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An experimental study of CI Reactive Blue 25 removal from aqueous solution by electrocoagulation using Aluminum sacrificial electrode: kinetics and influence of parameters on electrocoagulation performance

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ABSTRACT

Electrocoagulation (EC) has been employed for the removal of CI Reactive Blue 25 from aqueous solution using a batch stirred reactor with aluminum as sacrificial electrode. In the electrocoagulation of CI Reactive Blue 25, the effects of different electrolyte (NaCl and Na_2SO_4), electrolyte concentration (0.1–0.5 g/L), initial pH (5–11), initial concentration of dye (5–30 mg/L), electrodes spacing (2–6 cm), voltage applied (8–20 V), and electrode area (30–60 cm^2) exposed were investigated. The reaction rate constants were determined according to the pseudo-first-order kinetic model, which fits the experimental data. The highest rate constant of $0.0832\,min^{-1}$ was achieved for an electrolyte concentration of 0.2 g/L, pH 8, spacing of electrode 4 cm, current density of $2.33\,mA/cm^2$, an initial dye concentration of 10 mg/L, with a removal efficiency of 97.2% at 45 min of electrolysis. Electrocoagulation with aluminum electrodes degrades CI reactive Blue 25 effectively from aqueous solution.

Keywords: Electrocoagulation; CI reactive Blue 25; Aluminum electrode; Pseudo-first-order reaction rate

1. Introduction

Dyes are used in leather, pulp and paper, food processing, textile, cosmetics, and pharmaceutical industries. But textile industries are considered as the major consumer of the dyes in the market [1]. Over 10,000 dyes with an annual production of over 7×10^5 metric tonnes are commercially available [2]. Reactive dyes are the class of dye most widely used; industrially, they have a world market share of 60–70% [3]. Approximately, around 30% of the applied reactive dye is wasted because of dye hydrolysis in the alkaline dye bath [4,5]. The effluent discharged from textile dyeing mill forms the highly concentrated color wastewater, consisting of various types of colors [6]. Textile dyeing wastewater imposes serious environmental problems because of their high color, high chemical oxygen demand (COD), fluctuating pH and high temperature [7]. This colored and toxic wastewater released into the ecosystem undergoes chemical as well as biological changes, and consume dissolved oxygen from the stream. The degradations of dyes are the dramatic source of esthetic pollution and many

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dyes and their degradation products are carcinogenic toward aquatic life and mutagenic for humans [7–9]. Dyes are designed in such a way that they are resistant to light, water and oxidizing agents [10]. Conventionally, textile wastewater is treated by various methods like adsorption, precipitation, chemical degradation, advanced oxidation processes, ultrafiltration, ozonation, biodegradation, chemical coagulation, and electrocoagulation (EC) [11]. As the dyes are designed to resist light, water and oxidizing agents they cannot be treated by conventional treatment processes such as an activated sludge process [10]. The costs of adsorption, ultrafiltration, and ozonation exceed that of chemical coagulation. When chemical coagulation is used to treat dyeing wastewater, the pollution may be caused by chemical substance added at a high concentration [12]. In addition, when an excessive amount of chemicals are added, a huge volume of sludge and secondary pollution may result [7]. Excessive coagulant material and other disadvantages with other process can be avoided by EC. The EC process is an eco-friendly, effective, fast and economic technique for the treatment of water and wastewater [13,14]. Treatment of wastewater by EC has been practiced for most of the twentieth century [15]. The EC systems can be effective in removing suspended solids, dissolve metals, tannin and dyes [16], foodstuff wastes [17,18], dye wastewater [19], oil wastewater [20], textile wastewater [21], heavy metal [22], fluoride and polyvinyl alcohol [23], paint wastewater [24]. The EC process is very effective in removing organic pollutants including dyeing wastewater and allows for the reduction of sludge generation. Additionally, the coagulants are generated by electrooxidation of a sacrificial soluble anode, so the addition of chemicals is either not necessary or is at least minimized [25].

The EC process involves many chemical and physical mechanisms [26]. According to Canizares et al. [27,28], the various stages involved in the EC process are as follows: (i) migration to an oppositely charged electrode (electrophoresis) and aggregation due to charge neutralization; (ii) the cation or hydroxyl ion (OH^-) forms a precipitate with the pollutant; (iii) the metallic cation interacts with OH^- to form a hydroxide, which has high adsorption properties thus bonding to the pollutant (bridge coagulation); (iv) the hydroxides form larger lattice-like structures and sweeps through the water (sweep coagulation); (v) oxidation of pollutants to less toxic species; (vi) removal by electro floatation or sedimentation and adhesion to bubbles.

The sacrificial electrodes usually used are iron and aluminum [29–32]. Also, a mild steel plain plate, stainless steel plain plate [31], carbon steel plates [33], metallic iron [34], stainless steel mesh [35] are also used as electrodes for EC. During EC, several reactions take place at the surface of electrodes, especially dissolution of aluminum occurs via oxidation of the anode with simultaneous reduction of water to form hydrogen.

The electrolysis of water at the cathode surface is:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (1)$$

At the anode:

$$Al \rightarrow Al^{3+} + 3e^- \quad (2)$$

$$2H_2O \rightarrow 4H^+ + O_2 + 4e^- \quad (3)$$

The electrochemical reactions at the cathode and anode are hydrogen evolution and oxygen evolution reactions, respectively. At high pH values, the OH^- generated at the cathode reacts to form various monomeric species with Al^{3+} generated at the anode. The anodic metal ions and hydroxide ions generated at the electrode surfaces react in the wastewater to form various hydroxides and built up polymers. The nascent Al^{3+} ions are very efficient coagulants for particulates flocculating. The hydrolyzed aluminum ions can form large networks of Al–O–Al–OH that can chemically adsorb pollutants [36]. Because of the amphoteric character of aluminum, the significant increase in the local pH at the cathode vicinity, due to hydrogen evolution (Eq. (4)), induces “chemical” attack of aluminum and its hydroxide film according to the following reaction [37,38].

$$2Al + 6H_2O + 2OH^- \rightarrow Al(OH)_4^- + 3H_2 \quad (4)$$

$$2Al + 3H_2O \rightarrow 2Al^{3+} + 3/2H_2 + 3OH^- \quad (5)$$

$Al(OH)_4^-$ can react with cationic to remove some pollutants from wastewater thus neutralizing their charge and reducing their solubility or can be transformed in the bulk solution into amorphous $Al(OH)_3$ which coagulate with colloid pollutants and precipitate [39].

$$[Al(OH)_4]^- \rightarrow Al(OH)_3 + OH^- \quad (6)$$

RB25 is one of the most often used dyes in dyeing textile industry in the Erode and Tirupur regions of Tamilnadu India. RB25 belongs to the phthalocyanine reactive dyes group. Phthalocyanine reactive dyes are metallic complexes and most of these dyes are copper phthalocyanines. They are potentially mutagenic and of special toxicity concern because of their metal Cu content [40]. It is essential to remove RB25 from the effluent as it is very mutagenic and toxic. In the present study, the removal of CI Reactive Blue 25 (RB25) from aqueous solution has been carried out by EC process. The factors affecting EC process such as pH, electrolyte and its concentration, electrode spacing, electrode area, applied voltage, and initial concentration of dye also have been investigated.

2. Materials and methods

2.1. Material

The CI Reactive Blue 25 was obtained from a textile industry, Erode, Tamilnadu. The properties of dye are given in Table 1. Supporting electrolytes such as NaCl and Na_2SO_4 (Merck, 99.9% purity) were used for the study. Commercially available aluminum plate of 99% purity was used as both anode and cathode.

2.2. Electrolytic cell

EC batch reactor made of acrylic tank measuring 10 cm × 10 cm × 15 cm with a working volume of 1.0 L was used to conduct the experiments. Two parallel facing aluminum plates of 5 cm × 7.5 cm × 0.4 cm were used as both anode and cathode electrodes for EC. The anode and cathode was connected to a DC (Direct Current) supply unit (30 V/5A, Jayam Electronics, Chennai, Tamilnadu, India). The total effective electrode area of 60 cm^2 and the spacing between electrodes was fixed as 4 cm. The reactor setup was kept above a magnetic stirrer for giving proper agitation during the process. The EC batch setup is shown in Fig. 1.

2.3. Experimental method

A measured quantity (1.0 L) of aqueous RB25 dye at a desired pH and concentration was transferred into the electrochemical cell. Electrolytes such as NaCl and Na_2SO_4 were used to adjust the initial conductivity of the solution. The required initial pH of the working solution was adjusted by 1 N HCl and 0.5 N NaOH. The anode and cathode were connected, respectively, to the positive and negative ends of the DC power supply and required voltage was fixed. The voltage was controlled throughout the study. The electrode spacing and electrode area was adjusted by changing the position of electrodes. The samples were collected for every 5 min up to 100 min and allowed to settle for 30 min. Then, the dye concentration was analyzed using UV–visible spectrophotomer (Lambda 25 Perkin Elmer, USA).

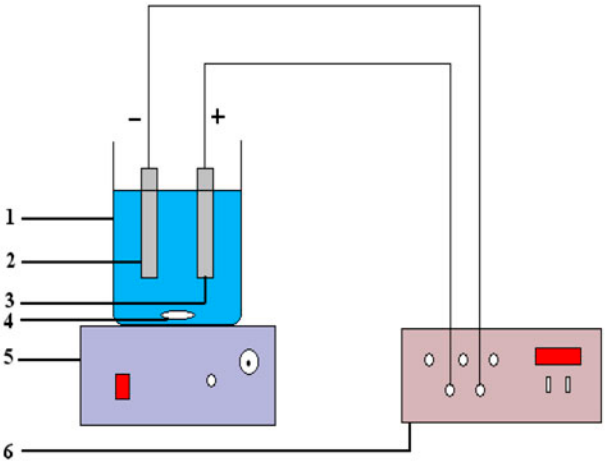
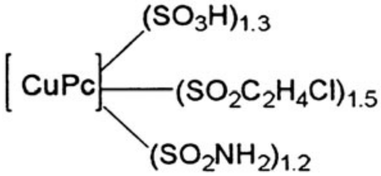


Fig. 1. Electrocoagulation reactor setup. (1) reactor, (2) cathode, (3) anode, (4) magnetic stirrer cork, (5) magnetic stirrer, (6) AC to DC convertor.

2.4. Determination of the concentration of CI Reactive Blue 25

The color removal efficiency (Y_t, %) was calculated from equation,

Table 1
Properties of CI Reactive Blue 25

Chemical name	CI Reactive Blue 25
CB number	CB41033614
Class	Phthalocyanine class
Molecular structure	 $[\mathrm{CuPc}]\text{–}(SO_3H)_{1.3}(SO_2C_2H_4Cl)_{1.5}(SO_2NH_2)_{1.2}$
Peak wavelength	663.66 nm

$$Y_t = \frac{(C_0 - C_t)}{C_0} \times 100 \tag{7}$$

where C_0 is the concentration of dye before EC (mg/L), and C_t is the concentration of dye after "t" min of EC (mg/L).

2.5. Kinetics

The rate of reaction for the degradation of RB25 has been studied using pseudo-first-order kinetics. The pseudo-first-order reaction rate for the degradation of dye is given in Eq. (8),

$$\frac{-dC}{dt} = k_1 C \tag{8}$$

The rate constant k_1 at time t can be estimated from the plot log (C/C_0) vs. EC time t [7,31]. The integration of Eq. (8) results in:

$$\log \frac{C}{C_0} = -k_1 t \tag{9}$$

where C = concentration of dye mg/L at time "t" min, C_0 = initial concentration of dye mg/L, k_1 = first order reaction rate constant min^{-1}.

3. Results and discussion

3.1. Effect of electrolyte and its concentration

Electrolytes are added to the EC process to increase the conductivity [39]. In this study, the effect of two different electrolytes such as NaCl and Na_2SO_4 on RB25 removal was investigated. The experiments were conducted at a constant voltage of 14 V, electrode spacing of 4 cm and effective electrode area of 60 cm^2. The results are shown in Fig. 2. From Fig. 2, it can be observed that the addition of NaCl gives better removal of RB25 from aqueous solution when compared to the addition of Na_2SO_4. Hence, further studies have been carried out using NaCl as supporting electrolyte. The addition of NaCl also contribute chloride ions to the solution that removes the insulating layer formed by Ca^{2+} and Mg^{2+} on electrode and also it acts as a good disinfectant [41,42].

The influence of NaCl concentration on the conductivity and current density were studied and is shown in Fig. 3. From Fig. 3, it was found that the conductivity of the dye increased with increase in concentration of the electrolyte. It was also observed that with increase in conductivity increases the current density linearly, thus reducing the ohms resistance [31]. Electrolytes employed to increase the conductivity of the water or wastewater to be treated, and increase the conductivity rather with increase in electrolyte concentration, the voltage decreases and the solution conductivity increases that leads to the decrease in power consumption [39].

The influence of NaCl concentration on the removal of RB25 dye is shown in Fig. 4. A maximum removal of 97.7% of dye was observed with 0.5 g/L NaCl concentration (Fig. 4). When the electrolyte concentration was increased from 0.1 to 0.2 g/L the removal efficiency increases from 40.9 to 89.7%. Further increase in electrolyte concentration from 0.2 to 0.5 g/L increases the dye removal only by 8%. The NaCl concentration of 0.2 g/L was considered throughout the study, as the increase in dye removal

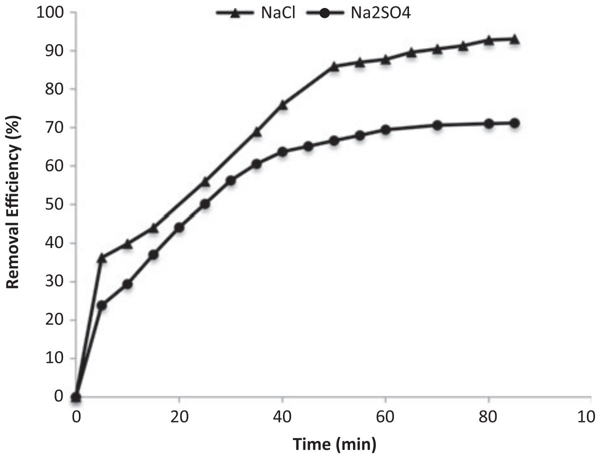


Fig. 2. Effect of supporting electrolyte on RB25 dye removal efficiency at an initial concentration 10 mg/L, pH 6.9 and constant voltage supply of 14 V.

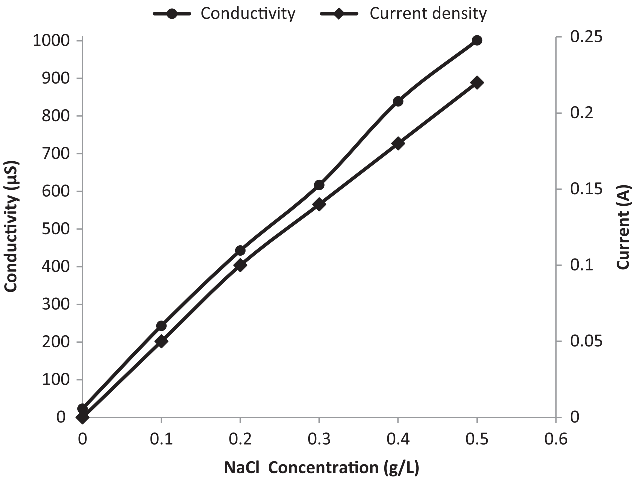


Fig. 3. Effect of electrolyte concentration on conductivity and current, at an initial concentration 10 mg/L, pH 6.9 and constant voltage supply of 14 V.

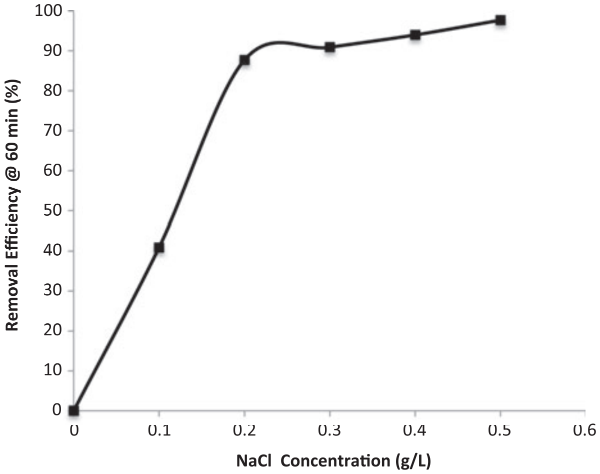


Fig. 4. Effect of electrolyte concentration on dye removal efficiency at 60 min at an initial dye concentration of 10 mg/L, pH 6.9 and constant voltage supply of 14 V.

with increase in conductivity for higher concentration of electrolyte is not significant [31].

3.2. Effect of initial pH

The pH is an important variable that influences the EC process [29]. The initial pH was adjusted by adding 1 N HCl or 0.5 N NaOH. The decolorization rate at the EC of aqueous solutions of RB25 dye was investigated in the initial pH range from 5 to 11. The influence of pH on RB25 removal efficiency is shown in Fig. 5. The removal efficiency increases rapidly up to 40 min and remains constant thereafter. It was found that with aluminum electrode the dye removal is not effective at pH 5 and pH 11. The most favorable initial pH value for the EC process is eight. At pH 7 the removal efficiency of 98.3% was observed at an electrolysis time of 80 min, whereas at pH 8, 97.2% of removal efficiency was observed at 60 min of electrolysis time. In the case of pH 5 and pH 11, the removal of dye is following a different trend with low removal efficiency when compared to pH 7–10. When the pH increases, the OH^- generated at the cathode reacts to form various monomeric species with Al^{3+} generated at the anode. When the pH increases above 10 due to high hydrogen evolution, induces "chemical" attack of aluminum [37]. At pH between five and seven these monomeric cations can evolve to polymeric species such as $Al_2(OH)\ _2^{4+}$ and $A_{16}(OH)_{15}^{3+}$. Soluble monomeric and polymeric cations are also converted into insoluble $Al(OH)_3$ flocs by the complex precipitation kinetics [28].

The pseudo-first-order kinetics plot for different pH is shown in Fig. 6. The reaction rate constant was derived from the plot and is reported in Table 2. A maximum first order reaction rate constant of 0.0674 min^{-1} was obtained at pH 8. The reaction rate was increased up to pH 8 and then decreased, significant reduction in reaction rate was reported for pH 5, 6 and for pH 11. It indicates that the removal of CI Reactive Blue decolorization is effective in the pH range from 7 to 10.

3.3. Effect of spacing of electrode

There is direct relationship between removal efficiency and electrodes spacing. With the increase in electrode distance, current intensity gets lower at a

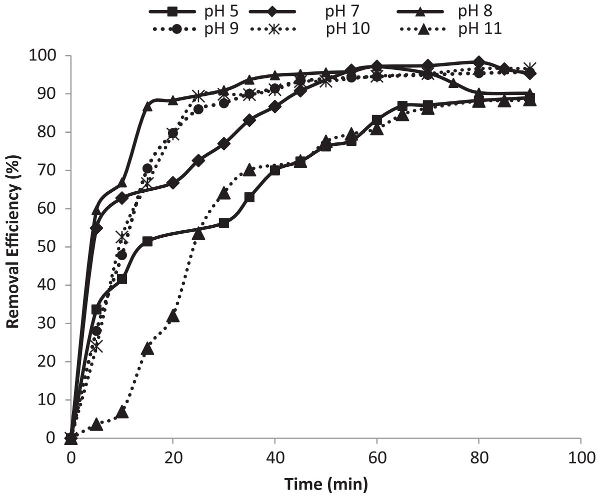


Fig. 5. Effect of initial pH in removal efficiency, at an initial concentration 10 mg/L, and constant supply of 14 V (current density 1.5 mA/cm^2), NaCl concentration of 0.2 g/L and spacing of 4 cm.

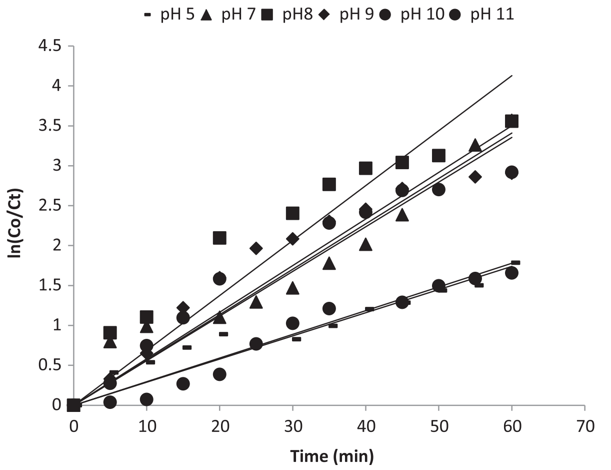


Fig. 6. Pseudo-first-order kinetics plot for pH variation at an initial concentration 10 mg/L, and constant supply of 14 V (current density 1.5 mA/cm^2), NaCl concentration of 0.2 g/L and spacing of 4 cm.

Table 2
Values of pseudo-first-order constant at different pH conditions

pH	Reaction rate constant k_1 (min^{-1})	R^2
5	0.0282	0.9229
7	0.0539	0.9579
8	0.0674	0.9148
9	0.0584	0.8874
10	0.05	0.8761
11	0.0271	0.9626

certain voltage and in order to achieve a certain current density, the voltage has to be increased [43]. The spacing between the electrodes was varied from 2 to 6 cm. The influence of electrode spacing on removal efficiency of RB25 dye is shown in Fig. 7. The results indicated that the increase in spacing between the electrodes decreases the removal efficiency of RB25 dye by EC process. It was also observed that the current density increases with decrease in spacing. When the spacing is increased from 4 to 6 cm, the removal efficiency of dye decreased to 90% from 97.2% at 60 min of electrolysis. The same trend was followed for 3 and 4 cm spacing, respectively. With increase in spacing between electrodes, less interaction between ions and hydroxyl polymers is expected [43]. The removal efficiency decreased when the spacing was reduced from 3 to 2 cm, this may be due to lack in agitation and lack in opportunity to aggregate and produce flocs [44]. A maximum reaction rate was observed for spacing 3 and 4 cm as 0.075 and 0.0674 min^{-1}, respectively (Table 3).

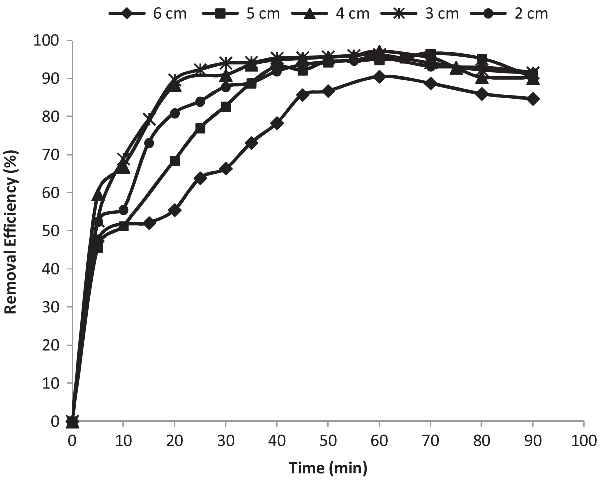


Fig. 7. Effect spacing of electrodes in removal efficiency, at an initial concentration 10 mg/L, constant supply of 14 V, NaCl concentration of 0.2 g/L, area 60 cm^2 and pH 8.

Table 3
Values of pseudo-first-order constant at different electrode spacing

Spacing (cm)	Reaction rate constant k_1 (min^{-1})	R^2
2	0.0596	0.8919
3	0.075	0.8841
4	0.0674	0.9148
5	0.0533	0.9363
6	0.0401	0.9734

3.4. Effect of electrode area

There is direct relationship between removal efficiency and total area of electrodes. The literature states that when the area of electrode increases, the distribution of the coagulation agent's density is more effective. This can produce the related coagulation and completes the removal of dye particles [43].

The area of electrode exposed to EC process was varied from 30 to 60 cm^2. The influence of electrode area on removal efficiency of RB25 dye is represented in Fig. 8. A maximum removal efficiency of 97.2% was obtained with 60 cm^2 of electrode area at 60 min of process. The removal efficiency increases from 85.3% to 97.2%, when the electrode exposed area increases from 30 to 60 cm^2, as the increase in electrode area increases the amount of Al ions exposed for coagulant activity. The removal efficiency at 20 min with 30 cm^2 electrode is 55.5% and at 60 min time is 85.3% and that with 60 cm^2 electrode is 88.3% at 20 min and 97.2% at 60 min. It was examined that increase in electrode area increases the removal efficiency of RB25 dye. Daneshvar et al. 2003 [43] also reported the same trend for orange II dye removal.

3.5. Effect of applied voltage

Current density is an important factor that influences the EC process and a strong factor on the color removal. The addition of the electrochemically generated reagent can be controlled by adjusting the supplied power and process can be optimized [35].

The applied voltage was varied from 20 to 8 V with the following experimental conditions: dye concentration of 10 mg/L, electrode area of 60 cm^2, NaCl concentration of 0.2 g/L, pH 8 and spacing 4 cm. The removal efficiency variation with voltage is shown in Fig. 9. It was observed that the increase in current voltage increases the removal efficiency of RB25. At low current voltage, the removal efficiency was comparatively low, and an increase in removal efficiency of 92% from 69.3% was observed, when the current voltage was increased from 8 to 14 V at 30 min. The

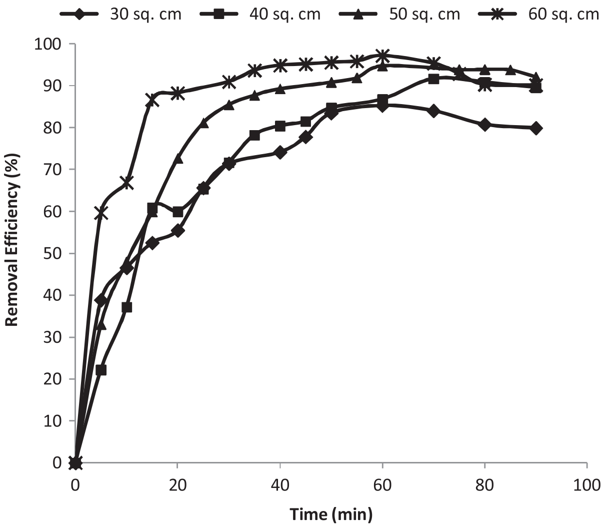


Fig. 8. Effect electrode area in removal efficiency, at an initial concentration 10 mg/L, and constant supply of 14 V, NaCl concentration of 0.2 g/L, pH 8 and spacing 4 cm.

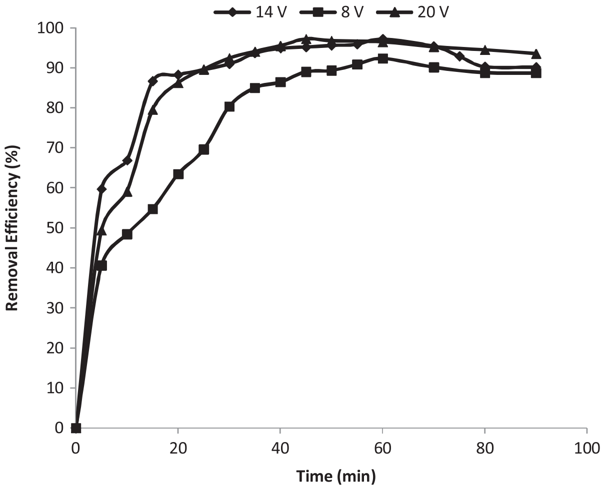


Fig. 9. Effect initial voltage supply in removal efficiency, at an initial concentration 10 mg/L, electrode area of 60 cm^2, NaCl concentration of 0.2 g/L, pH 8 and spacing 4 cm.

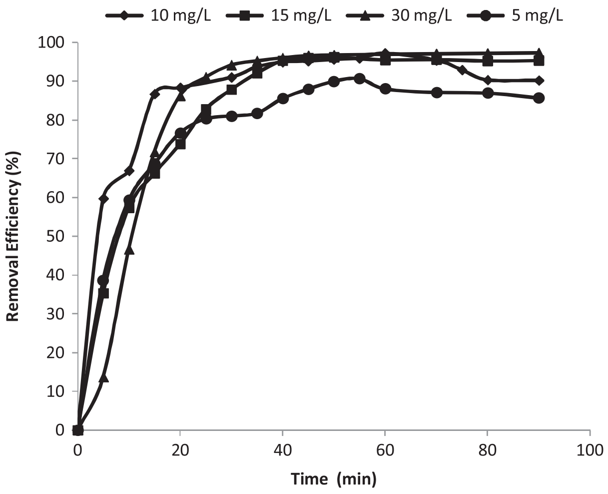


Fig. 10. Effect of initial dye concentration in removal efficiency at 60 min at an, electrode area of 60 cm^2, voltage 14 V, NaCl concentration of 0.2 g/L, constant pH 8 and spacing 4 cm.

further increase in current density from 14 to 20 V does not produce any significant change in removal efficiency of RB25. It represents that with increase in current density the removal efficiency of dye increases up to a certain extent.

In case of aluminum configuration 20% of the total electrode mass dissolved was due to chemical dissolution and remaining 80% by electrochemical dissolution. Faraday's law provides the relationship between current density and the amount of aluminum electrochemically dosed into solution as [27,45],

$$w = \frac{itM}{ZF} \tag{10}$$

where w = anode dissolving (g/cm^2), i = current density (A/cm^2), t = time (s), M = molecular weight of anode material, Z = number of electrons involved in the oxidation/reduction reaction and F = Faraday's constant (96,487 C/eq). It is clear from above equation that, at higher values of current density the anodic dissolution rate increases and the resulting iron hydroxides produce more flocs thus enhancing the coagulation process [46]. Also, the bubble generation rate increases which separates the pollutants by floatation [47].

3.6. Effect of initial concentration of dye

The initial concentration of the dye is a major factor that affects the removal efficiency. The initial concentration was varied from 5 to 30 mg/L. The influence of dye concentration on the removal efficiency is shown in Fig. 10. The removal efficiency remains almost same with the higher dye concentration when compared to that of low concentration of dye within the range considered. Maximum removal efficiency of 90.6% was observed for 5 mg/L dye concentration.

4. Conclusions

The results have clearly demonstrated that EC process can be a viable technique for the removal of RB25 dye-containing wastewater. The effects of the main operating parameters, such as pH, initial concentration, operating time, current density, electrolyte type, electrolyte support concentration, electrode spacing, and area on the RB25 dye removal were investigated. The treatment of RB25 dye in a batch EC reactor can lead to almost complete decolorization when the experimental setup is operated at optimal values of parameters. EC process of RB25 obeys the pseudo-first-order kinetic model. The results indicated that the RB25 getting almost complete decolorization in the NaCl concentration of 0.2 g/L, pH 8, spacing 3–4 cm, electrode area exposed of 60 cm^2, and current density of 2.33–1.5 mA/cm^2 with a reaction rate constant of 0.0832–0.0674 min^{-1}.

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