International Journal of Environment and Bioenergy, 2013, 6(2): 96-116

Modern Scientific Press

International Journal of Environment and Bioenergy

Journal homepage: www.ModernScientificPress.com/Journals/IJEE.aspx

ISSN: 2165-8951

Florida, USA

Article

Removal of Reactive Dyes from Textile Wastewater by Electrocoagulation Process: An Effective and Clean Approach

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Article history: Received 8 April 2013, Received in revised form 27 May 2013, Accepted 5 June 2013, Published 8 June 2013.

Abstract: The removal of dye pollutants from industrial effluents is a rising concern in recent years. In this paper, the decolorization of strong colored solutions containing the reactive textile dyes was done by electrocoagulation process using aluminum electrodes in a lab scale reactor operated in batch mode. The effects of applied voltage, pH variation with time, current density, dye and COD concentrations, electrode connection mode, inter electrode distance and retention time on the decolorization efficiency have been investigated. Also, an optimum condition was defined based on effect of electrical energy consumption and electrode consumption. Dye removal followed good agreement with first order kinetics and the reaction rate constant and dye removal was found to be strong function of operational conditions and type of dye. As per experimental outcomes, dye and chemical oxygen demand removals were 99.0 and 82.7%, respectively and electrode consumption, energy consumption and operating cost were 0.0637 kg/m^3, 4.6878 kWh/m^3 and 0.665 US\$/m^3 correspondingly.

Keywords: electrocoagulation; reactive dye; aluminum; operational condition; cost; kinetics; batch process.

1. Introduction

Over helming speed of industrialization and urbanization is the main culprit behind the present crucial situation of water pollution due to release of industrial wastes into the aquatic system without treatment of the hazardous contaminants. Among these excreted contaminants, releases of dyes are mainly transpiring from textile, lather, paper, printing, cosmetics industries etc. after the respective dying processes. The removal of dyes from textile wastewater is of great concern, not only for coloring impact but also as they are responsible for the destruction of ecological balance of the aquatic system. They interfere with the transmission of sunlight and inhibit the photosynthesis process, cause the depletion of the dissolved oxygen and a great harm on the food chain. Also, many reactive dyes produce carcinogen compounds upon dissociation which not only destroying the aquatic life but also entering & causing the serious health risk to human beings including causing many serious diseases in many provision of the globe, especially in developing countries via the action of food chain. Presently about 700,000 tonnes and 10,000 different types of dyes and pigments are being produced annually across the world (Gong et al. 2007; Mane et al. 2007) and it has been reported that up to 15% of the dyes used are released into wastewaters (Daneshvar et al., 2006). Considering both volume and composition, effluent from the textile industry was declared as one of the major sources of wastewater in ASEAN countries (Jumasiah et al., 2005). As the global natural resources are limited, and as such the use and reuse of water is becoming an escalating issue, effective and clean methods are required for treating the wide range of textile wastewater pollutant so as to save the climate and lives for the upcoming generations.

Reactive dyes are the most common dyes used due to their advantages, such as bright colors, excellent colorfastness and ease of application (Yang et al., 2001; Mahony et al., 2002). Though reactive dyes have good technical characteristics and consequent industrial preference but they cause severe environmental effects on landscape and water land near industrial area with genotoxic health risk and environmental genotoxicity. Acute exposure to reactive dye can cause allergic dermatoses and respiratory diseases (Estlander et al. 1988; Nilsson et al. 1993; Hatch et al. 1995; Manzini et al., 1996; Wilkinson et al., 1996), dermatitis and asthma (Thoren et al., 1980), change in immunoglobulin levels (Park et al., 1991), increased risks of colon and rectum cancers (Roos et al., 2005), high risk of contracting bladder cancer (Gonzales et al., 1988) etc. Moreover, experimental study showed these sorts of dyes as responsible for mutagenicity (Keneklis et al., 1981), genotoxicity when studied with HaCaT cells which are human keratinocytes (Wollin et al., 2004), and bears teratogenic potential (Birhanli et al., 2005). Also, reactive dyes contain aromatic rings in chemical structures (Robinson et al., 2001; Muthukumar et al., 2007) and therefore, have high stability against light, oxidants, and biological degradation (Robinson et al., 2001) and thus suspected to continue the severe impact on the respective for prolong period of time. Thus reduction of dye and chemical oxygen demand (COD) from textile wastewaters are difficult (Sanroman et al., 2004). So, maximum removals of such dyes from effluent are of topmost priority for the environment and life in the earth.

There are many processes available for the removal of dye by conventional treatment technologies including biological and chemical oxidation, chemical coagulation, foam flotation, biodegradation, advanced oxidation, photocatalysis and adsorption (Lin et al., 1996; Kim et al., 2004; Nakagawa et al., 2004; Papic et al., 2004). Conventional biological processes are less efficient in degrading synthetic dyes due to the complex structures, molecular size and nature (Shaul et al., 1991; Jiang et al., 1994; Lin et al., 1996). Also biological treatment process requires large land area and is constrained by the sensitivity towards diurnal variation as well as toxicity of some chemicals, and less flexibility in design and operation (Bhattacharyya et al., 2003). Also in such process, though many organic molecules are degraded, many others are recalcitrant due to their complex chemical structure and synthetic organic origin (Kumar et al., 1998). As a reference, the cell uptake was shown to be inversely proportional to the number of sulfonate groups of 18 azo dyes studied (Thebault et al., 1981; Carliell et al., 1995), and found that 11 passed practically unchanged through the activated sludge system, 4 were absorbed by the activated sludge and only 3 were biodegraded, resulting in the release of these substances into bodies of water. Physicochemical methods include adsorption (e.g. on active carbon), coagulation–flocculation (using inorganic salts or polymers), chemical oxidation (chlorination, ozonisation, etc.) and photo-degradation (UV/H_2O_2, UV/TiO_2, etc.) (Churchly et al., 1994; Gahr et al., 1994; Kang et al., 1997; Hachem et al., 2001; Chu et al., 2002; Neppolian et al., 2002; Pala et al., 2002; P´erez et al., 2002; Robinson et al., 2002; Zanoni et al., 2003; Zielinska et al., 2003). However, these technologies usually need additional chemicals which sometimes produce a secondary pollution and a huge volume of sludge (Kim et al., 2002; Daneshvar et al., 2006; Bayramoglu et al., 2007). Also many of them are very costly for the developing country to adopt effectively. Membrane separation processes (ultra-filtration, nano-filtration, micro-filtration, reverse osmosis etc.) are found of good outcome in such regards by many researchers, but high cost of membrane and severe fouling with high control measures while operation makes it tough to implement by the developing nations. All of these crucial limitations lead the necessity of an easy to operate, effective and easily adoptable approach for treatment of the textile effluents.

Electrocoagulation is a vital candidate with prominent capability to play an imperative function for water treatment as reported by many researchers so as to solve the present situation. Electrocoagulation is a process in which coagulants are produced in situ by the electro-oxidation (and thus dissolution) of sacrificing anodes followed by the formation of aluminum or iron hydroxide flocs which destabilize and aggregate the suspended particles or precipitates and adsorb dissolved contaminants. Also, electrocoagulation has been proposed in recent years as an effective method to treat wastewater streams from dyeing factories. The electrogenerated flocs separate rapidly and remove color and turbidity from dyeing wastewaters (Lin et al., 1994; Picard et al., 1994). According to Rajeshwar et al. (1994) benefits from using electrochemical techniques include environmental compatibility, versatility, energy efficiency, safety, selectivity, amenability to automation, and cost effectiveness. Also this process is more preferred from various strands as allowing controlled and rapid reactions, smaller systems to make viable and employment of only electrons instead of using chemicals and micro-organisms for water treatment.

In this study, efficiency of electrocoagulation treatment using aluminum electrodes to remove Novacron Red C4B from aqueous solution has been studied in a laboratory scale reactor. The effects of various parameters such as applied voltage, time of reaction, inter electrode distance, initial dye concentration; electrolyte concentration and electrode connection mode on dye removal percentage have been appraised. Also, an optimum operation condition was defined with respect to electrical energy consumption and electrode consumption and efficiency of the process at this condition was analyzed. Also, kinetics study and the efficiency of electrocoagulation reactor for different dyes were investigated.

2. Materials and Methods

Stock solutions of the Novacron Red C4B dye waste water was prepared by dissolving pre-defined amount of dye in distilled water and all chemicals used were of analytical grade (Merck, Germany). The pH of the solution was adjusted by use of HCl (10%) and/or NaOH (10%) solution. The measurement of pH and the conductivity of waste solution were done by a digitally calibrated pH-meter (Hanna, Model HI 2211) and conductivity meter (Hanna, Model HI 8733), respectively.

The electrocoagulation reactor was made by locally available materials. A rectangular glass jar of 2.5 L effective volume was used as the reactor. Inside it, aluminum electrodes (12.0 mm × 10.30 mm × 3.0 mm) were arranged in situ mode by spacer of insulation material (PVC). The required voltages were applied by a DC power supply (KENWOOD, Model: PR-601, Japan) and the current density and voltage were controlled by a sunt (Model: OSK 11669, Japan) and a digital multimeter (SANWA, Model: PM-3) was used for measuring the voltage and current. Four impellers (100 rpm rotation) equipped with motor was used to maintain the proper agitation in the reaction media. In order to maintain the flow of waste water into the electrocoagulation reactor, a Peristaltic Pump (Model: Simplex ENDP50) with level control mechanism was used. A schematic diagram of the experimental setup is represented in Fig. 1. The analytical determination of dye was carried out with the standard procedure using an UV spectrophotometer (Shimadzu Model UV-1601) after centrifuging for 10 min at 10,000 rpm. COD of the solution before and after electrocoagulation was determined by the standard procedure (5220 B. Open Reflux Method) described elsewhere (American Soc. Testing & Materials, 1995) after filtering treated solutions by using Whitman No. 40 filter paper.

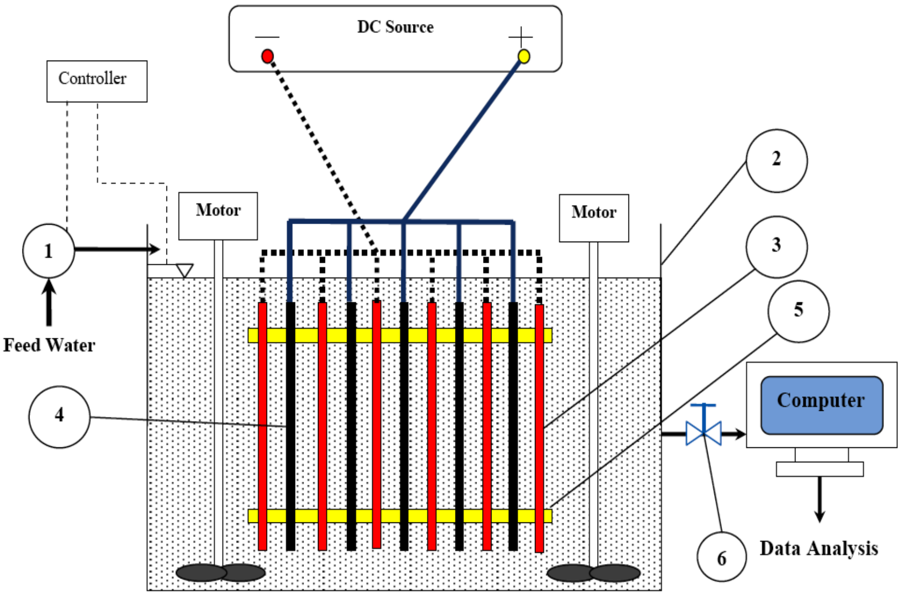

Figure 1. Experimental setup for the electrocoagulation (1 = peristaltic pump, 2 = reactor vessel, 3 = anode, 4 = cathode, 5 = spacer (PVC), and 6 = sampling valve)

The all operations were done in batch mode. Four different applied voltages of 5, 15, 30 and 45 V were used to assess the effect of applied voltage on the dye removal efficiency. To evaluate the effect of reaction time, inter electrode distance, initial dye concentration, electrode connection mode etc., each batch was operated for sufficient reaction time of (60 min) so as to obtain the maximum dye removal at desired conditions. All the experiments were uniformly done with the different operational conditions using Al electrodes (both cathode and anode) procured from local market. During the process, samples were withdrawn from the reactor at different reaction time intervals and settled for a time of 30 min, centrifuged (10,000 rpm, 10 min) and tested for further analysis. After each experimental run, electrodes were weighted after immerging in HCl solution (1.0 M) for 45 min followed by rubbing with a plastic brush, washing with water and drying in air. All the experiments were carried out at room temperature (25 ^{o}C). The calculation for percentage of dye removal, COD removal and electrical energy consumption were carried out as per equations (1), (2) and (3) respectively.

$$R_c = [\frac{C_o - C}{C_o}] \times 100 \tag{1}$$

$$R_{COD} = [\frac{COD_o - COD}{COD_o}] \times 100 \tag{2}$$

$$R_{COD} = [\frac{COD_o - COD}{COD_o}] \times 100 \tag{3}$$

where R_C and R_{COD} are dye and COD removals (%) respectively. C_0 and C are initial and final (after electrocoagulation) dye concentration (ppm), respectively. COD_0 and COD are chemical oxygen demand before and after reaction (mg/L), respectively. E, U, I, t, and V are electrical energy consumption (kWh/m^3), applied voltage (volt), current (ampere), time of reaction (hour), and the volume of solution (L), respectively.

3. Mechanisms of Electrocoagulation

Electrocoagulation is a complex and interdependent process performs via dissolving of Fe/Al in the electrolyte to generate corresponding metal ions of prescribed amount (depending on the operating conditions) which further hydrolyzes to polymeric iron or aluminum hydroxide and these are excellent coagulating agents. The sacrificial anode incessantly produces polymeric hydroxides by anodic dissolution and coagulation occurs when these metal cations combine with the negatively charged particles carried toward the anode by electrophoretic motion. At the same time electrolytic gases are generated at the cathode. Three main processes which occurred during electrocoagulation are:

(1) Electrolytic reactions at surface of electrodes;

(2) Formation of coagulants in aqueous phase;

(3) Adsorption of soluble or colloidal pollutants onto coagulants and removal of them using sedimentation or flotation of flocs when H_2 bubbles were produced at the cathode (Aleboyeh et al., 2008).

In the present experimental study aluminum has been used as anode and thus the electrode reactions can be considered as (Chen et al., 2004):

Anode:

$$Al(s) = Al^{3+}\,(\mathrm{aq}) + 3^{\mathrm{e}^-} \tag{4}$$

Cathode: Al^{3+} and OH^- ions generated during the electrochemical reactions react to form various monomeric and polymeric species and transform into $Al(OH)_3$ according to the following reaction.

$$Al^{3+}(aq) + 3H_2O(l) = Al(OH)_3(\mathrm{s}) + 3\mathrm{H}^+(\mathrm{aq}) \tag{5}$$

These $Al(OH)_3$ flocs capture the dye molecules present in the solution by the following reaction mechanism:

Precipitation:

$$DYE + Al_m = [DYE\ Al_m]_s \tag{6}$$

$$DYE + Al_p = [DYE\ Al_p]_s \tag{7}$$

Adsorption:

$$DYE + Al(OH)_{3,s} = Al(OH)_{3,s}(DYE) \tag{8}$$

$$[DYE\ Al_p]_s + Al(OH)_{3,s} = [Al(OH)_3 DYE\ Al_p]_s \tag{9}$$

4. Results and Discussion

4.1. Effect of Applied Voltage and Reaction Time

The applied voltage and time are two important parameters playing critical role in the electrocoagulation process. The corresponding effects for the present study are shown in Fig. 2. According to the obtained result, with the increment of applied voltage from 5 to 45 V, the dye removal percentage at the operating condition rises from 97.1 to 99.9%. Also with the increment of time from 5 to 60 min, dye removal percentage increases from 84 to 98.5% while applied voltage was kept constant at 5 V. Moreover, though the dye removal at initial state is low, the removal efficiency gradually increases with the increment of time and applied voltage. The similar fashion is found in preceding works (Lin et al., 1996; Daneshvar et al., 2003).

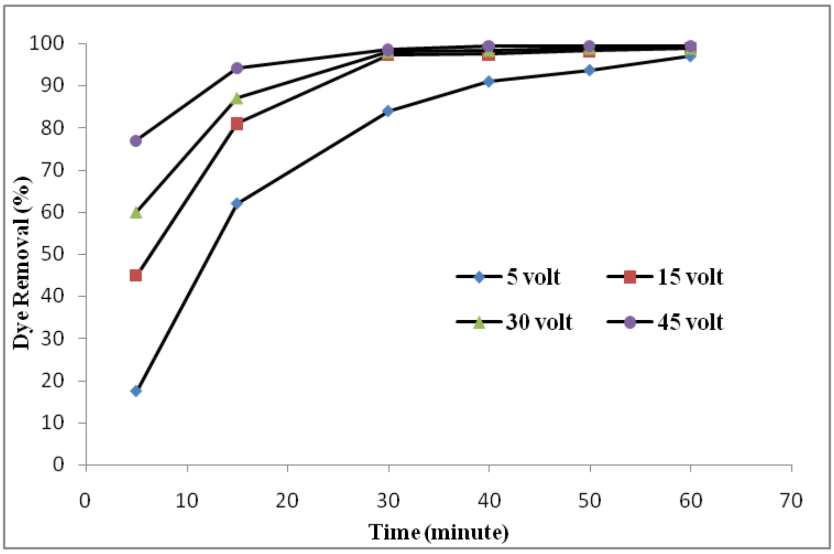

Figure 2. Dye removal efficiency with time at different applied voltage (dye concentration: 50 ppm, inter-electrode distance: 1 cm)

4.2. Effect of Inter Electrode Distance on Dye Removal

In order to assess the effect of inter-electrode distance on dye removal efficiency by the electrocogulation process, the experiment was carried out at constant voltage of 15 V with inter electrode distances of 0.5, 1, 1.5, and 2 cm. As shown in Fig. 3, inter electrode distance was found with negative correlation with the dye removal efficiency. Here, with the increment of inter electrode distances from 0.5 to 2 cm, dye removal decreases from 98.59 to 90.43% after 30 min electrolysis at 15 V. It can be attributed that with increasing of distance between electrodes at constant voltage, electrical resistance between electrodes increases and current passed through electrodes decreases. The decreasing of current, lead to lower production of aluminum and hydroxyl ions and dye removal efficiency decreases. Results showed that with an increasing of gap between electrodes, electrical energy consumption decreased. On the other hand, with increasing of inter electrode distance, less interaction of the dye with the hydroxyl polymers is expected, thus local concentration and electrostatic attraction would decrease and dye removal efficiency reduces (Song et al., 2007). But in case of 0.5 cm distance, the energy consumption for getting the desired purification was found to be much higher. As per Phalakornkule et al. (2010), a possible explanation is that at the electrode distance of 5 mm, the gap between the anode and cathode was too close for easy solid and fluid transfer, resulting in accumulated solid particles and bubbles between the anode and cathode and consequent higher electrical resistance.

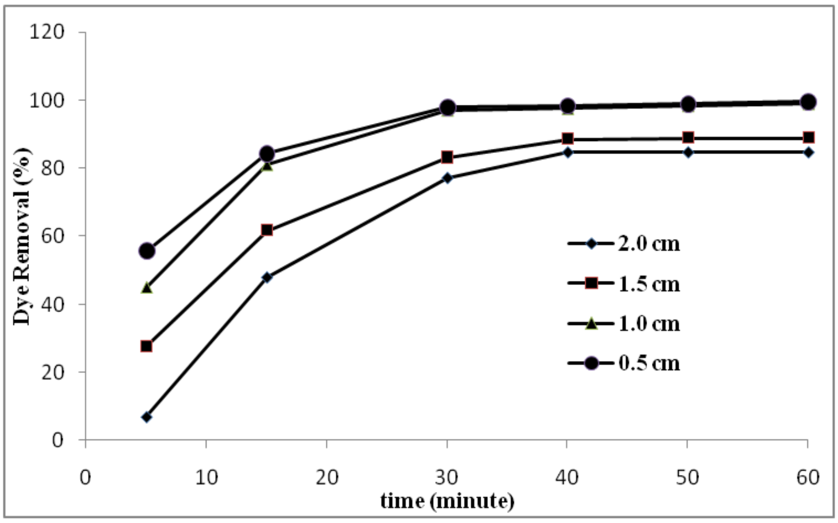

Figure 3. Effect of inter electrode distance on dye removal (dye concentration: 50 ppm, inter-electrode distance: 1 cm, voltage: 15 V)

4.3. Effect of Time and Voltage Variations on Electrical Energy Consumption

In order to determine an optimized voltage, time and inter electrode distance, dye removal percent and energy consumption were considered as the main strands of analysis. The effects of voltage and time on electrical energy consumption is shown in Fig. 4 showing the linear increment of electrical energy consumption from 0.18 to 3.94 kWh/m^3 wastewater at 15 V and 1 cm of inter-electrode distance when reaction time was increased from 5 to 105 min. In addition to this, the energy consumption for the 15 V of applied voltage was found of 10 times higher for all the inter electrode distances than that for 45 V. So, 15 V was chosen to be more effective than 45 V for high dye removal efficiency and low electrical energy consumption. Moreover, as per Fig. 3, at 15 V of applied voltage maximum dye removal of 99.90% was accomplished after 60 min, while dye removal was 98.14% at 30 min. And the electrical energy consumption at 60 min was 2.0 times higher than that of 30 min. As an outcome of all of these results, we can conclude the optimum condition as: voltage: 15 V, inter electrode distance: 1 cm and reaction time: 30 min.

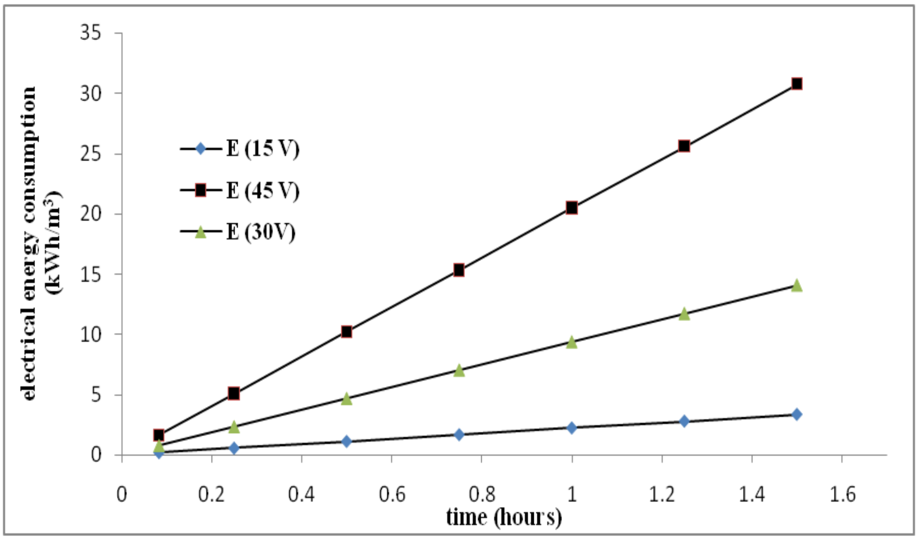

Figure 4. Effect of time and voltage variations on electrical energy consumption (dye concentration: 50 ppm, inter-electrode distance: 1 cm)

4.4. Effect of Voltage Variations on Final pH of Effluent during Electrocoagulation

The pH is one of the significant stricture affecting the electrocoagulation process. But many of the former investigations showed the efficient pH for electrocoagulation with aluminium electrode lied in the range of 5-6 (Arslan et al., 2008). So, as the initial pH of dye solution used in this investigation was 5.5, this parameter was not evaluated in this study and only effects of voltage and time on final pH of effluent during process were appraised. As shown in Fig. 5, pH gradually increases with the increment of reaction time which is due to the formation of OH^- ions in the cathode with the evolution of H_2 gas.

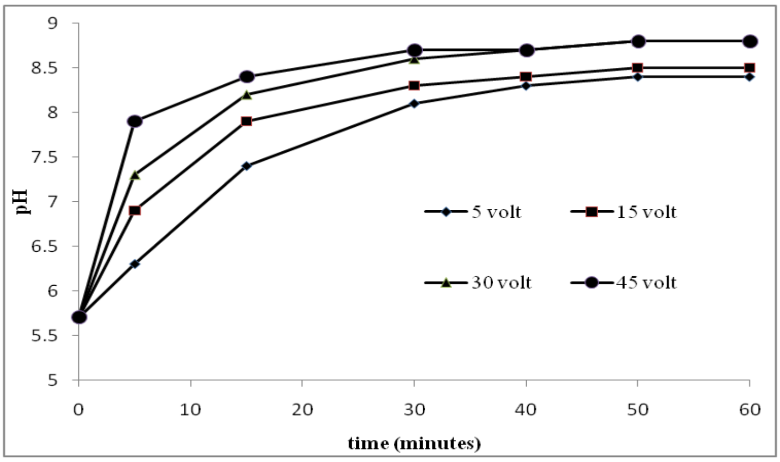

Figure 5. Effect of voltage variations on final pH of effluent during electrocoagulation (dye concentration: 50 ppm, inter-electrode distance: 1 cm, voltage: 15 V)

4.5. Effect of Initial Dye Concentration on Removal of Dye and COD

The effect of initial dye concentration is shown in Fig. 6 which are obtained by electrocoagulation of 50, 100, 150, 200, 250 and 300 ppm dye containing solution in electrocoagulation reactor for optimum condition (Voltage: 15 V, inter-electrode distance: 1.0 cm and reaction time: 30 min) with zero electrolyte concentration. The plot shows that, with the increment of concentration of dye solution from 50 to 300 ppm, the removal percentage of dye and COD at the specific condition declines from 98.9 to 63.4 and 84.7 to 50.3, respectively. As per the mechanism shown in section 3, the dye is removed by the adsorption of dye molecule by the aluminum hydroxides produced by the reaction of produced Al^{3+} ions in anode with OH^- ions. Now, since as per Farady's low, the mount of Al^{3+} released at constant current, voltage, and time for all dye concentration of dye solution are same, the gradual decline of percentage of dye removal in electrocoagulation process is due to limiting the adsorption of higher dye concentration due to correspondingly lower amount of aluminum hydroxide coagulant.

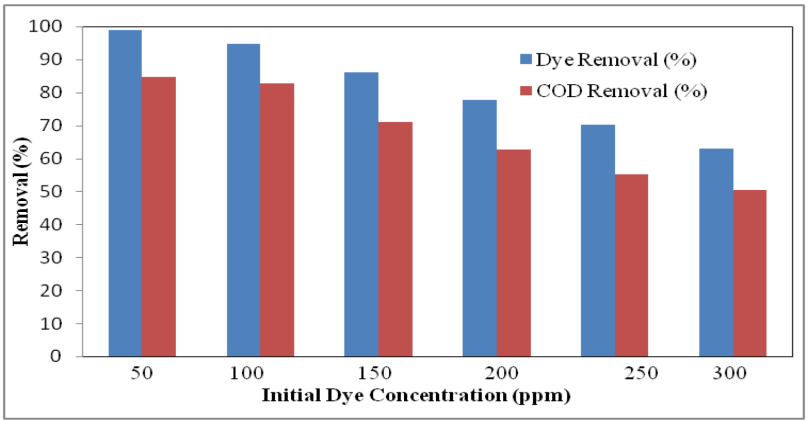

Figure 6. Effect of initial dye concentration on removal of dye and COD (inter-electrode distance: 1 cm, voltage: 15 V)

4.6. Effect of Electrolyte Concentration on Dye Removal

In order to investigate the effect of electrolyte concentration on the dye removal in the electrocoagualtion process, sodium chloride (NaCl) solution of different concentrations (0.0, 0.25, 0.5 and 1.0 mM) were added to the dye solution prior to experimental run at optimum operational condition (15 V, 1 cm, 30 min). Outcome of these runs are demonstrated in Fig. 7 indicating the increment of dye removal efficiency from 45.2 to 92.7% after 5 min contact time when the electrolyte concentration increased from 0 to 1 mM. Also with increasing of electrolyte concentration from 0 to 1 mM, the required time to achieve dye removal percentage of 98.5% decreases from 30 to 15 min. So, it can be concluded that, at constant applied voltage and inter electrode distance, higher dye removal efficiency are resulted with the increment of electrolyte concentration in the dye containing waste water. The reason behind this is the increment of conductivity (increased from 57 to 222 µS/cm by the NaCl concentration increment of 0 to 1.0 mM) of solution at higher electrolyte concentration which leads the higher current passing through the solution and corresponding formation high amount of aluminium hydroxide coagulant and thus the dye removal percentage increases. Also, as per the experimental result, with the increment of electrolyte concentration from 0.0 to 1.0 mM, the energy consumption also rises rapidly indicating the more anodic reaction with higher coagulant formation and dye reduction. In addition to this, when NaCl is used as electrolyte, hypochlorite ion forms in the anode which causes the additional destroy of dye molecules by oxidation and increases the overall performance.

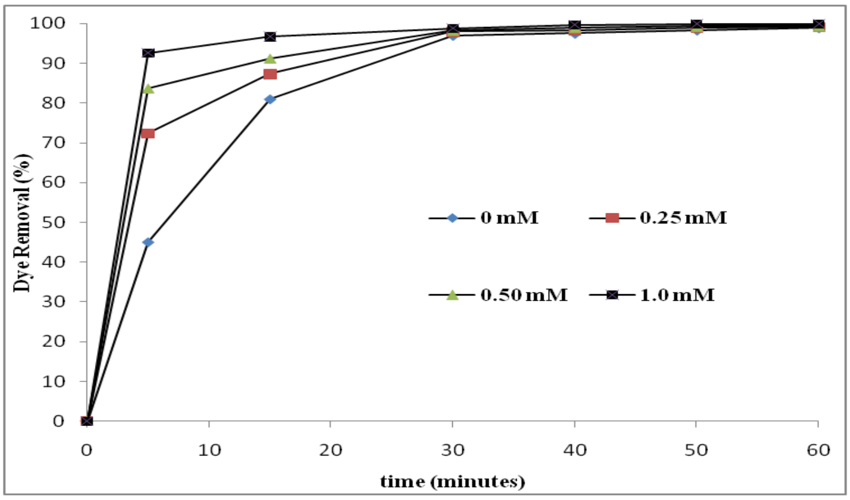

Figure 7. Effect of electrolyte (NaCl) concentration on dye removal (dye concentration: 50 ppm, inter-electrode distance: 1 cm, voltage: 15 V)

4.7. Effect of Mode of Electrode Connection on Dye Removal

Experiments were executed in three configurations as monopolar parallel (mp-p), monopolar series (mp-s), and bipolar for determining the consequence of electrode connection mode on dye removal percentage in optimum condition. The results are illustrated by Fig. 8 indicating monopolar parallel connection mode as the more effective connection for such process than those for monopolar series and bipolar counterparts. Dye removal of 98.01, 85.70, and 71.16% was achieved for mp-p, mp-s, and bipolar respectively for electrocoagulation of 50 ppm dye solution for 30 min. For monopolar parallel operation, dye removal was 99.00% after 60 min of run of the reactor where for mp-s and bipolar connections, the maximum was 91.2% and 88.6% respectively even after 85 min reaction time. The reason behind this is passing of higher current thorough the electrodes at mp-p connection mode than those for mp-s & bipolar connection modes, which releases more Al^{3+} ions and OH^- ions in the solution leading formation of more flocs and thus improve the ability of system to remove more dye molecules in contrast to other connections.

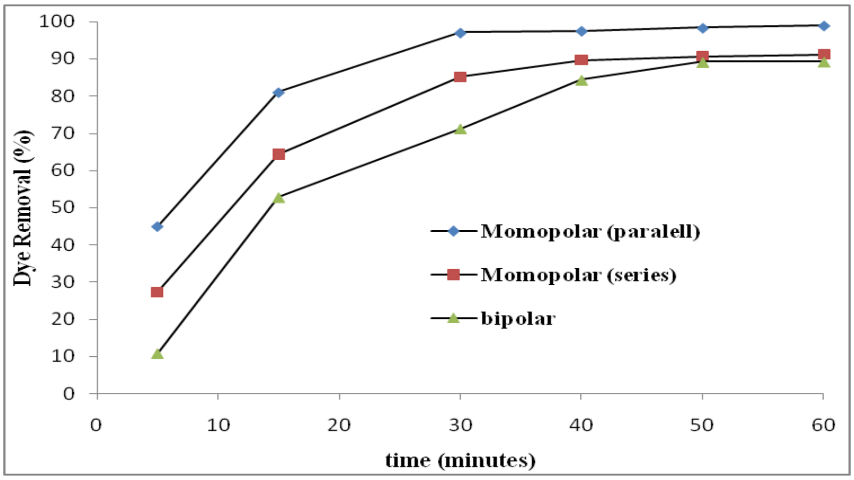

Figure 8. Effect of mode of electrode connection on dye removal (dye concentration: 50 ppm, inter-electrode distance: 1 cm, voltage: 15 V)

4.8. Effect of Current Efficiency on Dye Removal

Current efficiency (φ) is defined as the ratio of experimental aluminum dosage (ΔM_{exp}) to theoretical aluminum dosage ($\Delta M_{theoritical}$) and represented by equation 10.

$$\varphi = (\Delta M_{exp}/\Delta M_{theoritical}) \times 100 \qquad (10)$$

The theoretical anodic dissolution of aluminum is calculated by Faraday's low by equation 11.

$$Al^{3+}_{theoritical} = (MIt)/ZF \qquad (11)$$

where, M, I, t, Z, and F are molecular mass of aluminum (26.98 g/mol), the electrical current (Ampere), time of reaction (second), the number of electron moles (3), and Faraday's constant (96487 c/mol) respectively.

For calculating the ΔM_{exp}, after electrocoagulation, the aluminum electrodes were weighed subsequent to washing with acid, cleaning with polymeric brush and drying in air oven. As per the obtained data, the theoretical and practical anodic dissolution of aluminum are 0.05033 g and 0.127 g respectively for current density of 0.30 Ampere. The possible reason for this mass consumption may be the chemical hydrolysis of the cathode. Also it can be mentioned that, during electrocoagulation, the cathode is chemically attacked by OH^- ions generated during H_2 evolution at high pH (Xueming et al., 2000; Khemis et al., 2005). So, for treating dye waste water of high concentration, high current efficiency will gradually decrease.

4.9. Kinetics Studies

Kinetics studies of treatment process have an important role in determining the hydraulic retention time in any reactor system to achieve desired removal (Prasanna et al., 2005). The kinetics study expresses the solute uptake rate which consecutively controls the residence time required by the sorbent for a reactor in order to reach the desired extent of solute (dye) removal. For designing appropriate treatment systems, it makes us able to predict the rate at which pollutant is removed from aqueous solution and respective flow rates and reactor size needed to obtain the desired quality of the treated water. Also, kinetics studies in wastewater treatment offer cavernous understanding of the reaction pathways and the mechanism of treatment reactions. Moreover, as rate constant describes the rates of change in concentration of reactant per unit time, it is very important to calculate the main strands for design of a wastewater treatment unit.

Experimental results of the present work showed the removal of dye by electrocoagulation process to exhibit first order kinetics with high correlation coefficients (>0.97 ± 0.02). The first order kinetic reaction can be expressed as:

$$ln\frac{C}{C_0} = -kt \tag{12}$$

where, C_0, C, t, and k are the dye concentration before reaction (ppm), dye concentration after reaction (ppm), time of reaction (min), and reaction rate constant (min^{-1}) respectively.

The results of the kinetics study with various operating condition are given in Table 1. From this data, it is evident that the time efficiency of electrocoagulation process is in positive correlation with the applied voltage and electrolyte concentration while it decreases with the increment of introduced dye concentration and inter-electrode distance. Also, the efficiency of mono-polar parallel connection was found higher than those for mono-polar series and bi-polar connection modes.

Table 1. Dye removal status by electrocoagulation process with various operating condition

Connection Mode	**Voltage (V)**	**Initial Dye Concentration (ppm)**	**Inter Electrode Distance (cm)**	**Electrolyte Concentration (mM)**	**Dye Removal (%)**	**k (min^{-1})**
Monopolar parallel	5.0	50	1.00	0.00	97.11	0.580
Monopolar parallel	15.0	50	1.00	0.00	98.48	0.093
Monopolar parallel	30.0	50	1.00	0.00	98.96	0.114
Monopolar parallel	45.0	50	1.00	0.00	99.93	0.152
Monopolar parallel	15.0	100	1.00	0.00	82.74	0.051
Monopolar parallel	15.0	150	1.00	0.00	71.89	0.026
Monopolar parallel	15.0	50	1.00	0.25	99.13	0.107
Monopolar parallel	15.0	50	1.00	0.50	99.38	0.126
Monopolar parallel	15.0	50	1.00	1.00	99.99	0.257
Monopolar parallel	15.0	50	0.50	0.00	84.73	0.104
Monopolar parallel	15.0	50	1.50	0.00	88.92	0.057
Monopolar parallel	15.0	50	2.00	0.00	84.70	0.038
Monopolar Series	15.0	50	0.50	0.00	91.21	0.081
Bipolar	15.0	50	0.50	0.00	89.27	0.042

4.10. Analysis from Economic Viewpoint

The operational cost for electrocoagulation process can be analyzed with various strands as price of electricity, chemical reagents, sludge disposal, labors, maintenance and equipments. But in this study, for electrocoagulation process, cost of electricity and cost of electrode have considered as the main strand for the determination of the economically feasible optimum condition since the balance issues are negligible comparing these. Then, operating cost for electrocoagulation process is:

$$C = x.\mathrm{C_e} + \mathrm{y.C_c} \tag{13}$$

Where, C=specific cost of water treatment (US\$/m^3 of waste water), Ce=specific electrode consumption (Kg/m^3 of waste water), Cc=specific energy consumption (kWh/m^3 of waste water), x= cost of electrode (US\$/kg) and y=cost of electrical energy (US\$/kWh).

The effect of various applied voltage on specific energy consumption, E (kWh/m^3) and specific electrode consumption (kg/m^3) is shown in Fig. 9 from which it is evident that both the consumption and thus cost of treatment increases with increment of applied voltage. As per the plot, operating cost for treatment of one cubic meter colored wastewater at optimum operational condition of 15 V and inter electrode distance of 1cm is 0.665 US\$ for approx. 99.00% dye removal at 30 min which rises to 1.79 US\$ at higher voltage of 45 V at equivalent operating condition (costs are calculated against local prices in Bangladesh).

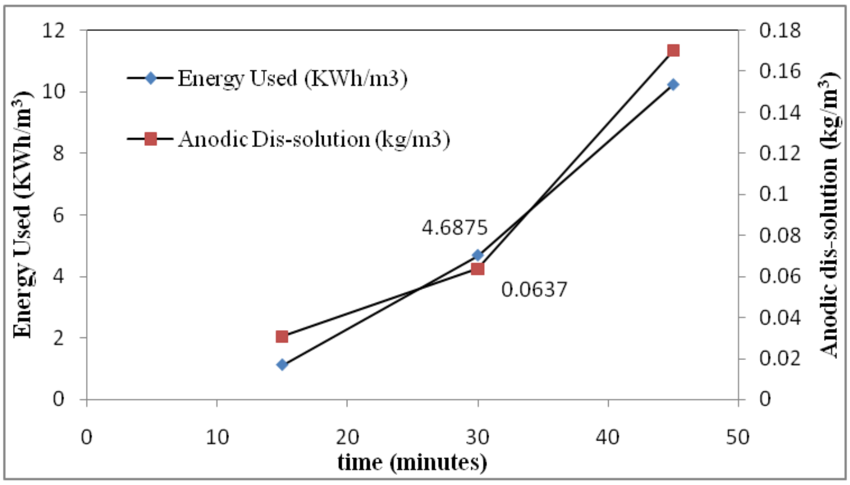

Figure 9. Effect of current efficiency (dye concentration: 50 ppm, inter-electrode distance: 1 cm, voltage: 15 V)

4.11. Efficiency of Electrocoagulation to Remove Different Reactive Dye

The electrocoagulation reactor was operated at a defined state for 5 reactive dyes (as Novacron Red C4B (λ_{max}=580nm), Novacron Ruby S3B (λ_{max}=560nm), Novacron Deep Red CD (λ_{max}=600nm), Novacron Blue CD (λ_{max}=640nm) and Novacron Blue SGL (λ_{max}=680nm)) for investigating the efficiency of electrocoagulation process with different dyes and results are shown in Fig. 10. The result is in quite good agreement with that obtained in section 4.1 where the removal efficiency is increases with electrocoagulation time. Also, it is evident that time requirement for 98% of dye removal as 30, 40, 40, 30 and 60 min respectively with reaction rate constant as 0.093, 0.097, 0.1018, 0.1411 and 0.0721 min^{-1} in sequence. The possible reason behind these is the existence of different functional group, aromatic ring and various bonding arrangements in different dyes which affect the reaction mechanism and rate to give different outcomes.

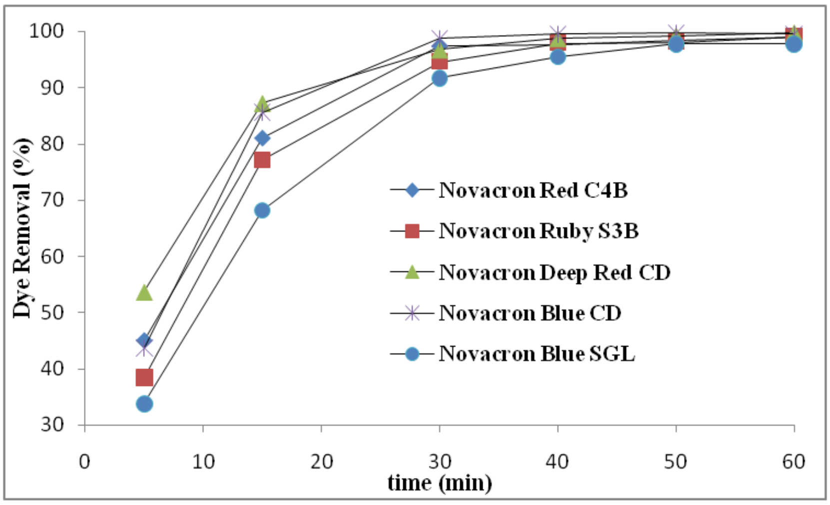

Figure 10. Efficiency of electrocoagulation to remove different reactive dye (dye concentration: 50 ppm, inter-electrode distance: 1 cm, voltage: 15 V)

5. Conclusions

The following conclusions are drawn based on the results: (1) Electrocoagulation is a fast, efficient and clean process for the removal of colour and total organic carbon in reactive dyes from textile waste water. Such system allows higher control with rapid reactions, make a smaller to be viable with employment of only electrons instead of using chemicals and microorganisms to make adoptable. (2) The efficiency of the process for treating of colored wastewater using aluminum electrodes is strongly influenced by the applied voltage, reaction time, inter-electrode distance, electrode connection mode, initial electrolyte concentration and dye concentration. (3) Dye removal, COD removal, electrode consumption, energy consumption and operating cost, were approx 99.00%, 82.7%, 0.0637 kg/m^3, 4.6878 kWh/m^3, and 0.665 US$/m^3, respectively (50 ppm Novacron Red C4B, 15 V, inter-electrode distance of 1 cm and retention time of 30 min). (4) For present study, dye removal followed first order kinetics and the reaction rate constant sturdily depends on the operating condition. (5) Monopolar parallel connection mode is found to be the most efficient connection for dye removal from wastewater rather than monopolar series and bipolar counterparts. (6) In this paper, an experimental representation of the effect of fundamental electrochemical factors in treatment process is presented. For developing countries like Bangladesh where access to safe drinking water is not guaranteed for a majority of the population, such a process is of great importance for maintaining the quality of surface water sources and electrocoagulation process can play a vital role in the water treatment arena due to quite a lot of advantages and the nature of the changing strategic water needs in the world.

Acknowledgments

A line of special gratitude is given to the Holcim (Project BISTAAR) team for the cordial support and advice. Author also wishes to acknowledge for assistance from Tanveer Ahmad (KUET, Bangladesh) for help in experimental run and taking data.

References

Aleboyeh, A., Daneshvar, N., and Kasiri, M. B. (2008). Optimization of C.I. acid red-14 azo dye removal by electrocoagulation batch process with response surface methodology. *Chem. Eng. Process*, **47**: 827-832.

American society for testing and materials. (1995). *Standard Test Methods for Chemical Oxygen Demand (Dichromate Oxygen Demand) of Water*, 1st ed. Philadelphia, Pennsylvania, pp. 1252-1295.

Arslan-Alaton, I., Kabdas, I., Hanbaba, D., and Kuybu, E. (2008). Electrocoagulation of a real reactive dye bath effluent using aluminum and stainless steel electrodes. *J. Hazard. Mater.*, **150**: 166-173.

Bayramoglu, M., Eyvaz, M., and Kobya, M. (2007). Treatment of the textile wastewater by electrocoagulation: Economical evaluation. *Chem. Eng. J.*, **128**: 155-161.

Bhattacharyya, K. G., and Sharma, A. (2003). Adsorption characteristics of the dye, Brilliant Green, on Neem leaf powder. *Dyes Pigments*, **57**: 211-222.

Birhanli, A., and Ozmen, M. (2005). Evaluation of toxicity and teratogenity of six commercial textile dyes using the frog embryo teratogenesis assay–Xenopus. *Drug Chem. Toxicol.*, **28**: 51-65.

Carliell, C. M., Barclay, S. J., Naidoo, N., Buckley, C. A., Mulholland, D. A., and Senior, E. (1995). Microbial decolorisation of reactive azo dye under anaerobic conditions. *Water (SA)*, **21**: 61-69.

Chantaraporn, P., Suprangpak, P., Warangkana, T., Benjawan, K., and Thanawin, N. (2010).

Electrocoagulation of blue reactive, red disperse and mixed dyes, and application in treating textile effluent. *J. Environ. Manag.*, **91**: 918-926.

Chen, A. G. (2004). Electrochemical technologies in wastewater treatment. *Sep. Purifi. Technol.*, **38**: 11-41.

Chu, W., and Tsui, S. M. (2002). Modeling of photodecoloration of azo dye in a cocktail photolysis system. *Water Res.*, **36**: 3350-3358.

Churchly, J. H. (1994). Removal of dyewaste color from sewage effluent—the use of full-scale ozone plant. *Water Sci. Technol.*, **30**: 275-284.

Daneshvar, N., Sorkhabi, H. A., and Tizpar, A. (2003). Decolorization of orange II by electrocoagulation method. *Sep. Purif. Technol.*, **31**: 153-162.

Daneshvar, N., Oladegaragoze, A., and Djafarzadeh, N. (2006). Decolorization of basic dye solutions by electrocoagulation: An investigation of the effect of operational parameters. *J. Hazard. Mater.*, **129**: 116-122.

De Roos, A. J., Ray, R. M., Gao, D. L., Wernli, K. J., Fitzgibbons, E. D., Ziding, F., Astrakianakis, G., Thoma, D. B., and Checkoway, H. (2005). Colorectal cancer incidence among female textile workers in Shanghai, China: A case-cohort analysis of occupational exposures. *Cancer Causes and Control*, **16**: 1177-1188.

Estlander, T. (1988). Allergic dermatoses and respiratory diseases from reactive dyes. *Contact Dermatitis,* **18**: 290-297.

Gahr, F., Hermanutz, F., and Oppermann, W. (1994). Ozonation—an important technique to comply with German laws for textile wastewater treatment. *Water Sci. Technol.*, **30**: 255-263.

Gong, R., Jin, Y., Chen, J., Hu, Y., and Sun, J. (2007). Removal of basic dyes from aqueous solution by sorption on phosphoric acid modified rice straw. *Dyes Pigments*, **73**: 332-337.

Gonzales, C. A., Riboli, E., and Lopez-Abente, G. (1988). Bladder cancer among workers in the textile industry: Results of a Spanish case-control study. *Amer. J. Indus. Med.*, **14**: 673-680.

Hachem, C., Bocquillon, F., Zahraa, O., and Bouchy, M. (2001). Decolourization of textile industry wastewater by the photocatalytic degradation process. *Dyes Pigments*, **49**: 117-125.

Hatch, K. L., and Maibach, H. I. (1995) Textile dye dermatitis. *J. Amer. Acad. Dermatol.*, **32**: 631-639.

Jiang, H., and Bishop, P. L. (1994). Aerobic biodegradation of azo dyes in biofilms. *Water Sci. Technol.*, **29**: 525-530.

Jumasiah, A., Chuah, T. G., Gimbon, J., Choong, T. S. Y., and Azni, I. (2005). Adsorption of basic dye onto palm kernel shell activated carbon: Sorption equilibrium and kinetics studies. *Desalination*, **186**: 57-64.

Kang, S. F., and Chang, H. M. (1997). Coagulation of textile secondary effluents with Fenton's reagent.

Water Res., **36**: 215-222.

Keneklis, T. B. (1981). Fiber reactive dye toxicological profiles, In: *Environmental Control*, 1st ed. Incorporated, Washington, D.C., U.S.A., p. 271.

Khemis, M., Tanguy, G., Leclerc, J. P., Valentin, G., and Lapicque, F. (2005). Electrocoagulation for the treatment of oil suspensions relation between the rates of electrode reaction and efficiency of waste removal. *Process Saf. Environ. Prot.*, **83**: 50-57.

Kim, T. H., Park, C., Shin, E. B., and Kim, S. (2002). Decolorization of disperse and reactive dyes by continuous electrocoagulation process. *Desalination*, **150**: 165-175.

Kim, T. H., Park, C. H., Shin, E. B., and Kim, S. Y. (2004). Decolorization of disperse and reactive dye solutions using ferric chloride. *Desalination*, **161**: 49-58.

Kumar, M. N. V. R., Sridhari, T. R., Bhavani, K. D., and Dutta, P. K. (1998). Trends in color removal from textile mill effluents. *Colourage*, **45**: 25-34.

Lin, A. S. H., and Peng, C. F. (1994). Treatment of textile wastewater by electrochemical method. *Water Res.*, **28**: 277-282.

Lin, S. H., and Peng, C. F. (1996). Continuous treatment of textile wastewater by combined coagulation, electrochemical oxidation and activated sludge. *Water Res.*, **30**: 587-592.

Mahony, T. O., Guibal, E., and Tobin, J. (2001). Reactive dye biosorption by *Rhizopus arrhizus* biomass. *Enzyme Microb. Technol.*, **31**: 456-463.

Mane, V. S., Mall, I. D., and Srivastava, V. C. (2007). Use of bagasse fly ash as an adsorbent for the removal of brilliant green dye from aqueous solution. *Dyes Pigments*, **73**: 269-278.

Manzini, B. M., Motolese, A., Conti, A., Ferdani, G., and Seidenari, S. (1996). Sensitization to reactive textile dyes in patients with contact dermatitis. *Contact Dermatitis*, **34**: 172-175.

Muthukumar, M., Karuppiah, M. T., Raju, G. B., and Thalamadai, **F**. (2007). Electrochemical removal of CI acid orange 10 from aqueous solutions. *Sep. Purif. Technol.*, **55**: 198-205.

Nakagawa, K., Namba, A., Mukai, S. R., Tamon, H., Ariyadejwanich, P., and Tanthapanichakoon, W. (2004). Adsorption of phenol and reactive dye form aqueous solution on activated carbons derived from solid wastes. *Water Res.*, **38**: 1791-1798.

Neppolian, B., Sakthivel, S., Arabindoo, B., and Murugesan V. (2002). Solar/UV induced photocatalytic degradation of three commercial textile dyes. *J. Hazard. Mater*., **89**: 303-317.

Nilsson, R., Nordlinder, R., Wass, U., Meding, B., and Belin, L. (1993). Asthma, rhinitis, and dermatitis in workers exposed to reactive dyes. *British J. Indus. Med.*, **50**: 65-70.

Pala, A., and Tokat, E. (2002). Color removal from cotton textile industry wastewater in an activated sludge system with various additives. *Water Res.*, **36**: 2920-2925.

Papic, S., Koprivanac, N., Bozic, A. L., and Metes, A. (2004). Removal of some reactive dyes from synthetic wastewater by combined Al(III) coagulation/carbon adsorption process. *Dyes Pigments,* **62**: 293-300.

Park, H. S., Lee, M. K., Kim, B. O., Lee, K. J., Roh, J. H., Moon, Y. H., and Hong, C. S. (1991). Clinical and immunologic evaluations of reactive dye-exposed workers. *J. Aller. Clin. Immunol.,* **87**: 639-649.

P´erez, M., Torrades, F., Dom´enech, X., and Peral, J. (2002). Fenton and photo-Fenton oxidation of textile effluents. *Water Res.,* **36**: 2703-2710.

Picard, T., Cathalifaund-Feuillade, G., Mazet, M., and Vandensteendam, C. (2000). Cathodic dissolution in the electrocoagulation process using aluminum electrodes. *J. Environ. Monit.,* **2**: 77-80.

Prasanna, D. B., and Sanjeev, C. (2005). Electrochemical denitrificaton of simulated ground water. *Water Res.,* **39**: 4065-4072.

Rajeshwar, A, K., Ibanez, J., and Swain, G. M. (1994). Electrochemistry and the environment, *J. Appl. Electrochem.,* **24**: 1077-1091.

Robinson, T., McMullan, G., Marchant, R., and Nigam, P. (2001). Remediation of dyes in textile effluent: A critical review on current treatment technologies with a proposed alternative. *Bioresour. Technol.,* **77**: 247-255.

Robinson, T., Chandran, B., and Nigam, P. (2002). Removal of dyes from a synthetic textile dye effluent by biosorption on apple pomace and wheat straw. *Water Res.,* **36**: 2824-2830.

Sanroman, A. M. A., Pazos, M., Ricart, M. T., and Cameselle, C. (2004). Electrochemical decolourisation of structurally different dyes. *Chemosphere,* **57**: 233-239.

Shaul, G. M., Holdsworth, T. J., Dempsey, C. R., and Dostal, K. A. (1991). Fate of water soluble azo dyes in the activated sludge process. *Chemosphere*, **22**: 107-119.

Song, S., He, Z., Qiu, J., and Chen X. L. (2007). Ozone assisted electrocoagulation for decolorization of C.I. reactive black 5 in aqueous solution: An investigation of the effect of operational parameters. *Sep. Purif. Technol.,* **55**: 238-245.

Thebault, P., Cases, J. M., and Fiessinger, F. (1981). Mechanism underlying the removal of organic micropollutants during flocculation by an aluminium or iron salt. *Water Res.,* **15**: 183-189.

Thoren, K., Meding, B., Nordlinder, R., and Belin, L. (1980). Contact dermatitis and asthma from reactive dyes. *Contact Dermatitis,* **15**: 186-192.

Wilkinson, S. M., and McGechaen, K. (1996). Occupational allergic contact dermatitis from reactive dyes. *Contact Dermatitis,* **35**: 376-378.

Wollin, K. M., and Gorlitz, B. D. (2004). Comparison of genotoxicity of textile dyestuffs in Salmonella mutagenicity assay, in vitro micronucleus assay, and single cell gel/comet assay. *J.*

Environ. Pathol. Toxicol. Oncol. **23**: 267-278.

Xueming, C., Guohua, C., and Po, L. Y. (2000). Separation of pollutants from restaurant wastewater by electrocoagulation. *Sep. Purif. Technol.*, **19**: 65-76.

Yang, X., and Al-Durf, B. (2001). Application of branched pore diffusion model in the adsorption of reactive dyes on activated carbon. *Chem. Eng.*, **83**: 15-23.

Zanoni, M. V. B., Sene, J., and Anderson, M. A. (2003). Photoelectrocatalytic degradation of Remazol Brilliant Orange 3R on titanium dioxide thin-film electrodes. *J. Photochem. Photobiol. A: Chem.*, **157**: 55-63.

Zielinska, B., Grzechuslka, J., and Morawski, A. W. (2003). Photocatalytic decomposition of textile dyes on TiO_2 Tytanpol A11 and TiO_2-Degussa P25. *J. Photochem. Photobiol. A: Chem.*, **157**: 65-70.