

*Original Article****Textile Wastewater Treatment by Electrocoagulation Process Using Aluminum Electrodes***Edris Bazrafshan¹ Amir Hossein Mahvi² ***Mohammad Ali Zazouli**³

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Abstract

Background and purpose: Textile industries are among the most polluting industries regarding the volume and the complexity of treatment of its effluents discharge. This study investigated the efficiency of electrocoagulation process using aluminum electrodes in basic red 18 dye removal from aqueous solutions.

Materials and Methods: This study was performed in a bipolar batch reactor with six aluminum electrodes connected in parallel. Several important parameters, such as initial pH of solution, initial dye concentration, applied voltage; conductivity and reaction time were studied in an attempt to achieve higher removal efficiency.

Results: The electrochemical technique showed satisfactory dye removal efficiency and reliable performance in treating of basic red 18. The maximum efficiency of dye removal which was obtained in voltage of 50 V, reaction time of 60 min, initial concentration 50 mg/L, conductivity 3000 μ S/cm and pH 7 was equal to 97.7%. Dye removal efficiency was increased accordance to increase of applied voltage and in contrast electrode and energy consumption was increased simultaneously.

Conclusion: As a conclusion, the method was found to be highly efficient and relatively fast compared to conventional existing techniques for dye removal from aqueous solutions.

[Bazrafshan E. Mahvi A. *Zazouli MA. *Textile Wastewater Treatment by Electrocoagulation Process Using Aluminum Electrodes*. 2014; 2(1):16-29] <http://jhs.mazums.ac.ir>

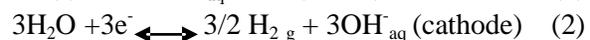
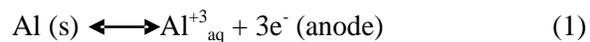
Key words: Textile Wastewater, Electrocoagulation, Aluminum Electrodes, Basic Red 18

1. Introduction

Textile industries usually use large amount of water and various chemicals for finishing and dyeing processes. Dye wastewater typically consists of a number of contaminants including acids, bases, dissolved and suspended solids, toxic and non-biodegradable compounds, which are noticeable even at low concentrations and need to be removed before the wastewater can be discharged to environment (1,2). Presently, there are several processes available for the removal of dyes by conventional treatment technologies, such as chemical coagulation and adsorption, biological methods (anaerobic reduction and aerobic oxidation), advanced oxidation processes, such as ozonation and UV/H₂O₂ have been employed so far in order to effectively purify dyeing effluents; decolorization being among the main objectives to achieve (3), but each of mentioned above methods have some limitations and problems. Biological treatment by activated sludge offers high efficiencies in COD removal, but does not completely eliminate the color of the wastewater and frequently operational problems such as bulking appear. Chemical oxidation by ozone, or a combination of UV-radiation and ozone and H₂O₂, has great interest, but the costs are still very high due to the high doses required (4). Coagulation–flocculation process has been found to be cost effective, easy to operate and energy saving treatment alternatives, but the coagulation process does not work well for soluble dyes. Furthermore, in coagulation process, large amount of sludge is created which may become a pollutant itself and

increase the treatment cost. The electrocoagulation (EC) technique is considered to be potentially an effective tool for treatment of textile wastewaters with high removal efficiency. In fact, this process is an alternative of the conventional coagulation process in which coagulant agents are generated in situ through the dissolution of a sacrificial anode by applying current between the anode-cathode electrodes. The electrocoagulation process has several advantages that make it attractive for treating various contaminated streams. This technique has been applied successfully for the treatment of many kinds of wastewater such as landfill leachate, restaurant wastewater, textile wastewater, petroleum refinery wastewater, dairy wastewater, slaughterhouse wastewater, tannery wastewater, carwash wastewater and for removal of fluoride, humic acid, phenol, pesticides and heavy metals from aqueous environments (5-19).

An examination of the chemical reactions occurring in the electrocoagulation process shows that the main reactions occurring at the aluminum electrodes are:



Al³⁺ and OH⁻ ions generated by electrode reactions (1) and (2) react to form various monomeric species, which finally transform into Al(OH)_{3(s)} according to complex precipitation kinetics:



Freshly formed amorphous Al(OH)_{3(s)} “sweep flocs” exhibit large surface areas which are beneficial for a rapid adsorption of soluble organic compounds and for trapping colloidal particles. Finally, these flocs are removed easily from aqueous

medium by sedimentation or by flotation induced by the H₂ bubbles generated at the cathode (6,20), which is referred to as electroflotation. The main purpose of this work is to study of the electrocoagulation process efficiency for basic red 18 (BR-18) removals as an anionic dye from aqueous environments with aluminum electrodes and determination of the important variables

such as electrical potential, initial pH, dye concentration, electrode distance, electrical conductivity and reaction time on the removal efficiency. Since BR-18 has an azo group band (-N=N-), then it is in the group of azo dyes. The molecular structure of BR-18 is shown in Fig. 1. This dye is soluble in water (Table 1).

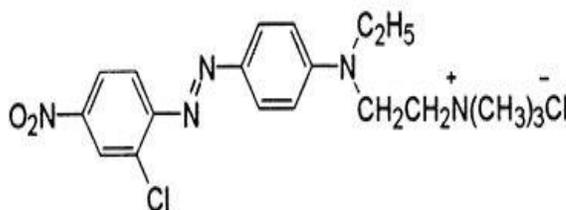


Fig 1. Chemical structure of the Basic red 18

Table 1. Some characteristics of the investigated dye

Characteristic	Basic red 18 (BR-18)
Molecular formula	C ₁₉ H ₂₅ ClN ₅ O ₂
Color index name	Basic Red-18
Molecular weight	390.887 g/mol
H-Bond Donor	0
H-Bond Acceptor	5
λ_{\max}	530 nm
Class	Monoazo (-N=N- bond)

2. Materials and methods

The BR18 dye was made with high purity of dye and was applied without further purification. The dye stock solution (1000 mg/L) was prepared by dissolving 1g of basic red 18 (BR18) in 1000 mL deionized water. Desired concentrations of dye solutions (25, 50, 75, 100, 150, 200 mg/L) were prepared by diluting proper amount of stock solution with deionized water.

All other chemicals including sodium hydroxide (NaOH), concentrated sulfuric acid (H₂SO₄) and potassium chloride (KCl) were used as analytical grade (Merck Company). In order to survey the effect of electrical conductivity of the solution on dye removal efficiency, the experiments were performed at various values of conductivity (1000, 1500, 2000, 2500, 3000, 3500, 4000 μ S/Cm). Also, the pH of initial solution was adjusted to a desired value (3, 5, 7, 9, 11) using H₂SO₄ and

NaOH solutions (0.1 M). The electrochemical system used in this study was of bench-scale. Experiments were performed in a bipolar batch reactor (Fig. 2), with six aluminum electrode connected in parallel. Electrodes were made from aluminum plates with dimensions of 11 mm×14 mm×2 mm. The total submerged surface area of each electrode was 792 cm². The volume (V) of the solution of each batch was 1 L. A power supply pack having an input of 220V and variable output of 0–60V (10, 20, 30, 40, 50 and 60 V for this study) with maximum current of 5 ampere was used as direct current source. The pH values in influent and reactor unit were measured using E520 pH meter (Metrohm Herisau, Switzerland Ultra Basic, U.S.). A Jenway Conductivity Meter (Model 4200) was employed to determine the conductivity of the solution. Different samples of 25ml were taken at 10 min intervals for up to 60 min and analysed to determine the residual of dye. The residual dye concentration in the solutions was analyzed by using UV-VIS spectrophotometer (Shimadzu, Tokyo, Japan; Model 1601) at wavelength 530nm. During the runs, the reactor unit was stirred at 120 rpm by a magnetic stirrer to allow the

chemical precipitate to grow large enough for removal. Before and at the end of each run, the electrodes were washed thoroughly with water, dipped in HCl solution (5% v/v) for at least 20 min and rinsed again with distilled water. Typical runs were conducted at 23±2°C. Dye removal efficiency was calculated as:

$$E, (\%) = \frac{(C_i - C_f)}{C_i} \times 100 \quad (4)$$

Where C_i is the initial dye concentration (mg/L) and C_f is the final dye concentration (mg/L). In this study, each treatment was repeated twice and the absorbance measurement of each sample was repeated three times and all of the data in the Figures were the average ones. The standard errors were all within 10% of the mean values. Electrical energy consumption and current efficiency are very important economical parameters in EC process that calculated in this study. Electrical energy consumption was calculated using the Eq. (5):

$$E = UI t \quad (5)$$

Where E is the electrical energy in Wh, U the cell voltage in volt (V), I the current in ampere (A) and t is the time of EC process per hour.

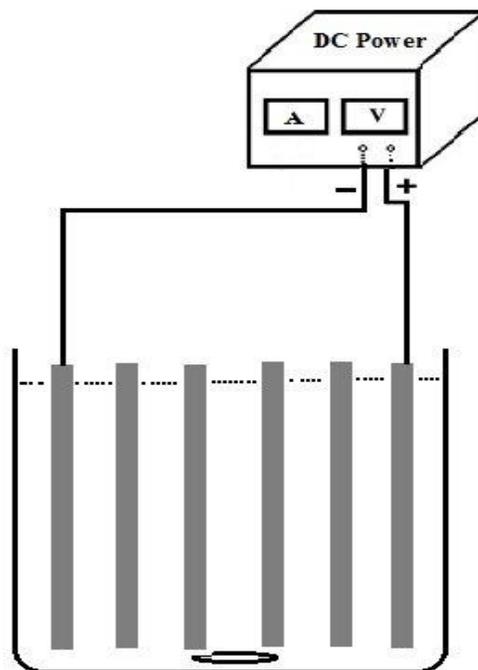


Fig 2. The schematic view of electrochemical reactor

3. Results

3.1. Effect of applied voltage on BR-18 dye removal efficiency

In all electrochemical processes applied voltage is the most important parameter for controlling the reaction rate within the electrochemical reactor (21). It is well known that this variable determines the production rate of coagulant, adjusts also bubble production, and hence affects the growth of formed flocs. To investigate the effect of applied voltage on BR-18 removal efficiency, electrocoagulation process was carried out using various applied voltage at fixed initial concentration of dye (50 mg/l) with pH value 7 ± 0.3 for 60 min operation. Fig. 3 shows that the time required

achieving steady-state conditions decreased when applied voltage increased from 10 to 60 V and then became nearly constant at about 40 min. In fact, as the applied voltage was increased, the required time for the electrocoagulation process decreased. On the other hand, for a given time, the removal efficiency increases significantly with an increase in applied voltages. After 60 min of electrolysis, it can be seen from Fig. 3 that 46.16%, 65.32%, 73.89%, 83.24%, 97.68% and 98.28% of the BR-18 were removed for applied voltages of 10 V, 20 V, 30 V, 40 V, 50 V and 60 V, respectively. In addition, an increase in applied voltage from 10 to 60 V yielded an increase in the efficiency of dye removal

from 46 to 98%. This could be expected: when the voltage increases, the amount of Al^{3+} cations released by the anode and therefore of $\text{Al}(\text{OH})_3$ particles also increases. On the other hand, this is ascribed to the fact that at high voltages, 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 applied voltage, which are both beneficial for high pollutant removal efficiency by H_2 flotation (22). Similar results were reported by other researchers (23). Energy consumption is a very important economical parameter in the electrocoagulation process. Therefore, for the same operating conditions, after 60 min of electrocoagulation, consumption of energy and electrode material was calculated using the related equations (24). Fig. 4 shows the effect of applied voltage on the energy and electrode consumptions. It can be seen from Fig. 4 that electrical

energy and electrode consumption were found to increase with increasing the applied voltage as would be expected in any other electrolytic process. An increase in applied voltage from 10 to 60 V causes an increase in energy consumption from 12.3 to 87.8 kWh/m^3 of dye which is in agreement with other studies. Also, an increase in applied voltage from 10 to 60 V causes an increase in electrode consumption from 0.14 to 0.56 kg/m^3 of BR-18 dye. When the applied voltage was increased from 40 V to 60 V, the removal efficiency of dye increased considerably, from 83.24% to 98.28%, whereas the corresponding specific energy consumption increased only slightly. Therefore, for the experimental conditions of Fig. 3 and 4, optimum applied voltage seemed to be between 50 and 60 V, as it can be considered that maximum efficiency 97-98% corresponds to the minimum acceptable value for EC process. Consequently, voltage 50 V was retained for further experiments.

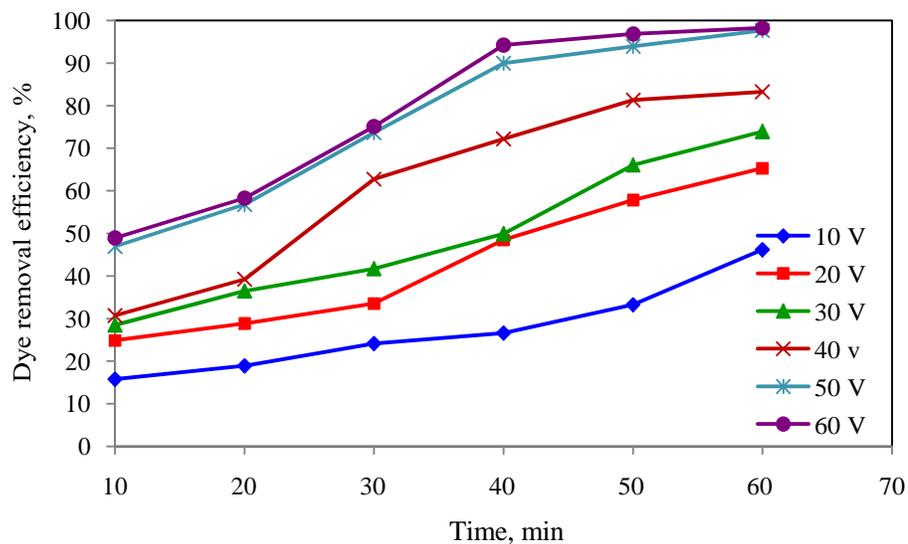


Fig 3. Effect of applied voltage on the decolorization efficiency (initial dye concentration = 50 mg/L, pH ~ 7, conductivity ~ 3000 μ S/cm)

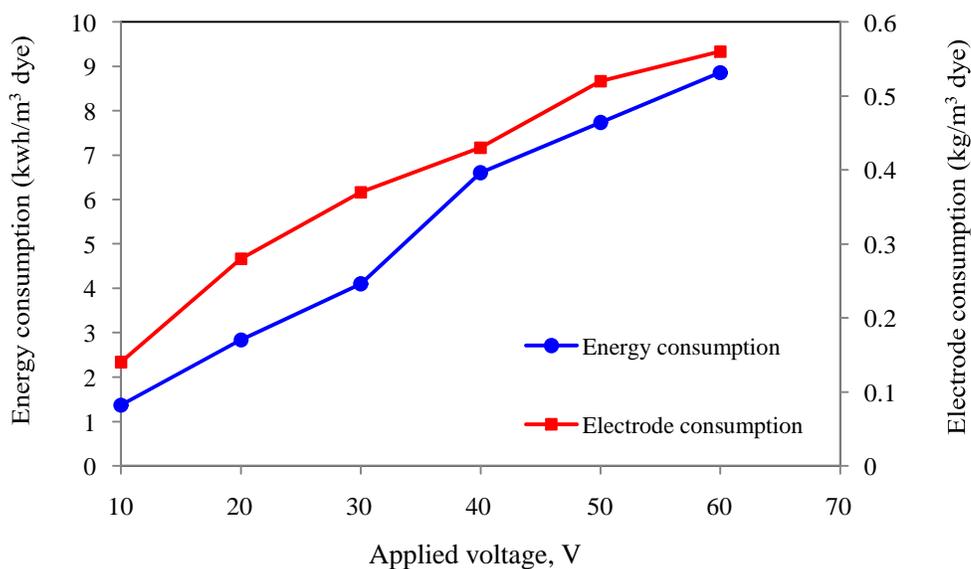


Fig 4. Energy consumption during electrocoagulation process (initial dye concentration = 50 mg/L, pH ~ 7, conductivity ~ 3000 μ S/cm)

3.2. Effect of initial pH on the BR-18 dye removal efficiency

Currently, it has been established that the pH is a critical operating parameter influencing the performance of electrocoagulation process (20,25,26). Hence, for examination the effect of initial pH of solution on the performance of process, the synthetic samples were adjusted to the desired value (3, 5, 7, 9, 11) for each experiment by adding either sodium hydroxide or sulfuric acid. As shown in Fig. 5. The efficiency of BR-18 removal was low either at low pH or at high pH. On the other hand, the experimental results showed that when pH of the dye solution was between 5 and 9, there was maximum color removal efficiency. The optimal pH was 7 (removal efficiency ~ 98%), at which higher dye removal efficiency could be reached. Also, according to the results (Fig. 5) the average BR-18 dye removal at reaction time 60 min increased from 84.67% to 98.43% when the pH was increased from 3 to 7. Further increasing the pH to 9 and 11 resulted in a reduction of dye removal efficiency to 97.38 and 87.91%, respectively. This behavior was ascribed to the amphoteric

character of aluminum hydroxide which does not precipitate at very low pH (27). Furthermore, high pH leads to the formation of Al(OH)_4^- , which is soluble and useless for adsorption of dye (28). Therefore, more increase of the initial pH would decrease the BR-18 dye removal efficiency. In addition, as shown in Fig. 6, the pH of the solution changes during the process. As observed by other researchers, a pH increase occurs when the initial pH is low (<7). Vik et al. ascribed this increase to H_2 evolution at cathodes (29). In addition, if the initial pH is acidic, reactions would shift towards which causes an increase in pH. In alkaline condition (pH > 8), the final pH does not vary very much and a slight drop was recorded. This result is in agreement with previously published studies (8,16). However, when the solution pH is above 9, a pH decrease occurs, therefore electrocoagulation process can act as pH buffer. At present study, the initial pH did not affect the removal efficiencies significantly over a wide range. Therefore, adjustment of initial pH before treatment is not required in practical applications.

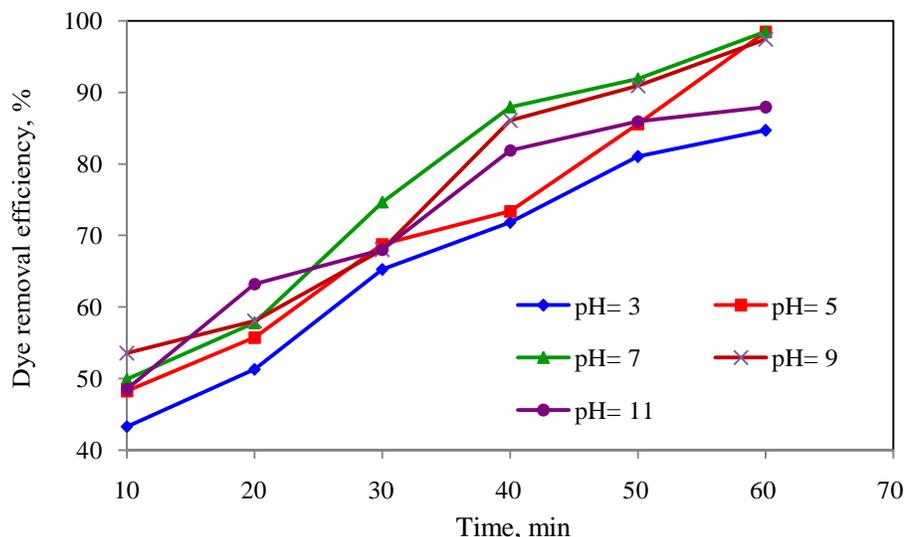


Fig 5. Effect of initial pH on the decolorization efficiency (Initial dye concentration = 50 mg/L, applied voltage = 50 V, conductivity ~ 3000 μ S/cm)

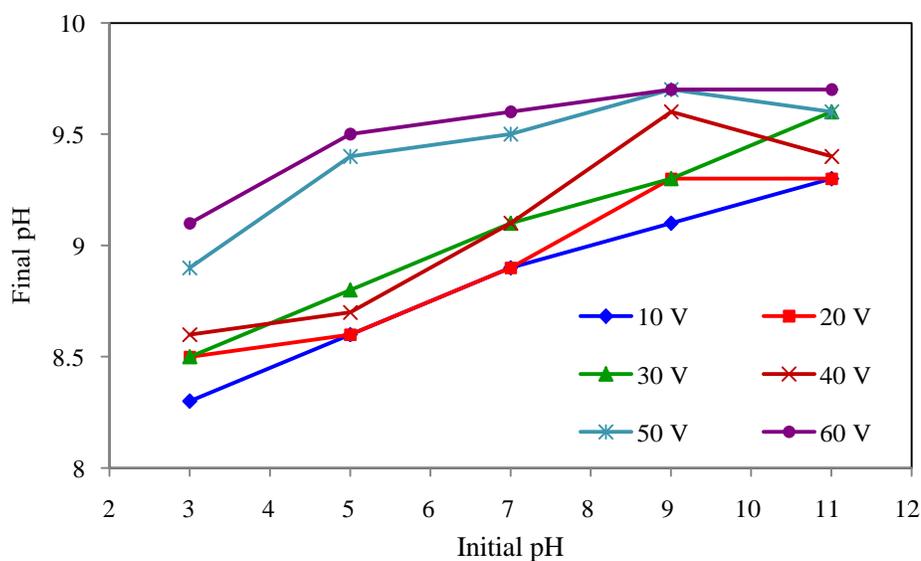


Fig 6. The variation of pH during the electrocoagulation process (Reaction time = 60 min, Initial dye concentration = 50 mg/L, conductivity ~ 3000 μ S/cm)

3.3. Effect of BR-18 dye concentration on dye removal

Fig. 7 shows the evolution of removal efficiency of color removal as a function of the initial dye concentration, using the optimum conditions obtained previously for applied voltage, reaction time and pH of solution. In this Figure, dye solutions with

different concentrations in the range 25–200 mg/L were treated by electrocoagulation process. As expected, the rate of colour removal decreases with the increase in initial dye concentration. On the other hand, the percentage dye removal was gradually decreased from 97.16 to 73.28 % as the dye concentration increased from 25 to 200

mg/L. This result is in accord with previously published findings (30). This may be attributed to the fact that at a constant current density or applied voltage the same amount of aluminum ions passes to the solution at different BR-18 concentrations. Consequently, the formed amount of aluminum hydroxide complexes were insufficient to coagulate the greater number of dye molecules at higher dye concentrations (31). On the other hand, the decrease in removal efficiency with

increasing initial dye concentration may be attributed to requiring more coagulant when increasing levels of pollutant. Therefore, it is quite clear that under the present experimental conditions, the lower is the dye concentration the better is the decolorization efficiency. Also, as it can be seen from Fig. 8 by increasing the dye concentration from 25 to 200 mg/L, the energy consumption was increased from 6.84 to 35.73 kWh/m³ dye.

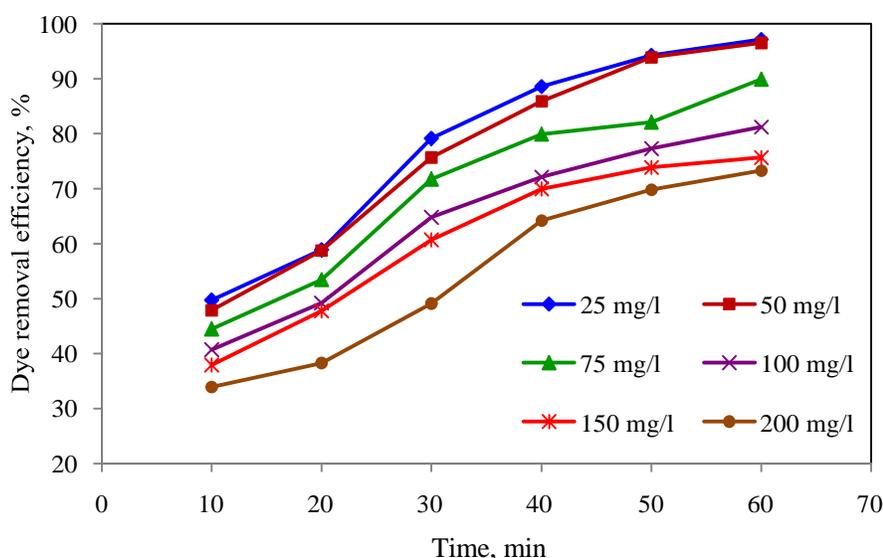


Fig 7. Effect of initial dye concentration on efficiency of colour removal (applied voltage = 50 V, pH ~ 7, electrical conductivity ~ 3000 μ S/cm)

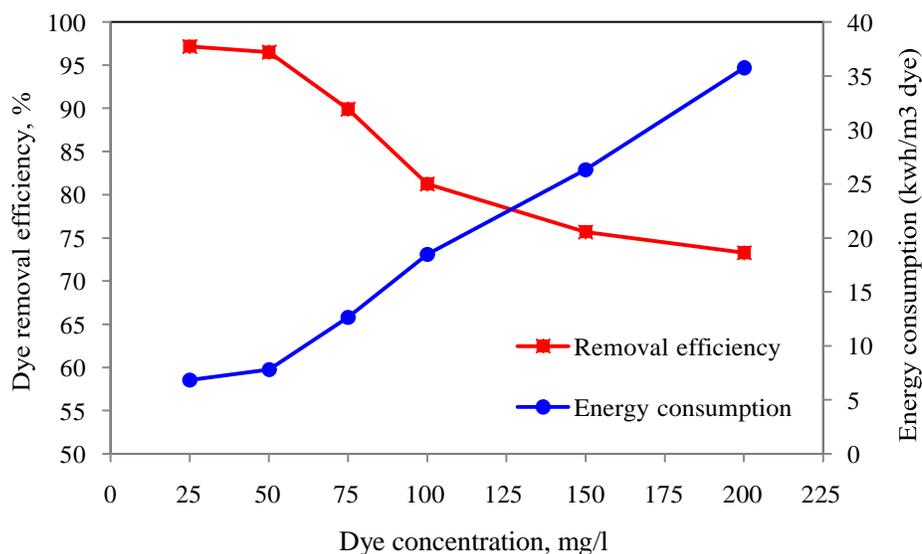


Fig 8. Effect of initial dye concentration on energy consumption (pH ~ 7, conductivity ~ 3000 μ S/cm, reaction time = 60 min)

3.4. Effect of electrical conductivity on dye removal efficiency

Solution conductivity affects the current efficiency, cell voltage and consumption of electrical energy in electrolytic cells. Typically, KCl is used to obtain the conductivity in EC process. Hence, a set of experiments was performed to determine the effect of electrical conductivity of solution on BR-18 dye removal efficiency. These experiments were performed using KCl as the electrolyte in the range of 1000–4000 $\mu\text{S}/\text{cm}$ at applied voltage of 50 V, initial dye concentration equal 50 mg/L and pH 7. As can be seen from Fig. 9, dye removal efficiency was increased with increase of solution conductivity. On the

other hand, as the solution conductivity increased from 1000 to 3000 $\mu\text{S}/\text{cm}$, the dye removal efficiency increased from 68.53 to 97.83%, but more increase of conductivity has not a considerable effect on dye removal efficiency, nevertheless, more increase of conductivity increased consumed energy. In fact, when the conductivity of solution increased, the current flow during electrocoagulation increased; as a result, the efficiency of BR-18 dye removal was improved. Golder et al. (32) stated that the availability of metal coagulants increases with increasing conductivity of solution.

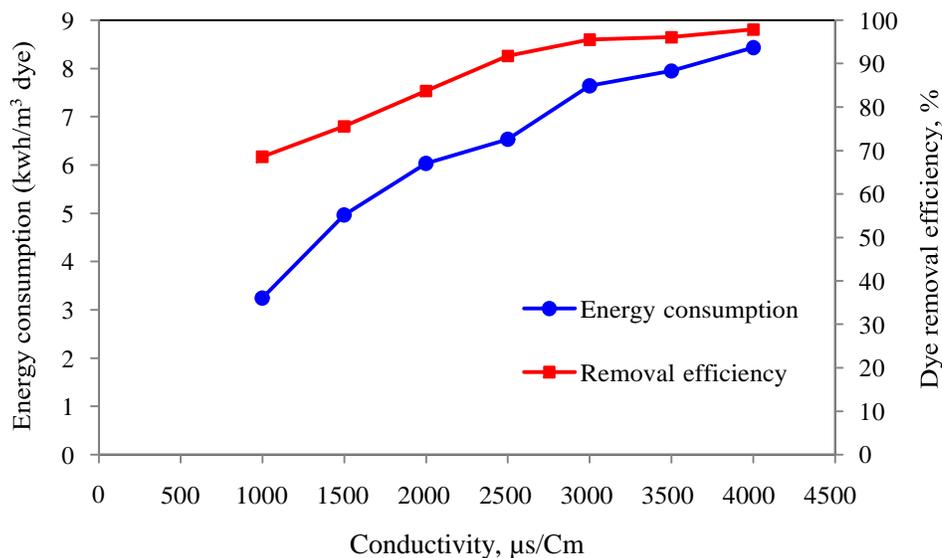


Fig 9. Effect of conductivity on the dye removal efficiency and energy consumption (applied voltage = 50 V, initial dye concentration = 50 mg/L, reaction time = 60 min, pH = 7)

Chen et al. reported that conductivity had little effect on the separation of pollutants from restaurant wastewater in the investigated range from 443 to 2850 $\mu\text{S}/\text{cm}$ (5). Kobya et al. found that the turbidity removal efficiency remained almost

unchanged in the conductivity range of 1000–4000 $\mu\text{S}/\text{cm}$ (26). But, it was in contrast to that given by Lin and Peng for electrocoagulation of textile wastewater using iron electrodes (33). Increasing solution conductivity resulted in the

reduction of cell voltages that caused a decrease in electrical energy consumption (34). But our results showed (Fig. 9) that at constant voltage, increasing of solution conductivity resulted in the increase of electrical energy consumption.

4. Discussion

Electrocoagulation process is one of the most effective methods to remove dye compounds from textile wastewater, which reduces the sludge generation. At present study a series of experiments was performed in order to find the effects of critical operating parameters for BR-18 dye removal by electrocoagulation from aqueous environments. Dye removal by batch EC process was affected by applied voltage, initial pH of solution, initial dye concentration and conductivity of solution. The findings obtained with synthetic solutions showed that the most effective removal capacities of dye achieved at 50 V electrical potential. In addition, for a given time, the increase of applied voltage, in the range of 10-60 V, enhanced the treatment efficiency. Nevertheless, electrical energy and electrode consumption were found to increase with increasing the applied voltage. The optimal pH was 7 (removal efficiency ~ 98%), at which higher dye removal efficiency could be reached. In addition, dye removal efficiency was increased with increase of solution conductivity. Finally, according to findings of this study it can be concluded that electrocoagulation process can effectively remove dye from aqueous environments.

Acknowledgements

The present work was financially supported by health research deputy of Zahedan University of Medical Sciences.

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