



Synthesis and characterization of Fullerene modified ZnAlTi-LDO in photo-degradation of Bisphenol A under simulated visible light irradiation[☆]



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ABSTRACT

In this study, ZnAlTi layered double hydroxide (ZnAlTi-LDH) combined with fullerene (C₆₀) was fabricated by the urea method, and calcined under vacuum atmosphere to obtain nanocomposites of C₆₀-modified ZnAlTi layered double oxide (ZnAlTi-LDO). The morphology, structure and composition of the nanocomposites were analyzed by Scanning Electron Microscopy, High-resolution transmission electron microscopy, X-ray diffraction patterns, Fourier transform infrared and specific surface area. The UV-vis diffuse reflectance spectra indicated that the incorporation of C₆₀ expanded the absorption of ZnAlTi-LDO to visible-light region. The photo-degradation experiment was conducted by using a series of C₆₀ modified ZnAlTi-LDO with different C₆₀ weight percentage to degrade Bisphenol A (BPA) under simulated visible light irradiation. In this experiment, the degradation rate of C₆₀ modified ZnAlTi-LDO in photo-degradation of BPA under simulated visible light irradiation was over 80%. The intermediates formed in the degradation of BPA process by using LDO/C₆₀-5% were 4-hydroxyphenyl-2-propanol, 4-isopropenylphenol and Phenol. Photogenerated holes, superoxide radical species, ·OH and singlet oxygen were considered to be responsible for the photodegradation process, among which superoxide radical species and ·OH played a predominant role in the photocatalytic reaction system. C₆₀ modified ZnAlTi-LDO catalysts for photocatalytic reduction shows great potential in degradation of organic pollutants and environmental remediation.

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1. Introduction

Bisphenol A (BPA, 2,2-bis(4-hydroxyphenyl)propane) is used extensively as the monomer to synthesize polycarbonate plastic utensils and epoxy resins for chemical applications (Wetherill et al., 2007; Hengstler et al., 2011). Due to its massive production and wide application, a large amount of BPA has been released into the

aquatic environment (Erler and Novak, 2010). BPA have many adverse effects on human beings and wildlife (Takeuchi et al., 2004; Zoeller et al., 2005; Keri et al., 2007). Thus, extreme attention has been paid to the treatment of BPA containing wastewater, and a number of techniques have been developed to remove BPA from water, such as physical absorption (Pan et al., 2008), chemical remediation (Cui et al., 2009; Rodriguez et al., 2010), and microbial degradation (Xie et al., 2011). Among these methods, photocatalytic degradation is attracting widespread attention due to its effectiveness and low toxicity (Seftel et al., 2008a,b; Mantilla et al., 2010). TiO₂, ZnO, and SnO₂ are well known semiconductors which can degrade organic pollutants in aqueous solutions under visible light or UV irradiation (Wang et al., 2002; Evgenidou et al.,

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2007; Han et al., 2009). Due to their high specific surface area, ideal anion exchange capacities, expansion properties, low cost and environmental friendliness, an increasing interest around layered double hydroxides (LDH) has been developed in numerous fields, such as catalysis (Zhang et al., 2008), photocatalysis (Seftel et al., 2008a,b), catalyst supports (He and Zhang, 2007), adsorbents (Pavan et al., 1999), anion exchange (Thomas and Rajamathi, 2011), electrochemical reactions (Yao et al., 1998), medicine (Choy et al., 2004). ZnAlTi-LDH (Wang et al., 2014a) and NiZnAl-LDH (Wang et al., 2014b) have been recently reported in photocatalytic applications, but degradation rate of BPA by LDH is not satisfying under visible light irradiation.

C₆₀ is well-known for its unique electronic properties and high electron affinity. Recently, extensive researches about the preparation and application of C₆₀ derivatives have been conducted in the new material science field (Li et al., 2007; Stevenson et al., 2000; Oh et al., 2007, 2008a,b). Addition of C₆₀ in semiconducting materials can promote the degradation and complete mineralization of various pollutants (Akiyama et al., 2000; Hasobe et al., 2006). One of the most remarkable properties in electron-transfer processes for C₆₀ is that it can arouse a rapid photo-induced charge separation and lead to a relatively low charge recombination (Yu et al., 1995). Generally, C₆₀ can act as an excellent electron acceptor because of its unique structure, which efficiently causes a rapid photo-induced charge separation. However, studies on the photocatalytic activity of C₆₀/ZnAlTi-LDO composites have not been reported.

In this work, C₆₀ modified ZnAlTi-LDO was prepared by the urea method. Photo-degradation experiments were conducted using a series of C₆₀ modified ZnAlTi-LDO to degrade BPA under visible light irradiation. The photocatalytic activity of C₆₀ modified ZnAlTi-LDO was enhanced compared with nanosized ZnAlTi-LDO. It is postulated that C₆₀ enhanced the photoactivity of ZnAlTi-LDO catalyst by its high migration efficiency of photo-induced electron. Photogenerated holes, superoxide radical species and singlet oxygen were considered to be responsible for the photodegradation by the C₆₀ modified ZnAlTi-LDO.

2. Material and methods

2.1. Materials

Bisphenol A (BPA, (CH₃)₂C(C₆H₄OH)₂) was bought from the Guangzhou Chemical Reagent Factory (Guangzhou, PRC). C₆₀ was purchased from Nanjing in XFNAN. Analytically pure ZnCl₂, AlCl₃·6H₂O, TiCl₄, HCl, Sodium oxalate (Na₂C₂O₄), C₈H₆O₄ (TAOH), NaOH, NaF, NaN₃, Benzoquinone (BQ, C₆H₄O₂), Isopropanol (ISO, (CH₃)₂CHOH) and urea were used without further purification and purchased from the Guangzhou Chemical Reagent Factory (Guangzhou, PRC). The deionized water was used in all the preparation processes.

2.2. Sample preparation by urea method

The ZnAlTi-LDH was synthesized by homogeneous precipitation method using urea hydrolysis. The synthetic procedure was as follows: 6×10^{-2} mol of ZnCl₂, 5.3×10^{-3} mol of AlCl₃·6H₂O, 1.20 mL of TiCl₄ solution (the solution was prepared by TiCl₄ and HCl with a volume ratio of 1:1, TiCl₄ was 1.06×10^{-2} mol), C₆₀ 3.4×10^{-4} mol (the mass ratio between the C₆₀ and ZnAlTi-LDH was 5:100) and 0.58 mol of urea were dissolved in 250 mL deionized water under vigorous stirring (Jing et al., 2010). The resulting dispersion was stirred for 48 h at 383.15 K refluxing condition. The precipitate was centrifuged and washed thoroughly with deionized water several times until the precipitate was free from chloride (AgNO₃ test) and then dried in a vacuum oven at 333.15 K

overnight. The dried products of ZnAlTi-LDH was noted as LDH. The mass ratio of C₆₀ in LDH was 0.1–5%. LDH/C₆₀-2% and the ZnAlTi-LDH were synthesized by the same method for comparison. These samples were denoted as LDH/C₆₀-r, where r represented the mass ratio of C₆₀ and ZnAlTi-LDH. The dried products were calcined for 5 h at 773.15 K under vacuum atmosphere. The dried products of ZnAlTi-LDH were vacuum calcined on the same conditions, denoted as LDO. These samples were denoted as LDO/C₆₀-r, where LDO— calcined sample and r — mass ratio of C₆₀ and ZnAlTi-LDH.

2.3. Characterization

The external surface microstructure of the samples was observed by a ZEISS Merlin Scanning electron microscopy (SEM, Carl Zeiss, Germany). The chemical composition of materials was characterized by EDS analysis (Bruker, Quantax EDS detector). High-resolution transmission electron microscopy (HRTEM) images were obtained by JEM-2100F field emission transmission electron microscope with an accelerating voltage of 200 kV. The X-ray diffraction (XRD) patterns were measured by an X-ray diffractometer (D/MAX-III A, Rigaku Ltd., Japan) equipped with Cu K α radiation. The measurement of particle specific surface areas and pore sizes was carried out by N₂ adsorption-desorption isotherm analysis with a Chemisorption Specific Surface Area (SSA) Analyzer (ASAP, 2010; Micromeritics, America). The Fourier transform infrared (FT-IR) spectra of the samples were collected by a PerkinElmer 1725X FT-IR spectrometer using KBr pellets, in the range of 4000–400 cm⁻¹ with 4 cm⁻¹ resolution. Inductively Coupled Plasma (ICP) analysis provides fast identification and detection of trace metals content in chemical samples. Solid state UV-vis diffuse reflectance spectra (UV-vis DRS) was recorded on Shimadzu UV-2501PC spectrometer within 200–800 nm wavelength range at room temperature. The X-ray photoelectron-spectra (XPS) analyses were obtained with an X-ray photoelectron spectrometer (AES430S, ANELVA, Japan). The decrease in total organic carbon (TOC) was evaluated by Apollo 9000 TOC analyzer. Photoluminescence data (PL) was acquired using an F-7000 fluorescence spectrophotometer (Hitachi, Japan). The Gas Chromatography-Micro Mass (GC-MS) was measured by QP2010Ultra GC-MS analyzer (Shimadzu, Japan). Transient photocurrent response was recorded in 0.1 M Na₂SO₄ electrolyte solution by using an electrochemical workstation (CHI660E, Shanghai Chenhua Instrument Co., China) with a conversional three-electrode system at room temperature. Ag/AgCl and a platinum foil served as reference and counter electrode, respectively. The working electrode was a glassy carbon disk, modified with catalysts to form the catalysts layer. The working electrode was made as follow: 2 mg catalyst was ultrasonically dispersed in 2 mL ethanol to form suspension and then 10 mL Nafion emulsion was added into the suspension. 10 mL of this suspension was deposited onto the surface of glassy carbon disk and dried at room temperature for electrochemical test. As for transient photocurrent response test, the light source was a 300 W Xe arc lamp and the transient photocurrent of the photocatalysts responded to the light on or off was recorded at 0.0 V to stimulate real photocatalysis reaction condition.

2.4. Photocatalytic experiment

The photocatalytic activity of the samples for the degradation of BPA was measured by exposing them to simulated visible light. Simulated visible light irradiation was provided by a 300 W xenon lamp. Appropriate amounts of catalyst (0.1 g) were added to 200 mL BPA solution (10 mg/L) in the reaction kettle and vigorously stirred in the dark for 60 min to establish the adsorption equilibrium. The suspension was then irradiated with simulated visible light for 4 h.

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