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Treatment of levafix orange textile dye solution by electrocoagulation

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Abstract

The decolorization of the levafix orange textile dye in aqueous solution by electrocoagulation using aluminum sacrificial anode has been investigated. The process performance is analyzed in terms of decolorization efficiency and the important cost-related parameters such as electrode and energy consumptions, as a function of initial pH, conductivity, current density, initial dye concentration and electrolysis time. The present study proves the effectiveness of electrochemical treatment for the textile dye solution. 95% decolorization efficiency may be obtained at suitable operating conditions such as; current density 100 A/m², operating time 12 min and initial pH 6.4. The corresponding electrode and energy consumptions during the electrolysis were found to be 1.8 kg Al/kg dye and 35 kWh/kg dye. © 2005 Elsevier B.V. All rights reserved.

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1. Introduction

Textile industries use large amount of water and chemicals for finishing and dying processes. Dye wastewater usually consists of a number of contaminants including acids, bases, dissolved solids, toxic compounds, and colored materials which are noticeable even at very low concentrations and need to be removed before the wastewater can be discharged.

Traditional methods for dealing with textile wastewater consist of various combinations of biological, physical and chemical methods [1,2]. Biological treatment of dying wastewater is cheaper than other methods, but it is less efficient for decolorization due to toxicity of the wastewater and the need for an aeration system. Although the dyestuff and colored materials in wastewater can be effectively destroyed by advanced chemical oxidation such as UV/H₂O₂, O₃ [3,4] and adsorption using activated carbon [5,6], the costs of these methods are relatively high for an economically feasible treatment of the textile wastewater. The electrocoagulation (EC) technique is considered to be potentially an effective tool for treatment of textile wastewaters with high removal efficiency.

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EC is a process consisting of creating metallic hydroxides flocs within the wastewater by electrodissolution of soluble anodes, usually made of iron or aluminum. Electrocoagulation has been applied successfully for treatment of potable water [7], food and protein wastes [8], textile wastewater [9,10], aqueous suspensions containing kaolinite, bentonite and ultrafine particles [11,12], fluoride containing water [13,14], restaurant wastewater [15,16], textile dyes solutions [17,18], and smelter wastewater containing harmful arsenic [19].

The objective of the present study is to investigate the decolorisation of a reactive textile dye (Levafix Orange E3 GA) in aqueous solution using aluminum as non-toxic and readily available electrode material. Several parameters, namely initial pH, conductivity, current density, dye concentration and electrolysis time, were investigated for their effects on the decolorisation efficiency. The corresponding electrode and energy consumptions are also determined as principal cost parameters.

2. Theoretical considerations

Electrocoagulation occurs via serial steps such as; electrolytic reactions at electrode surfaces, formation of coagulants in aqueous phase, adsorption of soluble or colloidal pollutants on coagulants, which are removed by sedimentation or flotation.

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The metal ions generation takes place at the anode; hydrogen gas is released from the cathode. The hydrogen gas would also help to float the flocculated particles out of the water. The main reactions occurring at the electrode are as follows:

Anode :
$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^{-}$$
 (1)

Cathode :
$$3H_2O_{(1)} + 3e^- \rightarrow 3/2H_{2(g)} + 3OH^-_{(aq)}$$
 (2)

 Al^{3+} and OH^- ions generated by electrode reactions (1) and (2) react to form various monomeric species such as $Al(OH)^{2+}$, $Al_{(OH)_2}^+$, $Al_2(OH)_2^{4+}$, $Al(OH)_4^-$, and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, $Al_{13}(OH)_{34}^{5+}$, which transform finally into $Al(OH)_{3(s)}$ according to complex precipitation kinetics [20–24].

$$Al_{(aq)}^{3+} + 3H_2O_{(1)} \rightarrow Al(OH)_{3(s)} + 3H_{(aq)}^{+}$$
 (3)

3. Materials and methods

3.1. Materials

Commercially available reactive dye, Levafix Orange E3 GA was obtained from DyStar and its molecular structure is shown in Fig. 1. Distilled water was used to prepare the desired concentration of dyestuff solution. The characteristics of the dye are summarized in Table 1.

3.2. Experimental apparatus

The experimental setup was the same as published previously [26]. The thermostated electrocoagulator was made of



Fig. 1. Molecular structure of Levafix Orange E3 GA.

Table 1	
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Characteristics	of Levafix	orange E3	GA
Characteristics	OI LOVAIIA	orange L5	011

Parameter	Value
Color index	Reactive Orange 64
CAS number	83763-57-9
Molar mass (non-hydrolyzed dye)	768
Percentage of pure dye	50-60%
Water solubility at 293 K (g/l)	30
Biological degradation (%)	<10
Thermal degradation (°C)	150
Bulk density (kg/m ³)	470-600
Acute oral toxicity LD50 (mg/kg)	>2000
Fish toxicity LC_0 (mg/l)	>100
pH value (100 g/l water)	6.5–7.5
λ_{max} (nm)	490

Plexiglas with the dimensions of $65 \text{ mm} \times 65 \text{ mm} \times 110 \text{ mm}$. There were four monopolar electrodes, two anodes and two cathodes of the same dimensions. Both aluminum (99.53%) cathode and anode were made from plates with dimensions of $46 \text{ mm} \times 55 \text{ mm} \times 3 \text{ mm}$. The total effective electrode area was 78 cm^2 and the distance between electrodes was 11 mm. The electrodes were connected to a digital DC power supply (Topward 6306D; 30 V, 6A) equipped with potentiostatic or galvanostatic operational options.

3.3. Experimental procedure

All the runs were conducted at constant temperature of 25 °C. Preliminary experiments showed that the stirring (magnetically) was beneficial for high removal efficiency, which was held constant at optimal value of 200 rpm in subsequent experiments. Before each run, the aluminum electrodes were washed with acetone to remove surface grease. The impurities on the surfaces of the electrodes were also removed by dipping for 5 min in a solution which was prepared by mixing 0.1 L of HCl solution (35%) and 0.2 L of hexamethylenetetramine $(CH_2)_6N_4$) aqueous solution (2.80%) [1]. In each run, 0.25 L of dye solution was placed into the electrolytic cell. The current density was adjusted to a desired value and then the experimental run was started. At the end of the run, the solution was filtered and the filtrate was centrifuged at 2000 rpm. The electrodes were washed thoroughly with water to remove any solid residues on the surfaces, dried and reweighed.

3.4. Analytical method

A UV-spectrophotometer (Shimadzu, Model UV-160) was employed to measure the maximum wavelength ($\lambda_{max} = 490$ nm), which was used to measure the concentration of the dye. The decolorization efficiency, *E*, is calculated as

$$E = \frac{C_{\rm i} - C_{\rm f}}{C_{\rm i}} \times 100 \tag{4}$$

where C_i is initial dye concentration (mg/L) and C_f is final dye concentration (mg/L).

4. Results and discussion

Decolorization efficiency, electrode and energy consumptions were investigated in terms of initial pH, conductivity, current density, dye concentration and electrolysis time in order to determine the optimum operating conditions for maximum decolorization efficiency of the reactive dye.

4.1. Effect of initial pH

pH is an important operating factor influencing the performance of electrocoagulation process, as observed also by other investigators [20–22,25]. The effect of the initial pH on decolorization efficiency for Al electrode is shown in Fig. 2a; the decolorization efficiency decreases slowly with increase in



Fig. 2. Effect of initial pH (a) on the decolorization efficiency, (b) on the energy and electrode consumptions for Al electrode (conditions: current density: 100 A/m^2 , Co: 250 mg/L, time: 15 min, conductivity: 750μ S/cm).

pH ranged from 3 to 9. At pH values above 9, the efficiency decreases rapidly. Maximum (99.9%) and minimum (76.6%) decolorization effiencies occurred at pH values of 3 and 11, respectively.

The effect of pH on the process performance is explained as follows: the dominant aluminum species are different according to the solution pH; Al^{3+} and $Al(OH)^{2+}$ are dominant in pH 2–3, and with pH between 4 and 9, various polymeric species such as $Al_{13}O_4(OH)_{24}^{7+}$ are formed and precipitated as $Al(OH)_3(s)$. As seen in Fig. 2a, the pH of the medium changes during electrocoagulation depending on, to some extent, the initial pH. For initial pH values between 4 and 9, the solution pH stabilizes very quickly at nearly constant value around 9 due to the buffering capacity of complex nature of aqua $Al^{3+}/Al(OH)_3$ system. When initial pH is higher than 9, the monomeric anion, $Al(OH)_4^-$ is formed at the expense of amphoteric $Al(OH)_3(s)$ according to the reaction (5) which causes a slight decrease in final pH by consuming OH⁻ ions.

$$Al(OH)_3 + OH^- \leftrightarrow Al(OH)_4^-$$
 (5)

Moreover, as $Al(OH)_4^-$ does not exhibit any decolorization capacity, a decrease is noted also in the removal efficiency above pH 9.



Fig. 3. Effect of conductivity (a) on the decolorization efficiency, (b) on the energy and electrode consumptions for Al electrode. (conditions: current density: 100 A/m^2 , Co: 250 mg/L, time: 15 min, initial pH: 6.4).

In conclusion, higher decolorization efficiency is obtained in acidic to neutral mediums, as the majority of aluminum complexes and the precipitates responsible for adsorptive decolorization are optimally formed in pH range 4–9.

Fig. 2b shows the electrode and energy consumptions in relation to initial pH values in the EC process. The electrode consumption values are 1.97–2.10 kg electrode//kg dye in the initial pH range 3–9, and 2.10–2.57 kg electrode//kg dye in pH 9–11, respectively. Values of the energy consumptions at pH 3–11 are 42.8–49.1 kWh/kg dye.

4.2. Effect of conductivity

The effect of conductivity was investigated between $250-4000 \,\mu$ S/cm by using NaCl as the supporting electrolyte. The decolorization efficiency and cell voltage decrease steadily as conductivity increases (Fig. 3a). The decrease in the decolorization efficiency may be attributed to a change in the ionic strength due to changing conductivity of the aqueous medium. Ionic strength affects the kinetics and equilibria of reactions like (3) and (5) between charged species occurring during EC [22,26].

Fig. 3b shows the effect of dye conductivity on the energy and electrode consumptions for Al electrode. The energy



Fig. 4. Effect of current density (a) on the decolorization efficiency, (b) on the energy and electrode consumptions for Al electrode. (conditions: Co: 250 mg/L, time: 15 min, initial pH: 6.4, conductivity: 750 μ S/cm).

consumption decreases from 102 to 13.3 kWh/kg dye with increasing conductivity. The electrode consumption varies from 1.93 to 2.20 kg electrode/kg dye in the conductivity range 250–4000 μ S/cm.

4.3. Effect of current density

Fig. 4a depicts the decolorization efficiency as a function of current density and charge loading. As the applied current density was increased from 100 to 250 A/m^2 , the decolorization efficiency did not change significantly. Minimum 100 A/m² is required for good decolorization efficiency (96%). This is ascribed to the fact that at high current densities, the extent of anodic dissolution of aluminum increases, resulting in a greater amount of precipitate for the removal of pollutants. Moreover, bubble generation rate increases, and the bubble size decreases with increasing current density, which are both beneficial for high pollutant removal efficiency by H₂ flotation [27].

The electrode consumption and the energy consumption increase from 0.18 to 0.75 kg electrode/kg dye and from 6 to 199 kWh/kg dye with increasing current density from 25 to 250 A/m^2 (Fig. 4b).



Fig. 5. Effect of time (a) on the decolorization efficiency, (b) on the energy and electrode consumptions for Al electrode material (conditions: current density: 100 A/m2; Co: 250 mg/L, initial pH: 6.4, conductivity: 750μ S/cm).

4.4. Effect of time

The effect of time was studied at constant current density of 100 A/m^2 and pH 6.4. According to Fig. 5a, 12 min of operating time is sufficient for nearly complete decolorization efficiency (92.3%). Decolorization efficiency remains almost constant above 14 min.

Both the energy and electrode consumptions increased almost steadily with increasing operating time (Fig. 5b). An increase in time from 5 to 20 min causes an increase in energy consumption from 22 to 53 kWh/kg dye, and an increase in electrode consumption from 1.1 to 2.6 kg Al/kg dye. This conclusion indicates that retention time is very important parameter as it affects the economic applicability of EC process.

4.5. Effect of initial dye concentration

The decolorization efficiency for Al electrode falls in a linear fashion from 99.6 to 88% with increase in concentration of the dye from 100 to 500 mg/l (Fig. 6a). This is likely due to the formation of insufficient number of aluminum hydroxide complexes produced by the electrode to coagulate the greater



Fig. 6. Effect of dye concentration (a) on the decolorization efficiency, (b) on the energy and electrode consumptions for Al electrode. (conditions: current density: 100 A/m^2 ; time: 15 min, initial pH: 6.4, conductivity: 750 μ S/cm).

number of dye molecules at higher concentrations. Therefore, it is quite clear that under the present experimental conditions, the lower is the dye concentration the better is the decolorization efficiency.

By increasing dye concentration, the values of both energy and electrode consumptions decrease from 98 to 23 kWh/kg dye and from 4.7 to 1.1 kg Al/kg dye, respectively (Fig. 6b).

5. Conclusion

The electrocoagulation process is successfully applied to remove the reactive textile dye from aqueous solution. The decolorization efficiency was found to be dependent on the initial pH, conductivity, the applied current density, the treatment time and the electrolyte concentration. Almost complete removal of pollutants (99.9%) was obtained with typical operating conditions; 100 A/m² current density, 15 min operating time and initial pH 3. The electrode and energy consumptions during the electrolysis were determined as 2.0 kg Al/kg dye and 43 kWh/kg dye, respectively. On the other hand, 95% decolorization efficiency may be obtained for the same current density in 12 min and at pH

6.4. The electrode and energy consumptions during the electrolysis were also found to be 1.8 kg Al/kg dye and 35 kWh/kg dye. The electrocoagulation setup described in this study is simple in design and operation and can be used as a convenient tool in the removal of textile dyes.

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