Original Article

Application of Electrocoagulation Process for Reactive Red 198 Dye Removal from the Aqueous Solution

Mansooreh Dehghani[1] Razieh Shabestari[2] Amir Anushiravani[3] ***Narges Shamsedini**[2]

1- Department of Environmental Health Engineering, School of Health, Shiraz University of Medical Sciences, Shiraz, Iran
2- Department of Environmental Health Engineering, School of Health, Student Research Committee, Shiraz University of Medical Sciences, Shiraz, Iran
3- Department of Internal Medicine, School of Health, Student Research Committee, Shiraz University of Medical Sciences, Shiraz, Iran

***shams8575@yahoo.com**

(Received: 7 Mar 2014; Revised: 12 May 2014; Accepted: 21 Jun 2014)

Abstract

Background and purpose: The main objectives of this research were to evaluating the application of electrocoagulation process for 198 dye from the aqueous phase and determining the optimum operating conditions to the dye removal using aluminum and iron electrodes.

Materials and Methods: The present study was conducted in bench-scale. The spectrophotometer DR 5000 was used to determine the dye concentration. The effects of pH, retention time, voltage, dye concentration on the efficiency of electrocoagulation process were investigated. Data were analyzed in SPSS for Windows 16.0 using Pearson's correlation coefficient to analyze the relationship between these parameters.

Results: The results showed that the optimal conditions for reactive red 198 (RR-198) dye removal from the aqueous solution are pH of 11, the voltage of 32 V, the initial dye concentration of 10 ppm, and the reaction time of 40 min. Pearson correlation analysis showed that there is a significant relationship between voltage and the reaction time with the removal efficiencies ($P < 0.01$).

Conclusion: It was revealed that the removal efficiency of dye was directly proportional to the voltage and reaction time, but inversely proportional to the initial dye concentration. In conclusion, electrocoagulation process using two-fold iron and aluminum electrodes is an appropriate method for reducing the RR-198 dye in the aqueous phase.

[Dehghani M, Shabestari R, Anushiravani A, *Shamsedini N. **Application of Electrocoagulation Process for Reactive Red 198 Dye Removal from the Aqueous Solution**. IJHS 2014; 2(2): 1-9] http://jhs.mazums.ac.ir

Key words: Electrocoagulation, Reactive red 198 Dyes, Aqueous Solutions, Textile Wastewater

1. Introduction

Reactive dyes are among groups of dyes that are largely used for the purposes of dying cellulosic fibers, especially cotton fabrics (1,2). Due to their greater tendency to react with water compared to the hydroxyls existing in cotton fibers, the dyes stabilization on the fibers is slight, while large amounts enter wastewater freely (3). Many different methods for the removal of dye from wastewater have been studied. These methods include sedimentation, ion exchange, adsorption on activated carbon, membrane processes (such as nanofiltration and reverse osmosis), and biodegradation processes (4-6). Although these methods seem to be effective, they are often not efficient and expensive (7).

In recent years, great attention has been paid to the electrochemical processes due to their simplicity of using and compatibility with the environment (8), low cost system, reduced sludge production due to the low water content of the sludge (9-11), simple dewatering sludge (12), and low space requirement to install the equipment (13-14).

Electrocoagulation is one of the most important processes effectively used to remove dyes from wastewater. In general, electro-dialysis is a process through which chemical reaction is occurred by transferring electron from one electrode interface through the other electrode. Electric flow is supplied by a direct electrical current and applied voltage between the two electrodes. The main mechanisms of coagulation include oxidation, electrochemical disinfection, electrochemical flocculation, and electrochemical flotation or combination of any of these mechanisms (15). This process takes place by applying a direct electric current. Applied voltages are from 10 to 60 V. The intensity of electric current depends on the contamination level. Aluminum, iron, and stainless steel, or carbon glass electrodes are used (16).

Several studies on the removal of dyes using electrocoagulation have been done, and high efficiencies were reported. Using electrocoagulation method, the dye removal efficiency for red 81 from aqueous solution (17), basic dyes (18), and effluent dye (19) were recorded 98%, 94%, and 96%, respectively.

Reactive red 198 (RR-198) dye widely used in textile and dye industries is frequently detected in water resources. Moreover, there is a concern regarding the contamination of water resources and its effect on people’s health and the environment. Due to non-biodegradation of reactive dyes in the environment and also the inability of conventional wastewater treatment to remove dye efficiently, a suitable technique to reduce and remove the dyes from textile industries wastewater is required. Therefore, the objectives of the study were to (i) evaluate the feasibility of using electrocoagulation process to remove RR-198 dye from the aqueous phase, (ii) determine its removal efficiency using aluminum and iron electrodes, and (iii) assess the optimum operating conditions to remove the dye effectively.

2. Materials and Methods

All tests were performed at a bench-scale batch reactor mode at room temperature and normal pressure. This study was conducted in a laboratory scale using two-fold iron and aluminum electrodes. A cube-shaped electrochemical Pyrex cell made of 10 mm thickness glass and 100 ml volume with iron and aluminum plate with dimensions of 1 cm × 5 cm × 2 mm was used as the electrode. The electrodes were vertically separated by 2 cm from each other. The end of each electrode was connected to a direct current power supply. The mixing was performed by a magnetic stirrer at a constant speed of 100 rpm (Figure 1). Control was also used for this study.

All chemicals were purchased from Merck (Germany). RR-198 dye ($C_{27}H_{18}ClN_7Na_4O_{15}S_5$) with a molecular weight 967.5 g/mole was supplied by Hoechst Company, Germany. Hydrochloric acid of 15% was used to clean the electrodes before starting the experiment. The pH was adjusted by sodium hydroxide (NaOH) and sulfuric acid (H_2SO_4).

The effects of different parameters (pH, voltage, reaction time, electrode type, and initial dye concentration) on the reduction rate of dye were determined at three replications. All data were presented based on the mean. Two-fold electrodes (aluminum and iron) were placed vertically inside the reactor. The studied parameters were operation time (10, 20, 30, and 40 min), voltage (10, 15, 20, and 32 V), and initial dye concentrations (10, 20, 30, and 40 ppm) at different pH (3, 7, 10, and 11). Dye absorption was determined at wavelength of 520 nm using spectrophotometer (DR 5000) according to the standard method (2120) (20).

The optimal conditions of different parameters were determined according to dye removal efficiency. The data were analyzed using the SPSS for Windows (version 16, SPSS Inc., Chicago, IL, USA) by Pearson's correlation coefficient to analyze the relationship between these parameters.

3. Results

First of all, the optimal voltage to achieve the maximum removal efficiency was determined. Figures 2-5 indicate the effects of voltage and reaction time on the efficiency of electrocoagulation process to remove dye at different pH. The results indicated that by increasing voltage, the removal efficiency was increased. Another parameter that affects the removal efficiency was the time of electrolysis. The same ascending trend was seen by increasing operation time as well. The maximum removal efficiency at the applied voltage of 32 V and 40 min operation time was 98.1%.

Figure 1. The schematic design of electrocoagulation

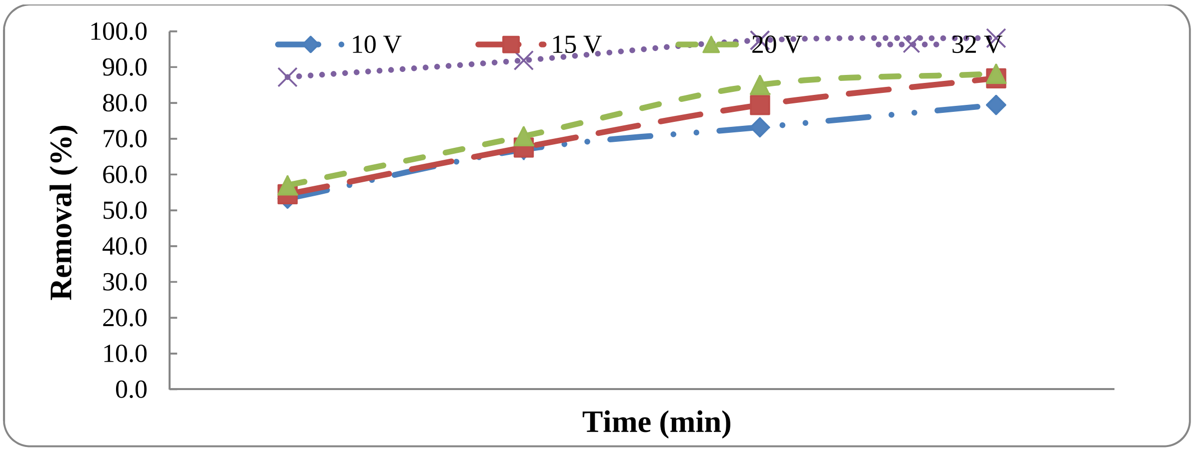

Figure 2. Removal efficiency of reactive red 198 dye using electrocoagulation process using two-fold iron and aluminum electrodes at different voltage and reaction time at pH = 11

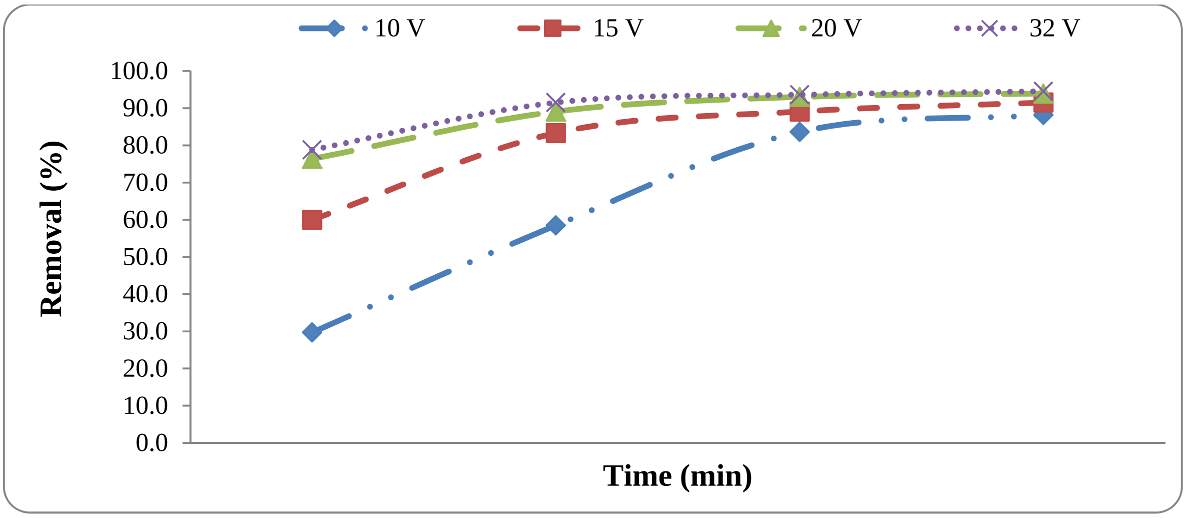

Figure 3. Removal efficiency of reactive red 198 dye using electrocoagulation process using two-fold iron and aluminum electrodes at different voltage and reaction time at pH = 10

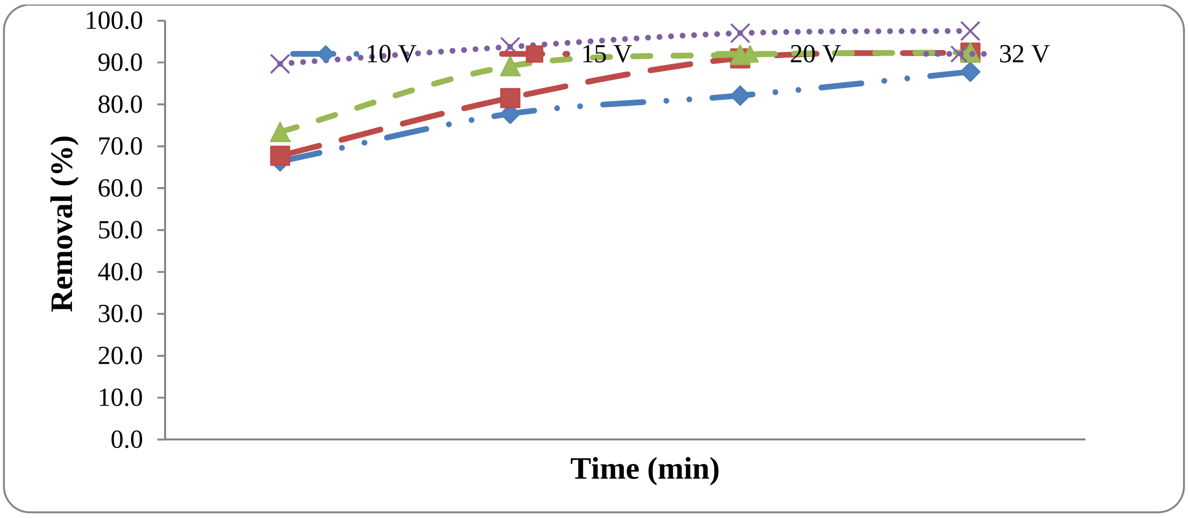

Figure 4. Removal efficiency of reactive red 198 dye using electrocoagulation process using two-fold iron and aluminum electrodes at different voltage and reaction time at pH = 7

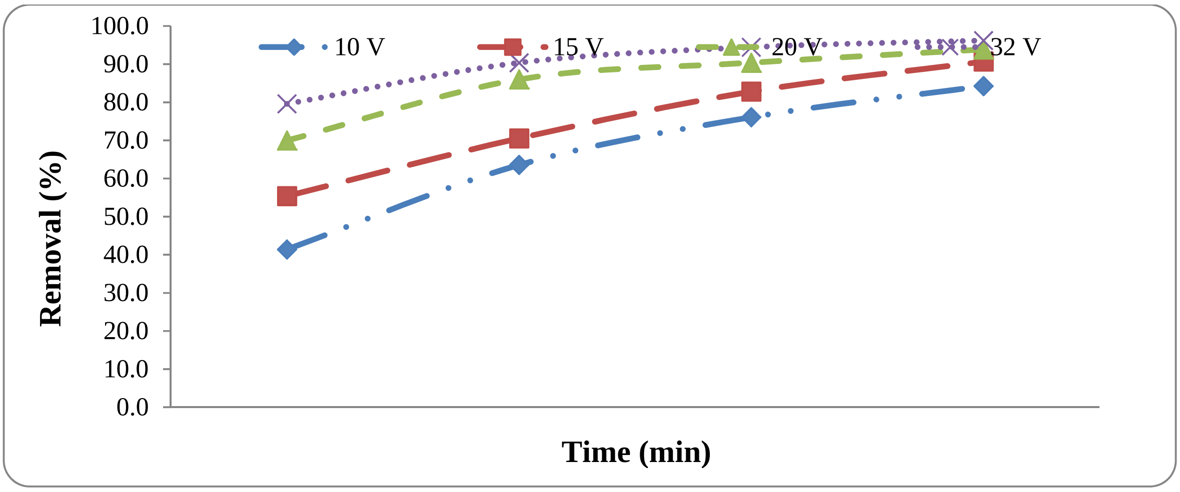

Figure 5. Removal efficiency of reactive red 198 dye using electrocoagulation process using two-fold iron and aluminum electrodes at different voltage and reaction time at pH = 3

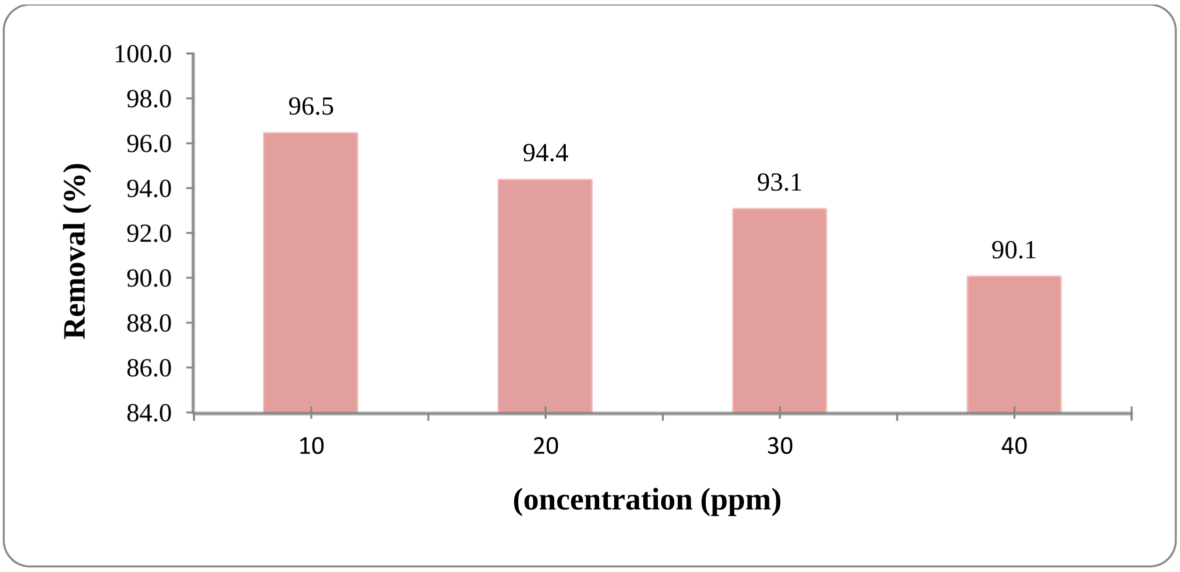

Figure 6. The effect of initial reactive red 198 dye concentration on dye removal efficiency at optimum conditions (pH = 7, reaction time = 30 min, and voltage = 15 V) using two-fold iron and aluminum electrodes.

Next, the effect of pH on the removal of dye from the aqueous phase was examined. In the current study, we used two-fold iron and aluminum electrodes, and the effect of pH on the removal efficiency was examined. The optimal pH to achieve the maximum removal efficiency was pH = 11.

After determining the optimal voltage and pH, the next attempt was to examine the effects of the initial dye concentration on the removal efficiency. The effect of initial dye concentration on the removal efficiency at optimum conditions is shown in figure 6. The maximum removal efficiency was occurred at initial dye concentration of 10 ppm. The dye removal efficiency was increased by decreasing the initial dye concentration. According to figures, the maximum removal efficiency was obtained at pH = 11, voltage = 32 V, and reaction time = 40 min.

4. Discussion

Electrocoagulation is being increasingly developed as a common dye removal method to treat wastewater. According to Pearson correlation coefficient, there is a significant relationship between the applied voltage and the removal efficiency of dye from the aqueous phase ($P < 0.01$). The results obtained in this study showed that by increasing the voltage at a constant electrolysis time, the removal efficiency is increased. In other studies, researchers have reached the same result (21-23). According to figures 2-5, by increasing the applied voltage, the current density is increased between the electrodes. Since the rate of electron flow is increased, the productions of ionic metals are accelerated and therefore the rate of the electrocoagulation process was enhanced. The reason for the direct relationship between the voltage and the removal efficiency can be due to the fact that faster chemical reactions take place on the anode and cathode electrodes. Then, the hydroxide ions produced by water reduction accumulated on the cathode electrode and react with aluminum hydroxide which can capture the dye pollutant in the aqueous phase either through surface complex mechanism and electrostatic adsorption or by physical sweeping mechanism during precipitation of aluminum hydroxide (21).

Bazrafshan et al. (24) applied the electrocoagulation process for the treatment of dairy wastewater. They concluded that the removal rate increased with increasing the applied voltage and reaction time. In general, the dye removal efficiency depended on the amount of aluminum ion produced in the solution. Basically, the amount of aluminum ions depended on the voltage and reaction time. Therefore, an increase in any of these mentioned parameters caused an increase in the formation of coagulant and higher dye removal observed as well. Low applied voltage and low reaction time result in low dye removal efficiency, which is due to sedimentation. Dalvand et al. (23) studies showed that the removal efficiency was 97% at 32 V and 60 min reaction time for RR-198 dye by electrocoagulation processing using iron electrode. Aoudj et al. (17) showed that the removal efficiency for red dye 81 was 98%. Jafarzadeh and Daneshvar[18] reported the removal efficiency of 94% for the basic dye using electrocoagulation process. Dalvand et al.[21] demonstrated that by the electrocoagulation process using aluminum electrode the removal efficiency of 99.1% at optimum conditions can be achieved (20 V and 30 min). Bazrafshan and Badli (25) showed that the optimal condition for the maximum removal efficiency of methylene blue dye from aqueous phase using electrocoagulation process (with four aluminum electrodes) was at pH = 11 and voltage = 32 V. By increasing the applied voltage and reaction time, the removal efficiency was increased as well. It was also reported by increasing the initial concentration of the pollutant, the reaction time required to achieve adequate removal efficiency was increased. Rastegarfar et al. (26) showed that the removal efficiency of phenol and color from pulping black liquor using electrocoagulation process was 98.5% at 70 min reaction time. Results obtained from Basiri Parsa and Nabizadeh Chianeh (27) also indicated that the removal efficiency of black dye 22 from synthetic wastewater was 92% at optimum conditions (2 V, 0.25 g/L electrolyte and pH = 4) by electrocoagulation processing using aluminum electrode. Can et al. (12) showed that the removal efficiency of 95% for textile wastewater using aluminum electrode. Massoudinejad et al. (28) showed that pH was the most important parameter affecting dye removal in electrocoagulation process. As pH increase from 7 to 9, higher removal efficiency was achieved. Increasing the operation time had a major role in the performance of the electrocoagulation process. There are many electrochemical reactions occurring simultaneously at the anodes and cathodes. The main reaction is the destabilization of pollutants. Electrodes which produce coagulants into water are made of either iron or aluminum. Iron and aluminum ions dissolve from the anodes. Released ions neutralized the charged particles and hence, the electrocoagulation process was performed. The removal efficiency was directly related to the concentration of ions generated on the electrodes. The ions concentration increased with increasing the time of electrolysis which in turn caused hydroxide flocks to increase. The results showed that the highest dye removal efficiency occurred at the operation time of 40 min. The effect of electrolysis time has been also considered as the main parameter in other studies. Many different studies demonstrated that increasing the electrolysis time resulted higher removal efficiency of phenol, heavy metals, and fluoride (29,30). Pearson correlation revealed that there was a significant relationship between dye removal and the electrolysis time ($P < 0.01$).

According to the results of this study, there is a significant relationship between pH and dye removal efficiency that is, the dye removal efficiency decreased with increasing pH from 3 to 11. The same results were obtained by other studies, which revealed that the removal efficiency of pollutant had an inverse relationship with increasing pH (19). Previous studies have shown that optimum pH was occurred in the range of 5-6 using aluminum electrode (21,22). In general, initial pH and final pH of the electrochemical cell have an effect on the dissolution of electrodes and the form of aluminum or iron species are mainly depend on pH of the solution. The results obtained in the current study revealed that the maximum dye removal efficiency was 98.1% at pH = 11 using two-fold iron and aluminum electrodes, but Rahmani and Samarghandi showed that at optimum conditions (30 V, 30 minutes, and pH = 3.5) the maximum efficiencies using iron and aluminum electrodes were 96% and 86%, respectively (19).

The result of the study showed that there is an inverse relationship between the initial dye concentration and the removal efficiency of dye ($P < 0.01$). Dalvand et al. also indicated at constant applied voltage and reaction time, the removal efficiency decreased with increasing initial dye concentration. Basically, a production of a certain amount of aluminum hydroxide ions at a constant voltage has a limited ability for adsorbing and removing only a certain amount of dye compound. Therefore, at a constant voltage, increasing dye concentration the removal efficiency was decreased due to the limitation for the production of aluminum hydroxide (21).

Since, economic evaluation is an important parameter in selecting an appropriate process for the removal of pollutant; optimization was performed. Operating cost calculations typically include the cost of chemicals, electrodes, and energy. Many studies showed that the cost of electrocoagulation was much cheaper than chemical precipitation (31). A comparative study showed that electrocoagulation was faster and more economic, consumed less material and produced less sludge, and pH of the medium was more stabilized than chemical coagulation. According to the results of this study, electrocoagulation can be an economically viable solution for the removal of dye from the aqueous phase (32-34). Based on the results obtained, pH = 7 was chosen as the optimum pH. Although increasing pH caused an increase in dye removal efficiency, it required more chemicals to adjust pH. Moreover, dye removal efficiency at pH = 7 was slightly lower than pH = 11. Therefore, pH = 7 was selected as the optimum pH. In addition, the applied voltage of 15 V and reaction time of 30 min was also selected as a proper condition to reduce the electrical energy consumption and cost.

Operating system parameters at the optimal condition can provide the dye removal efficiency of more than 98%. The results indicated the effectiveness of electrocoagulation for the treatment of dye wastewaters. Moreover, data obtained in the present study demonstrated the technical feasibility of the electrocoagulation process using aluminum and iron electrodes as a reliable method for the removal of dye from the aqueous phase. Due to the high efficiency of the electrocoagulation process and also the simplicity and relatively low cost, it might be considered as a reliable, flexible, fast, effective, and economical method for the removal of dye from the aqueous phase.

Acknowledgement

The authors would like to thank the Deputy of Research and Technology of Shiraz University of Medical Sciences for supporting the research project (92-01-21-6508).

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