

## Removal of Remazol Black B Dye by Electrocoagulation Process Coupled with Bentonite as an Aid Coagulant and Natural Adsorbent

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### ABSTRACT

Coloured discharge effluent into aquatic ecosystems may be carcinogenic and mutagenic besides aesthetic problems. In this work, the ability to remove (Remazol Black B) coloured water using electrocoagulation process is equipped with iron and aluminium electrodes with bentonite as a coagulant aid and natural adsorbent has been investigated. Remazol Black B was selected as a model dye and the treatment process was performed in a batch of electrocoagulation (EC) cell using iron and aluminium electrodes and bentonite. Experiments were conducted at 5 levels of various operational parameters at bench scale. The initial dye concentration was varied between 200-1200mg/L, pH ranging from 2-10; the retention time was between 0-60 minutes, the voltage studied was in the range of 5-30 V and the electrical conductivity was 0.25 to 2 Siemens/cm. Results showed that the use of electro-Aluminum and Iron, under optimum conditions of pH 2 and 4, the concentration of dye 1000mg/L, the voltage of 30V and 30 min have the maximum dye removal of about 99% and 91%, respectively. Results showed that both electrode voltages increase the efficiency of the above process in paint removal with decreasing pH from 10 to 2 and increasing the voltage from 5 to 30 V. Results show that by electrocoagulation Process Coupled with Bentonite as an Aid Coagulant could be used as well as an effective method for dye removal from colored wastewater.

**Key words:** Bentonite, Electrocoagulation, Iron Electrodes and Aluminum Electrodes, Color Removal, Remazol Black B.

### INTRODUCTION

Textile industry is one of the most water-consuming industries in the world which releases a remarkable amount of wastewater. This wastewater contains considerable amounts of organic dye compounds. Approximately 50% of the dyes utilized in the textile industry consist of dyes. These dyes are also used widely in industries such as paper and printing, pharmaceutical and toy industries [1, 2]. It is estimated that about 15% of the dye compounds used in dyeing processes are released to the environment through wastewater [3]. Releasing dye-containing wastewater into rivers and lakes results in a decreased water quality, oxygen transfer into water and gas solubility as well as high toxicity, carcinogenicity, and mutation for living creatures including humans [4-6]. Therefore, dye removal from this wastewater is of great importance [7, 8]. Usually 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 [9]. There are various methods to remove dyes from colored effluents such as adsorption, precipitation, chemical degradation, photodegradation, biodegradation, chemical coagulation and electrocoagulation [9, 10]. Some of these techniques suffer from other disadvantages such as high costs, production of huge amounts of sludge, and low efficiency [10, 11]. Electrochemical process is a biocompatible method which can compete with other water treatment methods due to its low costs [12, 13]. Therefore, electrocoagulation (EC) as an electrochemical method was established to dominate the weaknesses of conventional decolorization methods. Electrocoagulation is an optimal option for the removal of textile dyes [14-17]. EC process offered a simple, authentic and cost-effective method for wastewater treatment. To date, several studies have been conducted regarding the efficiency of electrocoagulation in dye removal from aqueous media. Some of them are as follows: Vidal *et al.*'s

study entitled "Acid Black 194 Removal from water by electrocoagulation using aluminum anode" [18], Shankar *et al.*'s study entitled "COD, TOC and dye Removal from pulp and paper industry wastewater through electrocoagulation" [19], and Mollah *et al.*'s study entitled "Treatment of orange II azo-dye by the electrocoagulation (EC) technique in a continues flow cell using sacrificial iron electrodes" [20, 21]. In recent years, research on the usage of electricity in the water treatment and purification for environmental compatibility and the possibility of liquids, gases and solids purification have been developed and continued as an attractive method for ligation or sedimentation, as the electrocoagulation / electrochemical methods [22-23]. Electric coagulation has been suggested as an effective option for removing contaminants from wastewater with high levels of organic matter. Electric coagulation process has been very interesting due to its compliance and environmental compromise [24]. The ability of this process in refining multiple types of wastewater is well documented including textile wastewater [25], domestic sewage [26], chemical industry waste [27], and basic color removal of textile wastewater [28]. One of the interesting methods of color removal is using physical adsorption [29]. One of the cheapest and most efficient aid coagulant is bentonite. Bentonite has the potential to act as an alternative low-cost aid coagulant because it is naturally available and possesses unique physicochemical properties [30]. Bentonite consists of the layers of two tetrahedral silica sheets sandwiching one octahedral alumina sheet. Due to the isomorphous substitution of silicon ions by aluminum or ferric cations in the tetrahedral sheets and the aluminum ions by magnesium or ferrous cations in the octahedral sheets, bentonite has net negative charges on its layer lattice [30]. Since there are some problems in both processes such as the lack of sedimentation, possible separation in the adsorption process and adjustment of feculence and sedimentation in the electrocoagulation process, this study generally aims to assess the remazol black b dye removal by an electrocoagulation process coupled with bentonite as an aid coagulant and natural adsorbent.

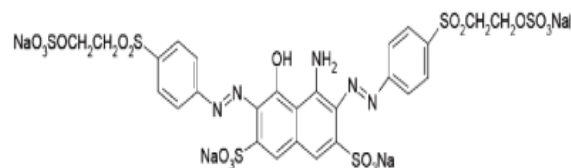
## MATERIALS AND METHODS

### *Dye solutions and Reagents*

The dye used in these experiments was Remazol Black B, which was provided by Alvan Sabet Co. of Iran. The molecular formula of Remazol Black B is (C<sub>26</sub>H<sub>21</sub>N<sub>5</sub>Na<sub>4</sub>O<sub>19</sub>S<sub>6</sub>). The chemical structure of this dye is given in Fig. 1.

Sulfuric acid and sodium hydroxide used to adjust the pH of solutions were purchased from Merck Co. (Germany). Stock solutions of Remazol Black B were

prepared by weighing the purified grade chemicals and dissolving them in deionized water without pH adjustment.



**Fig. 1:** Chemical structure of Remazol Black B

### *EC reactor*

The batch experimental was used, which consisted of an electrochemical reactor, a direct current (DC) power supply and iron and aluminium electrodes. The bench scale (BS) unit was made of Plexiglas with the reactor volume of 1L, which was equipped with four pairs of electrodes, made of iron and aluminium as anodes with the dimensions of 110mm × 110mm × 2 mm with the effective area of 121cm<sup>2</sup>. The distance between the electrodes was 20mm and the distance between the electrodes and the bottom of the reactor was 20mm. The electrodes were connected to a digital DC power supply (model-YX-360TR-EB) in the 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 each run, the specific concentration of dye in the aqueous solution was fed into the reactor and the pH and conductivity were adjusted. After each run, the electrode surfaces were removed by dipping in 50 cm<sup>3</sup> of the hydrochloric acid solution for 1 minute and were washed with deionized water to remove any solid residues on them and then were dried and weighted. In order to evaluate the effect of pH in the electrocoagulation process, pH adjustment was required by Sulfuric Acid and normal sodium hydroxide of the value of 0.1. In order to study the impact of the initial concentration of color of 200-1200mg/L, the pH of 2-10, within the range of 0-60 minutes, bentonite dose of 1g/L, a voltage in the range of 5-30V and the electrical conductivity of 0.25-2 Siemens/cm (potassium iodide), the performance of the compound process electrocoagulation and adsorption of Remazol Black B color removal at the wavelength of 597 Nanometer has been followed by spectrophotometry (DR5000 HACH Co. made in Germany).

## RESULTS AND DISCUSSION

Colour removal from natural wastewater by using the electrocoagulation process is a developing method. In this study, colour removal from colour water samples is used for comparing the efficiency of using two types

of iron and aluminium metals with the role of the sacrificial anode electrode in addition of maintaining

the exact similar design conditions, for both the metal types (Table 1).

**Table1:** The impact of pH, dye concentration, reaction time, voltage and the electrical conductivity on the efficiency of the electrocoagulation process with iron and aluminium electrodes with Bentonite

Electrocoagulation process with Bentonite		pH								
		2	3	4	5	6	7	8	9	10
Removal (%)	Fe	%83	%85	%91	%85	%84	%83	%83	%81	%85
	Al	%99	%96	%92	%85	%86	%85	%84	%80	%82
		Dye concentration								
		250		500		750		1000		
Removal (%)	Fe	%86		%87		%89		%91		
	Al	%88		%91		%96		%99		
		Reaction Time								
		5	10	20	30	40	50	60		
Removal (%)	Fe	%85	%86	%87	%91	%90	%90	%87		
	Al	%87	%92	%93	%99	%98	%96	%95		
		Voltage								
		5	10	20	30	40	50	60		
Removal (%)	Fe	%85	%86	%87	%91	%90	%88	%85		
	Al	%87	%92	%93	%99	%98	%97	%95		
		Electrical Conductivity								
		1000	1500	2000	2500	3000				
Removal (%)	Fe	%84	%86	%87	%88	%91				
	Al	%89	%88	%93	%95	%99				

Furthermore, the impact of the key parameters has been discussed and the results have been presented as follows:

#### *Effect of pH*

So far various studies regarding the impact of the initial pH on pollutants removal have been conducted by electrocoagulation process and each of them mentioned the significant effect of these parameters on the pollutants removal [5]. Fig. 2 shows the impact of pH on the process efficiency in terms of the reaction time of 30 minutes with a voltage of 30V. Results of many studies reported that in the electrocoagulation process using Al electrodes in units with the pH<2 and in units of pH >10, due to the amphoteric properties, the aluminum hydroxide particles did not have deposition on the pH<2 units and also in pH >10. The solubility of aluminum hydroxide to aluminum oxide as a non-use solution has increased and led to the decreased efficiency of the pollutant removal in this process [2]. Fig. 2 shows that the process efficiency is reduced while using both types of electrodes by increasing pH from 2 to 10. The best efficiency of color removal while using aluminum electrodes has been obtained at pH=2, 99.93%, and for iron electrode in pH=4 in about 91%. Also the results of this study showed that while using aluminum electrodes when the initial pH<8 was set, the final pH value increased constantly and it was always higher than the initial pH, while at the initial pH>8, the final pH value was less than the initial pH, while it has been observed by using the iron electrode that the final pH values had always an increasing trend and the pH was higher than the

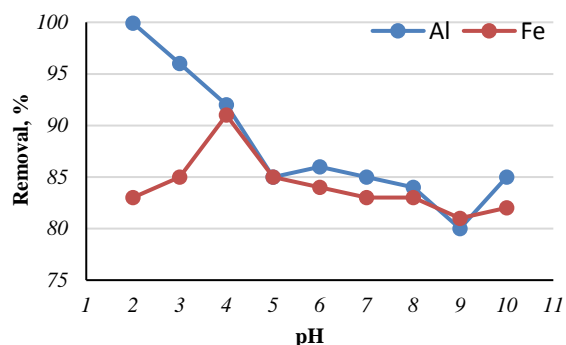
initial values. Similar results have been mentioned in other studies [32, 31]. Some researchers believe that the pH changes in the electrocoagulation process are due to the increments of the produced hydrogen from the cathode electrode and others believe in these changes by considering the fact that the acidity of many aquatic environments depends on the existence of compounds such as soluble carbon dioxide and the withdrawal of carbon dioxide (CO<sub>2</sub>) from the aquatic environment of the process and due to the incorporation of the produced hydrogen gas bubbles [33-35]. According to these results, researchers agree with each other on the buffering capacity of the electrocoagulation process, especially at moderate to alkaline pH values [36]. Therefore, the optimal pH in the process of electrocoagulation with bentonite was selected while using aluminum electrodes of pH 2 and using iron electrodes of pH 4.

#### *Effect of dye concentration*

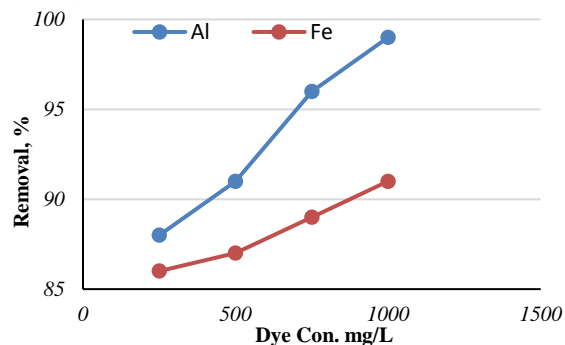
In this study, experiments of the synthetic solutions of Remazol Black B color with different initial concentrations between 250-1000mg/L have been conducted. The experiment results showed that in stationary conditions with different concentrations of the primary color, the initial color concentration can affect the removal efficiency so that while having a higher concentration, a higher voltage and a longer reaction time are needed. In this study, the impact of the examined concentrations of the initial colors on the process was insignificant and a slight reduction in the removal efficiency was observed by increasing the initial colors concentration. Fig. 3 shows the impact of

changes in the amounts of color concentration between 250-1000mg/L of the color removal efficiency.

Examination of Fig. 3 shows that the increment of color concentrations from 250 to 1000mg/L will increase the project efficiency. This is interpretable by the dilute solutions theory so that in dilute solutions, the formation of the sporadic layer near the electrode causes the reaction to slow down, but in concentrated solutions, the sporadic layer has no impact on the diffusion amount of metal ions on the electrode surface. Narooie *et al.*, in their study entitled "Evaluation of the efficiency of ashes from palm and pistachio wastes in the Reactive Red 120 Dye removal from aqueous solutions" reported that, the removal efficiency enhances by increasing the concentrated solutions, as by increasing the concentrated solutions from 10mg to 70mg, the dye removal efficiency increased from 85% to 99% [37]. In another study entitled "Electrolytic removal of hexavalent chromium from aqueous solutions", Chaudhary *et al.* stated that, the removal efficiency enhances by increasing the concentrated solutions [38].



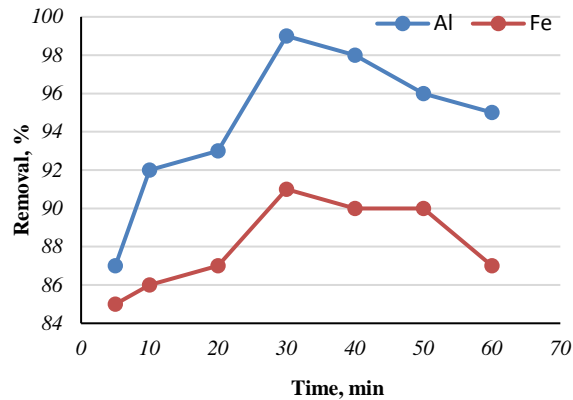
**Fig.2:** Impact of pH on the efficiency of the electrocoagulation process with iron and aluminum electrodes with Bentonite (Color concentration of 1000mg/L, Voltage of 30V, Electrical conductivity of 2 Siemens/cm, Bentonite dose of 1g/L, reaction time of 30 minutes)



**Fig. 3:** Impact of colour concentration changes on the electrocoagulation process efficiency equipped with iron and aluminium electrodes with bentonite (pH of 2 and 4, the voltage of 30V, the electrical conductivity of 2 Siemens/cm, Bentonite dose of 1g/L, and the reaction time of 30 minutes)

### Effect of the reaction time

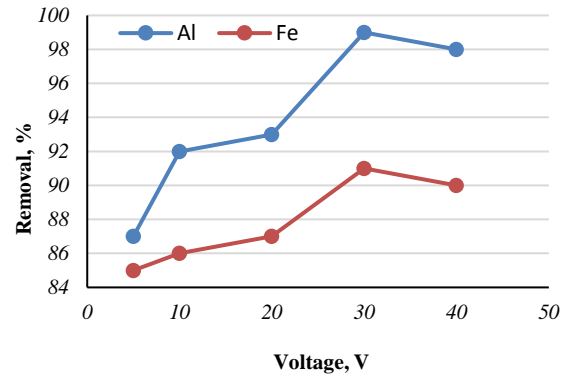
In almost all studies, the main parameter is an initial and important one in each study process, since in addition to the reduction of economic costs; the power and usability of the process would be increased. Many studies reported that by increasing the reaction time, the pollutants removal efficiency would be increased too, but the more response time increase, the less removal efficiency will be obtained. In fact, for each process, there are optimal and ideal reaction times for the removal of any contaminants that will determine the amount of economic boundaries of the process. Also In this study it was observed that in Fig. 3, by increasing the time, the efficiency will be increased and at the ending time of the process, the color values are absorbed from the absorber and suspension into the solution. Fig. 3 shows the impact of reaction time under the voltage of 30 V, concentration of dye 1000mg/L, pH 4 for the aluminum electrode and pH 2 for the iron electrode, respectively. Examination of Fig. 4 shows that in time durations of less than 20 minutes, color removal is less, but over time and as a result of producing more hydroxyl radicals, the color removal rate is increased significantly, so that in most experimental cases, after 5 minutes for aluminum and at different levels of saturation, the removal efficiency of more than 86.79% have been achieved and it is increasing too, so that the optimum conditions of the process which is about 30 minutes up to the 99.9% of color removal has been done. But after this time, the efficiency declines at a slower slope. Since after only 30 minutes of reaction time, the removal efficiency of about 99% and 91% in both types of electrodes have been achieved, it is expected that considering the reaction time, over 30 minutes for color removal is not appropriate and if more efficiency is needed, the result will be more favorable by increasing and setting other effective parameters such as voltage. Alizadeh *et al.* in their study entitled "The survey of electrocoagulation process for removal dye Reactive Orange 16 from aqueous solutions using sacrificial iron electrodes" reported that, the removal efficiency enhances by increasing the reaction time, as by increasing the reaction time from 5 to 30 minutes, the dye removal efficiency increased from 99.1% to 95.55% [39]. Salmani *et al.* in their study entitled "Removal of Reactive Red 141 from synthetic wastewater by electrocoagulation process" reported that, the removal efficiency enhances by increasing the reaction time, as by increasing the reaction time from 22.17 minutes to 28.10 minutes, the dye removal efficiency increased from 89.74% to 95.18% [1].



**Fig. 4:** Impact of the reaction time changes on the electrocoagulation process efficiency equipped with iron and aluminum electrodes with Bentonite (pH 2 and 4, Voltage of 30V, electrical conductivity of 2 Siemens/cm, Bentonite dose of 1gr/L, and Color concentration of 1000mg/L)

#### Effect of voltages

In this study, the impact of potential difference of 5-30V on Remazol Black B color removals from aqueous synthetics has been examined. As expected, it was observed that at a constant reaction time, the color removal efficiency increased significantly by the increments of potential difference. Fig. 4 shows the impact of voltage changes in the range of 5-30 V color analyses. Examination of Fig. 5 shows that the most rapid removal efficiency of about 91% are achieved while using iron electrodes and 99% while using aluminum electrodes in the potential difference of 30 V and the reaction time of 30 minutes and on the other hand, the least dye removal efficiency of about 84% is achieved while using iron electrodes and 89% while using aluminum electrodes in the condition that the potential difference is at its lowest level of about 5 V. The removal efficiency of potential difference at reaction times higher differences were significant and gradually the removal efficiency in both types of electrodes has significant differences. This issue expressed the fact that at high voltage levels, the metal-oxide levels increases and more hydroxide clots are produced for removing pollutants. In addition, it has been proven that by increasing the potential difference, the density of bubbles will increase and their sizes will reduce which will cause the faster and more removal of pollutants [40].

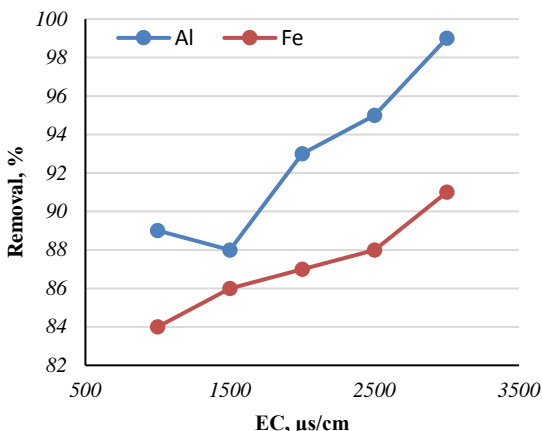


**Fig. 5:** Impact of voltage changes on the electrocoagulation process efficiency equipped with the iron and aluminum electrodes with Bentonite (pH 2 and 4, electrical conductivity of 2 Siemens/cm, Bentonite dose of 1g/L, and color concentration of 1000mg/L)

#### Impact of electrical conductivity

Natural waters and sewage and waste waters all contain compounds and different ions that in the case of establishing electric bridge in aquatic environments, the existing ions are responsible for passing electrical current between the bridges. The more ionic strength of these species are larger, the more stream flow transmission of constant voltage increases and in a uniform stream by increment of the electrical conductivity the voltage level will decrease (28). Therefore, in the current study, the impact of ions on factors like efficiency, energy levels and consumed electrodes' weights in the electrocoagulation process for dye removal has been investigated. In Fig. 6, the impact of electrical conductivity on the removal efficiency, energy and consumed electrodes' weights while using both iron and aluminum types of electrodes are shown. The presence of chloride ions in water caused that the destruction passive oxide layer formed from the electrode surfaces so the level of energy consumption in constant potential difference increased and the efficiency of dye removal improved [40]. This figure shows that by increasing the electrical conductivity from 1000 to 3000 Micro-Siemens/cm, the dye removal efficiency while using both types of electrodes also increased significantly. In fact, consuming more energy with constant potential difference in these two points, in addition to increasing the coagulants production, will cause the increment in the production of bubbles of the evaporated hydrogen gases, increasing the size and growth of the produced clots that increased the efficiency of electric coagulation process [41, 31].





**Fig. 6:** Impact of conductivity on the electrocoagulation process efficiency equipped with iron and aluminum electrodes with bentonite (pH 2 and 4, voltage of 30V, reaction time of 30 minutes, Bentonite dose of 1g/L, and color concentration of 1000mg/L)

## CONCLUSION

In this study, the usage of electrocoagulation process with bentonite has been evaluated in order to reduce the color of Remazol Black 5 in colored water samples. Results showed that the impacts of operating parameters are effective on the performance of the solution of pH, voltage, reaction time, concentration changes of color and the process bentonite value changes. The best efficiency of color removal of about 99% at optimal operating conditions while using of the aluminum electrode was obtained with the pH of 2, voltage of 30V, reaction time of 30 minutes, and color concentration of 1000mg and bentonite dose of 1g. The Consolidated electrocoagulation and Bentonite processes can be used as an environmentally method in the purification of colored wastewater, especially in order to reduce the color concentration of wastewater in textile industry.

## ETHICAL ISSUES

Ethical issues such as plagiarism were strictly observed while writing.

## CONFLICT OF INTEREST

The authors declare that there is no conflict of interest and nothing in contrary to the facts.

## AUTHORS' CONTRIBUTION

Seyed Ali Sajjadi was the supervisor of this research project and his comments at all stages were highly helpful. Ali Pakfetrat prepared the research proposal and designed the study. Data collection and drafting the article were done by Ali Pakfetrat and Morvarid Irani

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