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Decolorization and degradation of Ponceau S azo-dye in aqueous solutions by the electrochemical advanced Fenton oxidation

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ABSTRACT

Oxidation (decolorization/degradation) of Ponceau S azo-dye in aqueous solutions by electro-generated Fenton's reagent (Fe^{2+}/H_2O_2) was optimized. This was carried out in a reactor containing 0.05 M Na_2SO_4 solution of pH 2.5 and a catalytic quantity of 0.1 mM $FeSO_4$ while a cathode potential of −1.0 V (vs. SCE) was applied. The reactor was an undivided glass electrochemical cell with a reticulated vitreous carbon (RVC) cathode and a platinum gauze anode. Progress of oxidation of various concentrations of Ponceau S with time of electro-Fenton reaction was monitored by UV–visible absorption measurements. Mineralization of the dye solutions was examined by estimation of the chemical oxygen demand removal and HPLC analysis at various times of electro-Fenton oxidation. The complete color removal of 0.05, 0.1 and 0.3 mM Ponceau S was achieved by electro-Fenton oxidation for 30, 60 and 90 min, respectively. However, approximately 98% mineralization of 0.05, 0.1 and 0.3 mM Ponceau S was achieved due to electro-Fenton oxidation for 40, 60 and 120 min, respectively. HPLC analyses showed also that almost no aromatic compounds were remaining in the treated solutions indicating the efficiency of the electro-generated Fenton's reagent for complete decolorization and significant mineralization of the Ponceau S azo-dye in aqueous solutions.

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1. Introduction

Synthetic azo-dyes are widely used in textile, printing, cosmetic, food colorants and pharmaceutical industries. Some azo-dyes are also used in laboratories as either biological stains or pH indicators. The strong electron withdrawing character of the azo-group stabilizes the aromatic substances against conversion by oxygenases. The durability of azo-dyes, however, causes pollution of surface water and consequently the soil and groundwater once the dye discharged into the environment as effluent. Most of the synthetic azo-dyes and their biodegradation products, sulfonated and unsulfonated aromatic amines, are toxic against aquatic organisms and suspicious of being carcinogenic and mutagenic for humans [1–8]. Therefore, these dyes necessitate proper treatment before discharge into the environment.

Various methods for removal of synthetic azo-dyes from wastewaters have been reported in the literature. These include adsorption on inorganic or organic matrices, biological activation, coagulation, chemical oxidation and electrochemical oxidation methods [1–3,7–9]. Nevertheless, some of these methods are usually non-destructive, inefficient, costly, and resulted in production of secondary waste products such as sludge which may need further disposal. Electro-Fenton oxidation method as an indirect electrochemical advanced oxidation process was developed and widely applied for oxidation of various organic pollutants [9–25]. In this method, H_2O_2 is continuously generated by reduction of the dissolved molecular O_2 in mildly acidic aqueous medium (Eq. (1)) using different cathodes (e.g. reticulated vitreous carbon [14], mercury pool [15], carbon-felt [16] and O_2-diffusion [17,18]):

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \qquad (E^o = 0.69\,V/NHE) \tag{1}$$

The oxidizing power of H_2O_2 is enhanced in presence of Fe^{2+} ions (as catalyst) in mildly acidic solution. Hydroxyl radical ($OH^\bullet$) and Fe^{3+} ions are then generated from the classical Fenton's reaction between Fe^{2+} ions and H_2O_2 [14–19,26,27]:

$$Fe^{2+} + H_2O_2 + H^+ \rightarrow Fe^{3+} + H_2O + OH^\bullet (\text{Fenton's reaction}) \tag{2}$$

Fe^{2+} ions consumed by Fenton's reaction in the homogeneous medium (Eq. (2)) are regenerated at the cathode by reduction of Fe^{3+} ions (Eq. (3)) which would induce the Fenton chain reaction efficiently [16].

$$Fe^{3+} + e^- \rightleftharpoons Fe^{2+} \qquad E^o = 0.77\,V/NHE \tag{3}$$

On the other hand, molecular oxygen necessary for production of H_2O_2 (Eq. (1)) is also regenerated at the anode by oxidation of water in the reactor (Eq. (4)):

$$H_2O \rightleftharpoons 1/2\ O_2 + 2H^+ + 2e^- \qquad E^o = 1.23\,V/NHE \tag{4}$$

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The sum of the above cathodic and anodic processes gives the overall reaction:

$$1/2\ O_2 + H_2O \rightarrow 2OH^{\bullet} \quad (5)$$

The indirect electro-Fenton oxidation method is environmentally friendly since it does not involve the use of harmful chemical reagents due to the fact that Fe^{2+} ions, H_2O_2 and the produced hydroxyl radicals ($OH^{\bullet}$) are non-toxic, besides the method is easy to handle and its reactor is simple. Besides, the indirect electro-Fenton oxidation method is very promising since it achieves high reaction yields with low treatment cost and has been efficiently applied to degrade many organic compounds [9–25]. Moreover, there is no production of iron sludge in the reactor and consequently no subsequent disposal problems were found. On other side, the hydroxyl radicals ($OH^{\bullet}$) are non-selective very powerful oxidizing agent that react with organics yielding dehydrogentated or hydroxylated derivatives until their overall mineralization (conversion into CO_2 and H_2O).

Ponceau S (3-Hydroxy-4-(2-sulfo-4-[4-sulfophenylazo] phenylazo) -2,7-naphthalenedisulfonic acid sodium salt), Scheme 1, has been used in the present study as a model of sulfonated azo-dye. Ponceau S azo-dye (also known as Acid Red 112) is used in dying industry of textile, leather and paper [28–30] and also used in clinical laboratories as a protein-binding dye for staining of blood serum proteins [31]. However, its biotransformation products have toxic effects against aquatic organisms and suspicious of being carcinogenic for humans [3,29].

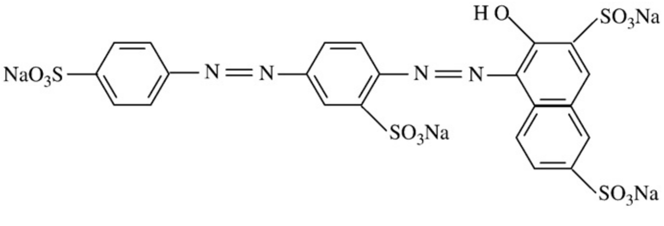
To our knowledge there is no information reported in the literature to date concerning decolorization/degradation of Ponceau S azo-dye by electro-generated Fenton's reagent. However, its complete decolorization was achieved using a developed methylene blue immobilized resin dower-11 photocatalyst [28] for 3 h treatment in solutions of pH 9. Besides, two biological methods were reported [32,33] for decolorization of Ponceau S azo-dye in aqueous solutions. The bacterium, *Kerstersia* sp. strain VKY1 could only decolorize Ponceau S azo-dye under aerobic condition with 100% decolorization efficiency at varying initial dye concentrations of 200, 400, 600 and 800 mg L^{-1} during 9, 12, 15, and 18 h incubation [32]. On other hand, the aerobic reduction of 0.2 mM Ponceau S azo-dye by Gram-positive bacterium *Enterococcus faecalis* strain ATCC 19433 to about 4.1% was achieved after 20 h cultivation in brain heart infusion medium [33]. Therefore, there is a need to optimize the optimal conditions for degradation of Ponceau S azo-dye, either to less harmful products or, more desirable, to complete mineralization by an effective and environmentally friendly method.

The present study was designed to optimize the oxidation (decolorization/degradation) of Ponceau S azo-dye in aqueous solutions by electro-generated Fenton's reagent (Fe^{2+}/H_2O_2) in an undivided glass electrochemical cell using a reticulated vitreous carbon (RVC) cathode and a platinum gauze anode.

2. Experimental

2.1. Materials and reagents

A commercial Ponceau S azo-dye (Aldrich Chemical Co. Inc, St. Louis, MO, USA) was used in the present study without further purification. Solutions of H_2SO_4 (1 M), anhydrous Na_2SO_4 (1 M), NaCl (1 M), KCl (1 M), $FeSO_4 \cdot 7H_2O$ (5 mM), $K_2Cr_2O_7$ (0.025 N) and $FeSO_4(NH_4)_2SO_4 \cdot 6H_2O$ (0.025 N) (all are analytical-grade reagents) were prepared in deionized water. Desired lower concentrations were obtained by the accurate dilution of the standard solutions by deionized water. Ferrion indicator solution (which was used in analysis of chemical oxygen demand, COD, by the standard open reflux method [34]) was prepared by dissolving 1.485 g of 1,10-phenanthroline monohydrate and 695 mg $FeSO_4 \cdot 7H_2O$ in 100 mL deionized water.



Scheme 1. Chemical structure of Ponceau S azo-dye molecule.

2.2. Electrochemical apparatus

A Potentiostat Model 362-PAR (Princeton Applied Research, Oka Ridge, TN, USA) was used in the present study. An undivided glass electrochemical cell (reactor) of 600-mL volume with a three electrode system was used. The cathode was a reticulated vitreous carbon (RVC) sheet (60 PPI) of the dimensions 5 cm, 7 cm and thickness of 0.9 cm (ARG Materials and Aerospace Corporation, Oakland, CA, USA). The anode was a platinum gauze of an area of 3.8 cm^2 placed at the center of the electrochemical cell. A reference saturated calomel electrode (SCE) was placed in a glass-luggin capillary at a position of 3 mm from the cathode surface. RVC was used as a cathode in the present electrochemical cell based on the fact that it is chemically and electrochemically inert over a wide range of potentials and with a broad variety of chemicals. It has a high specific surface area within the porous structure that is accessible to electrochemically active species. RVC has also a high fluid permeability, and it is easily shaped as required by cell design considerations. Furthermore, RVC has a good selectivity for H_2O_2 electrosynthesis. This is because it exhibits a range of electrochemical activities towards oxygen reduction, high overpotential for hydrogen evolution and low catalytic activity for decomposition of hydrogen peroxide.

2.3. Procedure

Oxidation of Ponceau S azo-dye by electro-generated Fenton's reagent was carried out in 450 mL of 0.05 M Na_2SO_4 solution of pH 2.5 (the pH of solution was adjusted by addition of H_2SO_4 solution) in the presence of a catalytic quantity of 0.1 mM $FeSO_4$ at a RVC cathode applied potential of −1.0 V vs. SCE using the described undivided glass electrochemical cell (reactor). Pure oxygen gas was bubbled (before starting the electro-Fenton oxidation) into the supporting electrolyte for 30 min to saturate the aqueous solution. The investigated solution was stirred magnetically in a rate of 400 rpm and the solution pH remained practically constant (2.5 ± 0.1). For monitoring the decolorization/degradation of the examined azo-dye in aqueous solution by electro-Fenton oxidation, 1.5 mL of the examined solutions were withdrawn from the reactor at regular time intervals (0, 10, 20, 30, 45, 60, 90, 120, 150, 180, 240, 300, 360, 420, 480, 540 and 570 min) then analyzed.

2.4. Monitoring of oxidation reaction

Progress of oxidation (decolorization/degradation) of Ponceau S azo-dye by electro-generated Fenton's reagent was monitored in the present study by:

2.4.1. UV–visible absorption measurements

The UV–visible electronic absorption spectra of Ponceau S azo-dye in aqueous solutions were recorded at ambient temperature (25 ± 2 °C) within the wavelength range of 200–800 nm using a Shimadzu UV–visible spectrophotometer Model 160A (Kyto, Japan) with a spectrometric quartz cell (1 cm path length). Quantitative analysis of Ponceau S azo-dye in aqueous solution was monitored spectrophotometrically by measuring its absorbance at $\lambda_{max.} = 515$ nm at different time intervals of electro-Fenton oxidation and then computing the corresponding concentration C_t of the dye from a

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