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Sonocatalytic degradation of Rhodamine B catalyzed by β-Bi_2O_3 particles under ultrasonic irradiation

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ABSTRACT

β-Bi_2O_3 particles were synthesized by a polyacrylamide sol–gel method. The sonocatalytic activity of β-Bi_2O_3 particles was evaluated by degrading Rhodamine B (RhB) under the ultrasonic irradiation, revealing that β-Bi_2O_3 particles exhibit a good sonocatalytic activity. The effects of various experimental factors including ultrasonic frequency (f), solution temperature (T), catalyst dosage ($C_{catalyst}$) and initial RhB concentration (C_{RhB}) on the sonocatalysis efficiency were investigated. The optimum conditions for sonocatalytic degradation of RhB are obtained to be f = 60 kHz, T = 40 °C, $C_{catalyst}$ = 3 g L^{-1}, and C_{RhB} = 5 mg L^{-1}. The percentage degradation of RhB after sonocatalysis for 90 min is 98.7%. Detected by the photoluminescence technique that of using terephthalic acid as a probe molecule, hydroxyl radicals (·OH) are found to be produced on the irradiated by β-Bi_2O_3 particles. Based on the experimental results, ·OH radicals are suggested to be the major active species which are responsible for the degradation reaction.

1. Introduction

It is estimated that about 15% of produced dyes in the world are released into the natural environment during their synthesis and processing [1]. The dyes which release in the textile, rubber, plastics and other industries are harmful to environment and human health [2,3]. Rhodamine B (RhB) is widely used in industrial processes as a basic dye of the xanthene class. But it is toxic, which is capable to cause irritation to the skin, eyes, gastrointestinal tract which is respiratory tract [4]. Therefore, degradation of dyes (i.e. Rhodamine B) is necessary before they were discharged into the natural environment.

In recent years, ultrasonic degradation as one of the advanced oxidation process (AOP) has been studied and extensively used to treat organic dye effluents [5,6]. The main mechanism of ultrasonic process is acoustic cavitations, namely, the process of the formation, growth and implosive collapse of gas bubbles in liquids and releasing a mass of energies [7]. Subsequently, many local hot spots with high temperatures (5000 K) and high pressures (1000 atm) are generated for short period of time. Under such extreme conditions, organic compounds are mineralized and water molecules are cleaved to highly reactive free radicals species. These free radicals are also capable of degrading organic compounds [8,9].

However, the degradation of organic dye pollutants by ultrasonic usually needs long reaction time [10] and has limited efficiency [11]. In order to overcome these drawbacks, the methods of sonocatalytic degradation are developed [12–14]. Sonocatalytic process contains ultrasonic treatment by catalyst application. Application of catalysts with good adsorption ability, in the sonocatalytic process can increase the formation rate of cavitation bubbles by providing additional nuclei and increase the degradation efficiency of dyes [15–17].

Bi_2O_3 is an important metal oxide semiconductor catalyst, which has been identified as a promising visible light photocatalyst [18,19]. It has been used in the degradation of organic dyes due to its chemical structure, non-toxicity, optical and electrical properties. However, very few reports are available to help know about the performance of Bi_2O_3 particles on the ultrasonic-assisted degradation of organics dyes in previous publications.

In this paper, β-Bi_2O_3 particles as a kind of sonocatalysts were synthesized by a polyacrylamide sol–gel method. Its sonocatalytic activity to degrade Rhodamine B (RhB) was studied. The influences of various experimental factors including ultrasonic frequency, solution temperature, catalyst dosage, initial RhB concentration on the sonocatalysis efficiency were systematically investigated. The major active species which is responsible for the degradation reaction was also examined.

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2. Experimental

2.1. Materials

All chemicals used in this work were analytical grade reagents. These chemicals were used without further purification. Bismuth nitrate, citric acid, acrylamide and Rhodamine B were all purchased from Tianjin Kemiou Chemical Reagent Co. Ltd. Nitric acid was purchased from Xilong Chemical Industry Co. Ltd. The solution was prepared with water purified by a Millipore Milli Q system.

2.2. Synthesis of β-Bi_2O_3 particles nanoparticles

0.01 mol of bismuth nitrate was dissolved in 100 mL aqueous nitric acid solution. Then, 0.02 mol of citric acid and 0.1 mol of acrylamide monomers were added successively. The constant magnetic stirring was used during procedure to produce Homogenous additives solution. The solution was heated at 80 °C for 10 min to produce a viscous polyacrylamide gel. The gel was dried at 120 °C, calcined at 600 °C for 2 h to obtain β-Bi_2O_3 particles.

2.3. Characterization

XRD patterns were obtained using Bruker D8 ADVANCE diffractometer with Cu Kα radiation. Transmission electron microscopy (TEM) analyses were conducted on a JEOL JEM-2010 transmission electron microscope. The UV–vis absorption spectra were recorded using a Hitachi U-2001 UV–vis spectrophotometer. Photoluminescence spectra was operated on a PerkinElmer LS-55 luminescence spectrometer.

2.4. Sonocatalytic activity testing

Sonocatalytic degradation of RhB was investigated in the presence of β-Bi_2O_3 particles using a commercial ultrasonic cleaning machine (BK-240), operating at an ultrasonic frequency of 40 or 60 kHz and output power of 240 W. In a typical process, a certain amount of the catalyst was added to 20 mL RhB aqueous solution in a glass beaker. The obtained solution was magnetically stirred for 30 min in the dark to reach the absorption–desorption equilibrium between the catalyst and RhB, and then submitted to the ultrasonic irradiation. The sonocatalytic experiment was performed in the dark to avoid photoexcitation of β-Bi_2O_3 particles. At the regular time intervals, 3 mL of the irradiated suspensions was withdrawn and centrifuged to remove the catalyst and then, remaining RhB absorption was recorded at RhB maximum absorbance wavelength of 554 nm by a UV–vis spectrophotometer. The degradation efficiency is defined as $(C_0 - C_t)/C_0 \times 100\%$, where C_0 is the initial concentration of RhB and C_t means the concentration of RhB at distinct process time. During the experiment, the ultrasonic bath temperature was controlled by circulating running water through the ultrasonic bath.

2.5. Hydroxyl radicals (·OH) analysis

By using terephthalic acid (TPA) as a probe molecule, the photoluminescence (PL) technique was used to examine the ·OH radicals produced by the ultrasonic-irradiated β-Bi_2O catalyst. TPA can readily react with ·OH radicals to produce a highly fluorescent compound, 2-hydroxyterephthalic acid (TAOH). The PL intensity of TAOH at 429 nm is proportional to the amount of produced ·OH radicals. Terephthalic acid was added to sodium hydroxide solution (1.0×10^{-3} mol L^{-1}), to obtain a 2.5×10^{-4} mol L^{-1} TPA solution. The catalyst was added to the solution with a concentration of 1.0 g L^{-1}. After magnetically stirred for 30 min in the dark, the mixed solution was placed in the ultrasonic bath for ultrasonic irradiation. The reacted solution was centrifuged to remove the catalyst and analyzed by recording the PL measurements at a luminescence spectrometer with the excitation wavelength of 315 nm.

3. Results and discussion

3.1. The XRD pattern and the TEM image of β-Bi_2O_3 particles

Fig. 1 shows the XRD pattern of β-Bi_2O_3 particles, and the standard diffraction pattern of tetragonal crystal structure of the β-Bi_2O_3 (JCDPS 27-0050). It is seen that all the diffraction peaks are related to the tetragonal β-Bi_2O_3 phase, and no traces of second phase are detected in the XRD pattern. Inserted in Fig. 1 is a TEM image of β-Bi_2O_3 particles. It can be seen that β-Bi_2O_3 particles are relatively uniform and oval in shape with an approximately diameter of 150 nm.

3.2. Effect of ultrasonic frequency on the degradation of RhB

Fig. 2 shows the effect of the ultrasonic frequency on the sonocatalytic degradation of RhB separately at 40 and 60 kHz, at different temperatures for 90 min, where the initial concentration of RhB is C_{RhB} = 5 mg L^{-1}, the loading of the catalyst is $C_{catalyst}$ = 1 g L^{-1}. This indicates that the sonocatalysis efficiency at the ultrasonic frequency of 60 kHz is larger than that at 40 kHz at different temperatures. The intensity of temperatures and pressures (or sonoluminescence) produced at higher frequencies is enough to generate electron–hole pairs regarding to β-Bi_2O_3 narrow bandgap energy of 2.37 eV. The main reason for the increased sonocatalysis efficiency at higher frequencies is ascribed to the increase in cavitation. In addition, high frequency ultrasound can greatly enhance the diffusion mass transfer [20], which is beneficial to the sonocatalytic degradation of RhB.

3.3. Effect of temperatures on the degradation of RhB

Fig. 3 shows the sonocatalytic degradation of RhB over β-Bi_2O_3 particles versus of time (t) at different temperatures under ultrasonic irradiation of 60 kHz, where C_{RhB} = 5 mg L^{-1}, $C_{catalyst}$ = 1 g L^{-1}. The blank experiment reveals a good stability of RhB under ultrasonic irradiation at 60 kHz without catalyst, which indicates that ultrasonic irradiation alone hardly lead to the degradation of RhB. Combination of ultrasonic irradiation with β-Bi_2O_3 particles

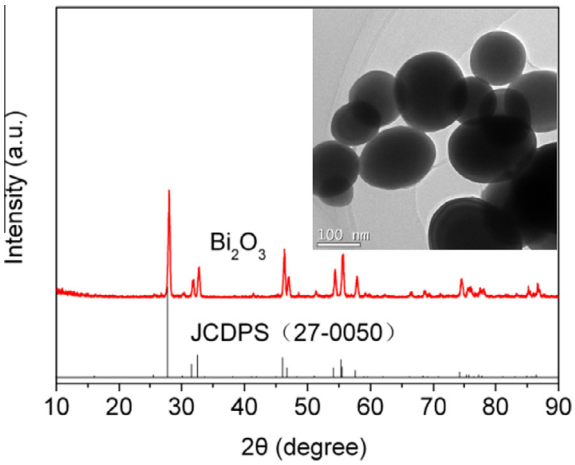

Fig. 1. XRD pattern of β-Bi_2O_3 particles, and the standard diffraction pattern of tetragonal crystal structure of the β-Bi_2O_3 (JCDPS 27-0050). The insert shows a TEM image of β-Bi_2O_3 particles.