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The application of iron mesh double layer as anode for the electrochemical treatment of Reactive Black 5 dye

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ABSTRACT

In this work a novel anode configuration consisting of an iron mesh double layer is proposed for the electrochemical treatment of wastewater. The removal of Reactive Black 5 dye (RB5) from synthetic contaminated water was used as a model system. At a constant anode surface area, identical process operating parameters and batch process mode, the iron mesh double layer electrode showed better performance compared to the conventional single layer iron mesh. The double layer electrode was characterized by RB5 and chemical oxygen demand (COD) removal efficiency of 98.2% and 97.7%, respectively, kinetic rate constant of 0.0385/min, diffusion coefficient of 4.9×10^{-5} cm^2/sec and electrical energy consumption of 20.53 $kWh/kg_{dye\ removed}$. In the continuous flow system, the optimum conditions suggested by Response Surface Methodology (RSM) are: initial solution pH of 6.29, current density of 1.6 mA/cm^2, electrolyte dose of 0.15 g/L and flow rate of 11.47 mL/min which resulted in an RB5 removal efficiency of 81.62%.

Introduction

Wastewater discharge from dye manufacturing and textile industries has become a global concern and is one of the major causes of environmental pollution. According to the Malaysian Department of Environment (DOE), approximately 160,000 tons of textile industrial wastewater were discharged within the years 2011 to 2012 and the amount continues to increase annually (DOE, 2012). Thus, the treatment of textile wastewater to a permissible concentration is essential before discharge to the aquatic environment.

Reactive dyes have complex chemical structures, which consist of organic ring, vinyl sulfonic groups and chloride atoms (Klemola et al., 2007). These dyes are widely used in the textile industry as their vivid colors and reactive groups easily attach to the textile fibers (Hassan et al., 2009). However, the relatively low fixation of reactive dyes on cellulose fibers during the dyeing process, resulted in the high amount of dyes released into dyebath effluent. Excessive amounts of reactive dyes discharged into receiving waters are dangerous and may pose a serious threat to the aquatic ecosystem. Moreover, these dyes are mutagenic and carcinogenic, and thus may cause a severe damage to liver, digestive and central nervous system of human beings (El-Zawahry et al., 2016). Reactive dyes in dyeing wastewater are known as recalcitrant compounds as they have high alkalinity, high concentration of organic compounds and strong color compared with other dyes (Barka et al., 2010).

Many methods have been developed and investigated for the removal of dyes from effluents such as membrane

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filtration, adsorption, photocatalytic oxidation and biological treatment (Mook et al., 2016; Zeng et al., 2014; Szlachta and Wójtowicz, 2013; Rivera et al., 2011). The chemical coagulation is a conventional (and one of the most popular) technique used for the treatment of textile dyeing wastewaters. However, the costs of required coagulants (aluminum(III) or iron (III) salts) and generation of the considerable amount of sludge cause that in some cases, the employment of chemical coagulation for decolorizing wastewater is not economically justified.

An alternative method to the chemical coagulation is electrocoagulation, which based on the electrochemical technique. During the electrocoagulation process, the coagulant is produced *in situ* by electrolytic oxidation of sacrificial anode (Merzouk et al., 2011). Thus, the advantage of application of the electrochemical treatment, apart from the high efficiency of the process and its versatility, includes low cost and minimal sludge generation (Mook et al., 2012). The potential of electrocoagulation in dye removal from wastewater has already been proven by other researchers (Nandi and Patel, 2013; Zodi et al., 2013). The process is characterized by low sludge production, easy floc separation, low level of total dissolved solids (TDS) and no secondary pollution caused by external added coagulant (Mollah et al., 2001).

However, the sacrificial electrode used in the process needs to be monitored and replaced frequently as it participates in the anodic oxidation. Moreover, the electrocoagulation requires the energy input, thus the operating costs related with the price of electric current may limit its application. Hence, the optimum of electric current needs to be investigated as it is one of the important parameters that affect the lifetime of the electrodes and operating cost.

The configuration of the electrode used in the process is a crucial feature to increase anodic oxidation and hence, induce higher dye elimination. Several studies have been conducted for dye removal that used different anode shapes such as a plane (Mohan et al., 2007; Nandi and Patel, 2013; Pajootan et al., 2012; Steter et al., 2014; Thiam et al., 2015) and mesh (del Río et al., 2012; Méndez-Martínez et al., 2012). A mesh type electrode produces a higher discharge current than a plane type due to a higher electric field intensity at the edge of the mesh holes (Kuroda et al., 2003). Moreover, the mesh produces a larger specific surface area than a flat sheet which could boost the formation of iron hydroxides (Ambler and Logan, 2011; Zhang et al., 2010). Therefore, in this work, a mesh shape was selected as the anode configuration.

In typical multifactor experiments, one-factor-at-a-time (OFAT) is used, *i.e.* varying one parameter while others are fixed. This approach is slow, requiring large numbers of experiments and it is difficult to estimate the interaction between the parameters. Response Surface Methodology (RSM), on the other hand, is a valuable tool for designing experiments. This tool can assist researchers in building models, reducing the number of experiments, exploring the interactions between parameters and obtaining optimum conditions for desirable responses (Mook et al., 2013). To date, no literature is available on Reactive Black 5 (RB5) treatment using electrocoagulation with RSM design. Hence, Central Composite Response Surface Design (CCD) was utilized to investigate the interaction between parameters and optimize the treatment process.

RB5 dye is a diazoacidic reactive dye, which consists of two sulfonates and two sulfato-ethylsulfon groups. It is typically used in textile industries for dyeing of cotton, woolen and nylon fabrics (Elwakeel et al., 2016). In this study, RB5 has been utilized as a model dye because of its recalcitrant nature and the fact that it contributes to 50% of the total world demand for reactive dyes (Schumacher, 2012).The first part of this work is to investigate the performance of new anode configuration (iron mesh double layer electrode) on RB5 removal, by comparing it to the conventional single layer design. The configuration with superior removal efficiency was subsequently used in comparing performance of the system mode and RSM studies.

1. Mechanism of electrocoagulation

Electrocoagulation (EC) is an electrochemical process using iron or aluminum as the anode in which Fe^{2+} or Al^{3+} ions are released into the solution through anodic dissolution and their hydroxides then react with pollutants (Körbahti et al., 2011). Iron has been proven by several researchers to be a more effective electrode material than aluminum (Akanksha et al., 2013; Chafi et al., 2011). EC has been successfully applied for the removal of different types of dye from textile industry wastewater (Khandegar and Saroha, 2013).

Iron hydroxides can be produced through two mechanisms (Eqs. (1)-(8)) and they remove the pollutant molecules *via* surface complexation or electrostatic attraction. In surface complexation, the pollutant acts as ligand to bind with hydrous iron through precipitation and adsorption mechanisms (Eqs. (9)–(12)) (Körbahti et al., 2011).

Mechanism I:

$$\text{Anode}: Fe_{(s)} \rightarrow Fe^{2+}{}_{(aq)} + 2e^- \tag{1}$$

$$\text{Chemical}: Fe^{2+}{}_{(aq)} + 2OH^-{}_{(aq)} \rightarrow Fe(OH)_{2\ (s)} \tag{2}$$

$$\text{Cathode}: 2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-{}_{(aq)} \tag{3}$$

$$\text{Overall reaction}: Fe_{\ (s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)}. \tag{4}$$

Mechanism II:

$$\text{Anode}: 4Fe_{(s)} \rightarrow 4Fe^{2+}{}_{(aq)} + 8e^- \tag{5}$$

$$\text{Chemical}: 4Fe^{2+}{}_{(aq)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H^+{}_{(aq)} \tag{6}$$

$$\text{Cathode}: 8H^+{}_{(aq)} + 8e^- \rightarrow 4H_{2(g)} \tag{7}$$

$$\text{Overall reaction}: 4Fe_{(s)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 4H_{2(g)} \tag{8}$$

$$\text{Precipitation}: \text{dye} + \text{monomeric Fe} \rightarrow [\text{dye\_monomeric Fe}] \tag{9}$$

$$\text{dye} + \text{polymeric Fe} \rightarrow [\text{dye\_polymeric Fe}]c \tag{10}$$

$$\text{Adsorption}: \text{dye} + Fe(OH)_n \rightarrow [\text{particle}] \tag{11}$$

$$[\text{dye\_polymeric Fe}] + Fe(OH)_n \rightarrow [\text{particle}]. \tag{12}$$

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