Original Research Article
Electrochemical Decolorization of Maxilon Red GRL Textile Dye

ABSTRACT

Aims: The aim of this research paper is to examine the efficiency of electrooxidation and electrocoagulation techniques to remove the textile dye Maxilon Red GRL from polluted water.

Study Design: The study was performed by using two graphite electrodes as anode and cathode for electrooxidation (EO) and a stainless steel electrode as anode and graphite as cathode for electrocoagulation (EC).

Place and Duration of Study: Chemistry Department, Faculty of Science, Benha University, 13518 Benha, Egypt, between April 2013 and July 2013.

Methodology: The experiments were carried out in a glass beaker containing 100 ml of dye solution. The two electrochemical techniques; EO and EC were performed to optimize both methods for maximum dye removal. The removal process was studied at different conditions of pH, treatment time, electrolyte concentration, current density, potential difference between electrodes and initial dye concentration.

Results: The results indicate that the optimum pH for EO is 5, while that for EC is 9. The increase of current density and cell voltage results in a corresponding increase in the removal efficiencies. Complete decolorization was achieved after 120 and 90 minutes for the EO and EC processes, respectively.

Conclusion: High levels of elimination of Maxilon Red GRL dye by EO or EC reached nearly 100% were detected by optimizing the experimental conditions.

Keywords: Maxilon Red GRL, electrooxidation, electrocoagulation, graphite, stainless steel

1. INTRODUCTION

Textile wastewater has a high content of pollutants, the sources of which are the natural impurities extracted from cotton fiber, processing chemicals and dyes. The discharge of this wastewater to the environment causes aesthetic problems due to the color and also damages the quality of the receiving water because many of dyes released and their breakdown products are toxic, carcinogenic or mutagenic to life forms [1]. Azo dyes are the largest group of dyes used in the textile industry [2] constituting 20–40% of the dyes used for dyeing cotton, rayon, silk, wool, nylon and leather [3,4]. They have one or more azo groups (R_1–N=N–R_2) having aromatic rings mostly substituted by sulfonate groups. These complex aromatic conjugated structures are responsible for their intense color, high water solubility and resistance to degradation under normal conditions [5,6]. Therefore, most of traditional methods of wastewater treatment are becoming inadequate. Electrochemical technology and its applications on wastewater treatment have become increasingly interesting; because of its advantage especially for color removal [7,8]. Electrochemical techniques are considered to be powerful means for the treatment of dyeing wastewater. Indeed, electrochemical methods have been successfully tested [9] and proved significant advantages such as easy operation and lower operating cost [10–12]. Electrochemical process requires significantly less equipment than conventional biological treatment processes [13,14]. Electrochemical treatment is relatively a new trend in the azo dyes wastewater management with promising results [15,16]. The electric current induces redox reactions resulting in the transformation/destruction of the organic compounds and their virtually complete oxidation to CO_2 and H_2O. For electrochemical treatment of dyeing wastewater, direct and indirect oxidation processes are being employed. In a direct oxidation process, pollutants are first absorbed on the anode surface and then destroyed by the electron transferring reaction. In an

indirect oxidation process, strong oxidants, such as hydrogen peroxide, metal peroxide, hypochlorite, chlorine, ozone and Fenton reagents, are electrochemically generated [17] and then destroy the pollutants by the oxidation reactions. All of the oxidants are generated in situ and are utilized immediately [18]. In the past, graphite was frequently used as an anode for the electrochemical degradation of textile dye as it is relatively cheaper and gives satisfactory results [19]. Pollutants can be destroyed electrochemically by conducting a direct anodic or indirect oxidation process [20]. The direct electro-oxidation rate of organic pollutant is dependent on catalytic activity of the anode and the applied current intensity. Indirect electro-oxidation of pollutants, which was observed to be most efficient method on decolorization and mineralization, can be conducted when chloride, ferric or silver are present. The first being the most important from the point of view of practical application, as chlorides are common constituent of several industrial textile waste water. The indirect electro-oxidation rate is dependent on the diffusion rate of strong oxidants electro-formed into solution which are able to completely convert all organics into water and carbon dioxide [21].

Electrocoagulation is an alternative technology for wastewater treatment in addition to its other conventional applications. The main advantages of electrocoagulation over other conventional techniques, such as chemical coagulation and adsorption, are “in situ” delivery of reactive agents, no generation of secondary pollution, and compact equipment. The earlier studies have reported the potential of electrocoagulation to treat a variety of industrial and domestic wastewater [22-24].

The decolorization of MxR has been performed using photoelectron-Fenton process [25] and photocatalysis in the presence of TiO_2 under UV or sunlight irradiation [26] and in single and binary mixture with Basic Yellow 28 under TiO_2/periodate ion system using UV irradiation [27]. Peroxi-coagulation process has been used for treatment of aqueous solution of MxR using carbon nanotube–PTFE cathode [28]. The aim of the present work is to test two electrochemical methods for the decolorization of a cationic azo dye; Maxilon Red GRL (MxR) using graphite or stainless electrodes as anodes. Different parameters affecting dye color removal as current density, pH, salinity, potential difference and stirring speed were examined to optimize the electrochemical methods. The efficiencies of the two processes were compared and evaluated.

2.MATERIALS AND METHODS

2.1 Chemicals

The dye used in the present work was Maxilon Red GRL (MxR), a textile dye also known as Astrazon Red FBL; C.I. Basic Red 46; Cationic Red GRL, it’s chemical structure is 1,2-dimethyl-3-((4-(methyl(phenylmethyl)amino)phenyl)azo)-1,2,4 triazolium bromide. The chemical structures of the dye is given in Fig. 1. A stock solution of dye (100 mg/L) was prepared by dissolving an accurate quantity of the dye in deionized water and suitably diluted to the required initial concentrations. The pH of the working solution was adjusted to the desired values with 0.1N HCl or 0.1N NaOH. All other chemicals used were of analytical grade.

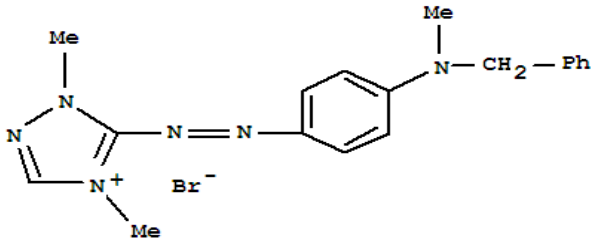

Fig. 1. Structural formula of Maxilon Red GRL

2.2 Equipment and procedures

Electrochemical methods for decolorization of the dye was carried out using an undivided electrochemical cell in a galvanostatic condition. The anode was either a graphite electrode

with an effective surface area of 11.10 cm^2 (Eveready Battery Co. Inc., Egypt) or stainless steel sheet (Stailess alloy 304, Tschno Steel, Egypt) with an effective area of 8.5 cm^2. The cathode used is a graphite cylinder in the two cases. The electrodes were positioned vertically and parallel to each other with an inter-electrode gap of 2 cm in a 100 ml of dye solution. A digital DC power supply (AD 302S: 30 V, 2A) was used as the source of constant electric current for the experiments. To enhance the mass transport and to maintain a uniform concentration of the electrolyte, the reactor solution was constantly stirred at 100 rpm using a magnetic stirrer. A schematic illustration of the experimental setup is shown in Fig. 2. The pH values were measured by using a pH-meter model HI 8014, Hanna Instruments (Italy). The initial pH of the dye solution was set appropriately using dilute solutions of either sodium hydroxide or hydrochloric acid. UV–visible spectra during the degradation of dye were recorded between 200 and 800 nm using a scanning UV–visible spectrophotometer (Jasco model V-530, Japan). The visible color removal for the dye was measured at optimum visible absorption wavelength of 530 nm in order to study the effects of various operating parameters. The dye (20 mg/L) was studied for complete color removal with a supporting electrolyte concentration of 2 g/L sodium chloride and a current density of 0.90 mA/cm^2, which was equal to 20 mA current input. Samples were drawn at convenient time intervals during the course of electrolysis and the color removal determination was done up to 210 min of electrolysis.

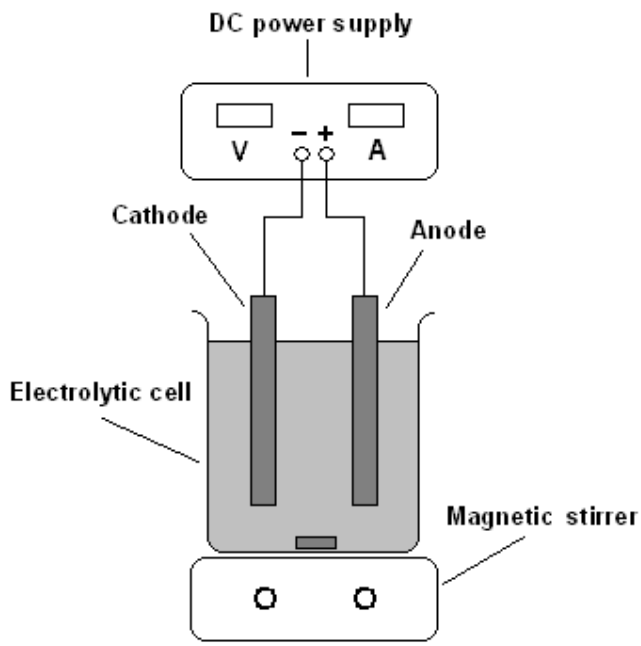

Fig. 2. Schematic diagram of the experimental setup for the electrochemical degradation

2.3. Analytical method

Dye concentration was determined spectrophotometrically by measuring the absorbance of the remaining dye at maximum wavelength 530 nm. Samples were withdrawn from the dye solution at certain time intervals (30, 60, 90, 120, 150, 180, and 210 min), centrifuged at 3000 rpm and analyzed. The percentage removal of dye and amount adsorbed (%E) were calculated using the following relationship

$$\%E = \frac{A_i - A_f}{A_i} \times 100 \quad (1)$$

where A_i and A_f are absorbance values of dyes solutions before and after treatment with respect to their λ_{max} [29].

3. RESULTS ANA DISCUSSION

3.1 Electrooxidation

Organic dye can be anodically destroyed in two different mechanisms: direct and indirect electrooxidation. In direct oxidation, the pollutants are adsorbed on the anode surface and destroyed by the electron transfer reaction and in indirect oxidation the dye oxidized in the liquid bulk by the electrochemically mediated oxidants, such as chlorine.
The direct electro oxidation rate of organic pollutants (when no supporting electrolyte is used) depends on the catalytic activity of the anode, while the indirect electro oxidation rate of organic pollutants (when a supporting is used) depends on the diffusion rate of the oxidants into the solution, temperature and the pH [30].

3.1.1 Effect of electrolyte concentration

Experiments were carried out using graphite electrodes at four different NaCl concentrations keeping the other parameters constant. The effect of NaCl on the dye decolorization is illustrated in Fig. 3. The results showed that the increase in NaCl concentration (from 1 to 4 g/L) leads to a corresponding increase in the decolorization rate. The presence of chloride ions plays an important role in the electrooxidation process by two ways. An increase in the concentration of the supporting electrolyte increases the hypochlorite ions production and the cell conductivity [31]. The former enhances the destruction of organics present in the solution and the latter can result in a rise of current density at the same voltage which provides more chance for the production of chlorine in the relatively higher current densities [32]. The electrochemical reactions that take place during the electrolysis are complicated and the following reactions are assumed to take place [33]:
At anode:
Main reaction $2Cl^- \rightarrow Cl_2 + 2e^-$ (2)
Side reaction $4OH^- \rightarrow O_2 + 2H_2O + 4e^-$ (3)
At cathode:
$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ (4)
In bulk of solution:
$Cl_2 + H_2O \rightarrow H+ + Cl^- + HOCl$ (5)
$HOCl \leftrightarrow H^+ + OCl^-$ (6)
$Dye + OCl^- \rightarrow CO_2 + H_2O + Cl^- + P$ (7)
The primary reaction occurs at the anode is the oxidation of chloride ion (2) with the liberation of Cl_2, which is a powerful oxidizing agent. In the same time a side reaction (3) takes place generating oxygen which is relatively weak oxidant leading to reduction of current efficiency. At the cathode, the counter reaction (4) would be the reduction of water when no other reducible species are present. In the bulk of solution, gaseous Cl_2 dissolves in the aqueous solutions due to ionization as indicated in (5). The rate of reaction is less in acidic solution due to OH^- instability and considerably more in basic solution due to ready formation of OCl^- ion in (5) which implies that the basic or neutral pH conditions are more favorable for conducting reactions involving Cl_2 [34].

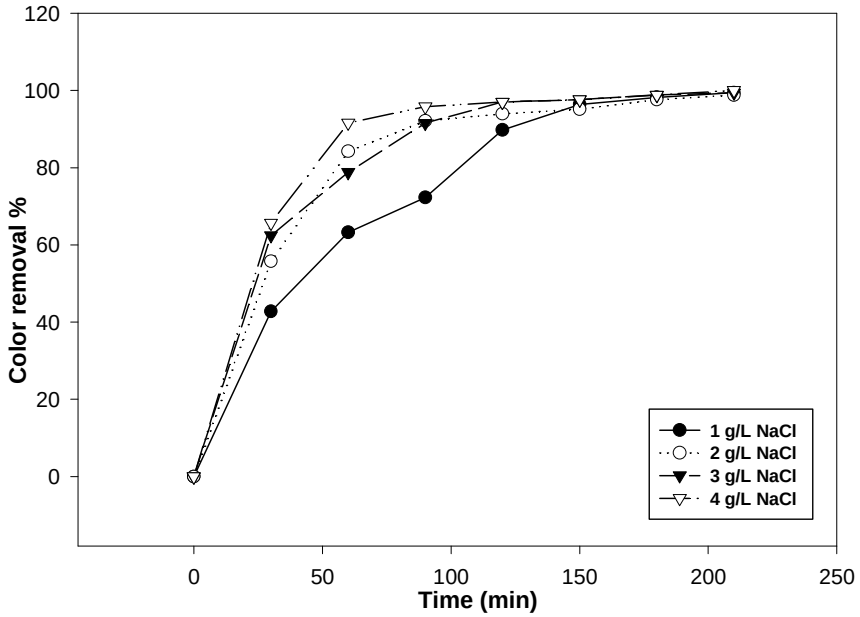

Fig. 3. Effect of electrolyte concentration on the color removal efficiency of MxR dye by EO (conditions: [dye]=20 mg/L, I=0.90 mA/cm^2, pH=5, stirring speed=100 rpm).

3.1.2 Effect of initial pH

To determine the role of pH in the decolorization process, the experiments were performed at different pH values (2.5, 5, 7, 9) in presence of a fixed amount of NaCl (2 g/L). Fig. 4 shows that the effect of pH on the decolorization efficiency of dye increased from 2.5 to 5 and then decreased gradually, which is in agreement with previous researches [35,36]. In acidic conditions, hypochlorous acid was predicted to be the most species in the solution. Therefore higher rate of decolorization in acidic condition may be due to higher oxidation potential of hypochlorous acid rather than hypochlorite. At neutral pH, the rate of reaction was lowest. This result can be attributed to undesirable side reactions such as oxidation of free chlorine to chlorate and perchlorate, formation of chlorate by chemical combination of HOCl and hypochlorite ions which are at equilibrium in water, electrolysis of water and cathodic reactions involving loss of hypochlorite [37]. At basic pH, a moderate rate of removal can be observed. This moderate value can be due to the presence of less potent hypochlorite ions which are the main oxidizing species in the bulk.

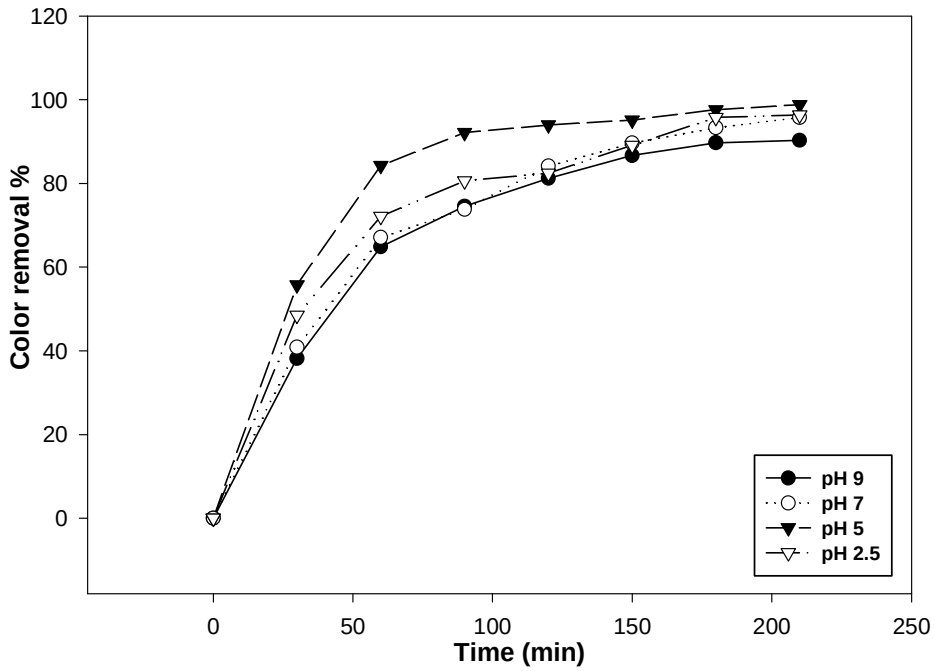

Fig. 4. Effect of pH on the color removal efficiency of MxR dye by EO (conditions: [dye]=20 mg/L, I=0.90 mA/cm^2, NaCl=2 g/L, stirring speed=100 rpm).

3.1.3 Effect of current intensity

Current density is an important variable in electrochemical applications. The effect of varying current density at constant electrolyte concentration (2 g/L NaCl) and pH 5 is shown on Fig. 5. It is remarkable that, as current intensity increases, the dye degradation efficiency increases. Color removal percent was 98.80%, 73.21%, 68.07%, 24.85% at current densities 0.90, 0.45, 0.23, 0.11 mA/cm^2, respectively after 180 min of electrolysis. This behavior is expected [34] because of the increased rate of generation of oxidants, such as chlorine/hypochlorite and hydroxyl radicals at higher current densities [38].

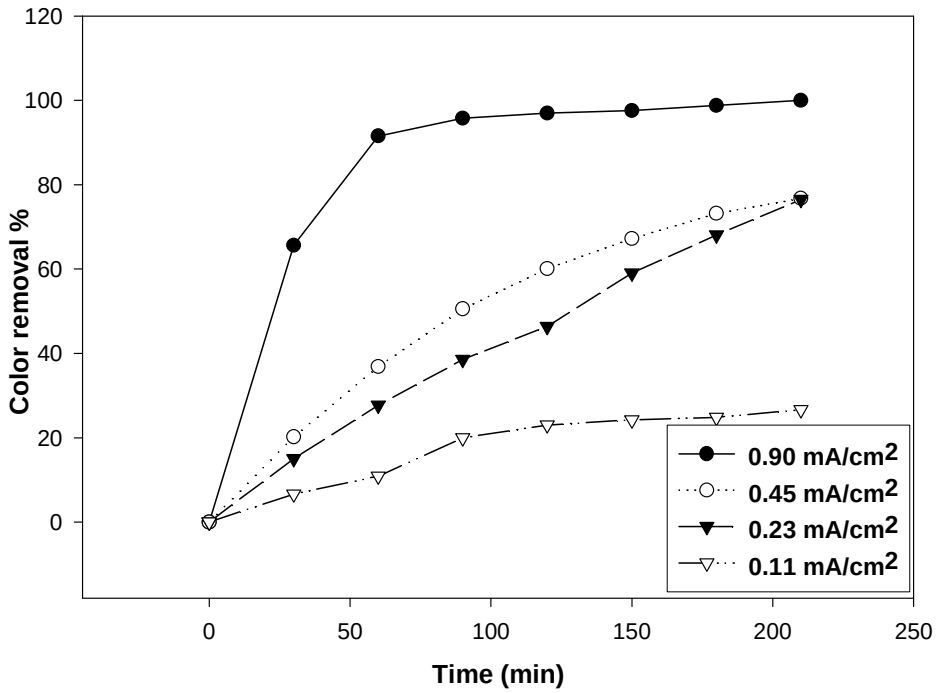

Fig. 5. Effect of current density on the color removal efficiency of MxR dye by EO (conditions: [dye]=20 mg/L, NaCl=2 g/L, pH=5, stirring speed=100 rpm).

3.1.4 Effect of cell voltage

It was found that dye degradation depends on the cell voltage; as voltage increased from 3 V to 12 V, the formation of chlorine increased that greatly increased the rate of dye degradation. Electrolysis gave 91.57 %, 95.15 %, 94.55 %, and 97.58 % of color removal after 60 min for 3V, 6V, 9Vand 12V, respectively. The steady increase in color removal efficiency with increasing cell voltage is in contact to other workers [39,40]. The difference between the color removal efficiency for applied voltage of 3 and 15 V is just about 6 %, which can not be an acceptable benefit for choosing high potentials as the best one because of energy consumption consideration.

3.1.5. Effect of initial dye concentration

It was of a particular interest to investigate the maximum loaded dye concentration on the effectiveness of the graphite electrode in treating MxR dye from wastewater. Fig. 6 displays the effect of different initial dye concentrations (20-80 mg/L) on the rate of dye degradation and corresponding color removal. Total color removal of the dyes can be achieved in the presence of initial dye load up to 80 mg/L. The highest removal rate of color removal was observed at dye concentration 20 mg/L. However, above that concentration of MxR dye the color removal rate did not changed. The results can be explained in terms of diffusion control, assuming that mineralization occurred on the electrode surface mediated by OCl^- ions. At low initial concentrations, the electrochemical reaction is faster than the diffusion. When the initial concentration increases, more organic molecules are transferred to the surface of electrode and the amount of dye reduction is increased. The OCl^- ions that are generated are limiting in this case, and the degradation efficiency would decrease with increased initial concentration of the dye [41].

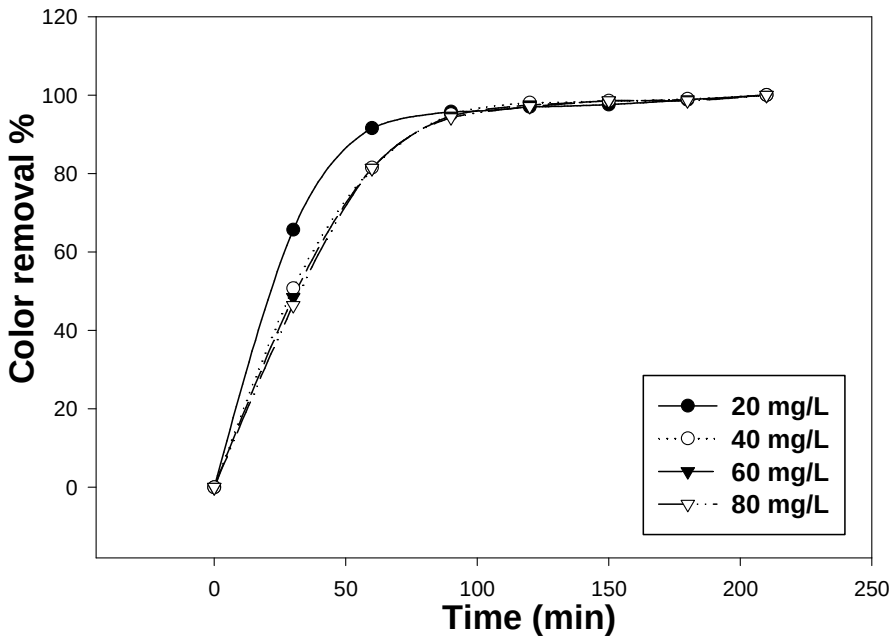

Fig. 6. Effect of initial dye concentration on the color removal efficiency of MxR dye by EO (conditions: I=0.90 mA/cm^2, NaCl=2 g/L, pH=5, and stirring speed=100 rpm).

3.2. Electrocoagulation (EC)

In the electrocoagulation process, coagulant is generated as a result of oxidation of the anode material by passing the electrical current. With iron anode is used, $Fe(OH)_2$ or $Fe(OH)_3$ is formed. Reaction mechanism of iron electrode at the anode and cathode are reported by many authors [42-44]:

(a) Mechanism 1:

Anode : $Fe\ (s) \rightarrow Fe^{2+}\ (aq) + 2\ e^-$

$Fe^{2+}\ (aq) + 2\ OH^-\ (aq) \rightarrow Fe(OH)_2\ (s)$

Cathode : $2\ H_2O\ (l) + 2\ e^- \rightarrow H_2\ (g) + 2\ OH^-\ (aq)$

Overall : $Fe\ (s) + 2\ H_2O\ (l) \rightarrow Fe(OH)_2\ (s) + H_2\ (g)$

(b) Mechanism 2:

Anode : $4\ Fe\ (s) \rightarrow 4\ Fe^{2+}\ (aq) + 8\ e^-$

$4\ Fe^{2+}\ (aq) + 10\ H_2O\ (l) + O_2\ (g) \rightarrow 4Fe(OH)_3\ (s) + 8\ H^+\ (aq)$

Cathode : $8\ H^+\ (aq) + 8\ e^- \rightarrow 4\ H_2\ (g)$

Overall : $4\ Fe\ (s) + 10\ H_2O\ (l) + O_2\ (g) \rightarrow 4\ Fe(OH)_3\ (s) + 4\ H_2\ (g)$

The generation of iron hydroxides Fe(OH)n is followed by an increase in the concentration of colloids (usually negatively charged) in the region close to the anode [45]. The produced ferrous ions hydrolyze to form monomeric hydroxide ions and polymeric hydroxide complexes that depend on the pH of the solution. The polymeric hydroxides, which are highly charged cations, destabilize the negatively charged colloidal particles allowing the formation of flocs. When the amount of iron in the solution exceeds the solubility of the metal hydroxide, the amorphous metal hydroxide precipitates is formed, which causes sweep-floc coagulation [46].

3.2.1 Effect of electrolyte concentration

It is important to investigate the effect of electrolyte since actual wastewater usually contains certain amount of salts [47]. The electrolyte concentration affects the conductivity. The increase in the conductivity allowed by the addition of sodium chloride is known to reduce the cell voltage V at constant current density due to the decrease of the ohmic resistance of wastewater [48-50]. Chloride ions could significantly reduce the adverse effects of other

anions, such as HCO_3^- and SO_4^{2-}, for instance by avoiding the precipitation of calcium carbonate in hard water that could form an insulating layer on the surface of the electrodes and increase the ohmic resistance of the electrochemical cell [51]. Fig. 7 depicts the effect of electrolyte concentration on the performance of the EC process for MR using stainless steel electrodes. With high salt (NaCl) concentration (4g/L), the system started to remove color and gave highest percent 96.97 % of the color removed in about 60 min. As shown in the figure, the decrease of salt content decreases the removal rate and removal efficiency.

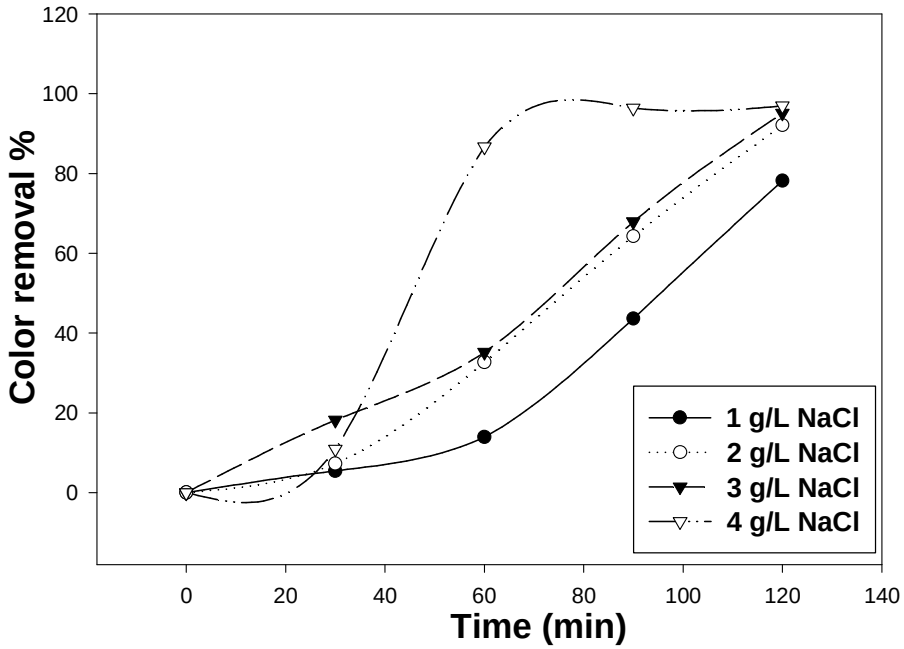

Fig. 7. Effect of electrolyte concentration on the color removal of MxR dye by EC (conditions: [dye]=20 mg/L, I=0.39 mA/cm^2, pH=5, stirring speed=100 rpm).

3.2.2 Effect of initial pH

It has been established in previous studies [52, 53] that initial pH has a considerable effect on the efficiency of the electrocoagulation process for removal of organic pollutants. Fig. 8 showed the effect of pH on dye removal efficiency using stainless steel electrode. From the figure, the removal efficiency is low in acidic electrolyte which is can only remove 77.58% at pH 2.5 after 120 min of continuous electrolysis. Meanwhile, in alkaline solution (pH 9.0), the removal efficiency is much higher reached 100% at the same period of electrolysis. So, from the Fig. 9, it can be concluded that the optimum pH values appeared between 7 and 9 which give the maximum removal percent.

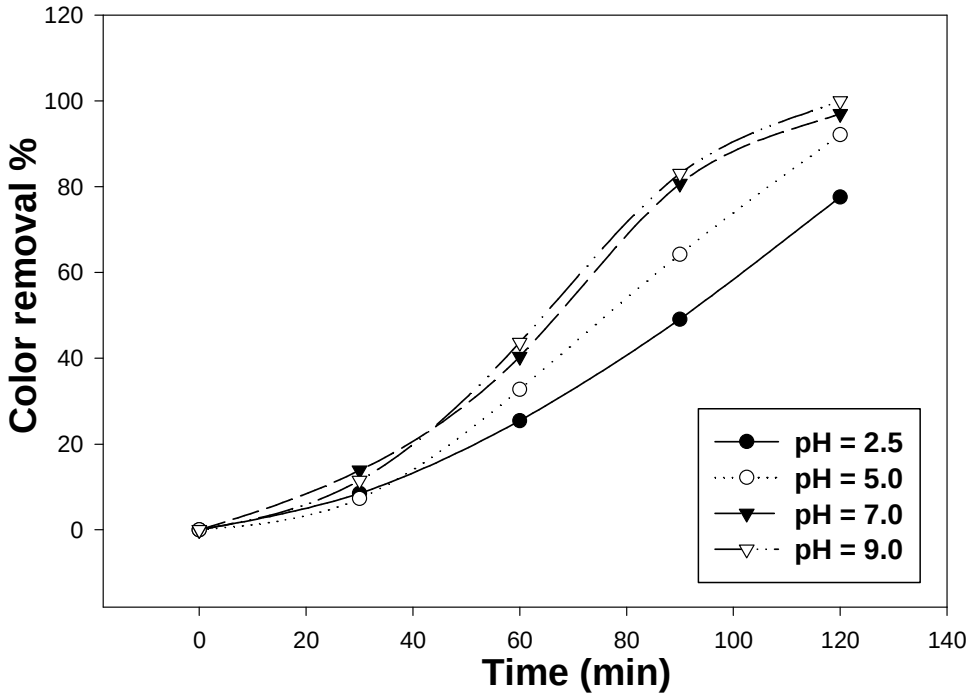

Fig. 8. Effect of pH on the color removal of MxR dye by EC (conditions: [dye]=20 mg/L, I=0.39 mA/cm^2, NaCl=4 g/L, stirring speed=100 rpm)

3.2.3 Effect of current density

The effect of current density on the removal of MxR dye was studied at four different current densities, i.e., 0.05, 0.10, 0.20 and 0.39 mA/cm^2 and the results were presented in Fig. 9. It is well known that anode dissolves both electrochemically and chemically [54]. As can be expected, the process efficiency, in terms of color removal and treatment time, is related to the current through the reactor. By increasing the current density, the anodic dissolution of metal will be increased resulting in a greater amount of precipitate generated in the solution and the time required for coagulation is reduced. Moreover, the rate of bubble generation increases and the bubble size decreases also with increasing current density, which are both beneficial for pollutant removal efficiency by hydrogen flotation [55].

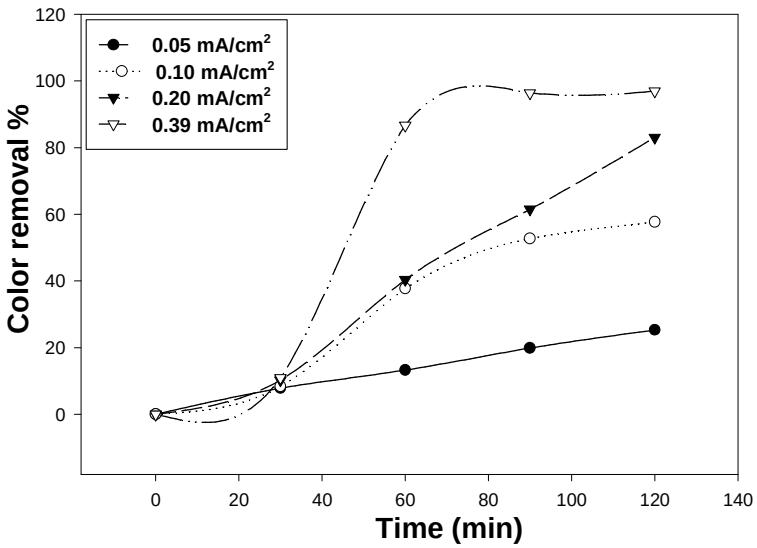

Fig. 9. Effect of current density on the color removal of MxR dye by EC (conditions: [dye]=20 mg/L, NaCl=4 g/L, pH=5, stirring speed=100 rpm).

3.2.4 Effect of cell voltage

Operating voltage as well as electric current is critical in electrocoagulation process. According to Faraday' s law, the amount of stainless steel dissolved electrochemically is proportional to charge loadings. Consequently, increasing current density will give rise to an increase in charge loading and leading to increased removal of the organic pollutant by generating much more oxidation products. After 120 min of electrolysis, color removal percents obtained were 25.30%, 84.85%, 79.58% and 99.39% by applying 3, 6, 9 and 12 V, respectively. It is clear from the obtained results that increasing the applied voltage cause an increase in both rate aand percent removal. According to Faraday's law, the amount of iron dissolved electrochemically forming $Fe(OH)_3$ is proportional to charge loadings. This behavior is expected as explained in literature [56].

3.2.5 Effect of initial dye concentration

The dye solutions with different initial concentrations in the range of 20–80 mg/L were treated by EC at current density of 0.39 mA/cm^2 for 60 min of continuous electrolysis. The initial dye concentration was plotted against related color removal percentage (Fig. 10). According to the results, up to the concentration of 50 mg l^{-1}, the removal capacity was not exhausted and the rate of color removal was relatively constant. However, the increase of initial dye concentration caused a steady increase in decolorization capacity. Tsantaki et al. [57] reported similar results for the decolorization of reactive dye solutions by EC process.

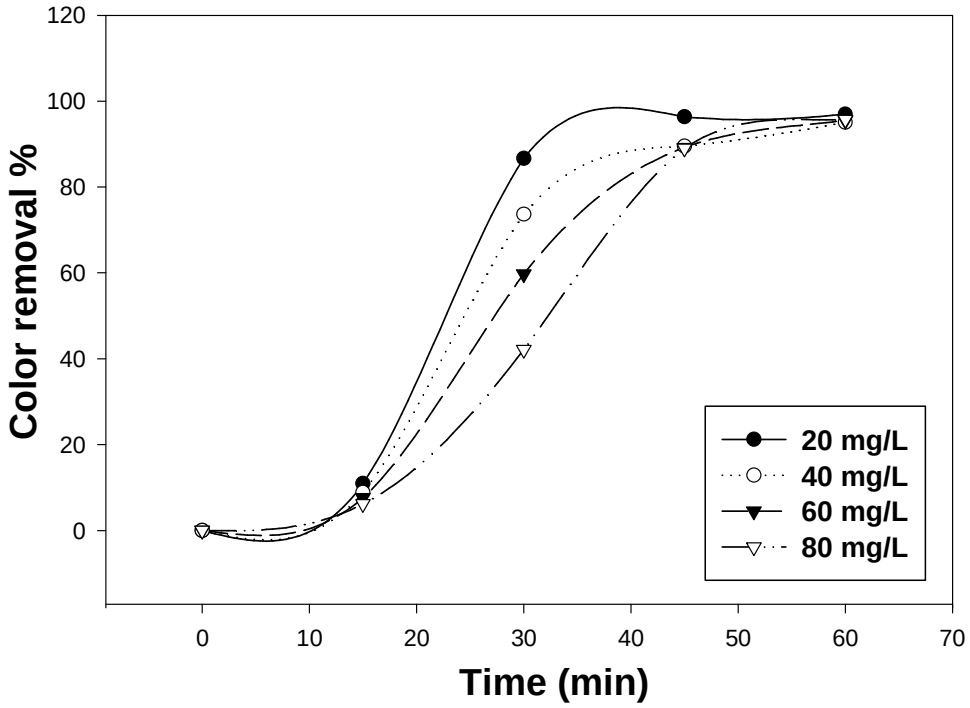

Fig. 10. Effect of initial dye concentration on the color removal of MxR dye by EC (conditions: I=0.39 mA/cm^2, NaCl=4g/L, pH=5, and stirring speed=100 rpm).

CONCLUSION

The present study demonstrated the applicability of indirect electrooxidation (EO) and electrocoagulation (EC) processes for removal of Maxilon Red GRL (MxR) dye from aqueous solution. Under optimal value of process parameters, high color removal (>96%) was obtained for dye solution containing 20 mg/L MxR for both processes. It was found that increasing current density or cell voltage has a great effect on the decolorization processes. The data indicated that increasing conductivity has a significant effect on dye removal efficiency. The results of removal percent showed that by increasing initial dye concentration, the dye removal efficiency decreased significantly in case of EC compared with EO process. The results indicated that the optimum pH for EO is 5, while that for EC is 9. It can be concluded that electrooxidation and electrocoagulation techniques are successful processes with high removal efficiency to remove dyes from polluted effluents.

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