effects of both Ag and MCM-41 upon the photocatalytic reaction of TiO_2, has never been reported.

There are several methods employed in the preparation of TiO_2, such as hydrothermal, ultrasound and sol–gel processes [18,19]. However, these techniques require heating to high temperatures, and so the processes are time consuming. To overcome these drawbacks, a new preparation of TiO_2, using a microwave assisted sol–gel technique, has been developed [20]. This method claims that microwaves can offer a faster and more convenient was to prepare nanocrystalline TiO_2, rather than use the traditional hydrothermal method, which requires longer synthesis time [21]. Microwave irradiation can promote nucleation of titania crystals, by providing homogeneous heating in a short time, and thus reduce energy costs [22,23]. Within the concept of green chemistry, a microwave assisted sol–gel has proven to be a promising method for TiO_2 synthesis.

This research aims to apply a microwave assisted sol–gel method to prepare visible light responsive nanocomposite films, by doping Ag and as-synthesized MCM-41 into TiO_2 thin films. The comparison of hydrothermal and microwave assisted sol–gel methods, on the characteristic of the prepared films, were studied. In addition, the influence of MCM-41 on the photocatalytic reactivity of the composite film, under both UV and visible light irradiation was investigated, by varying Si loading. The photocatalytic reactivity of the composite films was evaluated, using photocatalytic decolorization of methylene blue dye, under both UV and visible light. Finally, a kinetic reaction model was determined, and the reaction rate constants were then proposed.

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