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Electrocoagulation of Reactive Orange 16 Textile Dye Solution Using Steel, Aluminum, and Copper Metal Plates as Electrodes

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Abstract—This study presented the comparison of the electrocoagulation efficiencies for steel, aluminum, and copper electrodes for the chemical oxygen demand (COD) removal and decolorization from solutions comprising the reactive orange 16. The electrodes were used in similar couples in this research, namely, Fe (anode) to Fe (cathode), Al (anode) to Al (cathode), and Cu (anode) to Cu (cathode). The samples were analyzed using spectrophotometric techniques UV/VIS and a colorimetric method. Higher removal efficiencies for color and COD were found at the current density of 0.025 A/cm^2, i.e. Fe/Al/Cu electrodes gave 91, 62, and 51% in the COD removal and 93, 82, and 70% for color removal. Other parameters taken such as pH, electrolysis time, and the initial azo dye concentrations were also optimized at all of the electrodes. The decolorization and COD removal efficiencies were found decreased with increasing the initial dye concentration. Being simple and cost effective, this technique can be applicable for the treatment of real textile wastewater.

Keywords: reactive orange 16, electrochemical techniques, copper, steel, aluminum, textile

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INTRODUCTION

The toxicity, mutagenicity, and carcinogenicity of dyes have long been recognized. The textile wastewater containing dyes and other products may have toxic and mutagenic effects [1–4]. The wastewater discharge of textile industries into the water bodies can cause severe environmental and health problems [5–7]. Generally, the textile wastewater has intensive color, high pH and chemical oxygen demand (COD) values, and low biodegradability [8–11]. Textile industries wastewater discharge is dense colored, and therefore it is difficult for light to penetrate through it [12–15]. Color removal of such wastewater is arduous by using conventional wastewater treatment processes [16–19]. Most of the synthetic dyes are highly toxic and resistant to biodegradation because of their complex chemical structures [20–23].

In literature, various physical, chemical and biological treatments methods presented were used to treat dye or textile wastewater [24–29]. Physical treatment processes incline to transfer of pollutants to a different media, and biological methods are time consuming and low efficient [29–33]. Chemical methods are exciting and often effective in removing color; such techniques are considered to be advanced oxidation processes, e.g., ultrasound based ones, UV radiation, application of ozone and hydrogen peroxide, the Fenton processes, and photo-catalytic treatments [33–38]. Currently, electrochemical techniques have provoked a growing interest in the treatment of persistent organic pollutants and of textile wastewater [39–48]. Electrochemical degradation processes using reactive orange 16 (RO 16) present attractive advantages (Table 1). The azo dye RO 16 has striking dying characteristics, particularly for cotton and silk stuff. Though, it is very resilient to conventional wastewater treatment techniques.

This study presented the results of the investigations of the degradation and decolorization by the electrocoagulation using steel, aluminum, and copper electrodes. The effects of such variables as the current density, pH, the dye concentration, and electrolysis time were also studied.

MATERIALS AND METHODS

RO 16 Aqueous Solution Preparation

In this research, a stock solution of 1000 ppm of RO 16 was prepared by dissolving 1000 mg of RO 16 in 1 L of distilled water. Dilutions of 5, 10, 15, and

Table 1. Recent electrochemical studies for degradation of RO 16

Process	Electrodes	RO 16 Concentration	Efficiency	Ref.
Electrochemical oxidation	Platinum	0.5–10 mM	40%	[49]
Anodic oxidation	Platinum and boron–doped diamond	0.2–1.2 mM	80%	[50]
Electrochemical degradation	Charcoal-based metallic composite	200 mg/L	98.5% max	[51]
Electro-peroxone process	Gas diffusion electrode, graphite felt electrode	2.335×10^{-4} mol L^{-1}	99%	[52]
Microbial fuel cell	Manganese coated cathode	0.5 mM	98%	[53]
decolorization	Ti/PtOx anode	10–30 ppm	–	[54]
Electrochemical decolorization	Charcoal-based metallic composite	200 mg/L	83.3%	[55]
Continuous electrochemical process	Stainless steel	100–400 mg L^{-1}	20–80%	[56]
Electrochemical flow-cell	Platinum	35 mg L^{-1}	57 and 93%	[57]
Anodic oxidation	Boron-doped diamond	50 mg L^{-1}	40–60%	[58]
Electrochemical oxidation	Boron-doped diamond			[59]
Electrochemical decolorization and COD removal	Charcoalgraphite-tin	200 mg/L	80%	[60]
Electrochemical oxidation	$B_2H_6/CH_4/H_2$ with BDD	100 mg/L	45.6% COD removal	[61]
Electrocoagulation	Iron	50 mg/L	99% color removal	[62]

20 ppm were prepared from the stock solution. All of the required dilutions were prepared with distilled water.

Electrodes Types

Steel (Fe), aluminum (Al), and copper (Cu) electrode sheets were used for the electrocoagulation processes to study the decolorization and COD removal from synthetic wastewater containing RO 16. Dimensions of a single metal sheet for each electrode (Fe, Al, and Cu) are shown in Fig. 1. The thickness of each metal sheet was 1 mm. Standard electrode potentials for Al, Cu, and Fe were –1.66, 0.34, and –0.44 V, respectively, as mentioned elsewhere. Three electrode couples were used in the study: Al (anode) to Al (cathode), Cu (anode) to Cu (cathode), and Fe (anode) to Fe (cathode). The total active area of a single sheet electrode submerged into the solution was 11.6 cm^2. Three current densities were selected (0.013, 0.017, and 0.025 A/cm^2) to study their effect on the decolorization and COD removal.

The Electrolytic Cell

The couples of electrodes mentioned above were separately hanged into the reaction vessels. Each reaction vessel had 100 mL solution. Each couple of elec-

Fig. 1. Electrodes sheets with dimensions of the submerged surface area inside the dye solution in electrolytic cell.

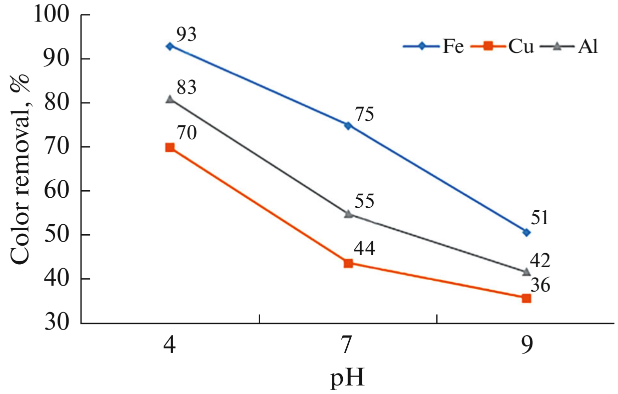

Fig. 2. Effect of initial pH on decolorization of RO 16 (current density 0.025 A/cm^2, dye concentration 5 ppm, and electrolysis time 15 min).

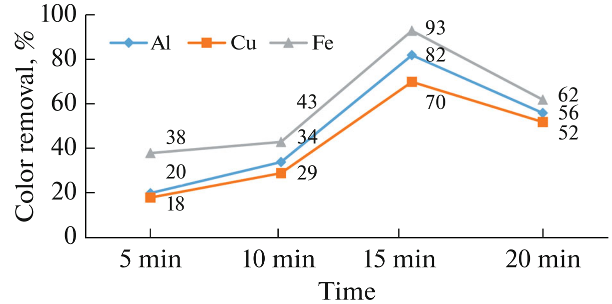

Fig. 3. Effect of electrolysis time on decolorization of RO 16 (current density 0.025 A/cm^2, RO 16 concentration 5 ppm, and pH 4).

trodes was connected with a power supply (PS-305D DAZHENG) for the continuous supply of current.

The pH Adjustment

The initial pH values of the RO 16 dye solutions were set to 4, 7, and 9, by adding molar solutions of H_2SO_4 and NaOH. The experiments were done in triplicates.

Electrolyte

In this study, Na_2SO_4 electrolyte was used by dissolving 1g per liter in each dye solution.

Analysis Details

A double beam spectrophotometer was used to determine the color analysis at a wavelength of 493 nm (λ_{max}). After electrocoagulation, it was necessary to wait for a complete settling of the flocks. COD was determined by a colorimetric method. The results obtained from the color and COD analyses were used to calculate the percentage of the removal efficiency.

Statistical Analyses

All of the statistical analyses were performed using MS-Excel. A two-way ANOVA was applied to determine the significance of the electrodes.

RESULTS AND DISCUSSION

Effect of pH on Decolorization

The result in Fig. 2 shows the efficiency of the electrocoagulation process for the decolorization of RO 16 solution against the difference in the initial pH (4, 7, and 9). By using Al electrodes, the maximum decolorization values were 82, 55, and 42%, respectively. While by using steel electrodes, 93, 75, and 51% decolorization obtained at pH 4, 7, and 9, respectively. The Cu electrodes gave 70, 44, and 36% decolorization at pH 4, 7, and 9, respectively. Other variables (i.e. the current density, the dye concentration, and electrolysis time) were set constant at 0.025 A/cm^2, 5 ppm, and 15 min. For the electrocoagulation process, as stated elsewhere, the pH of a liquid medium had a significant role in the separation process as its affect was double, i.e. making metal hydroxides and their solubility throughout electrocoagulation [63].

Effect of Time of Electrolysis on Decolorization

Figure 3 shows the effect of electrolysis time on decolorization of RO 16 solution at Al, Fe, and Cu electrodes. The maximum decolorization was obtained at 15 min electrolysis with all three electrodes. The Fe electrode showed 93% decolonization, which was the highest among electrodes. Similar results had been obtained in an earlier study by the authors for the reactive blue 2 dye [7]. The decolorization efficiency started to reduce after 15 and 20 min at all three electrodes. It was due to the formation of dense flocs that produced the turbidity in the RO 16 solution. While using Al electrodes, the decolorization efficiency of 20, 43, 82, and 56% was attained after 5, 10, 15, and 20 min, respectively. When using Cu electrodes, the decolorization efficiency of 18, 29, 70, and 52% was obtained after 5, 10, 15, and 20 min, respectively. From the obtained it can be concluded that the optimum time of electrolysis was 15 min. When the electrolysis time was 20 min, the percentage of decolorization efficiency started to decrease due to the formed denser flocks, which increased the turbidity. Thus the electrolysis time affected the treatment efficiency of the electrolytic procedure. Throughout electrolysis, anodic electro-dissolution led to the discharge of coagulating species. The decolorization efficiency directly depended on the concentration of metal ions formed on the electrodes.

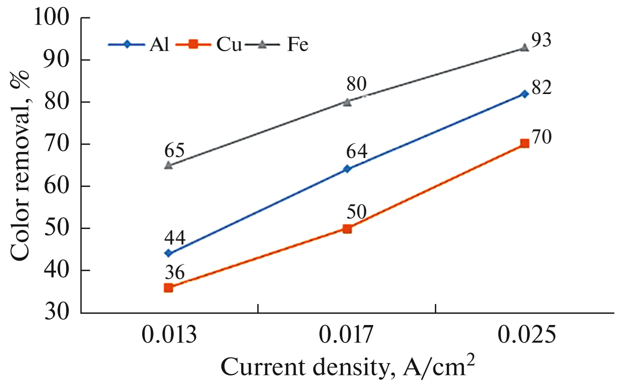

Fig. 4. Effect of current densities on decolorization of RO 16 (electrolysis time 15 min, RO 16 concentration 5 ppm, and pH 4).

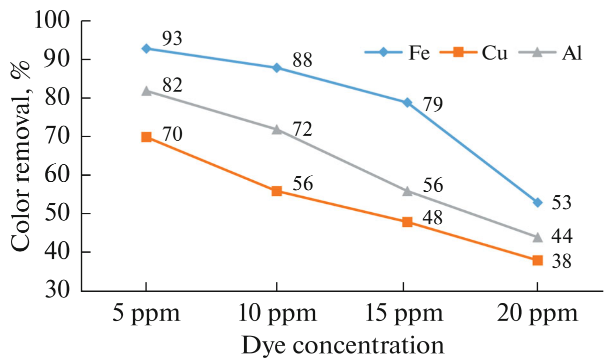

Fig. 5. Effect of initial dye concentration on decolorization of RO 16 solution (current density 0.025 A/cm^2, electrolysis time 15 min, and pH 4).

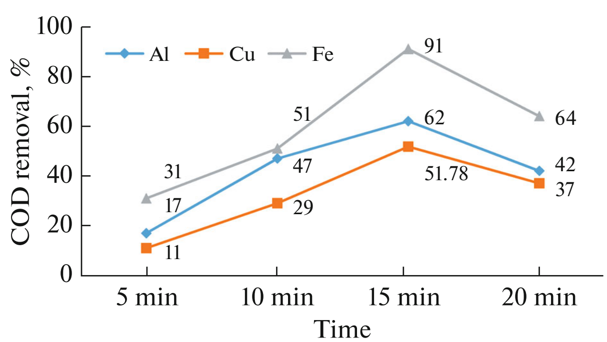

Fig. 6. Effect of electrolysis time on removal of COD in RO 16 solution (current density 0.025 A/cm^2, pH 4, and concentration 5 ppm).

Effect of Current Density on Decolorization

To study the effect of the current density (A/cm^2) on the decolorization efficiency, three different current densities of 0.013, 0.017, and 0.025 A/cm^2 were chosen (see Fig. 4). By using Fe electrodes, decolonization increased at increasing the current density as 65, 80, and 93% at 0.013, 0.017, and 0.025 A/cm^2, respectively. While using Al electrodes, the decolorization efficiencies were 44, 64, and 82% obtained at the current density of 0.013, 0.017, and 0.025 A/cm^2, respectively. With Cu electrodes, the removal efficiency increased from 36, to 50, and to 70% by increasing the current density as 0.013, 0.017, and 0.025 A/cm^2. A complete color removal could be obtained by a further increase in the current density.

Effect of Initial RO 16 Concentrations

Figure 5 shows the initial dye concentration effect on the decolorization of RO 16 the by electrocoagulation process. Four RO-16 dye concentrations (5, 10, 15, and 20 ppm) were subjected to 15 min reaction time at the current density of 0.0.025 A/cm^2 and pH 4. The results indicated that the RO 16 dye decolorization efficiency decreased by increasing the dye concentration. The decolorization at Fe electrodes gradually went down from 93 to 53% at 5 to 20 ppm of the RO 16 concentration. The same trend was observed for Al and Cu electrodes, but with different results. It was because of the number of flocs formed that were inadequate to absorb all color molecules, which also affected the COD removal.

COD Analysis

Effect of electrolysis time. Figure 6 shows the effect of electrolysis time at the current density 0.3 A/cm^2 and pH 4. Increasing the electrolysis time yields the COD removal from 51 to 91%. The optimum time of electrolysis is in between 5–15 min. When the electrolysis time increased, it was noticed that more dense flocks produce the turbidity in the RO 16 solution. So, the reaction time influences the treatment efficiency of the electrolytic process. By using Fe electrodes, the COD removal rose from 31 to 51% and from 91 to 64% at 5, 10, 15, and 20 min, respectively. Electrocoagulation was a two-step process: destabilization and aggregation. The first stage is usually short, whereas the second is typically long. The electrochemical processes create metal ions which function as destabilizes at the anode. When the electrolysis time is reduced, it results in a low charge loading, and the metal ion (Fe^{3+}) dose is inadequate to destabilize all colloidal and finely dispersed particles. The optimum efficiency of the electrochemical process was achieved at the treatment duration of 15 min, and increasing the treatment time did not result in a substantial increase in the removal efficiency of the examined parameters.

Effect of dye concentration. The RO 16 initial dye concentrations (5, 10, 15, and 20 ppm) were optimized at 0.025 A/cm^2, 15 min, and pH 4. Figure 7 shows that as the RO 16 dye concentration increased from 5 to 20 ppm, the COD removal for each electrode

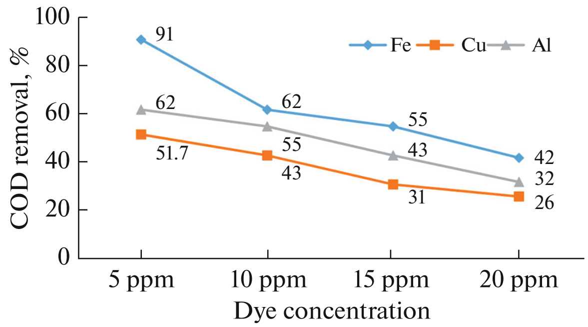

Fig. 7. Effect of initial dye concentration on COD removal of RO 16 solution (pH 4, current density 0.025 A/cm^2, and electrolysis time 15 min).

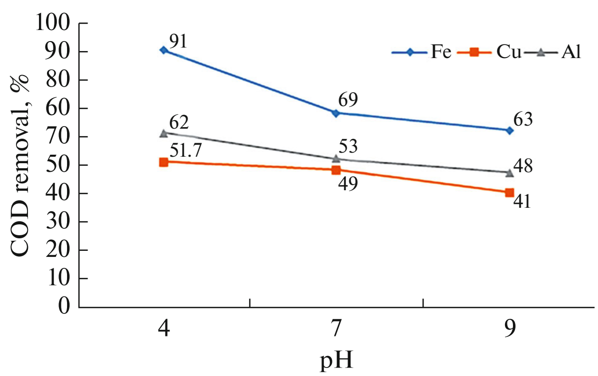

Fig. 8. Effect of initial pH on COD removal of RO 16 solution (current density 0.025 A/cm^2, dye concentration 5 ppm, and electrolysis time 15 min).

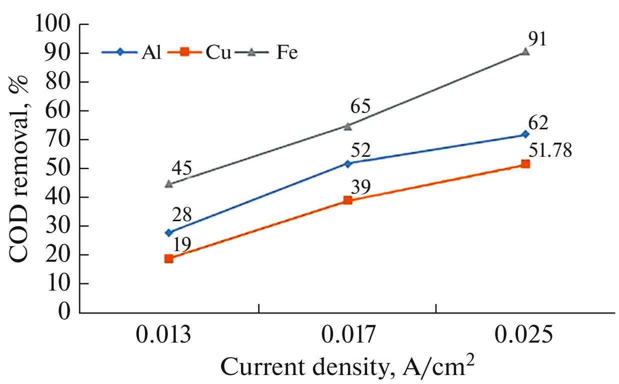

Fig. 9. Effect of current density on COD removal of RO 16 solution (electrolysis time 15 min, pH 4, and dye concentration 5 ppm).

decreased. When using Fe electrodes, the COD removal efficiency of 91 and to 42% were obtained at 5 and 20 ppm, respectively. When using Cu and Al electrodes, the maximum removal efficiency was achieved at 5 ppm while the minimum COD efficiency was achieved at 20 ppm.

Effect of pH. A comparative research was carried out at different pH levels of 4, 7, and 9 to better understand the influence of pH in the remaining variables (the dye concentration, the current density, and electrolysis time) which were held constant at 5 ppm, 0.025 A/cm^2, and 15 min, respectively. Figure 8 shows the effect of initial pH values (4, 7, and 9) on the removal of RO 16 during electrocoagulation process. By using Fe electrodes, the COD removal efficiencies of 91, 69, and 63% were obtained at pH 4, 7, and 9, respectively. At Al electrodes, 51, 49, and 41% of the COD removal efficiencies were obtained at 4, 7, and 9 pH, respectively. When using Cu electrodes, the COD removal efficiencies of 56, 53, and 48% were obtained at pH 4, 7, and 9 respectively. Variations of efficiencies among each electrode type impact the production of metal hydroxides and their solubility during the electrocoagulation process, the pH of the liquid medium also has a significant impact on the separation process [63].

Effect of current density on COD removal. To study the effect of the current density on the COD removal, three current densities of 0.013, 0.017, and 0.0250 A/cm^2 were chosen. Figure 9 shows that a higher current density increases the COD removal efficiency. The removal percentage increased from 45 to 65% at 0.017 A/cm^2 and to 91% at 0.0250 A/cm^2 for Fe electrodes. For Al electrodes, the removal percentage increased from 28% at 0.013 A/cm^2 to 52% at 0.0.017 A/cm^2, and to 56% at 0.025 A/cm^2. For Cu electrodes, the removal percentage increased from 19% at 0.013 A/cm^2 to 39% at 0.017 A/cm^2, and to 51% at 0.0250 A/cm^2 – the maximum value of the current load on the system, taking into account that the speed of the electrochemical process increases with increasing the current density. It is possible that an increase in the current load would lead to a greater process efficiency. The rate of the coagulant generation is determined by the current density, which effects the pollutant treatment by the electrocoagulation process [64].

Effect of electrodes. The effect of electrodes was explained with the help of ANOVA (Table 2). Results

Table 2. Effect of electrode material on removal of COD of RO 16 using steel, Cu, and Al via ANOVA

Source of variation	SS	df	MS	F_{obs}	P-value	F_{crit}
Between groups	1274	2	637	50.28947	0.000178	5.143253
Within groups	76	6	12.66667			
Total	1350	8				

in Table 2 reveal that electrodes types or materials significantly increased the removal efficiency.

CONCLUSIONS

Using an electrocoagulation process for color and COD removal of RO 16 dye solution, the effect of electrode material (Fe, Al, and Cu) on the removal efficiency was studied by varying operating parameters (current density, pH, electrolysis time and dye concentration). Best color and COD removal was obtained at pH 4, current density 0.025 A/cm^2, and electrolysis time 15 min for 5 ppm concentration. Fe shown higher removal efficiency among Al and Cu electrodes.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

AUTHOR CONTRIBUTIONS

All authors have accepted responsibility for the entire content of this manuscript and approved its submission.

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