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ARTICLE

REMOVAL OF REMAZOL BLACK B DYE FROM AQUEOUS SOLUTION BY ELECTROCOAGULATION EQUIPPED WITH IRON AND ALUMINIUM ELECTRODES

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ABSTRACT

Releasing wastewater of textile industry containing organic dye compounds such as Remazol Black B to the accepting water sources, results in severe damages to the environment and aquatic life. Since majority of the water treatment methods lack sufficient efficiency for removal of textile dye compounds, in this work, the efficiency of electrocoagulation method was investigated in treatment of synthetic water containing Reactive Black 5 dye using iron and aluminum electrodes in a batch reactor. First, a 1-liter reactor was equipped with iron and aluminum electrode by bipolar connection, and then the electricity source was turned on following adjusting the initial concentration to 5000 mg/L, electrical conductivity to 1000-3000 μSiemens/cm, pH to 2-9, and electrical potential to 30 V. In the next step, the sampling took place at 20-minute intervals and the efficiency of dye removal was determined during the maximum reaction time of 80 minutes, at 597 nm using a spectrophotometer. The results exhibited that, in this process, the maximum Reactive Black 5 dye removal efficiencies were obtained as 96 % and 89.45 % using iron and aluminum electrodes at pH values of 5 and 6, respectively, applying reaction time of 80 minutes, electrical conductivity of 3000 μSiemens/cm, and initial concentration of 5000 mg/l. Over time, the energy consumption rate increased as well as electrode mass consumption, pH, ultimate temperature of the wastewater, and the removal efficiency. The electrocoagulation process equipped with iron and aluminum electrodes could be used well as an efficient and cost effective method for treatment of wastewaters containing Remazol Black B dye.

KEY WORDS
Remazol Black B, Dye, Electrocoagulation, Aluminium Electrode

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INTRODUCTION

Textile industry is one of the most water consuming industries in the world and releases a remarkable amount of wastewater. These wastewaters contain considerable amounts of organic dye compounds. Approximately, 50% of the dyes utilized in textile industry consists of azo 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 the rivers and lakes results in decreasing water quality, oxygen transfers into the water, and gas solubility as well as high toxicity, carcinogenicity, and mutation for livings including humans [4, 5]. Therefore, removal of dye from these wastewaters is of great importance [6]. Following the advances of the science of textile dye production, novel dyes are introduced to the market which demonstrate better dyeing characteristics, however, this improvement of the quality has led to higher stability of dye structure, enhancing their resistance against biological and chemical degradation processes [7]. Majority of the dyes used in textile industry are not biodegradable and since they form strong complexes, common water treatment processes including coagulation, flocculation, and chemical precipitation are not considered effective methods for their degradation [6]. Furthermore, these techniques suffer from other disadvantages such as high costs, production of huge amounts of sludge, and low efficiency [7, 8]. Electrochemical process is a biocompatible method which can compete with other water treatment method due to the low costs [9, 10]. The advantage of this method include low costs of the instruments and application, low sludge production with low water content and high dewatering capability, and little room required for the instrument installation [11]. In electrochemical coagulation processes, electrical current is applied using electrodes of generally iron and aluminum type to destabilize water pollutant agents as well as suspended and colloidal material [12]. In electrocoagulation process, iron and aluminum electrodes are degraded though electrolysis process according to the following equations, forming metallic hydroxides capable of coagulating the clotting agent of the suspended material or other dissolved pollutants in the water, which in turn leads to destabilization of colloidal material and pollutant agents in the water by inducing floatation or precipitation [13].

$M_{[s]} \rightarrow M^{+n}_{[aq]} + [e^-]$

Anodic reactions

$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$

$n[H_2O] + n\,[e^-] \rightarrow \frac{n}{2} H_2 + n[OH^-]$

Cathodic reactions

$4H^+ + 4e^- \rightarrow 2H_{2\,[g]}$

$M^{+n} + n[OH^-] \rightarrow M[OH^-]_n$

Where, M is the anode metal and n is the number of electrons transferred in redox reaction [14]. To date, some 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 "Removal of Acid Black 194 from water by electrocoagulation using aluminum anode" [15], Shankar *et al*'s study entitled "Removal of COD, TOC, and dye from pulp and paper industry wastewater through electrocoagulation" [16], and Mollah *et al*'s study entitled "Treatment of orange II azo-dye by electrocoagulation (EC) technique in a continues flow cell using sacrificial iron electrodes" [17, 18]. Since many reactive dyes are not biodegradable and considering the lack of sufficient efficiency of common wastewater treatment methods in removal of these dyes, in this study, the possibility of application of electrocoagulation water treatment method, using iron and aluminum electrodes was investigated for removal of reactive Black 5 dye in laboratory scale and the optimum operational condition was determined for removal of the dye considering the energy and electrode consumption rates.

MATERIALS AND METHODS

Materials and instruments

All materials used in this study, including Reactive Black 5 dye (CASRN) C26H21Na4N5O19S6 (17095-24-8), sodium hydroxide, and hydrochloric acid were supplied from Merck Company of Germany. HPLC grade distilled water was used in all experiments. First, stock solution of the dye (1000 mg/l) was prepared by dissolving certain amount of RB5 powder with purity of 55% in water. The pH adjustment took place by a Denver Ultra basic-UB10 made in USA using 1 N hydrochloric acid and sodium hydroxide (19). The electrical conductivity was adjusted to 1000, 1500, 2000, 2500 and 3000 μs/cm using 1 normal KCl by a WTW-Cond 1310 made in Germany.

Performing the process

To start the experiments, electrode planes of iron and aluminum with dimensions of 11×10 cm and distance of 2 cm were connected to a Micro power supply of 30 v potential using bipolar connection method, in a glass container with height, depth, and width of 13×12×10, respectively and volume of 1 liter. The pH values of the samples containing synthetic wastewater with dye concentration of 5000 mg/l, were adjusted to 2, 3, 4, 5, 6, 8, and 9 before applying the electrical current. Furthermore, their conductivities were adjusted to 1000, 1500, 2000, 2500, and 3000 μs/cm. The samples were then added to the reactor which was placed on a magnetic stirrer. A gentle and equal rate of stirring was applied for all samples. The electrocoagulation experiments were performed by applying the electrical current with electrical potential difference of 30 V. The efficiency of dye removal was then determined using a Thermo Scientific Helios Epsilon UV/VIS spectrophotometer at 597nm by taking 20 ml of the samples from the middle of the container in 20-minute intervals and maximum reaction time of 80 minutes. Amounts of the iron and aluminum released in the reactor were calculated by Faraday's law in addition to comparing their weights.

RESULTS

pH effect

[Fig. 1] shows the pH effect on the efficiency of electrocoagulation process within the range of 2-9 as well as pH variations when using iron and aluminum electrodes. The maximum removal efficiency was obtained as 96% following using iron electrode at pH 6. The second best result was achieved as 89.45% removal efficiency by aluminum electrode at pH 5. Figure 1 furthermore demonstrates that, the ratio of ultimate pH increase rate to initial pH increase rate is greater when applying iron electrode than the aluminum. The average ratio of ultimate pH increase rates to initial pH increase rates are obtained as 3.8 and 3.03 unites for iron and aluminum electrodes, respectively. ;Fig. 2] provides the ultimate temperature variations and initial pH values of 5 and 6 through electro coagulation process over time.[Fig. 2] illustrates that, for both electrodes, the ultimate temperature of the process increases from 25.7 ˚C to 48.9 ˚C over time. The ultimate pH values also increase. In 80 minutes, when applying aluminum electrode, the pH value reaches 8.57 from 5 with correlation coefficient of 90%, and when applying iron electrode, it reaches 10.5 from 6 with correlation coefficient of 96%. Figure 2 also shows that the growth rate of pH is greater when the iron electrode is used rather than the aluminum electrode.

The initial pH of the environment induces a significant effect on the performance of all processes, especially electrocoagulation process for removal of organic contaminants from aqueous solutions. Considering the standard regulations and the possibility of releasing the wastewater to environment, the variations of ultimate pH of the wastewater is as much important as the initial pH [20]. Ghalwa Abu *et al* (2016) reported that in their studies about removal of Reactive Red 24 by electrocoagulation process, the removal efficiency was highly pleasant when using iron electrodes at neutral pH or alkaline condition. This happened due to decrease of the dissolution of iron ions and formation of iron hydroxides capable of adsorbing dye molecules. On the other hand, the aluminum electrode demonstrated better efficiency in acidic environment. The decrease of dye removal by aluminum electrode is attributed to decrease of protonated functional groups on the dye molecules[21]. Rastgarfar *et al* (2011), reported a removal efficiency of 98% for Liquor Black dye in their study of removal of phenol and dye from liquor black in pulp and papermaking process by electrocoagulation method using aluminum electrode at pH 7. They depicted the reason as the high surface area of hydrophobic aluminum hydroxide bulks at neutral pH, which leads to fast adsorption of the dissolved organic material and trapping colloidal particles. In addition, due to oxidation of hydroxide ions at the anode and oxidation of Fe^{2+} to Fe^{3+} and formation of $Fe(OH)_6^{3-}$ and $Fe(OH)^{4-}$, the removal efficiency declines. By application of aluminum electrodes at pH values below 2 and above 10, the removal efficiency of this process decreases since at pH values below 2, aluminum hydroxide particles do not precipitate considering their amphoteric characteristic, and at pH values above 10, the solubility of $Al(OH)_3$ increases as useless soluble AlO_2. In this study, the maximum efficiency was achieved when pH values were adjusted to 6 and 5 for iron and aluminum electrodes, respectively. This was in good agreement with the above studies. Hence, the optimum pH is determined as 6 and 5 for iron and aluminum electrodes in electrocoagulation process [22].

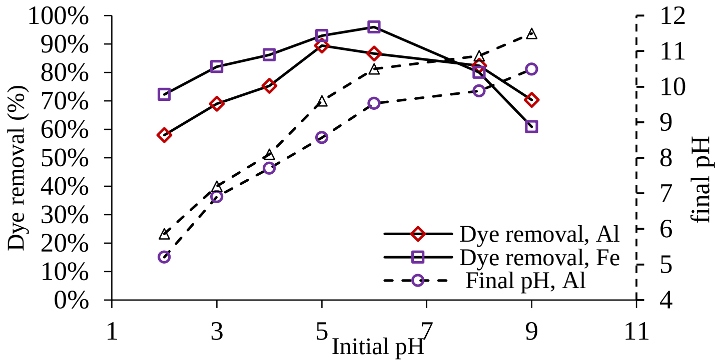

Fig. 1: Effect of pH on removal efficiency(time 80 min, 30 voltages, 5000 mg/l dye, 3000 μs/cm electrical conductivity).

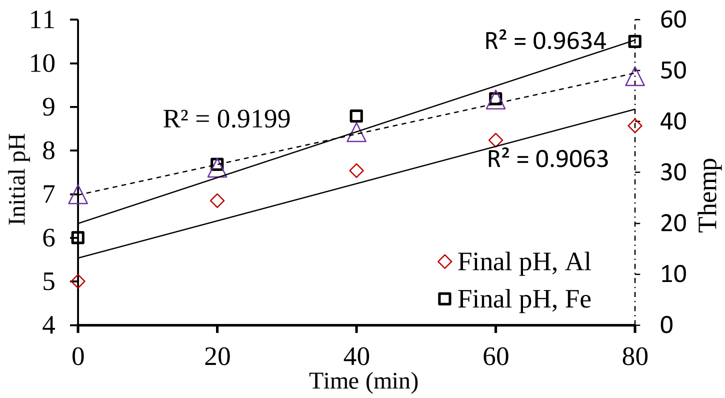

Fig .2: Temperature and pH change during process (30 voltages, 5000 mg/l dye, 3000 μs/cm electrical conductivity).

Reaction time effect

[Fig. 3] demonstrates the effect of reaction time on energy consumption in dye removal using aluminum and iron electrodes at pH of 5 and 6, respectively. Studying figure 3 shows that, with increasing the contact time, the remaining concentration of the dye decreased, whereas energy consumption increased. Application of aluminum and iron electrodes consume 0.26 and 0.36 kWh energy, respectively for removal of each gram of dye within 80 minutes. It further illustrates that, using aluminum and iron electrodes in electrocoagulation process decreased down the dye concentration from 5000 mg/l to 264 mg/l and 100 mg/l, respectively.

[Fig. 4] provides the effect of reaction time on iron and aluminum electrode weight per gram of dye removed. Studying this figure revealed that, about 1.94 g of aluminum and 2.39 g of iron electrodes were consumed or in other words, were produced in the system as coagulants per each gram of removed dye, after 80 minutes. Figure 4 shows that iron electrode is consumed more compared to aluminum electrode at the same time period and dye removal efficiency was higher by former one.

Almost in majority of the studies, the reaction time parameters is considered a primary and important one in investigation of the process, because it will improve the power and usability of processes in addition to reducing costs, in particular the current costs [23].

Dalvand *et al* (2011) in their study entitled as "treatment of wastewater containing Reactive Red 198 using electrocoagulation process" reported that by increasing the reaction time, energy consumption increases. They achieved 99.1% dye removal efficiency in their study, following voltage of 20 V, reaction time of 75 minutes, energy consumption of 1.516 kWh per cubic meter of treated wastewater. The increased electrical current from electrodes was the reason they explained for this achievement [13]. Salmani *et al* (2016), 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 ascended from 89.74% to 95.18%. They presumed the reason for this improvement was higher production of Fe ions and formation of sufficient iron hydroxide clots [24]. In this study, the rate of energy consumption, iron and aluminum consumption, and removal efficiency increased by increasing the reaction time. Thus, in this study, the maximum efficiency is introduced at 80 min reaction time. Economic reasons hindered further continuation of the process.

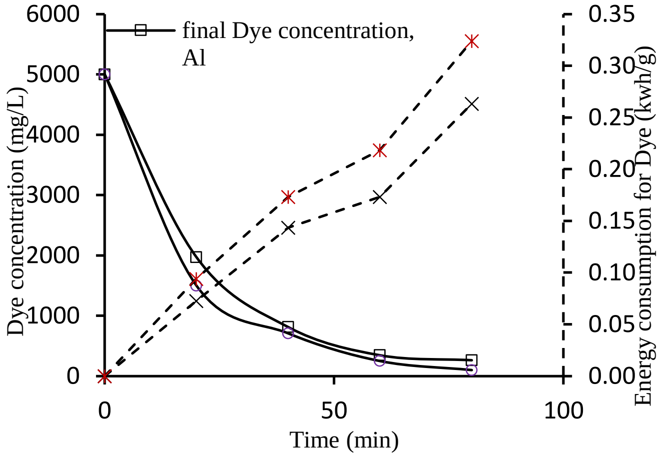

Fig. 3: Effect of time changing on removal efficiency (30 voltages, 5000 mg/l dye, 3000 μs/cm electrical conductivity, pH 5, 6).

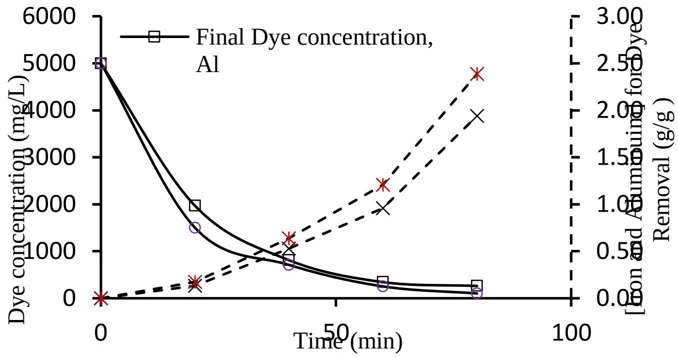

Fig. 4: Effect of time changing on removal efficiency and Wight of metal sacrificial (30 voltages, 5000 mg/l dye, 3000 µs/cm electrical conductivity, pH 5, 6).

Electrical conductivity effect

[Fig. 5] illustrates the effect of electrical conductivity on dye removal efficiency in electrocoagulation process when using iron and aluminum electrodes. Studying figure 5 shows the improvement of the efficiency by increasing the electrical conductivity. Following increasing the electrical conductivity from 1000 to 3000 µs/cm, the dye removal efficiency increased from 73.7% to 78.7% and from 89.45% to 96% using aluminum and iron electrodes, respectively. Figure 6 shows the impact of variation of initial electrical conductivity on energy and metal consumption rate in dye removal process. [Fig. 6] reveals that, energy consumption rate has increased by enhancing the electrical conductivity, and more metal is released in the solution and or consumed from the electrode planes subsequently resulting in higher dye removal efficiency. By increasing the electrical conductivity from 1000 to 3000µs/cm, energy consumption rate increased from 0.17 to 0.26 and from 0.2 to 0.32 when using aluminum and iron electrodes, respectively. For per gram of dye removed, the metal weight released to the solution increased from 1.44 to 2.38 and from 1.27 to 1.94 when using iron and aluminum electrodes, respectively. Natural water and all sewages and wastewaters contain various compounds and ions, which cause electrical conductivity between two electrodes, once an electrical pole is established. As the ionic strength grows, electrical conductivity will increase at constant voltage, or in a constant current, the voltage will decline by improving electrical conductivity [23]. Bazrafshan *et al* (2012), in their investigation regarding removal of humic acids from aqueous solutions by electrocoagulation process with iron electrode, reported that, at a constant voltage, enhancing electrical conductivity leads to increase of energy and electrode consumption rate. The rate of electrode consumption increased from 0.11 to 0.19 kg/g and humic acid removal efficiency increased from 76.95% to 92.69% following raising electrical conductivity from 1000 to 3000 µs/cm. They assumed that, increasing the electrical conductivity was the reason for this improvement in the efficiency (9). Bazrafshan *et al* (2012), in their study of removal of humic acid from aqueous media by electric coagulation process with the addition of hydrogen peroxide reported that, humic acid removal efficiency improved remarkably by increasing the electrical conductivity by addition of 1, 1.5, 2, and 3 g/l of potassium chloride. They claimed that, this occurred due to the increase of coagulant as well as increasing the size and growth rate of the produced clots which results in improvement of the efficiency [20]. In this study, the removal efficiency increased with increasing electrical conductivity and correspond with the results of the above studies. In this study, the optimal efficiency of the electrical conductivity was achieved as 3000 µs/cm.

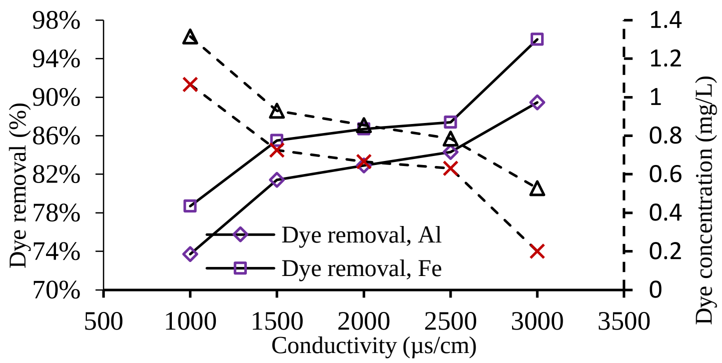

Fig . 5: Effect of electrical conductivity change on removal efficiency (30 voltages, 5000 mg/l dye, time 80 min, pH 5, 6).

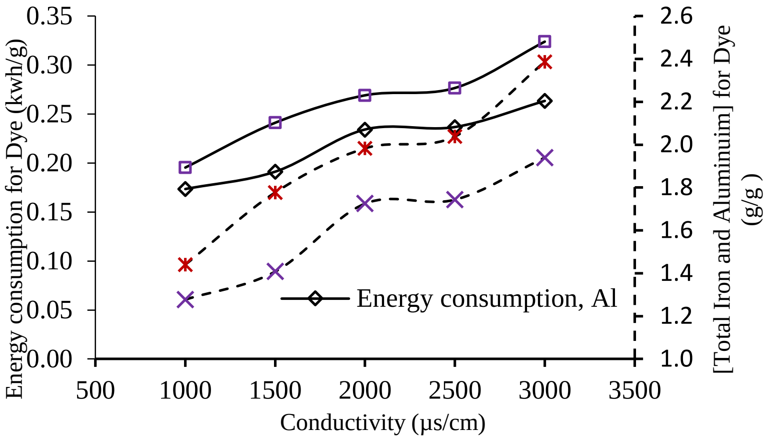

Fig . 4:Effect of electrical conductivity change on Wight of metal sacrificial and energy consumption (30 voltages, 5000 mg/l dye, 3000 µs/cm electrical conductivity, pH 5, 6).

on outer skin opening of fistula and knot was secured on the outer hard leather piece [Table 1].

CONCLUSION

This study was conducted to investigate the Remazol Black B dye removal efficiency by electrocoagulation process using iron and aluminum electrodes. The obtained results indicate that, in removal of Remazol Black B through electrocoagulation process, at a reaction time of 80 minutes, following enhancing electrical conductivity from 1000 to 3000 µs/cm, using iron and aluminum electrodes at pH of 6 and 5, leads to increases of energy and metal consumption rate which in turn increases the removal efficiencies to 96% and 89.45%, respectively. By increasing the contact time and electrical conductivity, the energy consumption rate and consequently the removal efficiency enhances. When using both iron and aluminum electrodes, the output wastewater is alkaline and needs to be concerned about once releasing to the environment. Furthermore, the results indicate higher dye removal efficiency of iron electrode from aqueous solution compared to the aluminum one with slightly more energy consumption and the same electrical conductivity. In general, the results of this research exhibited that, the electrocoagulation method using iron and aluminum electrodes is an appropriate method for removal of reactive dyes from wastewater in laboratory scale.

CONFLICT OF INTEREST

There is no conflict of interest.

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None

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