

The survey of electrocoagulation Process for removal dye Reactive Orange 16 from aqueous solutions using sacrificial iron electrodes

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ABSTRACT

Discharge of textile industries colored wastewaters without enough treatment into natural water supplies cause serious damages to the environment. This study was performed to investigate the effect of electrocoagulation for dye removal from synthetic wastewater using iron electrodes. Removal of dye reactive orange 16 (RO16) by electrocoagulation using iron electrode was conducted in a batch reactor with volume 1 liter. The effect of operating parameters such as current density, initial concentration of dye, pH and contact time was studied and the electrical energy consumption was calculated. The maximum efficiency of hardness removal which was obtained in current density of 20mA/cm², optimum concentration 50mg L⁻¹, optimum pH 5.5, reaction time of 30 min and NaCl concentration 1.5g/l are equal to 99.27%. Also COD removal efficiency is increased to 66%. Results show, electrocoagulation process by iron electrode is an effective method for reactive dye removal from colored wastewater.

Key words: Dye reactive orange 16 dye, Electrocoagulation, Decolourization, Iron electrode, Textile Wastewater

INTRODUCTION

Textile processes generate a large amount of wastewater which may include strong color, suspended particles, high pH and high chemical oxygen demand (COD) concentration. The discharge of these wastewaters into water supplies leads to serious pollution problems as well as endangering the lives of many aquatic wild lives [1]. The large quantity of organic colored wastewaters generated by textile, paper, plastic, leather, food, and mineral processing industries. Textile industry actually represents a range of industries with operation and processes as diverse as its products. It is expected that 1–15% of the dye is lost during dyeing and finishing processes and is released into wastewaters [2, 3]. Strong color of the effluents is especially troublesome because of its negative visual impact [4]. Dye bath effluents diffuse color to receiving streams and affect its aesthetic value. Color interferes with penetration of sunlight into waters, retards photosynthesis, inhibits the growth of aquatic biota and interferes with gas solubility in water bodies. Furthermore, dye effluent may include chemicals, which are toxic, carcinogenic, mutagenic, or teratogenicity in various micro-biologic, fish species [5].

Conventionally, biological, chemical, and physical methods have been performed for dye removal, but the biological techniques have not been very successful due to the non-biodegradable nature of most dyes [6]. There are many methods to remove dyes from colored effluents such as adsorption, precipitation, chemical degradation, photo degradation, biodegradation, chemical coagulation and electrocoagulation. When chemical coagulation is employed to treat colored wastewater, the pollution may be caused by chemical substance added at a high concentration [5, 6]. Many methods have been established for color removal from dye-containing wastewater. These contain adsorption (e.g. on active carbon), coagulation–flocculation, chemical oxidation (chlorination, ozonization, etc.) and photo degradation (UV/H₂O₂ or UV/TiO₂, etc.). Photo oxidation by UV/H₂O₂ or UV/TiO₂ and Fenton oxidation needs additional chemicals [5–11], and therefore leads to secondary pollution. The adsorption process is one of the useful methods applied to dye removal from aqueous solution.

Activated carbon is the most famous adsorbent for dye removal [12]. Electrocoagulation (EC) has been recommended in recent years as an appropriate method to various wastewater

treatments. Biodegradation process is cheaper than other methods; it is less effective because of the toxicity of dyes that has an inhibiting effect on the bacterial development [13, 14]. Hence, electrocoagulation (EC) as an electro chemical method was developed to overcome the drawbacks of conventional decolorization technologies. Electrocoagulation is an attractive option for the treatment of textile dyes [15–18].

EC process offered a simple, authentic and cost-effective method for wastewater treatment, without any need for additional chemicals, and thus the secondary pollution. It also decreases the amount of sludge, which needs to be disposed. The EC technique uses a direct current source between metal electrodes immersed in polluted water [2–6]. EC produces by anodic dissolution followed by hydrolysis, aluminum or iron hydroxide flocs which destabilize and aggregate the suspended particles or precipitates and absorb dissolved pollutants [19]. In recent years, the EC has been successfully tested to decolorization of dye-containing solutions. Applied electrodes in EC process are usually iron or aluminum [20]. The dye in colored wastewater is coagulated by iron (II) and aluminum hydrates or hydroxides produced from the sacrificial anode [16–19]. It should be noted that in water treatment processes which Fe^{3+} being used as a floc forming material, has significant benefits and it is harmless, compared to Al^{3+} which has some toxic effects (causing Alzheimer disease) [21]. EC technology, compared with other techniques, enjoys some advantages like plain equipment, easy functionality, short resistance time, no need of chemicals, low sludge production, sludge stability, suitable sedimentation of sludge, dewatering and environmental compatibility [22,23,24]. EC process is being used for the removal of ions, organic matters, colloidal and suspended particles, dyes, surfactants, oil and heavy metals from aqueous environments [21, 24].

In this study, the removal of a Reactive Orange16 (RO16) dye from a synthetic wastewater was investigated using iron electrodes and compared with available data from a previous work of the same authors on EC. Common operating parameters for EC were studied, such as the RO16 dye concentration in wastewater, pH, water conductivity, current density and contact time; the optimum values of these parameters for dye removal were determined.

MATERIALS AND METHODS

Dye solutions and Reagents

The dye used in the experiments was Reactive Orange 16 (RO16), which was provided by Alvan Sabet Company of Iran. The chemical structure and characteristics of this dye were given in Table 1. Sulfuric acid and sodium hydroxide used to adjust

the pH of solutions and sodium chloride used as supporting electrolyte, were purchased from the Merck Company (Germany). Stock solutions of RO16 were prepared by weighing the purified grade chemicals and dissolving them in deionized water without pH adjustment.

EC reactor

The batch experimental setup is schematically shown in Fig.1, which consists of an electro chemical reactor, a direct current (DC) power supply and iron electrodes. The bench scale (BS) unit was made of Plexiglas with reactor volume 1 L, which equipped with four pairs of electrodes, made of iron as anodes with the dimensions of 100 mm × 120 mm × 2 mm with effective area of 92 cm². The distance between the electrodes is 10 mm. Electrodes were connected to a digital DC power supply (model-YX-360TR-EB) in bipolar-parallel mode.

Experimental procedure

All experiments were performed at room temperature of 22±2°C. In each run, 1000 ml of dye solution was poured into the reactor. At the beginning of a run the specific concentration of dye in the aqueous solution was fed into the reactor and were adjusted the pH and conductivity. After each run, electrode surfaces were removed by dipping in 50cm³ of HCl solution for 1 minute and washed with deionized water to remove any solid residues on the surfaces, then dried and weighted. The sample was centrifuged (4000 rpm, 5 min).

Technical analysis

The COD of dye solutions was measured according to the standard methods for the examination of water and wastewater [25]. Dye concentrations were determined at maximum adsorption wavelength ($\lambda_{max} = 496$ nm), using a UV spectrophotometer (Shimadzu, Tokyo, Japan; Model 1601). The absorbance peak decreasing was directly proportional to the reduction of the dye concentration. The calculation of color removal efficiency after the EC process was performed using this formula:

$$CR(\%) = \frac{(C_0 - C)}{C_0} \times 100 \quad (1)$$

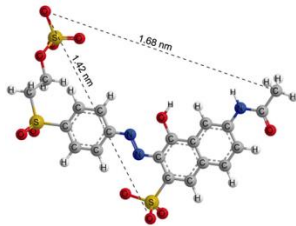
Where C_0 is the initial concentration of the dye ($mg L^{-1}$) and C is the final dye concentration ($mg L^{-1}$) after the EC process.

Electrical energy consumption was also calculated using the commonly used equation [26]:

$$E = \frac{UIt_{EC}}{V} \quad (2)$$

Where E is the electrical energy in kWh (m^3)⁻¹, U the cell voltage in volt (V), I the current in ampere (A), V is the volume solution (m^3) and t_{EC} is the of EC process per minute.

Table 1: Some characteristics of the investigated dye

Characteristic	Reactive (RO16)	Orange16
Structural		
Molecular formula	$C_{20}H_{17}N_3Na_2O_{11}S_3$	
Color index name	Reactive Orange16	
Molecular weight	601.54 g/mol	
λ_{max}	496 nm	

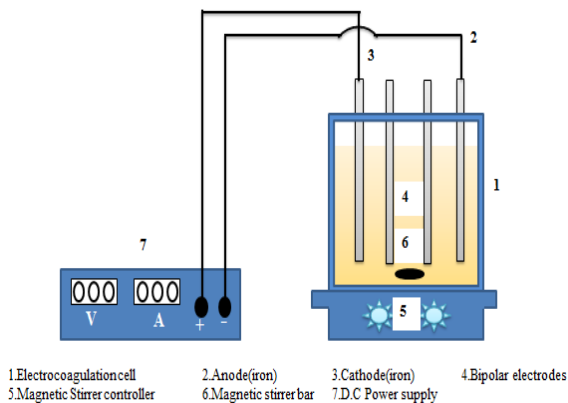


Fig. 1: EC reactor with bipolar electrodes in parallel connection

RESULTS

Effect of current density on the efficiency of dye removal

Fig. 2 shows the percent dye removal efficiency (RE %) during the electrocoagulation process, which initial dye concentration was 100 mg/L, KCl 1 gL⁻¹ and at pH 5.5. Experiments were carried out using various current densities (4, 10, 20 mA/cm²) by iron anode. As a consequence increases the amount of current density caused dye removal also increases.

Effect of initial dye concentration on the dye removal efficiency

Fig. 3 shows the effect of initial dye concentration on the dye removal efficiency in the range of 50-250 mg L⁻¹ and in optimized current density (20mA/cm²). It can be seen from the figure that the percentage dye removal decreases as the initial concentration of the dye increases.

Effect of NaCl concentration on the dye removal efficiency

Fig. 4 presents the effect of NaCl concentration under fixed current density (20mA/cm²) and dye concentration (mg L⁻¹). The increase in NaCl

concentration with the percentage dye removal increases.

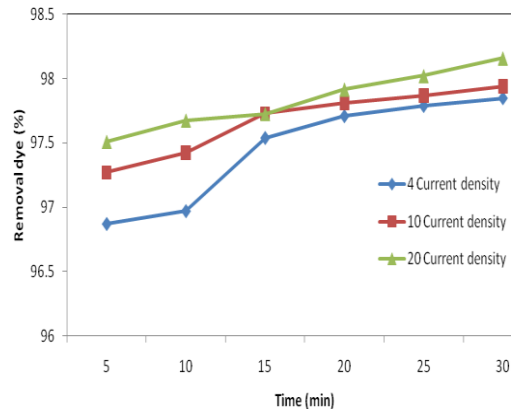


Fig. 2: Effect of current density on efficiency of dye removal ($C_0=100 \text{ mgL}^{-1}$, $\text{NaCl}=1 \text{ gL}^{-1}$, $d = 1 \text{ cm}$, $\text{pH}= 5.5$, $T= 22\pm 2^\circ\text{C}$)

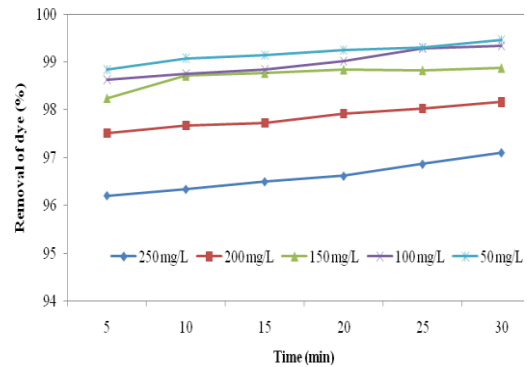


Fig. 3: Effect of dye concentration on efficiency of dye removal (current density=20mA/cm², $\text{NaCl}=1 \text{ gL}^{-1}$, $d = 1 \text{ cm}$, $\text{pH}= 5.5$, $T= 22\pm 2^\circ\text{C}$)

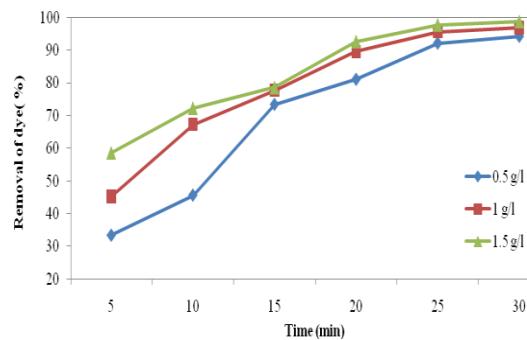


Fig.4: Effect of NaCl concentration on efficiency of dye removal ($C_0=50 \text{ mgL}^{-1}$, current density=20mA/cm², $d = 1 \text{ cm}$, $\text{pH}= 5.5$, $T= 22\pm 2^\circ\text{C}$)

Effect of initial pH on the dye removal efficiency

It has been established that pH is an important parameter influencing the performance of the EC process especially at low pH. This statement is confirmed by Fig. 5.

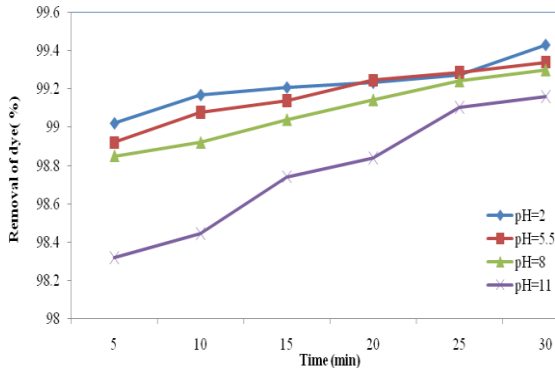


Fig. 5: Effect of initial pH on efficiency of dye removal ($C_0 = 50 \text{ mg L}^{-1}$, current density = 20 mA/cm^2 , $d = 1 \text{ cm}$, $\text{NaCl} = 1.5 \text{ gL}^{-1}$, $T = 22 \pm 2^\circ \text{C}$)

COD of the dye solution

The COD reduction percentages for COD solutions with the passage time were shown in Fig 6.

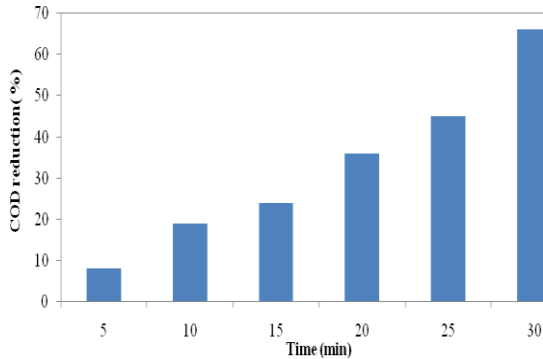


Fig.6: Effect of reaction time on COD reduction ($C_0 = 50 \text{ mg L}^{-1}$, current density = 20 mA/cm^2 , $d = 1 \text{ cm}$, $\text{NaCl} = 1.5 \text{ gL}^{-1}$, $\text{pH} = 5.5$, $T = 22 \pm 2^\circ \text{C}$)

Energy consumption

To achieve an optimum current density, both dye removal percent and energy consumption should be evaluated. Results showed that an increase in the current density and also increase dye concentration cause an increase consumption energy and increase in dye removal efficiency. Also the results showed that with increase NaCl concentration during the EC process and decrease in energy consumption (Figs. 7–10).

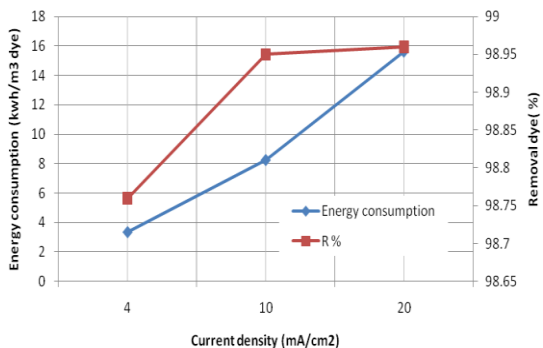


Fig.7: Effect of current density on efficiency of energy consumption and dye removal ($C_0 = 50 \text{ mg L}^{-1}$, $t = 25 \text{ min}$, $\text{pH} = 2$, $d = 1 \text{ cm}$, $\text{NaCl} = 1.5 \text{ gL}^{-1}$, $T = 22 \pm 2^\circ \text{C}$)

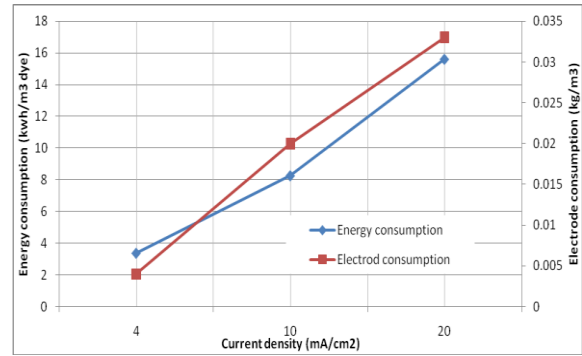


Fig.8: Effect of current density on efficiency of energy consumption and electrode consumption ($C_0 = 50 \text{ mg L}^{-1}$, $t = 25 \text{ min}$, $\text{pH} = 2$, $d = 1 \text{ cm}$, $\text{NaCl} = 1.5 \text{ gL}^{-1}$, $T = 22 \pm 2^\circ \text{C}$).

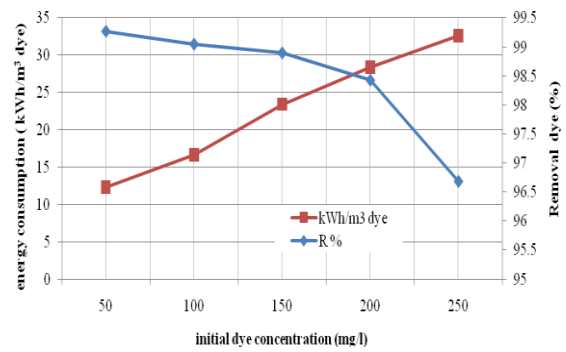


Fig.9: Effect of initial dye concentration on efficiency of dye removal and energy consumption (Current density = 20 mA/cm^2 ; $t = 25 \text{ min}$, $\text{pH} = 2$, $d = 1 \text{ cm}$, $\text{NaCl} = 1.5 \text{ gL}^{-1}$, $T = 22 \pm 2^\circ \text{C}$)

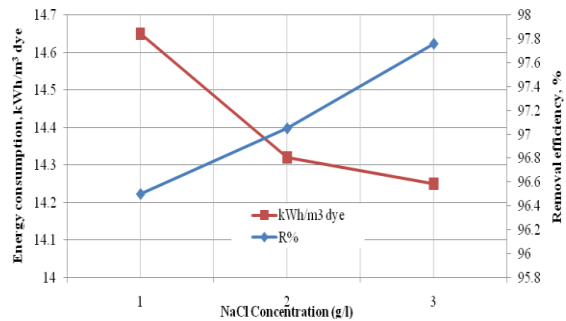


Fig.10: Effect of NaCl concentration on efficiency of dye removal and energy consumption ($C_0 = 50 \text{ mg/l}$, current density = 40 mA/cm^2 ; $t = 25 \text{ min}$, $d = 1 \text{ cm}$, $\text{pH} = 2$, $T = 22 \pm 2^\circ \text{C}$).

DISCUSSION

In all electrochemical processes, current density is the most important parameter for controlling the reaction rate within the reactor [27]. It is clear that, the rate of dye removal increases with increasing current density from 4 to 20 mA/cm^2 at the initial concentration of dye 100 mgL^{-1} . The color removal efficiency increased to 98.16% from 97.51% at 20 mA/cm^2 and for current density 10 mA/cm^2 from 97.27% to 97.94%. When current density increases, the amount of ion produced on the electrodes increases. The elimination efficiency shows that by

increase in the current density the elimination of dye increase. This process is related to the increase in the amount of absorbents (hydroxide iron ions) which are produced at site. Therefore, there is an increase in floc production and hence an improvement in the color removal efficiency [28, 29]. For a solution with a dye concentration of 100 mg L^{-1} , the optimum current density was 20 mA/cm^2 . Daneshvar's investigation showed that with increasing the initial current density in EC process, the C.I. Acid Yellow 23 removal efficiency was increased [30]. Also These results are in line with the results from Yilmaz *et al.* in separating out of warm subsurface waters by electrocoagulation and Ghosh *et al.* in eliminating iron (II) from tap water by using electrocoagulation techniques [31,32].

As the results indicated the rate of dye removal decrease with the increase in initial dye concentration. Results showed the flocs produced at a high dye concentration were sufficient to adsorb all of the dye molecules of the solution [18]. It is clear that higher concentrations of pollutant require longer time for complete removal, but for initial high dye concentration compared to low initial concentration a relatively short time was needed for complete removal. This can be explained by diluting solution theory. In dilute solutions, the formation of distributing layer around the electrode causes lower reaction speed but in concentrated solutions, distributing layer has no effect on distribution speed or immigration of metallic ions toward electrode surfaces [33]. When iron is used as an electrode material, removal efficiencies have reduced by increasing initial dye concentration. Because of this, reaction rates have decreased by increasing initial dye concentration and removal efficiencies are mainly due to pH using iron electrodes [34]. Phalakornkule *et al.* presented similar results for Electrocoagulation of blue reactive, red disperse and mixed dyes, and application in treating textile effluent; also Ghosh *et al.* had the same results for Decolorization of Crystal Violet Solution by Electrocoagulation [18,26,30].

The problem of electrode passivation is a serious shortcoming in the current application of electrocoagulation. To destroy the passivation layer and hence enhance the dissolution rate of Al or Fe electrodes is thus crucial. The addition of Cl^- to acidic and neutral solutions was to increase the anodic dissolution rate of Fe, either by the incorporation of Cl^- to the oxide film or by the participation of Cl^- in the metal dissolution reaction [35]. Hence, adding NaCl to the wastewater is probably a better choice for increasing the performance of the electrocoagulation technology. Also added salt also increases water conductivity and, consequently, decreases simultaneously the ohmic loss between

electrodes and power requirements [14]. It is clear that increasing chloride concentration from 0.5 to 1.5 gL^{-1} increases the efficiency dye removal. This may be explained by the fact that the higher Cl^- concentration the higher ability of Cl^- to destroy any passive oxide film which tends to form on iron anodes at relatively high potential and limit iron dissolution [30]. In addition to the coagulation process, an indirect electrochemical oxidation likely occurs if the solution contains Cl^- . The Cl^- will be discharged at the anode to generate Cl_2 , which will be immediately dissolved in the solution, chemically converted to ClO^- . The ClO^- can oxidize the pollutants effectively. Therefore the removal efficiency of COD will increase. However, the removal efficiency decreased when more NaCl was added to the solution [34]. The result of Ayhan Sengil's studies showed that high color removal percentage with low cell current densities and low energy consumption can be obtained in dye solutions with NaCl of around 3 g^{-1} [36].

The EC process is highly dependent on the pH of the solution. In order to evaluate the effect of pH of the solution on the percentage dye removal, a series of experiments were conducted and the dye solution was adjusted to the desired pH for each experiment by using sodium hydroxide or hydrochloric acid solutions. The maximum dye removal efficiency was observed from 99.02% to 99.43% at pH 2. Therefore, it can be concluded that at pH 2, the majority of iron complexes (coagulants) is formed and it is the optimum pH for carrying out the electrocoagulation. In the other to, the adsorption capacity of iron flocs toward the dye molecules decreases. In surface compound, it is assumed that the dye molecule can act as a ligand to bind a hydrous iron moiety with precipitation and adsorption mechanisms [10, 37, 38]. Also Dye removal efficiency decreased in higher pH values since hydroxide ions were oxidized at the anode. In addition, $\text{Fe}(\text{OH})_6^{3-}$ and $\text{Fe}(\text{OH})_4^{4-}$ ions may be present at high pH, which lacks a removing capacity [21]. Therefore the kinetics of Fe^{2+} conversion to Fe^{3+} is strongly affected by the pH; the surface charge of the coagulating particle also varies with pH [39].

For a study of final pH was growing during the time. According to previous studies that was examined by Moroak *et al.* about removal of disperse red dye from synthetic wastewater, the results showed that by increasing pH, the dye removal efficiency was decreased. Optimum pH was obtained 5.5. In other research by Phalakornkule *et al.* on removal of blue reactive, red disperse and mixed dyes, the finding proved that during the time, the final pH solution had enhanced [16,18]. Then, subsequent to decolorization in the optimized conditions (current density = 20 mA/cm^2 , $C_0 = 50 \text{ mgL}^{-1}$, pH=2,

NaCl=1.5 g l^{-1}) the COD of the treated solutions was measured again. On the other hand, for an electrolysis time of 5 min, COD removal efficiency of EC is 8% and for time of 30 min, COD removal efficiency is 66%. The results revealed that when the electrolysis time was 30 min, there was a maximum COD reduction percentage. Time of electrocoagulation is the most important parameter in the process of electrocoagulation. It is the time provided to the process to generate metal hydroxides and to complete coagulation of the impurities [28, 30]. The pervious studies' results were proved this fact that an increase in contact time can enhance COD reduction from solution [40].

Electrical energy consumption and removal efficiency of dye are very important economically parameters in EC process. The value of energy consumption as a function of treated solution volume was also calculated in different current density for each dye solution [41, 42]. The results showed that an increase in the current density causes an increase in color removal efficiency also increases energy consumption due to polarization and increase in the number of iron compounds [32, 38]. Energy consumption ranges from 3.36 to 15.61 kWh/m^3 dye removed depending on the operating conditions for electrochemical electrocoagulation. The same research results was determined which an enhancement in current density, can increase the removal efficiency and consumption energy [18, 26, 43].

CONCLUSIONS

The treatment of Reactive Orange 16 dye in a synthetic wastewater using EC cell described in the present experiment. The effect of various operational parameters on color removal efficiency was investigated and optimized. Results showed that applying optimum current density of 20mA/cm^2 could remove a high percentage of color from dye solutions. The dye removal efficiency for RO16 was 99.27% at the current density 20mA/cm^2 and when iron was used under the conditions of initial pH 2. The COD removal efficiency is increased from 8% at time of 5 min to 66% min at 30 min. Consequently energy consumption estimation had to be accountable for this, which is rarely the case in the literature where current efficiency is assumed to be constant. The EC process is promising with respect to the treatment of wastewaters containing azo dyes.

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