



Research article

Synchronized methylene blue removal using Fenton-like reaction induced by phosphorous oxoanion and submerged plasma irradiation process



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ABSTRACT

In this study, a combination of phosphorus (PP) oxoanions in a submerged plasma irradiation (SPI) system was used to enhance the removal efficiency of dyes from wastewater. The SPI system showed synergistic methylene blue removal efficiency, due to the plasma irradiation and Fenton-like oxidation. The ferrous ions released from the iron electrode in the SPI system under plasmonic conditions form complexes with the PP anions, which can then react with dissolved oxygen (O₂) or hydrogen peroxide (H₂O₂) via Fenton-like reactions. The experimental results revealed that a sodium triphosphate (TPP) combined SPI system has a higher dye removal efficiency than a tetrasodium pyrophosphate (DP) or a sodium hexametaphosphate (HMP) combined SPI system under similar dissolved iron ion concentrations. To confirm the accuracy of the proposed removal mechanism via Fenton-like oxidation, it was compared to SPI systems under an oxygen environment (TPP/SPI/O₂ ($k = 0.0182 \text{ s}^{-1}$)) and a nitrogen environment (TPP/SPI/N₂ ($k = 0.0062 \text{ s}^{-1}$)). The results indicate that the hydroxyl radical ($\cdot\text{OH}$) in the TPP/SPI/O₂ system is the major oxidant in methylene blue removal, because the dye degradation rates dramatically decreased with the addition of radical scavengers such as *tert*-butanol ($k = 0.0023 \text{ s}^{-1}$) and methanol ($k = 0.0021 \text{ s}^{-1}$). On the other hand, no change was observed in the methylene blue removal efficiency of the TPP/SPI/O₂ system when it was subjected to a wide range of pHs (3–9). In addition, it was proved that this system could be used to eliminate six different commercial dyes. The results of this study indicated that the TPP/SPI/O₂ system is a promising advanced oxidation approach for dye wastewater treatment.

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1. Introduction

The textile industry is considered one of modern society's major industries because of the exponential population growth in

the twenty-first century. The processes carried out in this industry, such as dyeing, bleaching, printing, and finishing, require large quantities of water, which results in the release of large amounts of wastewater contaminated with complex dyes to natural streams (Krishnan et al., 2017). This wastewater contains high concentrations of highly toxic dyes that can affect the environment and human health (Oladipo and Gazi, 2014; Wang et al., 2011). When inhaled, methylene blue can cause vomiting, increased heart rate, paralysis, dysentery, jaundice, limb paralysis, nausea, excessive sweat secretion, mental confusion, painful urination, anemia, and tissue necrosis (Avom et al., 1997; Ghosh and

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Bhattacharyya, 2002; Vadivelan and Kumar, 2005). Therefore, water polluted with methylene blue should be appropriately treated before disposal.

However, conventional biological or physico-chemical treatment methods are not effective in most cases of dye wastewater treatment because of the complex poly-aromatic structure of the pollutants, high salinity (> 6.0% NaCl or > 5.0% Na₂SO₄) of the water, and recalcitrant nature of the dyes (Chung et al., 1978; Lin et al., 2015; Pagga and Brown, 1986). For example, byproducts of aromatic amines formed by the reductive cleavage of azo bonds under anaerobic conditions are more toxic than pristine dye molecules (Weber and Wolfe, 1987). In addition, the biochemical oxygen demand (BOD) to chemical oxygen demand (COD) ratios (< 0.1) of dye wastewater indicate non-biodegradable characteristics (Pagga and Brown, 1986). Furthermore, biological degradation is known to be less effective when used to remove high concentrations of color from wastewater (Gogate and Pandit, 2004; Teh and Mohamed, 2011).

Therefore, the use of alternative treatment technologies, such as advanced oxidation processes (AOPs), which aim to mineralize the dye molecules, have been used for the treatment of dye wastewater. The hydroxyl radical ($\cdot\text{OH}$) produced during AOPs effectively removes the organic contaminants, because it is a reactive electrophile that can rapidly react with electron-rich organic compounds. AOPs include hydrogen peroxide/UV light, hydrogen peroxide/ozone, titanium dioxide/UV light, ozone/UV light, and Fenton's reaction (Fe(II)/H₂O₂, H₂O₂/Fe(II)/UV) (Locke et al., 2006; Sunka et al., 1999).

Recently, the use of submerged plasma irradiation (SPI) processes for the degradation of organic pollutants in wastewater treatment has been attracting attention because it does not produce secondary pollutants (Grymonpré et al., 2001, 2004; Sato et al., 1996; Sun et al., 1999). High electrical energy-induced plasma channels can generate a variety of oxidizing agents, e.g., $\cdot\text{OH}$, H \cdot , O₂, HO₂ \cdot , O₂ \cdot^- , H₂O₂, and O₃, around the metallic electrodes in the SPI reactor (Jiang et al., 2014). However, the SPI process has several drawbacks, such as high energy consumption, low efficiency, and a short lifespan for the metal electrode, which is degraded by the plasma channel (Holzer and Locke, 2008; Lukeš et al., 2006). Recently, an SPI system for generating a Fenton-like reaction by adding hydrogen peroxide and utilizing dissolved copper ions from the electrode was developed to improve the energy efficiency of the process (Parkansky et al., 2012, 2013, 2014; Son et al., 2016). In a Fenton-like reaction, the ferrous ion complexes, with suitable ligands, can activate oxygen to produce hydrogen peroxide (H₂O₂) or $\cdot\text{OH}$ (Keenan and Sedlak, 2008; Welch et al., 2002).

Therefore, we investigated a novel iron electrode-coordinated SPI system, which includes enhanced Fenton-like oxidation by ferrous-complexing phosphate (Fe(II)-PP). In this study, phosphate was used as a cost-effective electrolyte and to catalyze the Fenton-like reaction. Primary plasma irradiation and secondary Fenton-like oxidation were used to remove methylene blue, and were investigated by conducting a number of comparison experiments between the PP/SPI/O₂ and PP/SPI/N₂ systems. In addition, the optimum operating conditions for the removal of methylene blue dye were determined by examining the electrode material, initial pH, and input voltage of the TPP/SPI/O₂ system. Finally, to evaluate the potential practical application of the proposed system, the TPP/SPI/O₂ system was also applied to degrade indigo carmine, reactive black 5, chromotrope FB, methyl orange, and reactive black 4, which are the most-widely consumed dyes in the industrial sector.

2. Materials and methods

2.1. Chemicals

All reagents used in this study were purchased from Sigma-Aldrich and were used without further processing. Furthermore, all the solutions used in the experiments were prepared using distilled water. In addition, the following dyes were used without further purification as the organic dye pollutants: methylene blue (MB, C₁₆H₁₈N₃SCl, 95%), reactive blue 4 (C₂₃H₁₄C₁₂N₆O₈S₂, 35%), methyl orange (C₁₄H₁₄N₃NaO₃S, 85%), chromotrope FB (C₂₀H₁₂N₂Na₇O₇S₂, 50%), reactive black 5 (C₂₆H₂₁N₅Na₄O₁₉S₆, > 50%), and indigo carmine (C₁₆H₈N₂Na₂O₈S₂, 85%). A stock solution (5 mg L⁻¹) of the methylene blue was prepared and the PP anion solution was prepared using PP stock solutions such as sodium triphosphate (TPP, Na₅P₃O₁₀, 98%), tetrasodium pyrophosphate (DP, Na₄P₂O₇, 99%), and sodium hexametaphosphate (HMP, Na₆P₆O₁₈, 96%), with a 50 mM phosphate buffer at pH 7. Sodium dihydrogen phosphate (NaH₂PO₄) and disodium phosphate (Na₂HPO₄) were used to prepare the phosphate buffer. Methanol (CH₃OH, 99.5%) and *tert*-butyl alcohol (C₄H₁₀O, 99.5%) were used as the radical scavengers. For the pH adjustment, 0.1 N NaOH and 0.1 N HClO₄ solutions were used.

2.2. SPI configuration and operating conditions

All the experiments in this study were conducted at room temperature (20 ± 2 °C) using a 50 mL sample in a 55 mL acrylic reactor (Fig. 1). Underwater plasma generated by an alternating current power generator with 220 V input energy and it boost 8 times. The plasma generator has a maximum electrical potential output of 1900 kV was employed. This plasma generation system based on DBD (Dielectric Barrier Discharge) plasma. The electrodes of the SPI used in this study were 2 mm in diameter and made of iron, copper, tungsten, and aluminum. Each electrode was placed in the reactor with a 1–1.5 mm ceramic insulator. A 250 mg L⁻¹ stock solution of the methylene blue was prepared, and the desired concentration of 5 mg L⁻¹ was obtained by adding adequate volumes of the prepared PP stock solution with the phosphate buffer to a predefined volume of methylene blue. TPP was mixed with the phosphate buffer before being transferred to the reactor. Finally, the methylene blue stock solution was injected. The pH was measured at the beginning of every experiment using a pH meter (Orion 4 star, USA). In this study, various operation conditions (PP type, dissolved oxygen, electrode materials, applied voltage, pH, radical scavengers, and dye type) were investigated to evaluate the degradation rate of methylene blue. The total reaction time was about 2 min, the plasma reaction proceeded at predetermined intervals (0, 10, 30, 60, 90, and 120 s), and the concentration of methylene blue was measured immediately after the reaction. Three replicas were carried out for each experiment to reduce error and to improve the reliability of the data.

2.3. Instrumentation

The concentration of methylene blue was determined with a UV-VIS spectrophotometer (Humas, HS-3300). The absorbance of methylene blue was analyzed at 664 nm (Kavitha and Namasivayam, 2007), and the other dyes (methyl orange, reactive black 4 and 5, chromotrope FB, and indigo carmine) were measured at 462 nm, 595 nm, 515 nm, and 610 nm, respectively. Distilled water was used to prepare the blank used in the methylene blue measurements. The removal rate of methylene blue was calculated

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