



Preparation and photocatalytic activity of mesoporous-assembled nano-titania thin films derived from a laurylamine hydrochloride surfactant-aided sol–gel method



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ABSTRACT

The utilization of mesoporous-assembled nano-titania (TiO₂) thin films immobilized on glass plates for the photocatalytic degradation of Acid Black (AB) azo dye under UV light irradiation is investigated. The mesoporous-assembled TiO₂ nanoparticle photocatalyst is synthesized by a sol–gel process with the aid of laurylamine hydrochloride (LAHC) surfactant as an effective mesopore-forming agent. The effects of calcination temperature and number of TiO₂ coatings on the photocatalytic AB dye degradation performance are investigated. The experimental results reveal that an increase in calcination temperature in the range of 400–500 °C can improve the photocatalytic activity of the mesoporous-assembled TiO₂ films, but thick multilayer TiO₂ films cannot be prepared at a higher calcination temperature. In addition, an increase in number of TiO₂ coatings significantly enhances the photocatalytic activity to reach maximum and optimum three coatings.

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1. Introduction

Heterogeneous photocatalysis using a semiconductor photocatalyst has become an emerging technology for treatment of hardly degradable organic pollutant-containing wastewaters [1–3]. Titania (TiO₂)-based photocatalysis is one of the most promising methods for efficient degradation of hardly degradable organic pollutants [4–11]. In some recent works, dispersion/suspension of TiO₂ photocatalyst powders in simulated/real wastewater during photocatalytic treatment shows great photocatalytic activity, but requires

a troublesome filtering step to separate the photocatalyst from the treated wastewater for recycling purposes. Therefore, utilization of a TiO₂ photocatalyst in the immobilized mode is more practical because it eliminates a TiO₂ separation step [12–17].

The photocatalytic degradation of organic pollutants using immobilized mesoporous TiO₂ photocatalysts has been widely investigated. The most extensively used approach to prepare mesoporous TiO₂ photocatalysts for immobilizing on substrates is based on sol–gel methods comprised of the coating (e.g. dip coating, spray coating, and spin coating) of either TiO₂ sol or gel on substrates followed by heat treatment to form TiO₂ thin films [18–39]. To achieve mesoporous characteristics of the coated TiO₂ thin films, various kinds of high-molecular-weight surfactants have been commonly used as mesopore-directing

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molecules during the sol and/or gel preparation—Pluronic P-123 poly(ethylene oxide)–poly(propylene oxide)–poly(ethylene oxide) triblock copolymer [18–22,28–30,32,33,35–37], Pluronic F-127 poly(ethylene oxide)–poly(propylene oxide)–poly(ethylene oxide) triblock copolymer [23,24,27,31,38,39], Brij-58 polyoxyethylene cetyl ether [27,38], and Tween 80 polyoxyethylene sorbitan monooleate [25,26,34]. There is difficulty in removing these bulky surfactants during the calcination step. To our knowledge, the less-complex and low-molecular-weight laurylamine hydrochloride (LAHC) surfactant has been used successfully for the preparation of nanocrystalline mesoporous-assembled TiO₂ powders in previous works [40–42], but never been applied to the sol–gel-derived mesoporous-assembled TiO₂ thin films for photocatalytic degradation of organic pollutants in wastewater.

In this work, a mesoporous-assembled TiO₂ photocatalyst was prepared by a sol–gel method with the aid of a mesopore-directing LAHC surfactant and immobilized on glass plates by the doctor-blading method, followed by heat treatment under various calcination temperatures to form mesoporous-assembled TiO₂ thin films. The photocatalytic activity of the prepared mesoporous-assembled TiO₂ thin films was evaluated via the degradation of Acid Black (AB) azo dye. The important preparation parameters during the immobilization step, i.e. calcination temperature and number of TiO₂ coatings, were mainly investigated for photocatalytic AB dye degradation under ultraviolet (UV) light irradiation. AB dye was selected as a model contaminant in wastewater in this work because AB is an intensely colored azo compound widely used in dyeing and textile printing, and it is a chemically stable, non-biodegradable, and difficultly decomposed dye pollutant in wastewater. In addition, AB dye can be easily analyzed by UV–visible spectroscopy to monitor its degradation.

2. Experimental

2.1. Materials

Tetraisopropyl orthotitanate (TIPT; Ti(OCH(CH₃)₂)₄, Merck), acetylacetone (ACA; CH₃COCH₂COCH₃, Carlo Erba), laurylamine hydrochloride (LAHC; CH₃(CH₂)₁₁NH₂·HCl, Merck), Acid Black azo dye (AB; C₂₂H₁₄N₆Na₂O₉S₂, Nacalai Tesque), sodium hydroxide (NaOH; Lab Scan Asia), Triton X-100 (C₁₄H₂₂O(C₂H₄O)_n, n=9–10; Merck), ethanol (C₂H₅OH; Ajax Finechem), and propanol (CH₃CH₂CH₂OH; Ajax Finechem) were used in this work. All chemicals were of analytical reagent grade and used without further purification.

2.2. Synthesis procedure of mesoporous-assembled TiO₂ gel

The mesoporous-assembled TiO₂ gel was synthesized by a sol–gel method with the aid of the mesopore-directing LAHC surfactant [40–42]. During synthesis, a specific amount of ACA was first introduced into the TIPT with a molar ratio of 1:1. The mixed solution was gently shaken to be homogeneous. Next, a 0.1 M LAHC aqueous solution was added to the ACA-modified TIPT solution, in

which the molar ratio of TIPT to LAHC was adjusted to 4:1. The mixture was continuously stirring at 40 °C for 8 h to obtain a transparent yellow sol. Then, the sol was placed into an oven at 80 °C for a week to obtain a complete gel formation. The as-synthesized TiO₂ gel was washed by mixing with propanol and continuously stirred at room temperature for 2 h. Finally, the mixture was centrifuged to separate the wet TiO₂ gel for the next step of immobilization.

2.3. Immobilization of mesoporous-assembled TiO₂ thin films on glass plates

To prepare a TiO₂ paste, a specific amount of the obtained wet TiO₂ gel was mixed with ethanol and Triton X-100. The mixture was homogenized by continuously stirring at room temperature for 1 d. The surfaces of the glass plates were cleaned by soaking in a NaOH aqueous solution for 1 d to remove any impurities adsorbed on their surface. After that, the glass plates were sonicated in deionized water several times to remove residual NaOH and finally dried in an oven at 80 °C prior to use. The as-prepared TiO₂ paste (2 g for a TiO₂ layer) was coated on a pretreated glass plate by the doctor-blading method, and the glass plate with TiO₂ film was placed in an oven at 80 °C for drying. The resulting TiO₂ film-coated glass plates were finally calcined at various temperatures for 2 h in order to remove the remaining LAHC surfactant. Consequently, mesoporous-assembled TiO₂ thin films immobilized on glass plates were obtained.

2.4. Characterization techniques

The thermal decomposition behavior of the TiO₂ gel and its optimal calcination temperature were obtained by using a thermogravimetric/derivative thermogravimetric analyzer (TG-DTA, PerkinElmer, Pyris Diamond) operated at a heating rate of 10 °C/min in a static air atmosphere with α-Al₂O₃ as the reference. The BET specific surface areas, mean mesopore diameters, and total pore volumes of the TiO₂ photocatalysts were measured with a N₂ adsorption-desorption analyzer (Quantachrome, Autosorb I). A TiO₂ sample was outgassed to remove volatile species adsorbed on its surface under vacuum at 150 °C for 4 h prior to analysis. A UV–visible spectrophotometer (Shimadzu, UV-2550) was used to identify the light absorption ability of the prepared TiO₂ photocatalysts immobilized on the glass plates, with BaSO₄ as the reference. The UV–visible spectra were recorded in the wavelength range of 250–500 nm. X-ray diffraction (XRD) patterns of the TiO₂ film-coated glass plates were recorded by using a Bruker AXS system (D8 Advance) with a copper tube for generating CuKα radiation (1.54056 Å) at 40 kV and 30 mA with a nickel filter. The XRD patterns were obtained in the 2θ range of 10–80°. The morphology of the TiO₂ photocatalysts immobilized on glass plates was observed by a scanning electron microscope (SEM; Hitachi, S-4800) and a transmission electron microscope (TEM; JEOL, JEM 2100). The SEM was also used to determine the thickness of the TiO₂ films. The SEM and TEM analyses were performed at accelerating voltages of 2 kV and 200 kV, respectively. An atomic force microscope (AFM);

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