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Article

Effects of Operational Parameters on the Removal of Acid Blue 25 Dye from Aqueous Solutions by Electrocoagulation

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Abstract

Influence of several experimental parameters (e.g., initial dye concentration, pH, distance between electrodes, applied voltage, electrical conductivity, current density, and reaction time) on the performance of electrocoagulation (EC) process for the removal of acid blue 25 (AB25) was studied. A bipolar batch reactor was used to test the impact of the parameters. The removal efficiency (RE) of AB25 dye was promoted by increasing the contact time, voltage, electrical conductivity, and applied current density. In contrast, RE of AB25 decreased with higher level of AB25 and the longer distance between electrodes. The removal efficiency increased consistently until pH 7, but decreased above pH 7. The maximum efficiency of AB25 removal above 90% was obtained at a voltage of 60 V, reaction time of 90 min, distance between electrodes of 0.5 cm, initial concentration of 25 mg/L, conductivity of 3,000 μS/cm and pH of 7. These results imply that the high RE of AB25 dye from the aqueous solution can be achieved by EC process.

Keywords: *Electrocoagulation, Acid blue 25, Aluminum electrodes*

1. Introduction

Dyes normally exist in wastewaters from dyeing industrial complex which is the remarkable major source for color in its effluents[1-4]. Over 100,000 numbers of commercially available dyes have been produced[5]. The volume of water used in the textile industries has increased due to the expansion of the textile industries[6]. According to an estimate, about 40~65 L of wastewater is generated to produce 1 kg fabric[7]. Thus, considerable amount of wastewater including dye is directly discharged into streams and rivers through aqueous effluent[5].

Dye is major organic compounds which increase environmental issue[3,4]. The effluents containing dyes include chemicals and suspended solids which could be toxic materials[6]. The toxic materials can harm the aesthetic environment and can be toxic to aquatic life[2]. For instance, the dyes in the water can change the photosynthetic activity due to declining transparency of the water, resulting in affecting aquatic life[8]. Furthermore, the dyes could be carcinogenic and mutagenic; so, the dyes can damage internal organs, brain, and central nervous system[9].

The chemicals in the dyes are harmful for humans as well as for the aquatic environment. Therefore, dyes should not be discharged into rivers or any water bodies directly. It is difficult to remove dye from wastewater by conventional methods including chemical coagulation and flocculation, adsorption, ozonation, and fungal decolonization [10,11]. Electrochemical technique is alternative when removal of pollutants by chemical coagulation becomes difficult or impossible. Electrochemical technique such as electrocoagulation (EC) has several advantages compared to the other conventional methods. Firstly, supplementary equipment of EC is relatively cheaper compared to the other techniques. Second, there are small production of total dissolved solids and harmful substances during removal process[12,13]. Third, colloidal particles are also removed as dyes are eliminated[12,13]. Finally, EC reduces the amount of sludge which should be disposed [14].

There are more strong points described somewhere[13]. Generally, aluminum (Al) and iron (Fe) are used for electrode materials during EC process. When Al electrode is used, the electrolytic reactions cause metal dissolution and water reduction during EC[15-17]:

$$\text{Anode: } Al \rightarrow Al^{+3} + 3e^{-} \quad (1)$$

$$\text{Cathode: } 3H_2O + 3e^{-} \rightarrow \frac{3}{2}H_{2(g)} + 3OH^{-} \quad (2)$$

Al^{3+} and OH^- ions are generated by equation (1) and (2), and these ions react to form final products, $Al(OH)_3(s)$, by following reaction:

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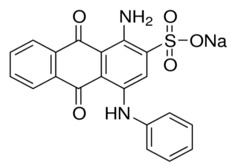

Figure 1. Molecular structure of AB25 dye.

$$Al^{+3} + 3H_2O \rightarrow Al(OH)_3(s) + 3H^+ \quad (3)$$

$Al(OH)_3(s)$ exists as a gelatinous suspension which can eliminate dye from wastewater by coagulation[17]. Sedimentation or electroflotation can remove the flocs from aqueous solution[18,19]. There are some factors that regulates the removal efficiency (RE) during dye removal process using the EC techniques. Initial dye concentration, pH, current density, applied voltage, distance between electrodes, conductivity and reaction time are ones of the examples. It has been reported that EC process had provided high RE for various dyes from wastewater[12,13].

Acid Blue 25 (AB25) has been used for dyeing wool, silk, and mixed fabric in dyeing industry which is the reason there have been many studies to remove AB25 from wastewater. However, usually adsorption has been used as removal mechanism for AB25, suggesting that there is lack of information about removal of AB25 by EC. Therefore, the objective of this study is to investigate various operating conditions to achieve higher removal capacity using EC. This study could allow us to design better dye removal system in the field.

2. Experimental

2.1. Materials

All of the chemicals were used without further purification in this study. The high pure AB25 ($C_{20}H_{13}N_2NaO_5S$, molecular weight: 416.38) was purchased from Alvan Co. (Hamedan, Iran), and the chemical structure is displayed in Figure 1. Hydrochloric acid (HCl) (0.1 mol/L) and sodium hydroxide (NaOH) (0.1 mol/L) were used to adjust pH of the solutions and purchased from Merck Co. (Darmstadt, Germany).

2.2. Experiments

The experiments were conducted using a bipolar batch reactor (2 L of volume), with four Al electrodes with 0.2 cm × 8 cm × 12 cm installed parallel to each other (Figure 2). The Al electrodes were placed in 2 cm distance, having a monopole electrode configuration. The electronic which can adjust the voltage from 0 to 60 V was supplied to the EC reactor. The 1.5 L of wastewater containing AB25 dye was introduced to the reactor and mixed by a magnetic stirrer. The wastewater of the reactor was settled for 45 minutes after each experiment, and then the dye concentrations were determined by UV-visible spectrophotometer (Shimadzu UV-2101PC) at the λ_{max} value, which is 602 nm for AB25.

RE was calculated by following equation:

$$RE\ (\%) = (C_i - C_o) / C_i \times 100 \quad (4)$$

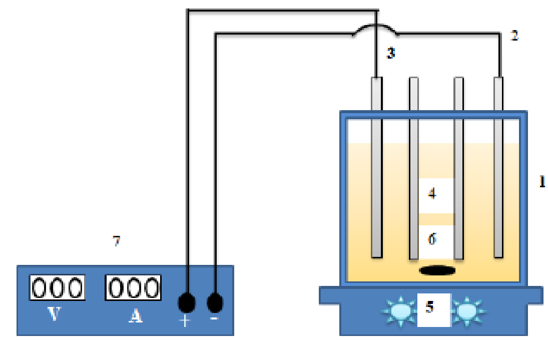

Figure 2. Bench-scale EC reactor with bipolar electrodes in parallel connection (1. EC reactor, 2. anode, 3. cathode, 4. bipolar electrodes, 5. magnetic stirrer, 6. DC power supply).

Where C_i and C_o stand for AB25 dye concentrations pre and post removal, respectively.

The electrodes were cleaned with deionized water to remove solid particles on the surface of the electrodes when each test was over. The effect of electrochemical factors (e.g., initial dye concentration, pH, electrical conductivity, voltage, distance between electrodes, current density and electrolysis time) on the AB25 dye RE were investigated with two replicates. The range of each condition was indicated in Table 1.

3. Results and Discussion

3.1. Effect of reaction time and initial dye concentrations

Reaction time is one of the important parameters which affects the performance of EC process[20]. Besides the reaction time and AB25 dye concentrations, the other conditions were fixed: current density of 3 mA/cm^2, distance between electrodes of 2 cm, voltage of 40 V, conductivity of 2,500 μS/cm, and pH 7. The range of the operating conditions and RE with the conditions are indicated in Table 1.

The highest REs were reached after 90 min of the reaction time and maintained until 120 minutes (Figure 3). These trends occurred at all different AB25 dye solutions. It was reported that production rate of hydroxyl and metal ions on the electrodes increased with reaction time[19,20]. However, there was no increase observed in the RE after 90 min of the reaction time. Therefore, the most favorable condition is 90 min and our results were similar with previous studies[19,20]. Taheri *et al.* (2015) also reported that RE by EC increased with the increase of reaction time and reaction time was one of the most important parameter to remove acid azo dyes. Larue *et al.* (2003) also concluded that removal of the organic matter by EC was improved when the reaction time was increased from 10 to 60 minutes.

The RE decreased as the initial AB25 dye concentrations were increased (Figure 3). It is known that amounts of coagulant are not changed even though initial dye concentrations were increased[16]. Thus, the coagulation capacity of sweep flocs decreases, and because of this reduction of dye RE occurs[16]. A similar result was previously reported[21,22]. For instance, besides AB25 dye removal, orange II removal was affected with the increased initial dye concentration[21].

Table 1. Removal Efficiency with the Operating Conditions (Dye Concentration: mg/L, Reaction Time: min, Voltage: V, Distance between Electrodes: cm, Current Density: mA/cm^2, Electrical Conductivity: μS/cm)

Dye concentration	Time	RE (%)	Voltage	Time	RE (%)	Distance	RE(%)
25	15	45.9	10	15	26.2	0.5	95.2
	30	60.8		30	40.7	1.0	92.4
	45	67.9		45	51.3	1.5	88.5
	60	81.3		60	58.3	2.0	86.1
	75	89.5		75	63.1	2.5	80.2
	90	95.5		90	69.0	3.0	71.5
	105	95.8		105	69.4		
	120	95.9		120	69.8		
50	15	40.3	20	15	29.9	Current density	RE (%)
	30	51.8		30	44.4	1	72.3
	45	64.3		45	55.1	2	78.3
	60	75.4		60	66.7	3	85.2
	75	81.4		75	72.5	4	89.2
	90	89.8		90	78.4	5	91.8
	105	89.9		105	78.9	6	93.2
	120	90.1		120	79.2		
100	15	34.2	40	15	34.2	pH	RE (%)
	30	47.4		30	49.4	3	82.2
	45	55.5		45	59.5	5	93.4
	60	69.3		60	69.3	7	98.2
	75	77.2		75	77.2	9	79.4
	90	86.1		90	86.1		
	105	86.3		105	86.3		
	120	86.3		120	86.3		
150	15	31.4	60	15	41.9	Conductivity	RE (%)
	30	45.2		30	57.8	500	70.7
	45	54.4		45	65.9	1000	78.8
	60	66.2		60	79.3	1500	86.5
	75	75.4		75	89.5	2000	91.3
	90	80.1		90	93.5	2500	93.5
	105	80.3		105	93.8	3000	95.7
	120	80.4		120	94.1		
200	15	30.2					
	30	41.2					
	45	48.6					
	60	62.2					
	75	68.4					
	90	70.4					
	105	70.6					
	120	71.7					

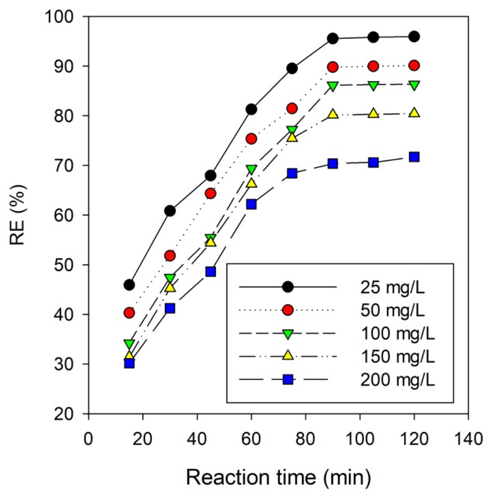

Figure 3. The RE of AB25 dye with reaction time and dye concentrations (distance between electrodes: 2 cm; voltage: 40 V, pH 7; conductivity: 2,500 μS/cm).

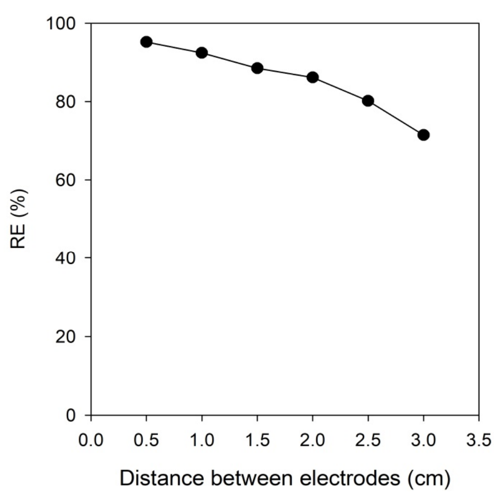

Figure 4. Influence of electrodes distance on RE of AB25 dye (voltage: 60 V, pH 7; conductivity: 2,500 μS/cm; reaction time: 90 min; dye concentration: 100 mg/L).

Mohammad *et al.* (2004) reported that the RE decreased by 34% when the initial orange II dye concentration increased from 10 to 100 mg/L.

3.2. Influence of the distance between electrodes

Usually, it is known that a closer distance between electrodes is advantageous for EC because the electrical resistance increases with inter electrode distance. To test influence of the distance between electrodes on AB25 dye removal, the other conditions were fixed: voltage of 60 V, pH 7, conductivity of 2,500 μS/cm, and AB25 dye concentration of 100 mg/L. Figure 4 shows that a decrease in removal of AB25 dye was observed when the distance between electrodes was increased. Similar phenomena were described previously[12,23].

Due to electrostatic effects of electrodes distance, dye removal decreases with the distance between electrodes[24]. That is, if distance between electrodes increases, the movement of Al^{+3} hydroxides can be slow down, and resulting in the reduction of mixing with the dye molecules and coagulant.

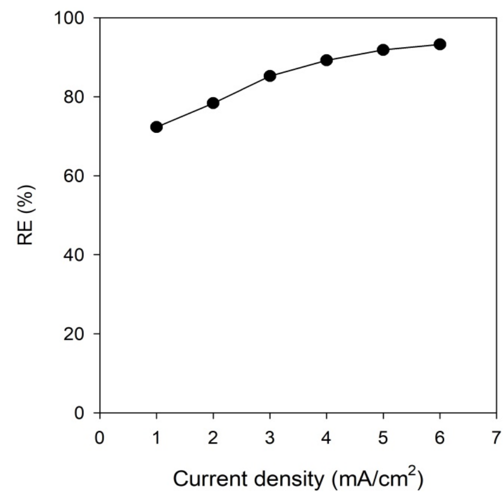

Figure 5. The effect of current density on AB25 dye RE (voltage: 60 V, pH 7; conductivity: 2,500 μS/cm; reaction time: 90 min; dye concentration: 100 mg/L; distance between electrodes: 2 cm).

3.3. Influence of current density

Current density can play an important role in controlling electrochemical rate[12]. Besides current density, the other conditions were fixed: voltage of 60 V, distance between electrodes of 2 cm, conductivity of 2,500 μS/cm, AB25 dye concentration of 100 mg/L, and pH 7. Figure 5 displays the effect of current density on AB25 dye removal, suggesting that the RE increased with the current density. Other studies also reported a similar trend[12,23,25,26]. Increase in current density can increase the amount of Al oxide and coagulation of the matrix; therefore, removal of dye can be improved[27,28].

3.4. Effect of initial pH

It is known that pH is a significant parameter affecting dye removal by EC[12,23,25,29,30]. pH can influence the ionic characteristic of Al hydroxides and the dye molecule in wastewater and hence affect dye RE[25]. To determine the effect of pH on AB25 dye removal, the other conditions were kept fixed: voltage of 60 V, distance between electrodes of 2 cm, conductivity of 2,500 μS/cm, reaction time of 90 min, and AB25 dye concentration of 25 mg/L.

The RE was highest at pH 7, and the RE was decreased at pH 9 (Figure 6). Many investigators have also reported that RE is found the highest at neutral pH[12,23,25,31,32]. At acidic, neutral, and basic conditions, $Al(OH)^{2+}$, $Al(OH)_3$, $Al(OH)^{4-}$ are the dominant species in the solutions, respectively[23,33-35]. As mentioned above, $Al(OH)_3$ has large surface areas which is available to adsorb dye molecule. This is because of the highest removal of AB25 dye which was observed at neutral pH in many studies including this study.

In contrast at low pH, $Al(OH)^{2+}$ and Al^{3+} were found dominated and are not beneficial in the dye removal[31]. Likewise, at high pH, the Al hydroxide is oxidized at anode; therefore, $Al(OH)_3$ is not pro-

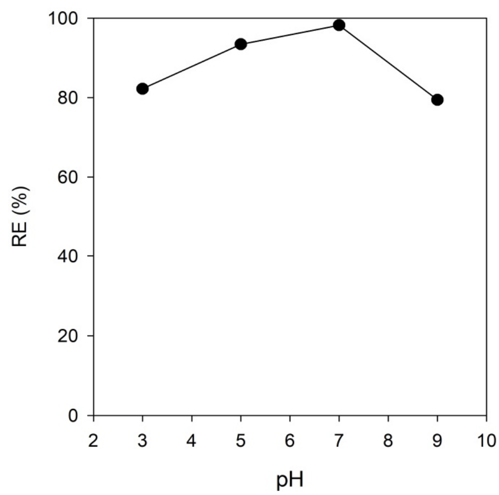

Figure 6. Effect of pH on the AB25 RE (voltage: 60 V, pH 7; conductivity: 2,500 μS/cm; reaction time: 90 min; dye concentration: 25 mg/L; distance between electrodes: 2 cm).

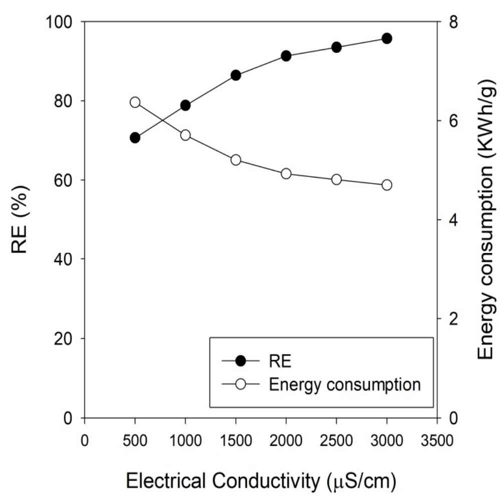

Figure 7. Effect of conductivity on RE of AB25 dye and energy consumption (voltage: 60 V, pH 7; conductivity: 2,500 μS/cm; reaction time: 90 min; dye concentration: 100 mg/L; distance between electrodes: 2 cm).

duced[23]. That is why AB25 dye REs was declined under alkaline pH[33,34].

3.5. Effect of electrical conductivity

To test the effect of electrical conductivity on AB25 dye removal, the other conditions were fixed: voltage of 60 V, distance between electrodes of 2 cm, reaction time of 90 min, and AB25 dye concentration of 100 mg/L. The RE of AB25 dye increased with electrical conductivity of the reactor (Figure 7). Our results also showed that decrease in the energy consumption was correlated to increase in electrical conductivity (Figure 7). For instance, when electrical conductivity increased from 500 to 3,000 μS/cm, energy consumption was reduced from 6.4 to 4.7 KWh/g (Table 1). The current flow increases with electrical conductivity of the solution during EC process. As a result of increase in the current flow, dye removal performance was enhanced.

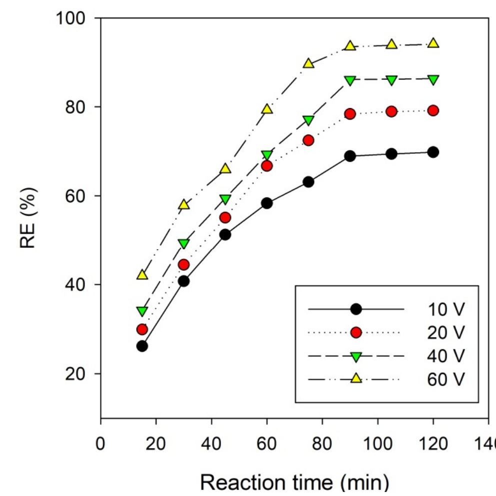

Figure 8. The RE of AB25 dye with voltage (distance between electrodes: 2 cm, pH 7; conductivity: 2,500 μS/cm; dye concentration: 100 mg/L).

Increase in electrical conductivity results in decreasing in the cell voltage at fixed current density, declining the power consumption in electrolytic cells[25]. Our result was similar with previous reports [32,36,37]. For instance, Bazrafshan *et al.* (2012) highlighted that increase in the electrical conductivity by addition of chloride potassium significantly enhanced the RE of humic acid because the formation of coagulator materials and the size of flocs was increased[36]. The study of Chafi *et al.* (2011) is another example to represent positive correlation between electrical conductivity and dye removal. It was also showed that energy consumption was decreased with the increase in electrical conductivity.

3.6. Effect of voltage

Besides voltage, the other conditions were fixed: current density of 3 mA/cm^2, distance between electrodes of 2 cm, pH 7, conductivity of 2,500 μS/cm, and AB25 dye concentration of 100 mg/L. The applied voltage ranged from 10 to 60 V to study influence of voltage on AB25 dye removal from the wastewater. AB25 dye removal increased with the increased voltage (Figure 8). This could be ascribed to the increased amount of metal hydroxide at high current which leads to greater amount of precipitates and removal of AB25 dye molecules[35].

4. Conclusion

Our results showed that the efficiency of the system can be promoted by increasing the contact time, voltage, electrical conductivity, and the applied current density. However, the efficiency of EC process decreased when higher level of AB25 ions and longer distance between electrodes were presented in the aqueous phase. In contrast, a consistent increase appeared until there was a prominent decrease after the high at pH 7. Based on our results, the maximum efficiency of AB25 removal was obtained with voltage of 60 V, reaction time of 90 min, distance between electrodes of 0.5 cm, initial concentration of 25 mg/L, conductivity of 3,000 μS/cm and pH 7. Under these conditions, the RE surpassed 90%. Therefore, these results showed that EC process can effectively reduce the dye contaminant at a very low level.

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