Efficient De-colorization of Methylene Blue by Electro-coagulation Method: Comparison of Iron and Aluminum Electrode

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ABSTRACT: In this study, removal of methylene blue by electro-coagulation method using aluminum and iron electrodes was investigated. The influence of the operating parameters such as contact time, current density, anode type, inter-electrodes distance, initial and final pH and energy consumption rate was determined. Dye removal was increased with increases in solution pH, current density and contact time and then decreased for increase in initial dye concentration and electrodes distance. The results show that the electrochemical method has significant efficiency in removal of methylene blue, higher efficiency was observed for iron (Fe) electrode; namely 100% and 95.78% of dye was removed by iron and aluminum electrode; respectively, after 24 min contact time. For a given current density, the removal efficiency and energy consumption rate showed that iron electrode was superior to aluminum in removal of methylene blue. In the case of iron as anode type, the required energy for complete dye decolorization was 3.8 kWh/m³; for 98% dye removal, the required energy was observed to be 4.3kWh/m³ in the case of aluminum as anode type. In general, complete methylene blue can be removed at operating parameters condition of iron as anode, distance between electrodes of 1cm, solution pH of 9 and current density of 50 A/m² for 24 min electro-coagulation time.

KEY WORDS: Electro-coagulation, Methylene blue, Iron electrode, Aluminum electrode.

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INTRODUCTION

Dyes are widely used in many industries such as textiles, printing, paper, plastic, leather and so on. Textile industry use large amount of produced dyes in world [1]. Textile industries produce wastewaters that are the source of important pollution problems in environment that need an urgent solution [2]. It is estimated that up to 15% of the dyes are released in wastewater due to their limited fixation efficiency [3]. Unused dyes from textile industries must be removed as they represent an undesirable color to water and in some cases they are harmful compounds and can develop dangerous by-products through oxidation, hydrolysis, and or other chemical reactions taking place in the waste solution [4]. In addition, such wastewater may contain chemicals, which are toxic, carcinogenic, mutagenic, or teratogenic in various microbiologic [5]. Azo dyes are the greatest group of synthetic dyes and used as coloring agents in the textile, paint, ink and plastic industries, accounting for 50% of all commercial dyes [6]. They are characterized by nitrogen double bond (-N=N-) that together, with other chromospheres, are responsible of the color [7]. Even at low concentration (1 mg/L), dyes are undesirable and could cause an aesthetic pollution and disturbance to the ecosystem and water sources. Dyestuff effluents are characterized as high or low pH, high temperature and a high concentration of coloring material [8]. In this view, various methods such as of biological treatment, chemical coagulation, activated carbon adsorption, ultrafiltration, ozonation and electro-coagulation has been used for removal of dyes from wastewater [9-12]. Electrocoagulation is a promising technology in the field of water and wastewater treatment. This technology rely upon the electrochemical dissolution of sacrificial metal electrodes (usually iron or aluminum) into soluble or insoluble species that enhance the coagulation, the adsorption or the precipitation of soluble or colloidal pollutants [13]. In the electro-coagulation process, coagulant is produced continuously in water by the electrochemical dissolution of electrode material [14].

In brief, the following reactions are occurred in the case of aluminum and iron electrodes [15, 16]:

$$Al^{3+} + 3e^{-} \rightarrow Al \qquad E_0 = -1.662V/NHE$$
 (1)

$$Fe^{2+} + 2e^{-} \rightarrow Fe \qquad E_0 = -0.447 \text{V/NHE}$$
 (2)

At pH higher than 6, ferrous ions would be precipitated. In addition, the ferrous ions (Fe (II)) will be oxidized easily to ferric ions in the presence of oxygen as follows:

$$2Fe^{2+} + 1/2O_2 + H_2O \rightarrow 2Fe^{3+} + 2OH^-$$
 (3)

At the cathode and the anode water will be hydrolyzed as follow: occurs at the:

$$2H_2O + 2^e \rightarrow H_2 + 2OH^- \qquad E_0 = -0.828V/NHE$$
 (5)

$$2H^+ + 1/2 O_2 + 2e^- \rightarrow H_2O \quad E_0 = 1.229V/NHE$$
 (6)

The dissolution of iron and aluminum in water will produce a gelatinous suspension, which can remove the pollutants from the wastewater by either complexation or electrostatic attraction followed by coagulation. The formation of gelatinous compound is as follow:

$$Fe^{3+} + 3H_2O = Fe(OH)_3 + 3H^+$$
 (7)

$$Al^{+3} + 3H_2O = Al(OH)_3 + 3H^+$$
 (8)

The formation of gelatin Fe(OH)₃ and Al(OH)₃ gelatinous compounds may be responsible for removal of pollutant by adsorption/complexation process. By this view, the present work investigated the removal of Methylene Blue by electro-coagulation with iron and aluminum electrode. Various experimental parameters such as current density, contact time, energy consumption rate, anode type, initial solution pH and initial dye concentration were investigated in term of dye removal efficiency.

EXPERIMENTAL SECTION

Electro-coagulation cell

The experimental equipment schematically is shown in Fig. 1. The electro-coagulation unit consisted of a 2 L plexiglas reactor. Four electrodes (iron and aluminum) connected in a bipolar mode in the electrochemical reactor, with dimensions of $11~\rm cm \times 12~\rm cm \times 0.2~\rm cm$ and at a distance of 2cm from the bottom of reactor. Total effective surface area of electrodes was 528 cm². Electrodes were connected to direct current power supply (ADAK PS808, Iran) with 30 V as maximal tension and 50 A as maximal intensity.

Dye Solution

Methylene Blue with $C_{16}H_{18}N_3SCl$ formula and 319.85 g/mol and molar mass as analytical grade

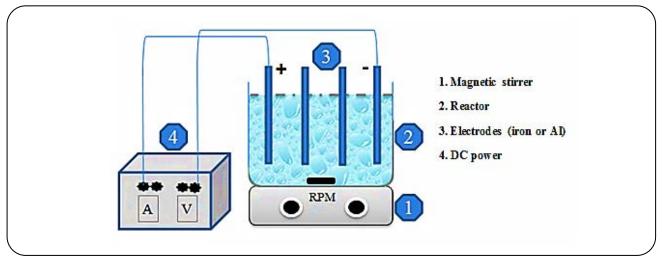


Fig. 1: Schematic of used eectro-coagulation.

Fig. 2: Chemical structure of used methylene blue.

was purchased from Merck Co (Germany). The chemical structure of used dye is showed in Fig. 2. Dye solution was prepared by adding appropriate amount of dye into deionized water. Practical solutions were prepared daily by dilution of stock dye solution. Initial electrical conductivity and solution pH of dye solution was 52-54 mS/cm and 5.5; respectively. Dye concentration was measured by plotting standard curve at maximum adsorption wavelength of 663 nm by using spectrophotometer (Shimadzu, Tokyo, Japan; Model 1601).

Electro-coagulation experiments

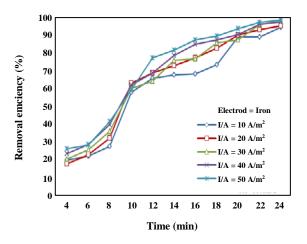
The solution pH was adjusted and controlled by adding 1NNaOH or H₂SO₄ during experiments (model E520, Metrohm Herisau, Switzerland). Before and after electro-coagulation, the electrodes were first washed with dilute HCl (5% v/v) for 15 min and then rinsed with deionized water for remove unused acid. For electro-coagulation experiments, about 2L of synthetic dye solution was introduced to the electro-coagulation

reactor. The electro-coagulation cell was kept in a magnetic starrier and then reactor content was mixed at 100 rpm. All experiments were conducted at room temperature (21±2°C). Several experimental parameters such as current density (10 to 50 A/m²), electrocoagulation time (4 to 24 min), electrodes distance (1 to 3 cm), solution pH (3 to 9) and initial dye concentration (50 to 200 mg/L) were studied. For electro-coagulation, as noted above, 2 L of dye solution was introduced to reactor and then the current dentistry was adjusted to the desired value. After that, the reactor content was magnetically stirred during experiments and at predetermined time interval, the sample was taken; filtered and final dye concentration was determined using a visible spectrophotometer at 663 nm. For example, in the investigation of influence of reaction time and current density on dye removal efficiency, about 2 L of dye solution with 50 mg/L concentration with pH = 5.5 was introduced to reactor. The electrodes distance was adjusted as 1 cm and in bipolar mode and then mixed at 100 rpm. At predetermined time interval, the samples were taken; filtered and final dye concentration was measured. For all other parameters, the same experimental arraignment was conducted.

RESULTS AND DISCUSSION

Influence of contact time and current density

The current density is one of the main important factors which can influence electro-coagulation process economics. Increases in current density will increase ions



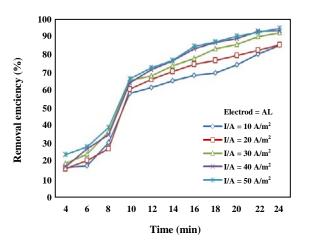


Fig. 3: Removal of methylene blue as a function of current density and electro-coagulation time [Dye concentration = 50 mg/L, electrode distance = 1 cm, pH=5.5, T= $21\pm2^{\circ}\text{C}$, 100 rpm agitation speed].

production on the anode/cathode, leading to an increase in flock production in the solution and hence an improvement in the efficiency of color removal [17]. In addition, the optimal current density will depend on the geographical as well as the economic situation where the electro-coagulation process is utilized [18]. Therefore, in the present work, a current density ranged from 10 to 50 A/m² was used. Fig. 3 shows the effect of current density and contact time on dye removal. As can see from Fig. 3, removal efficiency was increased for both electrodes with increases in current density and reaction time. After 24 min reaction time and with 50 A/m² current density, 98% and 100% of methylene Blue was removed with aluminum and iron electrode, respectively. In addition, at 50 A/m² current density and with increase in reaction time from 4 to 24 min, dye removal efficiency increased from 24% to 98% and from 28% to 100% for aluminum and iron electrode, respectively. An increase in dye removal percentage with increase in reaction time is accordance to Faraday's law. Based on Faraday's law, increases in electrolysis time leads to increase in Al+3 and Fe+3 species in the electrocoagulation reactor and their hydroxide flocs so the electro-coagulation process yield is enhanced. In addition, at higher current density the generated bubbles improve the mixing of Al+3 and Fe+3 hydroxides and dye molecules and further the separation of the flocs by flotation [19]. Therefore, the 50 A/m² current density and 24 min contact time were used for future experiment.

Similar work was reported for removal of dyes with

electro-coagulation. In removal of levafix brilliant blue using aluminum and iron electrodes, 97.8% of dye was removed at 200 A/m² current density and 10 min reaction time [20]. In another work for removal of Indigo Carmine dye with iron electrode after 3 h of electro-coagulation a 96% decolorization rate was reported at 10.91 A/m² and 99.9% removal efficiency at 54.57 A/m² after 60 min contact time [21]. In removal of acid red 14, after 24 min electro-coagulation time, about 91.12% of dye was removed with 60 A/m² current density [18]. By comparison with reported works, the results of present work well demonstrate that methylene blue will be removed at lower contact time and current density.

Influence of electrodes distance on dye removal and energy consumption

In the electro-coagulation, the ohmic potential drop is proportional to the distance between electrodes. Therefore, when the distance of the electrodes increases, the electrolysis energy consumption will be increased especially when the conductivity is low. For this reason, effect of electrodes distance raging from 1 to 3 cm on dye removal and energy consumption was investigated and results are shown in Fig. 4. Results reveals insignificant effect of electrodes distance on dye removal percentage and energy consumption; however, higher removal efficiency and lower energy consumption was observed at 1 cm electrode distance. At 1 cm inter-electrode distance, 100% of methylene blue was removed by iron electrode with 4.5 kWh/m³ energy consumption and 98% of dye

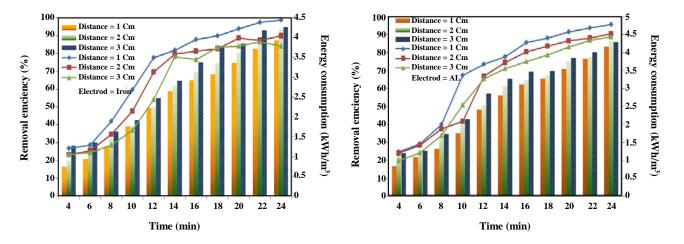


Fig. 4: Influence of electrodes distance on removal of methylene blue and energy consumption rate. [Current density: 50 A.m^{-2} , pH=5.5, $T=21\pm2^{\circ}C$, 100 rpm agitation speed, 50 mg/L dye concentration].

was removed by aluminum electrode with 4.9 kWh/m³ energy consumption. Slightly decreases in dye removal and increases in energy consumption with increases in electrodes distance may be due to electrostatic effects of electrodes distance. As electrodes distance increased, the movement of Al⁺³ and Fe⁺³ hydroxides would be slower and they would have lower opportunity to mix with dye molecules and they may be precipitated in near of electrodes. In addition and as expected, for removal of a given percentage of dye, higher energy will be consumed with increases in inter-electrode distance [18].

Various results were reported for dye removal in term of energy consumption. In removal of Levafix Brilliant Blue using aluminum and iron electrodes, higher energy consumption was observed for iron electrode rather than aluminum electrode. In addition, it is reported that for complete dye removal (100%), 34kWh/m³ was used at 200 A/m² current density [20]. In another work for removal of Acid Red 14, it is reported that with increases in inter-electrode distance no significant effect on the efficiency of color removal was observed. However, with increase in electrode distance, the efficiency of color removal decreased slightly [18]. In removal of orange II, increases in the distance of the electrodes from 1 to 4 cm leads to decreases in the removal efficiency by about 5.61% [22].

Therefore we can best conclude that increase in electrodes distance will reduce pollutant removal efficiency due to precipitation of Al^{+3} and Fe^{+3} hydroxides before adsorption/complexation with dye

molecules. In addition, as inter-electrode distance increased, the required energy/energy consumption rate for removal of constant dye molecules will be increased especially when electrolyte concentration is low.

Influence of initial solution pH on dye removal

Initial pH is an important parameter which influences the electro-coagulation performance. For that reason, dye solution with different initial pH (3-5-7-9) was prepared and introduced to electro-coagulation cell and results are shown in Fig. 5a and Fig. 5b for iron and aluminum electrodes, respectively. As it is shown in Fig. 5 (a-b), methylene blue removal is completely depends on the initial pH value and at the highest initial pH value; dye removal efficiencies were higher for both electrodes. At pH=9 and after 24 min contact time, 100% of dye was removed by both electrodes.

Increases in dye removal at higher pH values may be due to the nature of the reaction between Al⁺³ and Fe⁺³ ions and hydroxide ions. At higher pH values, more iron and aluminum electrodes will be oxidized to Al⁺³ and Fe⁺³. On the other hands, at alkaline environment (pH=9) more OH⁻ species will exist, therefore more Al⁺³ and Fe⁺³ hydroxides gelatinous compound will be formed; leading to increase in pollutant removal efficiency [18].

Similar results were reported for removal of Acid Red 14 [18], levafix brilliant blue E-B [20] and orange II [22]. In addition, the final solution pH was increased for both electrodes which this increases were higher for iron electrode rather than aluminum electrode. On the other hands,

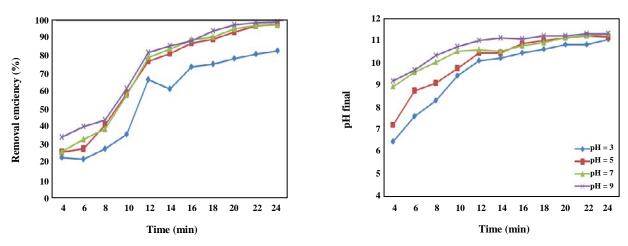


Fig. 5a: Dyeremoval efficiency as a function of initial solution pH for iron electrode. [Current density: 50 A/m^2 , $T = 21 \pm 2^{\circ}C$, 100 rpm agitation speed, 50 mg/L initial dye concentration].

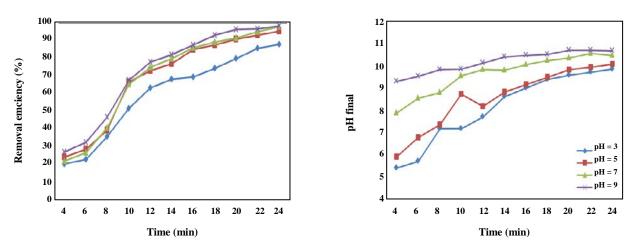


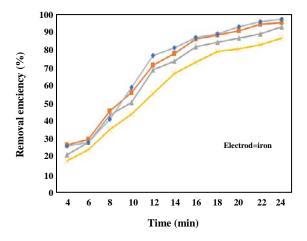
Fig. 5b: Dye removal efficiency as a function of initial solution pH for aluminum electrode. [Current density: 50 A/m^2 , $T = 21 \pm 2^{\circ}C$, 100 rpm agitation speed, 50 mg/L initial dye concentration].

the increases in final solution pH were highest for initial lower pH values. In other mean, this phenomenon well demonstrates that electro-coagulation act as pH naturalization. Higher increases for lower initial solution pH may be due to the excess of hydroxyl ions produced at the cathode by the liberation of OH⁻ due to the occurrence of a partial exchange of Cl⁻ with OH⁻[19]. The variation of final solution pH was insignificant for higher initial solution pH (pH 7-9). This phenomenon may be due to the consumption of the OH⁻ ion and the formation of Al (OH)⁻4 [23].

Influence of initial dye concentration

The effect of initial dye concentration on the removal efficiency has been studied in the range of 50 to

200 mg/L. As shown in Fig. 6, the removal efficiency of methylene blue is reduced as the initial dye concentration is increased. Also, the decrease in the removal efficiency is considerable with higher initial dye concentration. Therefore, it is clear that the removal efficiency of methylene blue at a given time is decreased by increasing the initial dye concentration. After 15 min contact time and with initial dye concentration of 50, 100, 150 and 200 mg/L about 98.24, 96.16, 93.72, 87.49% and 95.87, 93.73, 88.67 and 86.39 % of methylene blue were removed by iron and aluminum electrodes, respectively. The decreased in dye removal efficiency with increases in initial dye concentration is due to the fact that the amounts of dissolved Al⁺³ and Fe⁺³ ions are constant at the same constant current density and time for all initial dye



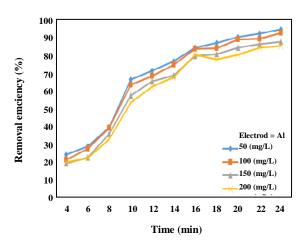


Fig. 6: RemovalMethylene Blueas a function of initial dye concentration. [Current density: 50 A/m^2 , pH=9, $T = 21 \pm 2^{\circ}\text{C}$, 100 rpm agitation speed].

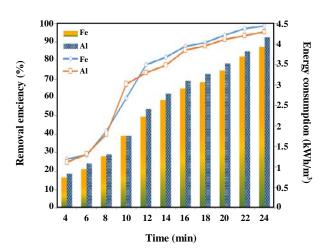


Fig. 7: The removal efficiency of methylene blue as a function of anode type with electro-coagulation time and energy consumption rate.

concentrations (according to Faraday's law). While the removal efficiency of dye has decreased, the amount of dye removed by increasing the initial dye concentration has increased. It means, after 24 min contact time and with initial dye concentration of 50, 100, 150 and 200 mg/L about 49.12, 96.16, 140.58, 174.98 mg and47.935, 93.37, 133.005 and 172.78 mg of methylene blue were removed from the solutions by iron and aluminum electrodes, respectively. The increases in amount of removed dye as increases in initial dye concentration can explained by the fact that; at constant amount of Al⁺³ and Fe⁺³ ions and higher amount of dye concentration, dye molecules

have more chances of bounding with Al⁺³ and Fe⁺³ions leading to increase in amount of removed dye [20,21]. Similar results were reported for removal of acid red 14 [18], acid brown 14 [24] and Indigo Carmine [21].

Influence of Anode type

Different connection of electrodes is very important factor affecting the performance of electro-coagulation process. The following reaction was proposed for iron and aluminum as anode type [25]:

For Al anode:

$$Al \rightarrow Al^{+3} 3e^{-}$$
 (9)

For Fe anode:

$$Fe \rightarrow Fe^{+2} 2e^{-}$$
 (10)

Therefore in present work, the influence of anode type on dye removal efficiency and energy consumption rate was studied and result is shown on Fig. 7. The percentage of removed dye was observed to be as high as 100% and 98% by iron and aluminum electrode, respectively. The higher removal percentage for iron electrode may be due to the higher oxidation potential of Fe (-0.447 V) than A1⁺³ (-1.662 V). In the case of iron metal as anode, the generation of Fe⁺³ ions is nearly three time greater than A1⁺³ which causes the higher methylene blue removal efficiency [26]. Many researchers have reported that the removal efficiency of dye molecules by Al(OH)₃ compounds is much lower than those using Fe(OH)₃ [27]. *Wei-Lung* reported, from the two reactions (9) and (10),

the electrochemical equivalent mass of 18.59 mmol/Ah and 12.43 mmol/Ah for Al and Fe, respectively [28]. Based on above comments, more coagulants will be theoretically produced by iron anodes when the same electrical charge is applied.

Similar results were reported by many researchers in the field of electro-coagulation. In removal of COD from distillery effluent, it is reported that removal percentage is higher in the case of iron electrode as anode [25]. In another work for removal of blue reactive, red disperse and mixed dyes, higher removal efficiency was reported for iron electrode as it act and anode type [26]. In removal of arsenic from water by electro-coagulation [27] and in the removal of polyvinyl alcohol from aqueous solutions using electro-coagulation [8] higher removal efficiency has reported in the case of iron as anode which was in accordance with our results.

In addition, lower energy consumption was observed for iron electrode as anode rather than aluminum. As can see from the left side of Fig. 7, the required energy for complete removal of methylene blue was ranged between 3.6-3.8 kWh/m³ in the case of iron metal as anode. For removal of 98 percentage of dye with aluminum electrode and anode, the required energy was 4.3kWh/m³.

CONCLUSIONS

In this study, the removal efficiency of methylene blue in term of contact time, current density, anode type, inter-electrodes distance, initial and final pH and energy consumption rate was investigated in a bipolar electrochemical cell. The overall data showed that iron was superior to aluminum in treating methylene blue dyes in term of removal efficiency and energy consumption rate. The suitable choices of values of some parameters were: iron as anode, distance between electrodes of 1 cm, solution pH of 9 and current density of 50 A/m for 24 min electro-coagulation time.

Acknowledgments

The authors gratefully thank Zahedan University of Medical Sciences for financial supporting of present work.

Received: Jan. 25, 2013; Accepted: Jan. 19, 2015

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