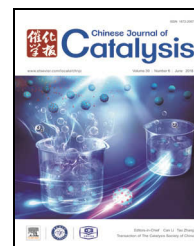


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Article

Preparation of a fullerene[60]-iron oxide complex for the photo-fenton degradation of organic contaminants under visible-light irradiation



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ABSTRACT

Iron oxide (Fe₂O₃) was doped onto fullerene[60] (C₆₀) to form a C₆₀-Fe₂O₃ composite using an easy and scalable impregnation method. The as-prepared C₆₀-Fe₂O₃ samples were characterized by powder X-ray diffraction, X-ray photoelectron spectroscopy, scanning electron microscopy, high-resolution transmission electron microscopy, UV-vis absorption spectroscopy, Raman spectroscopy, and Fourier transform infrared spectroscopy. The photocatalytic activity of the C₆₀-Fe₂O₃ catalyst was evaluated by examining the degradation of methylene blue (MB), rhodamine B (RhB), methyl orange (MO), and phenol under visible light ($\lambda > 420$ nm) in the presence of hydrogen peroxide. The results showed that the catalyst exhibited excellent catalytic properties over a wide pH range 3.06–10.34. Under optimal conditions, 98.9% discoloration and 71% mineralization of MB were achieved in 80 min. Leaching test results indicated that the leaching of iron from the catalyst was negligible and that the catalyst had a high photocatalytic activity after five reaction cycles. The catalyst was also efficient in the degradation of RhB, MO, and phenol. These findings could be attributed to the synergetic effects of C₆₀ and Fe₂O₃. We used active species trapping experiments to determine the main active oxidant in the photocatalytic reaction process and found that hydroxyl radicals played a major role in the entire process.

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1. Introduction

With rapid industrial development over the past few decades, large volumes of colored dye effluents have created severe environmental pollution problems by releasing toxic and potentially carcinogenic substances into the aqueous phase [1,2]. Waste accumulation has a serious adverse impact on the

environment and is dangerous to human health [3]. Advanced oxidation processes (AOPs) have emerged as promising alternative processes for wastewater treatment, especially for persistent and nonbiodegradable contaminants, such as dyeing wastewater [4–6]. As one of the most important AOPs, photo-Fenton degradation of organic dyes using hydrogen peroxide (H₂O₂)/visible light has been widely studied [7–9]. In this

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process, the photodegradation efficiency depends on the decomposition rate of H_2O_2 . Therefore, developing a photocatalyst to enhance the visible-light-driven activity is important.

Fullerene[60] (C_{60}), which has a unique three-dimensional structure because of its delocalized π electrons, has gained considerable attention; the delocalized conjugated structures of C_{60} molecules allow weak absorption in the visible range and high electron mobility ($>1.3 \text{ cm}^2/\text{V/s}$) [10,11]. Hence, C_{60} can be photochemically activated under visible-light irradiation, resulting in rapid photoinduced charge separation with a high quantum efficiency and relatively slow charge recombination [12–14]. Some recent studies have focused on investigating the utilization of visible light for the treatment of pollutants by coupling C_{60} with other semiconductors with low band gaps [15–18].

Iron oxide (Fe_2O_3) is an abundant, stable, cost-effective, nontoxic, and environmentally benign n-type semiconductor with a band gap of 2.2 eV [19]. Owing to its unique and fascinating physicochemical properties, several researchers have investigated the behavior of Fe_2O_3 in diverse fields such as water splitting [20], catalysis [21], and medicine [22]. Recently, Fe_2O_3 has been used to form hybrid materials with carbon nanomaterials, such as multiwall carbon nanotubes, graphene oxide, and graphene, for the degradation of organic pollutants under visible-light irradiation [23–25]. However, the application of a heterogeneous $\text{C}_{60}\text{-Fe}_2\text{O}_3$ catalyst for the photo-Fenton degradation of organic contaminants has not been reported to date.

In this study, $\text{C}_{60}\text{-Fe}_2\text{O}_3$ composites were prepared for the first time via an easy and scalable impregnation method at 60 °C. The photocatalytic activity of $\text{C}_{60}\text{-Fe}_2\text{O}_3$ was evaluated by examining the degradation of methylene blue (MB), rhodamine B (RhB), and methyl orange (MO) in the presence of H_2O_2 under visible light. The effects of operating parameters, such as initial pH, catalyst dosage, and H_2O_2 dosage, were investigated. A possible mechanism for photo-Fenton degradation was also discussed.

2. Experimental

2.1. Materials

Crystalline fullerene powder (99.9% purity) was purchased from Suzhou Dade Carbon Nanotechnology Co., Ltd. *m*-Chloroperoxybenzoic acid (MCPBA), H_2O_2 (30%, W/W), ferric chloride hexahydrate ($\text{FeCl}_3\cdot 6\text{H}_2\text{O}$), sulfuric acid (98%), sodium hydroxide, and benzene (99.5%) were purchased from Tianjin Damao Chemical Factory. MB, RhB, and MO were used as model dye pollutants without further purification. All the experiments were conducted using deionized (DI) water.

2.2. Oxidation of fullerene

For the oxidization of C_{60} , MCPBA was used as the oxidizing agent. MCPBA (ca. 1 g) was suspended in 50 mL of benzene, followed by the addition of fullerene (ca. 80 mg). The mixture was heated under reflux in air and stirred for 6 h in a 250 mL

distillation flask. The solvent was then removed at the boiling point of benzene (80 °C). After completion, the dark-brown precipitate was washed with ethyl alcohol and dried at 50 °C to obtain oxidized fullerene.

2.3. Preparation of $\text{C}_{60}\text{-Fe}_2\text{O}_3$

Oxidized C_{60} (ca. 1 g) was added to 100 mL of water and ultrasonically exfoliated in a bath sonicator for 15 min. Then, 20 mL of 0.2 mol/L FeCl_3 solution was added slowly into the aqueous solution of functionalized C_{60} and stirred for 2 h at 60 °C in a 250 mL distillation flask. After the obtained suspension was aged for at least 8 h at 60 °C, the obtained solids were collected and washed five times with DI water. After drying under vacuum at 60 °C for 8 h, a chemically bonded $\text{C}_{60}\text{-Fe}_2\text{O}_3$ composite was finally obtained.

2.4. Characterization

X-ray diffraction (XRD) analysis on a D8 Advance diffractometer (Bruker, Germany) was performed to identify the structure and crystallinity of the as-prepared samples with monochromatic high-intensity $\text{Cu } K_\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$). High-resolution transmission electron microscopy (HR-TEM) (Tecnai G220, USA) and scanning electron microscopy (SEM, JEOL, Japan) were conducted to analyze the morphology and microstructure of the prepared composites at an acceleration voltage of 200 kV. Fourier transform infrared (FT-IR) spectroscopy (Nicolet 6700; Thermo Ltd., Japan) was carried out to detect the chemical bonds on the surface of the catalyst. The mulls of $\text{C}_{60}\text{-Fe}_2\text{O}_3$ were supported by a KBr plate. UV-vis diffuse reflectance spectra were obtained using a UV-vis spectrophotometer (UV-3600, Shimadzu Ltd., Japan). Photoluminescence measurements were conducted on a fluorescence spectrophotometer (RF-5300PC, Shimadzu, Japan) at room temperature. X-ray photoelectron spectroscopy (XPS; XSAM 800) was employed to characterize the iron states. The binding energies were calibrated with respect to the C 1s peak at 284.6 eV. Peak deconvolution was performed using Gaussian-Lorentzian components after linear background subtraction.

2.5. Photo-Fenton degradation tests and analytical methods

The photocatalytic activity of the $\text{C}_{60}\text{-Fe}_2\text{O}_3$ catalyst was evaluated by examining the degradation of MB, MO, and RhB under visible-light irradiation ($\lambda > 400 \text{ nm}$). A 300 W UV-vis lamp (OSRAM, Germany) was used as a light source. The photo-Fenton degradation of organic compounds was performed in a 100 mL beaker at room temperature ($25 \pm 2 \text{ °C}$). The reaction beaker was located axially and held in a visible lamp box. The lamp was located at a distance of approximately 100 mm from the aqueous solution in a dark box. The wall of the beaker was shielded from surrounding light with tinfoil. Visible light was obtained with a $\lambda > 400 \text{ nm}$ cut-off filter, which covered the window of the beaker to absorb UV light and allow visible light of $\lambda > 400 \text{ nm}$ to pass through. Around 50 mL of test solution was typically used in the photo-Fenton experiments. The initial

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