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Application of Fe⁰ aggregate in ultrasound enhanced advanced Fenton process for decolorization of methylene blue

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ABSTRACT

A successful decolorization of methylene blue (MB) was achieved through advanced Fenton's process (AFP) coupled with sonolysis (sono-AFP) using zero-valent iron (Fe⁰) aggregate as a catalyst. The decolorization was dependent on the pH, Fe⁰, H₂O₂ doses, and acoustic power. Fe⁰ aggregate was reusable. A modified pseudo-first order equation was developed to describe the decolorization kinetics. To save electricity, short ultrasound irradiation was applied to the system. MB was completely degraded in the sono-AFP under optimized condition. The decolorization was obviously inhibited by the presence of Cl⁻, but it was unaffected by the presence of H₂CO₃, H₂PO₄⁻, ClO₄⁻, NO₃⁻, and SO₄²⁻.

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Introduction

Ultrasonic irradiation (sonolysis) within a liquid medium results in the collapse of cavitation bubbles. Consequently, oxidizing agents such as H•, •OH, O•, and H₂O₂ are produced. Organic pollutants can be decomposed by: (1) direct pyrolysis of volatile compounds in the cavitation bubbles; or (2) by oxidation of non-volatile compound with oxidization agents at the bubble/water interface or in the bulk solution [1]. Although attempts have made using ultrasonic irradiation to decolorize textile dyes, the complete mineralization of azo dyes was difficult to achieve from only ultrasound [2].

Fenton's process involves instantaneous production of Fe²⁺ when ferrous salts are rapidly converted to Fe³⁺ as it reacts with H₂O₂ to produce •OH [3]. The rate of Fenton's reactions decreases when the amount of Fe²⁺ decreases, because Fe³⁺ changes slowly back into Fe²⁺ during the chain reaction [4]. To avoid the insufficiency of Fe²⁺/Fe³⁺, a Fenton-like process (Fe³⁺/H₂O₂) has been developed. The Fenton-like process normally refers to the use of ferric salts or iron powder as a source of catalytic iron [5]. The adding iron salts to Fenton's reactions would decrease the activity

of reaction and cause unnecessary loadings of inorganic anions (SO₄²⁻, Cl⁻) into aquatic systems. The use of zero-valent iron (Fe⁰) to keep Fenton's reactions active is referred to the advanced Fenton Process (AFP, Fe⁰/H₂O₂). While the use of nano-sized Fe⁰ in the AFP process effectively degrades many persistent organic compounds, optimization of the Fe⁰ dose is difficult because of variations in contaminant concentrations. From a practical implementation perspective, the AFP that utilized iron metal powder [6] or Fe⁰ aggregate [7] instead of nano-sized Fe⁰ avoids such concentration disturbance and provides sufficient Fe²⁺ for the reactions. Fenton-based advanced oxidation technologies, such as photo-Fenton, electro-Fenton, sono-Fenton, and AFP in conjunction with hydrodynamic cavitation have been developed to remove recalcitrant organic contaminants from wastewater because they were easily degraded by •OH [7–15]. Iglesias et al. [11] reported that Y-zeolite embedded in alginate could be efficiently used in electro-Fenton process to degrade commonly pesticides, imidacloprid and chlorpyrifos, of aqueous medium with energy consumption of around 11 kW h/kg-pesticide. Photo-Fenton process enables to oxidize recalcitrant wastewaters and consequently produce biodegradable intermediates. The main concerns over photo-Fenton process are the high operating cost and optimization of main operational conditions. As to minimize cost and improve photo-Fenton efficiency, several strategies have been put in practice, such as the use of heterogeneous catalysts and/or chelating agents, application of solar energy and integrating

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biological treatment technologies in the treatment process [12]. In a treatment of Acid Red 17 study, Khataee et al. [13] revealed that using calcined laterite soil as iron source in heterogeneous photo-Fenton-like process under visible light led to higher decolorization efficiency than that of Fenton-like process. Another similar work [14] also showed that heterogeneous photo-Fenton oxidation using CdS/multi-walled carbon nanotube-TiO₂ composite catalyst under visible light was able to remove methylene blue effectively, and MB was oxidized more efficiently compared with other Fenton and photo-Fenton processes. Yue et al. [15] found that using heterogeneous Fenton-like catalysts (Fe₂O₃/γ-Al₂O₃) in electro-Fenton process was effective in degradation of Acid Red 3R. However, a 65% degradation efficiency means 100 min of reaction time for degradation of 500 mg/L Acid Red 3R using 20 V cell voltage.

Recent attention has been paid to the use of Fenton's reagent combined with ultrasonic irradiation (sono-Fenton) to treat dye-laden wastewaters [7,16,17]. Synergistic effects occurred when Fenton's reagent was combined with ultrasonic irradiation enabled Fenton's reactions to effectively decompose complex dye compounds. An attempt of using EF in conjunction with ultrasound has been made for degradation of reactive black 5 dye; however the combination of EF with sonication was negligibly improved in terms of COD and color removals, compared to electro-Fenton process [18]. However, the study of incorporating Fe⁰ aggregate into the sono-AFP system for decolorization of cationic dyes has not been documented.

Fe⁰ aggregate is commonly used to remove certain persistent organic and inorganic pollutants from contaminated groundwater [19–21]. In this study, the decolorization of cationic dye, methylene blue (MB, C.I. 52015) has been investigated with sono-AFP using Fe⁰ aggregate as catalyst. In the sono-AFP system, we expected that ultrasonic irradiation enhanced the corrosion of Fe⁰ aggregate and continuously released massive amounts of Fe²⁺. Knowledge of reaction mechanisms and optimum conditions are essential to operate sono-AFP while treating dye waste. We systemically characterized four key parameters (pH, H₂O₂ dose, Fe⁰ dose, and ultrasonic power intensity) that affect the system to identify suitable decolorization conditions. The effect of inorganic salts on MB decolorization and performance of recycled Fe⁰ in sono-AFP were evaluated. In order to save electricity, this process could firstly operate with short ultrasonic irradiation, and then continued with a subsequent US unassisted degradation period. MB was chosen as a target contaminant because it is structurally simple and it is a common basic dye used as a redox indicator in analytical chemistry.

Experimental

Materials

MB (C₁₆H₁₈N₃SCl·3H₂O) was purchased from Riedel-de Haën Co. (Germany). The MB concentration in the solution was analyzed by measuring the absorption at 660 nm using a spectrophotometer (Thermo Scientific Evolution 201, USA). Fe⁰ aggregate (Fig. 1) with particle size 0.297–2.380 mm and specific weight 2240–2560 kg/m³ was obtained from Connelly-GPM Inc., USA. H₂O₂ (35% w/w) was purchased from J.T. Baker (USA).

Methods

Sonication of the dye solution was conducted in air atmosphere by an ultrasonic generator (S-450A, Branson, USA) equipped with a titanium horn transducer. The tip of the probe was 1.2 cm in diameter; 6 cm of the tip's total length was submerged in the solution. The sonication was administered in pulses with a 60% duty cycle at 60 kHz. In a typical experimental set-up, a glass beaker was filled with 1000 mL dye solution. The solution pH was



Fig. 1. Fe⁰ aggregate catalyst with a flake-like shape obtained from Connelly-GPM Inc., USA.

adjusted to a predetermined value using either HNO₃ or NaOH solutions. The dye solution was then directly irradiated with ultrasound for 30 min upon adding appropriate amounts of Fe⁰ and H₂O₂. At desired time intervals, aliquots (5 mL) were withdrawn from the beaker and immediately filtered through 0.45 μm membrane filter to collect the supernatant, which was then analyzed to determine the residual MB concentration. Total organic carbon (TOC) content of the samples was determined using a TOC analyzer (Apollo 9000 Dohrmann, USA). The Fe²⁺ present in the solution after reaction was analyzed by measuring absorption at 510 nm using HACH FerroVer[®] iron reagent [22]. Each decolorization experiments were conducted in duplicate, and average values were used in analysis.

Results and discussion

Synergistic effect of combined sonication with Fenton's reagent

In order to evaluate the synergistic effect of AFP combined with sonolysis (sono-AFP), different experiments were conducted using the same operating conditions except: (1) in the absence of sonication (AFP), (2) in the absence of Fe⁰ aggregate (US/H₂O₂), and (3) in the absence of H₂O₂ (Fe⁰/US). Fig. 2 shows the depletion of

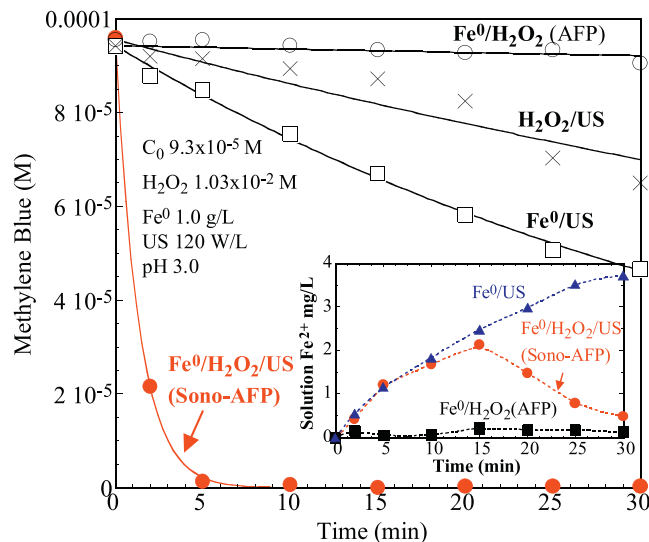


Fig. 2. Depletion of MB treated with different processes. The graph inset shows the variation of Fe²⁺ concentrations in the sono-AFP, AFP, and Fe⁰/US systems. The solid lines are the best fit of pseudo first-order equation.

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