

Sonocatalytic degradation of RhB over LuFeO₃ particles under ultrasonic irradiation



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HIGHLIGHTS

- LuFeO₃ particles exhibit a good sonocatalytic activity toward the RhB degradation.
- Various experimental factors on the sonocatalysis efficiency were investigated.
- •OH radicals are produced over the ultrasonic-irradiated LuFeO₃ particles.
- Ethanol leads to a quenching of •OH radicals and a simultaneous decrease in RhB degradation.
- •OH radicals are the primary active species responsible for the dye degradation.

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ABSTRACT

LuFeO₃ particles with an average particle size of ~200 nm were synthesized via a polyacrylamide gel route. The sonocatalytic activity of LuFeO₃ particles was evaluated by the degradation of Rhodamine B (RhB) under ultrasonic irradiation, revealing that they exhibit a good sonocatalytic activity. The effects of various experimental factors including ultrasonic frequency (f), reaction solution temperature (T), catalyst dosage (C_{catalyst}), initial RhB concentration (C_{RhB}), and pH value on the sonocatalysis efficiency were investigated. It is found that the former four factors have an important influence on the sonocatalytic degradation of RhB, where the best degradation conditions are obtained to be $f = 60$ kHz, $T = 40$ °C, $C_{\text{catalyst}} = 4$ g L⁻¹, and $C_{\text{RhB}} = 5$ mg L⁻¹. The pH value has a relatively small effect on the sonocatalytic degradation of RhB compared with other experimental factors. Hydroxyl (•OH) radicals were detected by fluorimetry using terephthalic acid as a probe molecule, revealing that they are produced over the ultrasonic-irradiated LuFeO₃ particles. The addition of ethanol leads to a quenching of •OH radicals and a simultaneous decrease in the RhB degradation. This indicates that •OH radicals are the primary active species responsible for the dye degradation.

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1. Introduction

Semiconductor-based photocatalysis has been an enduring research focus because it, as a green technology, offers a great potential to solve the problems of environmental pollution and energy crisis [1]. Especially nowadays, with the rapid growth of world population and economy, the environmental and energy problems that confront mankind are becoming more and more serious. Organic pollutants such as dyes in wastewater must be removed or destroyed to an acceptable level before being discharged into the natural environment. Among various dye wastewater treatment technologies, semiconductor-based photo-

catalysis allows the use of sunlight, a clean and renewable source of energy, for the destruction of dye pollutants. When a semiconductor is irradiated with light of energy greater than its E_g , valence band (VB) electrons are excited to the conduction band (CB), thus creating electron-hole ($e^- - h^+$) pairs. The photogenerated electrons and holes migrate to the particle surface and participate in a series of redox reactions to produce a number of active species like hydroxyl (•OH) radicals. However, the dye wastewater is usually non-transparent and highly-concentrated; as a result, the light penetrating ability is limited and the penetrating depth is only several millimeters. Therefore, it is hard for semiconductor photocatalysis to degrade non-transparent and highly-concentrated dye wastewater. There is a need to develop a new treatment method that is more effective in eliminating highly-concentrated dyes from the non-transparent wastewater.

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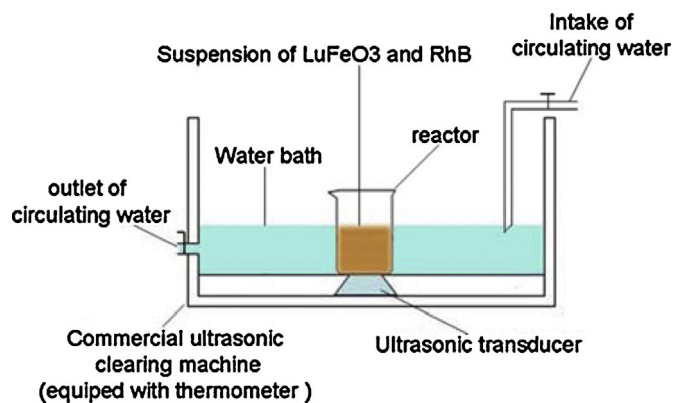


Fig. 1. Schematic illustration of the sonocatalysis apparatus.

Semiconductor-based sonocatalysis is an alternative method to degrade organic pollutants in wastewater, which can overcome the disadvantages of photocatalytic technology mentioned above because the ultrasonic wave has a strong penetrating ability for any water medium [2,3]. The sonocatalytic mechanism can be understood based on the acoustic cavitation in liquid induced by ultrasonic irradiation [4]. The acoustic cavitation involves the processes of formation (nucleation), rapid growth (expansion) and violent collapse (implosion) of cavitation bubbles in liquid. The sudden collapse of the cavitation bubbles can generate local high temperature (5000 K) and pressure (1000 atm) [5,6] (i.e., the so-called hot spots), which leads to the emission of light, i.e., the so-called sonoluminescence [7–9]. The sonoluminescence spectrum has a fairly wide range of wavelength, including a part of ultraviolet (UV) light. When a semiconductor (sonocatalyst) is present in an ultrasonic irradiation wastewater, its VB electrons are excited to the CB by the hot spot or sonoluminescence, leaving behind holes in the CB [10]. The sono-excited electrons and holes participate in redox reactions to degrade the organic pollutants. Moreover, the presence of the catalyst in the ultrasonic process can increase the formation rate of cavitation bubbles by providing additional nuclei [11,12], thus leading to an increase in the sonocatalysis efficiency. In fact, the sonocatalysis is very similar to the photocatalysis, both of which are initiated by exciting the semiconductor. The essential difference between sonocatalysis and photocatalysis is that the semiconductor is excited by two different kinds of energies. A number of studies have been devoted to the degradation of hazardous organic compounds by combining ultrasonic irradiation with UV or sunlight irradiation in the presence of semiconductor catalyst, i.e., sonophotocatalysis [13–17]. The results showed a synergistic effect between sonocatalysis and photocatalysis since the reaction rate constant of the combined process was greater than the summed value of the two individual processes.

It is well-known that the sonocatalytic efficiency depends highly on the type of catalyst. Development of novel sonocatalyst is important to further understand the sonocatalytic mechanism and promote the sonocatalysis applications. In this work, we report a novel sonocatalyst LuFeO_3 which exhibits a good sonocatalytic activity toward the dye degradation under ultrasonic irradiation. LuFeO_3 is a member of the rare-earth orthoferrite family, which can be formed in orthorhombic (space group: $Pbnm$) or hexagonal (space group: $P6_3cm$) structures depending on preparation methods [18]. In LuFeO_3 , there is almost no Lu^{3+} – Lu^{3+} or Lu^{3+} – Fe^{3+} interactions due to no localized magnetic moment on Lu^{3+} ions, but the Fe^{3+} – Fe^{3+} superexchange interaction is exceptionally strong, which results in antiferromagnetic ordering at a high temperature (~ 625 K) [19]. However, the alignment of Fe^{3+} magnetic moments is not completely antiparallel but there is a small declination from the collinear antiferromagnetic order of Fe^{3+} spins [20], generally

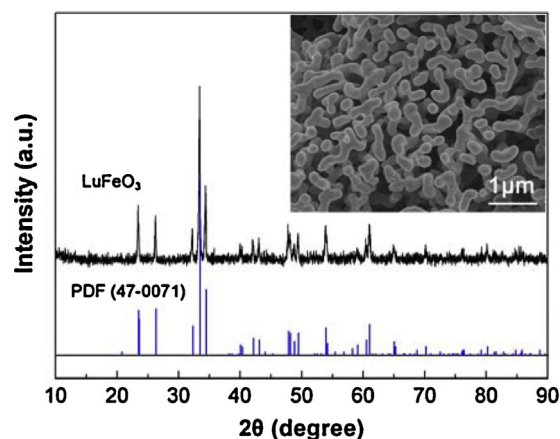


Fig. 2. XRD pattern of LuFeO_3 particles, along with the standard XRD line pattern for LuFeO_3 orthorhombic perovskite structure (PDF card no. 47-0071). The insert shows a SEM image of LuFeO_3 particles.

leading to a weak ferromagnetic behavior in a LuFeO_3 . Due to its interesting magnetic structure as well as large dielectric constant and multiferroic property [21–26], LuFeO_3 has attracted a great deal of attention during recent years. Herein we demonstrate that LuFeO_3 particles synthesized by a polyacrylamide gel route can be used as a promising sonocatalyst for the degradation of Rhodamine B (RhB). The influences of various experimental factors including reaction solution temperature, ultrasonic frequency, catalyst dosage, initial RhB concentration, and pH value on the sonocatalysis efficiency were systematically investigated. Ethanol was used as a hydroxyl ($\cdot\text{OH}$) scavenger [27] to investigate its effect on the sonocatalysis efficiency as well as the $\cdot\text{OH}$ radical yield with the aim of clarifying the role of $\cdot\text{OH}$ radicals in the sonocatalysis.

2. Experimental

2.1. Materials

Lutetium nitrate hydrate ($\text{Lu}(\text{NO}_3)_3 \cdot \text{H}_2\text{O}$, 99.9%) was purchased from Chengdu Gray West Chemistry Technology Co., Ltd. Iron nitrate nonahydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 98.5%) was purchased from Tianjin KaiXin Chemical Industry Co., Ltd. Ethylene diamine tetraacetic acid (EDTA, 99.5%) and acrylamide were purchased from Tianjin Kermel Chemical Reagent Co., Ltd. Glucose (analytical reagent) was purchased from Tianjinshi Baishi Chemical Co., Ltd. Rhodamine B (analytical reagent) was purchased from Shanghai ZhongTai Chemical Reagent Co., Ltd. Terephthalic acid (TPA, 99.0%) was purchased from Shanghai KEFENG Chemical Reagent Co., Ltd.

2.2. Synthesis of LuFeO_3 nanoparticles

LuFeO_3 nanoparticles used in this work were synthesized by a polyacrylamide gel route. Typically, equimolar amount (0.0075 mol) of $\text{Lu}(\text{NO}_3)_3 \cdot \text{H}_2\text{O}$ and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ was dissolved in 20 mL dilute nitric acid solution (1.6 mol L^{-1}). Then to the mixed solution were successively added 0.0225 mol of EDTA, 20 g of glucose, 20 mL of distilled water, and 0.135 mol of acrylamide. Every step mentioned above was accompanied by a constant magnetic stirring to make the additives dissolve completely. The mixed solution was filled up to 100 mL by adding distilled water and then adjusted to a pH of ~ 2 with aqueous ammonia. The final mixed solution was heated at 80°C to initiate the polymerization reaction. A deep brown gel was formed after about 2 h of polymerization reaction. The formed gel was dried at 120°C for 24 h in a thermostat drier. The obtained xerogel was ground into powder in an

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