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# Removal of acid green dye 50 from wastewater by anodic oxidation and electrocoagulation—A comparative study

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# ABSTRACT

The present work represents a comparative study for removing acid green dye 50 by anodic oxidation and electrocoagulation using a new batch self gas stirred electrochemical cell. The effect of operating parameters such as current density, initial dye concentration, NaCl concentration and pH on the efficiency of colour removal has been examined. The chemical oxygen demand (COD) reduction under suitable operating conditions was also calculated and found to be reduced by 68% and 87% in case of electrocoagulation is more economic than anodic oxidation, energy consumption ranged from 2.8 to 12.8 kWh/kg dye removed in case of electrocoagulation while in case of anodic oxidation it ranged from 3.31 to 16.97 kWh/kg dye removed. Although the mechanisms of electrocoagulation and anodic oxidation are different, results show that the first-order rate equation provides the best correlation for the decolourization rate of acid green 50 by the two methods.

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## 1. Introduction

The production of textiles represents one of the big consumers of high water quality. Textile industry actually represents a range of industries with operation and processes as diverse as its products. Fabrics, after its manufacturing, are subjected to several wet processes collectively known as "finishing" and it is in these finishing operations that the major waste effluents are produced [1]. Wastewater from textile dyeing and finishing factories is a significant source of environmental pollution because its high concentration of organic compounds can damage seriously aqueous ecosystems. Textile wastewater is characterized by high chemical oxygen demand (COD), low biodegradability and high-salt content. It is the source of aesthetic pollution related to colour. There are many processes to remove dyes from coloured effluents such as adsorption, precipitation, chemical degradation, photodegradation, biodegradation and chemical coagulation [2,3]. However, these processes are quite expensive and involve several operational problems. For these reasons there has been increasing interest in the use of new methods. Electrochemical methods have advantages such as they require of no chemicals before and after treatment, thus producing no sludge, requirement of small area and low investment cost [4]. One of these methods is the electrochemical oxidation of wastewater containing organic compounds [5–15]. In the electrochemical oxidation process, the pollutants are destroyed by either the direct or indirect oxidation process. In a direct anodic oxidation process, the pollutants are first adsorbed on the anode surface and then destroyed by the anodic electron transfer reaction. In an indirect process, strong oxidants such as hypochlorite/chlorine, ozone, and hydrogen peroxide are electrochemically generated. The pollutants are then destroyed in the bulk solution by oxidation reaction of the generated oxidant [8]. In presence of chloride, oxidation of organic compounds is mediated by active chloro species. The main reactions at the electrodes are [16,17]:

anode :	2Cl-	$\rightarrow Cl_2 + 2e^-$	(1	1)	)
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cathode: 
$$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$$
 (2)

In the solution bulk Cl<sub>2</sub> hydrolysis takes place as follows:

$$Cl_2 + H_2O \leftrightarrow HOCl + H^+ + Cl^-$$
(3)

$$HOCI \leftrightarrow H^+ + OCI^- \tag{4}$$

At the anode O<sub>2</sub> evolution competes with Cl<sub>2</sub> evolution according to the reaction:

$$H_2 O \rightarrow 2H^+ + \frac{1}{2}O_2 + 2e$$
 (5)

Electrocoagulation technique uses a direct current source between metal electrodes immersed in polluted water. The electrical current causes the dissolution of metal plates including iron or aluminum into wastewater. The metal ions, at an appropriate pH,

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Fig. 1. Electrochemical oxidation cell.

can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate the suspended particles or precipitate and adsorb dissolved contaminants [18].

Electrocoagulation has been reported to be efficient in removing dyes from wastewater [19–36]. The most widely used electrode materials in electrocoagulation process are aluminum and iron. Aluminum oxidation results in formation of  $Al^{3+}$ , which hydrolyzes to form hydroxides depending on the pH (Eq. (7)) [37].

$$Al_{(s)} \to Al^{3+} + 3e^{-}$$
:  $E^{\circ} = 1.662 V$  (6)

$$\mathrm{Al}^{3+} + n\mathrm{H}_2\mathrm{O} \to \mathrm{Al}(\mathrm{OH})^{(3-n)}_n + n\mathrm{H}^+ \tag{7}$$

At the cathode H<sub>2</sub> evolution takes place according to the reaction:

$$6H_2O + 6e \rightarrow 3H_2 + 6OH^-$$
 (8)

The predominate formed amorphous  $Al(OH)_{3(s)}$  "sweep flocs" have large surface areas, which are beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. Finally, these flocs are removed easily from aqueous medium by sedimentation or H<sub>2</sub> flotation [18].

In the present work, the performance of electrochemical oxidation and electrocoagulation systems for the removal of acid green dye 50 from wastewater has been investigated with the object of finding out which method is more cost effective in treating textile effluents. The two processes were carried out in a new gas stirred batch reactor where cathodically evolved  $H_2$  stirred the solution. This would reduce the capital and operating costs of the reactor as a result of eliminating mechanical stirring. Previous studies have shown that gas stirring is as efficient as mechanical stirring if the cell is designed properly [38]. Stirring is beneficial to both anodic oxidation and electrocoagulation because it improves the mixing conditions and eliminates concentration polarization [16].

# 2. Experimental technique

Commercially available acid green 50 dye was obtained from Isma Dye Company, Kafr El Dawar, Egypt. Its chemical structure is  $C_{27}$   $H_{25}$   $N_2$   $NaO_7S_2$  (M. wt. = 576.61). Distilled water was used to prepare the desired concentration of dyestuff solution.

The experimental setup is shown in Figs. 1 and 2 for electrochemical oxidation and electrocoagulation units, respectively. The electrochemical oxidation unit consists mainly of a plexiglass cylindrical vessel having a diameter of 15 cm and a height of 30 cm. The vessel was equipped with eight graphite rod anodes of diameter





1 cm, fixed to the bottom of container  $45^{\circ}$  from one another at a distance of 1.2 cm from the wall. Graphite was used as anode in view of the fact that O<sub>2</sub> overpotential is high on graphite while Cl<sub>2</sub> overpotential is low [16], this would increase current efficiency of Cl<sub>2</sub> evolution. A cylindrical stainless steel screen cathode (mesh no. 10) was placed at a distance 1 cm in front and behind of the anode. Stainless steel was used as a cathode in view of its stability in the solution. The electrocoagulation unit consists mainly of a plexiglass cylindrical vessel having a diameter of 10 cm and a height of 16 cm. A cylindrical aluminum sheet lining the inner wall and the bottom of the vessel was used as anode, while a cylindrical aluminum screen cathode having geometry similar to the anode was placed at a distance of 1 cm from the anode.

The electrodes in the two units were connected to a dc power supply (20 V, 10 A) with a voltage regulator and a multi-range ammeter connected in series.

Before the beginning of each run, 3 L of solution was placed into the electrochemical unit after mixing with the appropriate amount of a sodium chloride. The pH of the solution was measured by pH meter and adjusted by adding sodium hydroxide or hydrochloric acid solutions. The current density was adjusted to the desired value by using the power supply regulator. Samples were drawn at regular intervals of 3 min during the experiment period using 10 ml pipette. All the experiments were performed at room temperature. In electrocoagulation, after the end of each run, the electrodes were rinsed in dilute HCl solution to remove any oxide film formed during experiment and passivate it.

The dye concentration was determined using UV–vis spectrophotometer (Labomed, USA) with the calibration method at maximum wavelength of 640 nm. The colour removal efficiency in the treatment experiments was calculated as follows:

$$\% \text{ colour removal} = \frac{C_0 - C_t}{C_0} \times 100$$
(9)

where  $C_0$  and  $C_t$  are the initial dye concentration and concentration of dye at time *t* in solution (mg/L), respectively.

The COD was measured by a volumetric analytical method [39]. In both anodic oxidation and electrocoagulation no external

mechanical stirring was used depending on the fact that the cathodically generated  $H_2$  bubbles stir the solutions by inducing microconvection and macroconvection in the cell [16,40].

#### 3. Results and discussions

#### 3.1. Electrochemical oxidation

Fig. 3 presents the percentage colour removal during the electrochemical oxidation at different current densities. It is clear that, the rate of colour removal increases with increasing current density, this is attributed to the increase of Cl<sub>2</sub>, HOCl and OCl<sup>-</sup> concentrations in the cell solution, which eventually increase the dye degradation. Beyond 3.51 mA/cm<sup>2</sup> there is no significant increase



**Fig. 3.** Effect of current density on efficiency of colour removal ( $C_0 = 100 \text{ mg/L}$ , NaCl = 1 g/L, pH 6.9, temperature = 25 °C).

in the rate of colour removal, this may be attributed to the fact that the discharge potential  $Cl_2$  increases with current density and becomes close to the discharge potential of  $O_2$  [40]. Under such conditions simultaneous evolution of  $O_2$  along with  $Cl_2$  takes place with a consequent reduction in the current efficiency and the rate of  $Cl_2$  generation. Also increasing the current density would increase the rate of  $OCl^-$  reduction at the cathode according to the reaction:

$$NaOCl + H_2O + 2e \rightarrow NaCl + 2OH^-$$
(10)

Increasing current densities increases the rate of  $H_2$  discharge, the generated  $H_2$  bubbles induce turbulence in the cathode vicinity and increase the rate of diffusion controlled reduction of NaOCl at the cathode [16,40].

Fig. 4 shows the effect of initial dye concentration on the percentage colour removal. As expected, the rate of colour removal decreases with the increase in initial dye concentration, this may be explained by the fact that, increasing dye concentration consumes  $Cl_2$  and  $OCl^-$  rapidly with a consequent decrease in their concentration at a given current density, this decrease the rate of colour removal [41].

Fig. 5 presents the effect of NaCl concentration under fixed current density and dye concentration. It is clear that increasing chloride concentration increases the percentage colour removal. The reason is that increasing NaCl concentration lowers the discharge potential of  $Cl_2$  according to Nernst equation [40] and



Fig. 4. Effect of dye concentration on efficiency of colour removal (c.d. =  $3.51 \text{ mA/cm}^2$ , NaCl = 1 g/L, pH 6.9, temperature =  $25 \degree$ C).



**Fig. 5.** Effect of NaCl concentration on efficiency of colour removal ( $C_0 = 100 \text{ mg/L}$ , c.d. = 3.51 mA/cm<sup>2</sup>, pH 6.9, temperature = 25 °C).

therefore more current is consumed in  $Cl_2$  and hypochlorite generation at the expense of  $O_2$  evolution. Hence, the rate of colour removal increases.

Fig. 6 shows the effect of initial pH on the percentage colour removal. It is clear that varying the initial pH from 3 to 9 does not have significant influence on the colour removal. Many investigators explained this fact on the basis that the generation of chlorine/hypochlorite does not depend on the initial pH conditions [8]. This finding is consistent with previous studies on the use of Cl<sub>2</sub>/OCl<sup>-</sup> system to treat landfill leachate, tannery wastewater, textile wastewater, cresols and phenol [8].

#### 3.2. Electrocoagulation

Fig. 7 shows that the percentage colour removal increases with increasing current density. As the current density increases the amount of dissolved aluminum and the amount of cathodically generated  $H_2$  bubbles increase according to Faraday's law. As a consequence the amount of Al hydroxyl polymers available for adsorption of the dye molecule also increases.

The variation of rate of colour removal with the initial dye concentrations is shown in Fig. 8. It can be seen from the figure that the percentage colour removal decreases as the initial concentration of the dye increases. This is may be attributed to the fact that, as the concentration of dye increases, the adsorption capacity of metallic hydroxide flocs becomes exhausted early. Besides, increasing dye concentration leads to increased association of dye molecules [42],



**Fig. 6.** Effect of pH on efficiency of colour removal ( $C_0 = 100 \text{ mg/L}$ , NaCl = 1 g/L, c.d. = 3.51 mA/cm<sup>2</sup>, temperature = 25 °C).



**Fig. 7.** Effect of current density on efficiency of colour removal ( $C_0 = 100 \text{ mg/L}$ , NaCl = 1 g/L, pH 6.9, temperature = 25 °C).

this decreases the adsorbability of the dye molecules on  $Al(OH)_3$  because of the unavailability of the polar functional groups which are consumed in dye association.

As shown in Fig. 9, the percentage colour removal increases with the increase in NaCl concentration. This may be explained by the fact that the higher Cl<sup>-</sup> concentration the higher the ability of Cl<sup>-</sup> to destroy any passive oxide film which tends to form on aluminum anodes at relatively high potential and limit aluminum dissolution [43].

In order to evaluate the effect of pH of the solution on the percentage colour removal, a series of experiments were conducted and the dye solution was adjusted to the desired pH for each experiment by using sodium hydroxide or hydrochloric acid solutions. Fig. 10 shows that the maximum colour removal efficiency was observed at pH ranging from 6.9 to 11. At low pH, such as 2, cationic monomeric species  $Al^{3+}$  and  $Al(OH)_2^+$  predominate which are not beneficial in colour removal. When pH is between 4 and 11, the  $Al^{3+}$  and  $OH^-$  ions generated at the anode and cathode respectively react to form various monomeric species that finally transform into insoluble amorphous  $Al(OH)_{3(s)}$  through complex polymerization/precipitation kinetics [44] and consequently the adsorption capacity of aluminum flocs toward the dye molecules increases.



Fig. 8. Effect of initial dye concentration on efficiency of colour removal (NaCl = 1 g/L, pH 6.9, c.d. =  $1.67 \text{ mA/cm}^2$ , temperature =  $25 \degree$ C).



**Fig. 9.** Effect of NaCl concentration on efficiency of colour removal ( $C_0 = 100 \text{ mg/L}$ , c.d. = 1.67 mA/cm<sup>2</sup>, pH 6.9, temperature = 25 °C).



**Fig. 10.** Effect of pH on efficiency of colour removal ( $C_0 = 100 \text{ mg/L}$ , NaCl = 1 g/L, c.d. = 1.67 mA/cm<sup>2</sup>, temperature = 25 °C).

## 3.3. COD of the dye solution

In view of the fact that the mechanism of the two processes are different it would be more plausible if the comparison between the two method is made on the basis of COD removal efficiency rather than dye removal efficiency. COD measurements by the standard technique [39] have shown that the %COD reduction is 68 and 87 for electrochemical oxidation and electrocoagulation respectively (Fig. 11). The relatively low COD reduction in case of anodic oxidation calls for further future studies using direct anodic oxidation on highly active anodes such as SnO<sub>2</sub> or boron doped diamond [45]. Anodic incineration of dyes on a highly active anode is more attractive than electrocoagulation which suffers from the formation of a solid waste (sludge) which has to be disposed.

#### 3.4. Energy consumption

To further assist in assessing the economic feasibility of electrochemical oxidation method in comparison with electrocoagulation method, the energy consumption was calculated as follows [46,47]:

energy consumption (kWh/kg dye removed)

$$=\frac{VIt \times 10^{3}}{60(C_{0}-C_{t}) \times \text{treated volume}(L)}$$
(11)

#### Table 1

Values of rate constant at different dye concentrations.

Method	Electrochemical oxidation					Electrocoagulation					
Initial dye concentration (mg/L)	75	100	125	150	200	100	125	150	200	250	300
Rate constant (min <sup>-1</sup> )	0.1802	0.1392	0.1302	0.1176	0.103	0.1767	0.1689	0.153	0.1435	0.1376	0.123



**Fig. 11.** Effect of electrochemical treatment method on COD reduction (NaCl = 1 g/L, pH 6.9,  $C_0$  = 100 mg/L, time = 21 min, temperature = 25 °C, c.d. = 1.67 and 3.51 mA/cm<sup>2</sup> for electrocoagulation and electrochemical oxidation, respectively).

energy consumption (kWh/kg COD removed)



**Fig. 12.** Effect of current density on efficiency of colour removal and energy consumption ( $C_0 = 100 \text{ mg/L}$ , NaCl = 1 g/L, pH 6.9, temperature = 25 °C, time = 21 min). (a) Electrochemical oxidation; (b) electrocoagulation.



**Fig. 13.** Effect of initial dye concentration on efficiency of colour removal and energy consumption (NaCl = 1 g/L, pH 6.9, time = 21 min, temperature =  $25 \degree$ C, c.d. = 1.67 and 3.51 mA/cm<sup>2</sup> for electrocoagulation and electrochemical oxidation respectively). (a) Electrochemical oxidation; (b) electrocoagulation.

where *V* is the cell voltage (V), *I* is the current (A), *t* is the electrolysis time (min),  $C_0$  and  $C_t$  are the initial dye concentration and concentration at time *t* (in mg/L), COD<sub>0</sub> and COD<sub>t</sub> are the initial chemical oxygen demands and chemical oxygen demands at time *t* (in mg<sub>O<sub>2</sub></sub>/L) respectively. Figs. 12–14 show that energy consumption ranges from 3.31 to 16.97 and from 2.8 to 12.8 kWh/kg dye removed depending on the operating conditions for electrochemical oxidation and electrocoagulation respectively. Energy consumption based on COD removal was found to be 3.82 and 5.73 kWh/kg COD removed for electrocoagulation and electrochemical oxidation respectively.

#### 3.5. Kinetic study

The rate of removal of acid green 50 can be represented by the following first-order mechanism:

$$\ln\left(\frac{C_0}{C_t}\right) = kt \tag{13}$$

where  $C_0$  is the initial dye concentration (mg/L), k is the rate constant (min<sup>-1</sup>) and t is the time (min). According to the above equation, a plot of  $\ln(C_0/C_t$  against t will yield a straight line with a



**Fig. 14.** Effect of NaCl concentration on efficiency of colour removal and energy consumption (NaCl = 1 g/L, pH 6.9,  $C_0$  = 100 mg/L, time = 21 min, temperature = 25 °C, c.d. = 1.67 and 3.51 mA/cm<sup>2</sup> for electrocoagulation and electrochemical oxidation, respectively). (a) Electrochemical oxidation; (b) electrocoagulation.



**Fig. 15.**  $\ln(C_0/C)$  vs time at different dye concentration (NaCl = 1 g/L, pH 6.9, temperature = 25 °C, c.d. = 1.67 and 3.51 mA/cm<sup>2</sup> for electrocoagulation and electrochemical oxidation, respectively). (a) Electrochemical oxidation; (b) electrocoagulation.



**Fig. 16.** log k vs log C<sub>0</sub>.

slope of *k*. Fig. 15. reveals reasonably good fit of pseudo first-order kinetic model for electrochemical oxidation and electrocoagulation data. Fig. 16 and Table 1 show that the rate constant for both methods decrease with the increase in initial dye concentration. The decrease in the rate constant of both electrocoagulation and anodic oxidation may be ascribed to the increase in dye molecule association with increasing dye concentration; this gives rise to (i) decrease of the diffusivity of the dye in the solution (ii) decrease in the activity of the dye molecules owing to the consumption of the dye active functional groups in the process of association.

#### 4. Conclusions

Electrochemical oxidation and electorocoagulation were used to remove colour from dye solutions containing acid green 50. The effect of various operational parameters on the efficiency of colour removal was investigated. The results showed that the colour removal increases with the increase in current density, NaCl concentration, while it was found to decrease with the increase in initial dye concentration. The pH was found to be an important parameter for the electrocoagulation process, the maximum efficiency of colour removal was pH ranged from 6.9 to 11, while there is no effect of pH tested in electrochemical oxidation process. The percentage COD reduction was found to be 68 and 87 for electrochemical oxidation and electrocoagulation methods respectively. From the economic point of view, it can be concluded that electrocoagulation seems to be more cost efficient compared to anodic oxidation. To confirm the present conclusion that electrocoagulation is more economic than anodic oxidation, further studies are needed on different dyes to delineate the role of dye structure and the functional groups it contains on the efficiency of the process of dye removal. The results show that the present data fit a pseudo first-order kinetic model for both processes.

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