Decolorization of disperse dye solutions by electrocoagulation

N.K. Amin [a], E-S.Z. El-Ashtoukhy [a] and O. Abdelwahab [b]

a Chemical Eng. Dept., Faculty of Eng., Alexandria University, Alexandria, Egypt
b Environmental Division, National Institute of Oceanography and Fisheries, Alexandria, Egypt

In this study, the decolorization of strong colored solutions containing dispersed dye by electrocoagulation was investigated. The experiments were carried out in a simple electrochemical reactor using iron electrodes in a batch mode. The effect of operating parameters such as electrolysis time, current density, pH, NaCl concentration and temperature on the color removal efficiency have been examined.The results indicate that electrocoagulation is very efficient technique and is able to achieve 100% color removal under suitable operating conditions. The energy consumptions was calculated and found to range from 3.3 to 27 kWh/kg of dye removed. Furthermore, the optimum operating conditions were applied to treat textile carpet wastewater of textile carpet plant (Alexandria, Egypt), 100% color removal was achieved after 50 min of treatment at c.d of 5.76mA/cm^2 and pH equal 9.

في هذه الدراسة فان إزاله اللون للمحاليل ذات القوه اللونية المحتوية علي صبغه مشتته بالتجميع الكهربي تم تحقيقها. تمت التجارب في مفاعل كهر وكيميائي بسيط من النوع الدفعي باستخدام الحديد كانود.تم اختبار تأثير عوامل التشغيل مثل زمن التحليل الكهربي، كثافة التيار، الرقم الهيدروجيني، تركيز كلوريد الصوديوم و الحرارة علي كفاءة ازاله اللون. أوضحت النتائج أن التجميع الكهربي تقنيه فعاله جدا و قادرة علي تحقيق ازاله للون بنسبه ١٠٠% عند ظروف تشغيل مناسبة.وبحساب استهلاك الطاقه وجد انها تتراوح بين ٣,٣ و٢٧ كيلو وات ساعه لكل كيلو جرام من الصبغه المزاله . بالاضافه لذلك فان ظروف التشغيل المثلي تم تطبيقها لمعالجه الماء المتخلف من نسيج مصنع السجاد الموجود بمدينه الاسكندريه بجمهوريه مصر العربية ، تحققت الازاله للون بنسبه ١٠٠% بعد خمسين دقيقه من المعالجة عند كثافة تيار ٥,٧٦ مللي أمبير/سم ^{٢} و أس هيدروجيني ٩.

Keywords: Electrocoagulation, Disperse dye, Color removal, Textile wastewater

1. Introduction

Dye wastewater from textile mills is a series pollution problem because it is high in both color and organic content. Effluents from the dyeing industry are not only aesthetically displeasing but also hinder light penetration and may in consequence disturb biological processes in water bodies. In addition, dyes are toxic to some organisms and hence harmful to aquatic animals. Some dyes are carcinogenic and others after transformations or degradation yield compounds such as aromatic amines, which may be carcinogenic or otherwise toxic. Furthermore, dyes accumulate in sediments at many sites, especially at locations of wastewater discharge, which has an impact on the ecological balance in the aquatic system. Ground water systems are also affected by these pollutants because of leaching from soil [1]. Thus, dyes in wastewater have to be removed before it is discharged into a water body or on land.

Conventionally textile wastewater is treated through biological, physical and chemical methods [2]. Biological methods cannot be applied to most textile wastewater due to the toxicity of most commercial dyes to the organisms used in the process [3]. However, various physical-chemical techniques, such as chemical coagulation [4], chemical oxidation [5], photocatalysis [6], electrochemical [7], adsorption [8] and electro coagulation techniques have been examined.

Most of these methods are becoming inadequate due to the enforcement of new/ or tighter regulations, concerning the discharge of wastewater in many countries.

A host of very promising techniques based on electrochemical technologies have regained the importance during the past two decades. Nowadays, electrochemical technologies have reached such a state that they are not only comparable with other technologies in terms

of cost but also are more efficient and more compact. Among the electrochemical technologies, electrocoagulation is considered to be potentially an effective tool for the treatment of dye wastewater [9]. Electrocoagulation is the process of destabilizing suspended, emulsified or dissolved contaminants in an aqueous medium by introducing an electrical current into the medium, which provides the electromotive force to drive the chemical reaction. In the process a metallic hydroxide flocs is created by electrodissolution of sacrificial anodes, usually made of iron or aluminum. The electrocoagulation process has a great advantage over the traditional chemical coagulation method for wastewater treatment. In chemical coagulation a large volumes of sludge with high bound water content that can be slow to filter and difficult to dewater is generated, it also tends to increase the total dissolved solids content of the effluent, making it unacceptable for reuse within industrial applications. Otherwise, electrocoagulation can often neutralize ion and particle charges, thereby allowing contaminants to precipitate, reducing the concentration below that possible with chemical precipitation, and can replace and/ or reduce the use of expensive chemical agents (metal salts, polymers). Although the electrocoagulation mechanism resembles chemical coagulation in that the cationic species are responsible for the neutralization of surface charges, but the characteristics of the electrocoagulation floc differ dramatically from those generated by chemical coagulation, as it tends to contain less bound water and is more readily filterable. Therefore, electrocoagulation process has proven to be able to cope with a variety of wastewater such as paper pulp mill waste [10, 11], tanneries [12], removal of phosphate from wastewater [13], textile wastewater [14, 15], laundry wastewater [16] and restaurant wastewater [17]. Disperse dyes have an organic composition, almost insoluble in water. They can be applied to most synthetic fibers using simple immersion techniques. The performance of dye removal by electrocoagulation depends upon the solubility of the dyes, the final floc formation and its settling quality. Dyes with low solubility, such as disperse dyes, have

been generally reported to be well removed by flocculation and/ or the electrocoagulation method. Disperse dyes are often azo, anthraquinone and sulfide structures, they form collides in solution, so they tend to be adsorbed by metal hydroxide particles.

In the present work, the decolorization of disperse dye by electrocoagulation has been investigated. Experiments were conducted to examine the effects of the operating parameters, such as pH, initial dye concentration, current density, NaCl concentration, temperature and electrolysis time, on the efficiency color removal. The corresponding energy consumption was also determined as principle cost parameters. The optimum conditions were applied to decolorize textile wastewater obtained from textile carpet factory (Alexandria, Egypt) using electrocoagulation.

2. Experimental

2.1. Materials and methods

Commercially available disperse blue SBL dye was obtained from Isma dye company, Kafr El Dawar, Egypt. Its molecular structure is shown in fig. 1. Distilled water was used to prepare the desired concentration of dyestuff solution.

The experimental setup is shown in fig. 2. The electrocoagulation unit consisted of a rectangular plexiglass container of dimensions $20 \times 20 \times 30$ cm equipped with two parallel vertical iron plate electrodes (20×30 cm) with electrode separation of 1 cm. The electrodes were connected to a digital d.c. power supply (20V, 10A) with a multi-range ammeter connected in series and a voltmeter connected in parallel with the cell. The temperature of the solution was controlled by using a thermostated water bath.
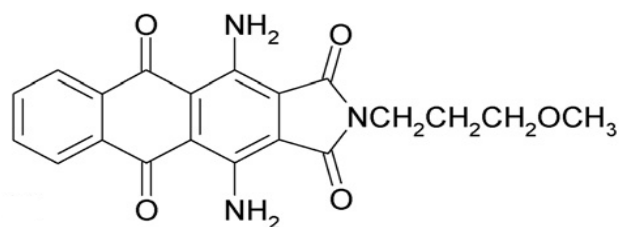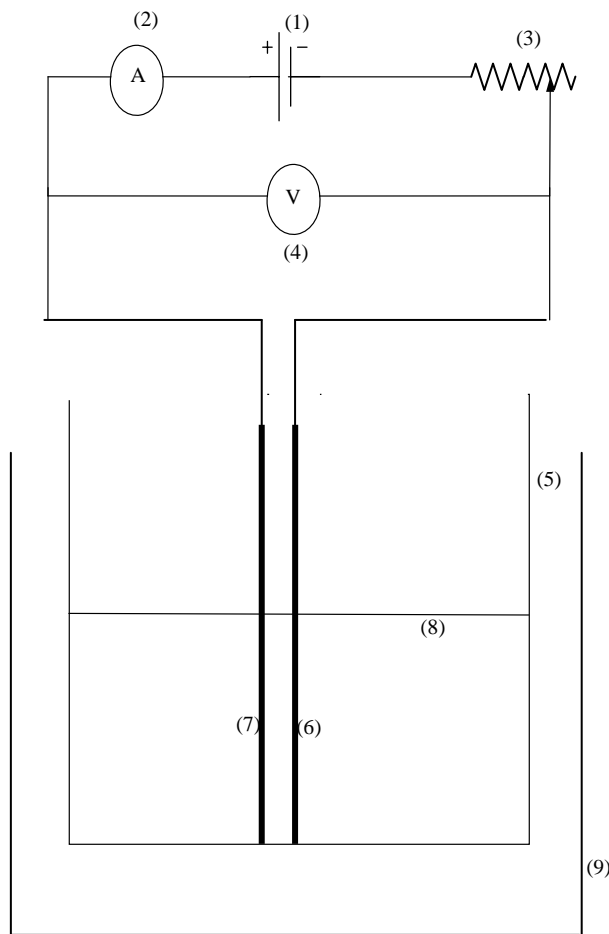

Fig. 1. Chemical structure of disperse blue SBL dye.

1. D.C power supply.
2. Ammeter.
3. Variable resistance.
4. Voltmeter.
5. Rectangular plexiglass container.
6. Vertical iron plate (cathode).
7. Vertical iron plate (anode).
8. Dye solution level.
9. Thermostated water bath.

Fig. 2. Schematic diagram of the experimental set-up.

Before each run, the impurities on the electrodes surface were removed by dipping for 15 min in HCl solution (20%). In a typical run, 6 L of dye solution was placed into the electrocoagulation unit. After the temperature, pH and current density were adjusted to the desired value the experimental run was started, samples were periodically taken from the reactor. All samples were filtered then analyzed.

2.2. Chemical analysis

The dye concentration before and after electrolysis was determined using UV-vis spectrophotometer (Labomed, USA) with the calibration method at maximum wavelength of 600 nm.

The dye removal efficiency was calculated as follows:

$$\% \ Color \ Removal = \frac{C_o - C}{C_o} \times 100 \ . \tag{1}$$

Where C_o and C are the initial dye concentration and concentration of dye at time t in solution (mg/l), respectively.
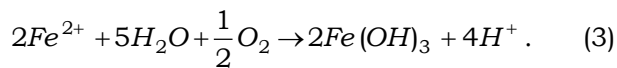
3. Results and discussions

In an electrocoagulation process the coagulating of ions involved three successive stages: (i) formation of coagulants by electrolytic oxidation of the "sacrificial electrode"; (ii) destabilization of the contaminants, particulate suspension and breaking of emulsions; (iii) aggregation of the destabilized phases to form flocs, depending on the pH of the aqueous medium. These compounds have strong affinity for dispered particles as well as counter ions to cause coagulation. The most common electrode materials for electrocoagulation are aluminum and iron as they are cheap, readily available and proven effective [13]. In the present study iron electrode was chosen, where it is upon oxidation in an electrolytic system monomeric ions, $Fe(OH)_n$ where $n=2$ or 3 and polymeric hydroxy complexes are generated. The mechanisms of electrocoagulation for wastewater treatment are very complex. It is generally believed that there are three possible mechanism involved besides electrocoagulation, i.e. electroflotation, electro chemical oxidation and adsorption [17].

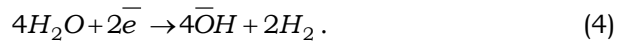Two mechanisms have been proposed for the production of monomeric ions ($Fe (OH)_n$):
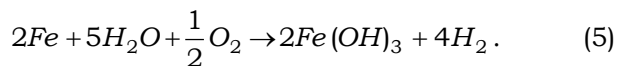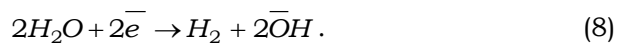(a) Mechanism 1:
• Anode:

$$2Fe \rightarrow 2Fe^{2+} + 4\bar{e} \ . \tag{2}$$

$$2Fe^{2+} + 5H_2O + \frac{1}{2}O_2 \rightarrow 2Fe(OH)_3 + 4H^+ . \quad (3)$$

• Cathode:

$$4H_2O + 2\overline{e} \rightarrow 4\overline{OH} + 2H_2 . \quad (4)$$

• Overall

$$2Fe + 5H_2O + \frac{1}{2}O_2 \rightarrow 2Fe(OH)_3 + 4H_2 . \quad (5)$$

(b)Mechanism 2:
• Anode:

$$2Fe \rightarrow 2Fe^{2+} + 2\overline{e} . \quad (6)$$

$$Fe^{2+} + 2\overline{OH} \rightarrow Fe(OH)_2 . \quad (7)$$

• Cathode:

$$2H_2O + 2\overline{e} \rightarrow H_2 + 2\overline{OH} . \quad (8)$$

• Overall

$$Fe + 2H_2O \rightarrow Fe(OH)_2 + H_2 . \quad (9)$$
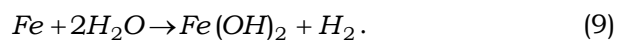
The gases evolved at the electrodes may impinge on cause flotation of the coagulated materials and enhancement in stirring of the solutions.

In all the experiments and during electrolysis the solution becomes green and gas bubbles were seen at the cathode. After electrolysis, the solution becomes clear, a green and yellow sludge were formed at the bottom of cell. The green and yellow colors can probably be attributed to *Fe* (II) and *Fe* (III) hydroxide. These hydroxide flocs have large specific area so it can remove the ions of the dispersed dye by adsorption and precipitation [18].

3.1. Effect of current density and electrolysis time on the efficiency of color removal

It is well known that current determines not only the coagulant dosage rate but also the bubble production rate and the size and growth of flocs which can influence the treatment efficiency of the electrocoagulation, as well as strongly influencing both solution mixing and mass transfer at the electrodes [11, 19]. Thus a set of experiments were carried out to study the influence of current density and electrolysis time on the electrocoagulation reactor. Fig. 3 shows the efficiency of color removal with electrolysis time and current density. The increase in current density and electrolysis time increases the efficiency of color removal, this can be attributed to the fact that the current density and electrolysis time increase the amount of hydroxyl and metal ions produced by the anodic dissolution of iron electrode which play an important role in the removal of dye. Besides, the amount of small sized cathodic H_2 bubbles which assist in floating the formed sludge increases with increasing current density.

3.2. Effect of initial dye concentration on the efficiency of color removal

The variation of rate of color removal with the initial dye concentrations is shown in fig. 4. It can be seen from the figure that the efficiency of color removal decreases considerably from 100 to 8% as the initial concentration of the dye increases from 50 to 1000 mg/L respectively. According to the results, up to a concentration of 100 ppm the adsorption capacity of flocs is not exhausted and the rate of color removal is relatively constant (at about 100% removal). However, beyond this concentration, the adsorption capacity of flocs becomes exhausted. This results is in agreement with many authors who studied the decolorization of dye solution by electrocoagulation [15, 20, 21].

3.3. Effect of NaCl concentration on the efficiency of color removal

In this study, NaCl was added as an electrolyte and the effects of electrolyte concentration on efficiency of dye removal and cell voltage were studied. NaCl is generally added as a supporting electrolyte and a source of chloride reactant for electrochemical processes due to its high conductivity, high
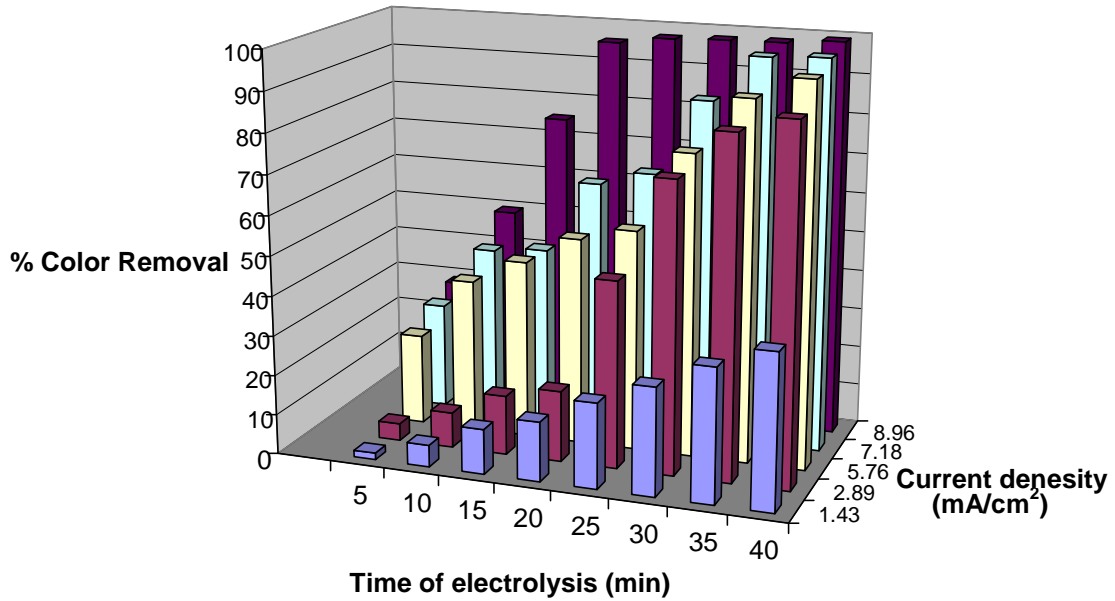
Fig. 3. Effect of current density and electrolysis time on the efficiency of color removal
(C_o= 300 ppm, NaCl = 1.5 g/l, pH=8, Temp. =25^{0}C).

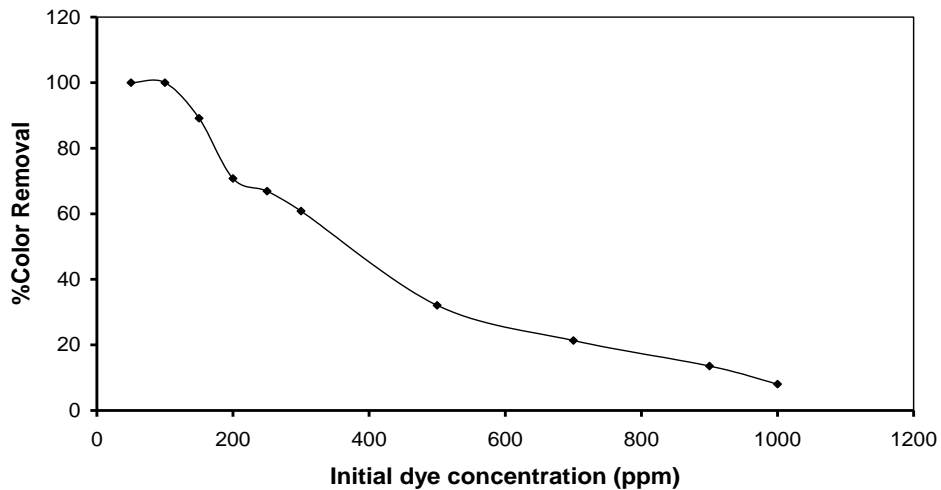Fig. 4. Effect of initial dye concentration on the efficiency of color removal
(Time = 40 min, c.d =5.76 mA/cm², NaCl = 1.5 g/l, pH=8, Temp.=25^{0}C).

solubility, low cost and strongly oxidizing properties of the active chlorines [22]. As shown in fig. 5, the efficiency of color removal increases with the increase in NaCl concentration. This may be explained by the fact that the higher Cl^- concentration the higher the ability of Cl^- to destroy any passive oxide film which tends to form on iron anodes at relatively high potentials and limit iron dissolution [23].

3.4. Effect of temperature on the efficiency of color removal

The electrochemical reaction rate increases when temperature of solution increases. As shown in fig. 6, the efficiency of color removal increase with the increase in temperature up to a certain limit, then the increase in temperature has a slight effect on the color removal. Temperature can affect the rate of dye removal in many ways, namely:
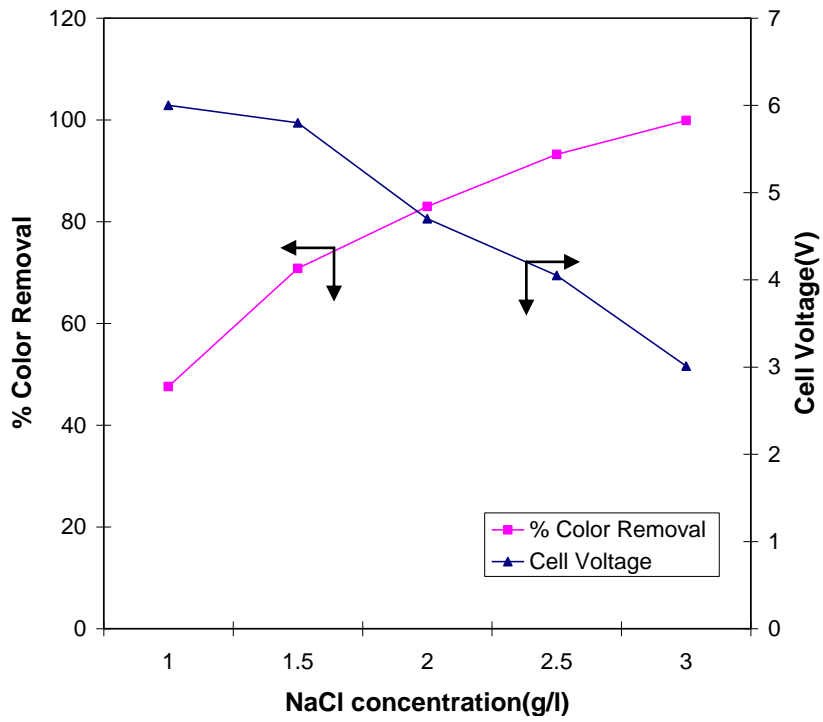
Fig. 5. Effect of electrolyte addition on the efficiency of color removal and cell voltage
(Time = 40 min, C_o= 300 ppm, c.d =5.76 mA/cm^2, NaCl = 1.5 g/l, pH=8, Temp. =25^{0}C).
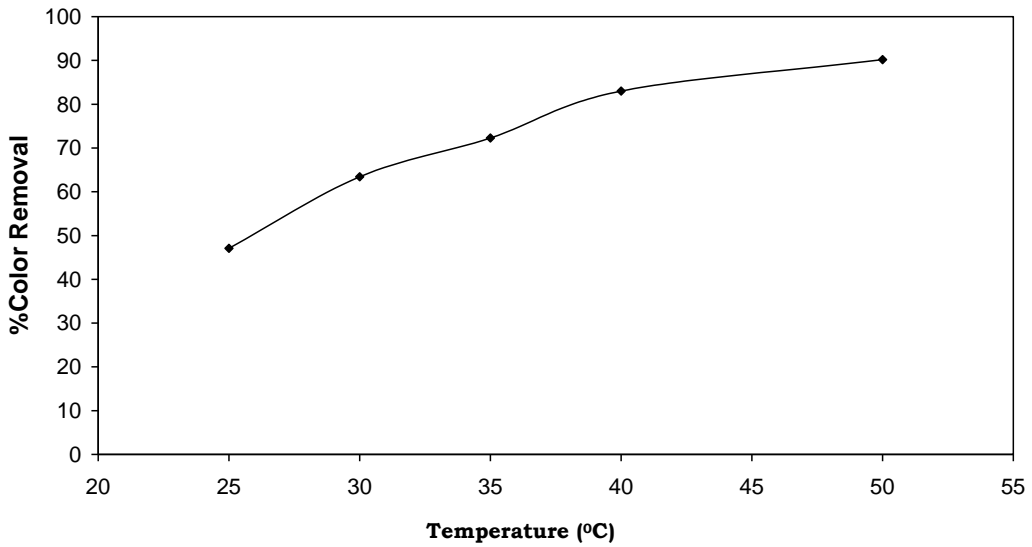

Fig. 6. Effect of temperature on the efficiency of color removal
(Time = 40 min, C_o= 300 ppm, c.d =5.76 mA/cm^2, NaCl = 1.5 g/l, pH=8).

(i) increase in temperature reduces concentration polarization at the dissolving iron anode with a consequent reduction in the tendency of iron to passivate, i.e., increase of temperature favours iron dissolution.

(ii) increase of temperature increases the diffusivity of the dissolved Fe^{++} and improves the mixing conditions in the solution bulk.

The above two factors tend to enhance the rate of dye coagulation. However further increase in temperature may have adverse effects on the rate of dye removal owing to:

(i) removal of dissolved O_2 from the solution with a consequent decrease in $Fe(OH)_3$ concentration which is more effective than $Fe(OH)_2$ in coagulating the dye.

(ii) desorption of the dye molecules from $Fe(OH)_2$ and $Fe(OH)_3$.

(iii) coalescence of the effective small sized H_2 bubbles to less effective large sized H_2 bubbles, as a consequence the floating ability of H_2 bubbles decreases [24] as a result of decreasing the area of H_2 bubbles. It seems that at sufficiently high temperatures, the enhancing effects become balanced by the adverse effects.

3.5. Effect of pH on the efficiency of color removal

It has been established that the pH is an important operating factor influencing the performance of electrochemical process [15, 17, 25].

To examine its effect, the dye solution was adjusted to the desired pH for each experiment by using sodium hydroxide or hydrochloric acid.

Fig. 7, shows the effect of pH on color removal efficiency. The maximum color removal efficiency was observed at pH ranging from 6-8, i.e., so higher removal efficiencies are obtained with iron electrode in neutral and weakly alkaline medium. This can be attributed to the several interaction mechanisms that are possible between dye molecules and hydroxide products and the rates of these depend on pH of the medium and types of ions present. Two major interaction mechanisms are being considered, precipitation and adsorption. In acidic and alkaline medium the iron hydroxide flocs are charged with either positive charge (in acidic medium) or negative charge (in alkaline medium), these charges flocs decrease the adsorption capacity of iron hydroxides to adsorb the dispersed dye molecules. While in neutral and slightly alkaline medium the iron hydroxide flocs are neutral so the adsorption capacity of it towards the dye molecules increases. These results are in agreement with many authors who studied the decolorization of dye solutions by electrocoagulation using iron electrode as a sacrificial anode [22, 26, 27].
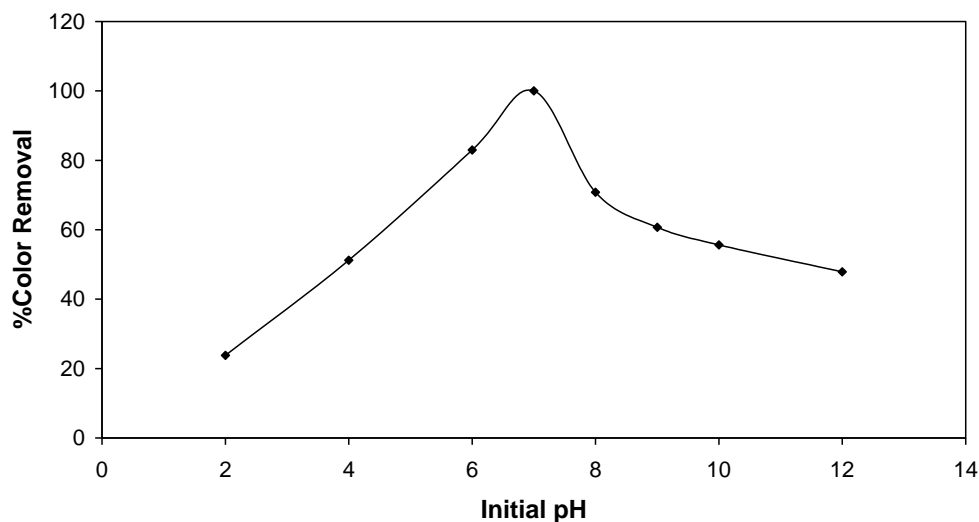

Fig. 7. Effect of initial pH on the efficiency of color removal
(Time = 40 min, C_o= 300 ppm, c.d =5.76 mA/cm², NaCl = 1.5 g/l, Temp. =25⁰C).

3.6. Energy consumption

Electrical energy consumptions are very important economical parameters in electrocoagulation process. The consumed electrical energy per Kg of dye removed was calculated using the following eq. [28, 29]:

Electrical energy consumption

$$(kWh) = \frac{VIt \times 1000}{60(C_o - C_t)}, \qquad (10)$$

where V is the cell voltage (V), I is the current (A), t is the electrolysis time (min), C_o and C_t are the initial dye concentration and concentration at time t (mg/l), respectively.

Figs. 8-10 show the effect of electrolysis time, current density and NaCl concentration on energy consumption respectively. The results show that the energy consumption increases with the increase in electrolysis time and current density while it decreases with the increase in NaCl concentration.

3.7. Application of the present results to the effluents of the textile carpet factory (Alexandria, Egypt)

The wastewater was obtained from the effluent of textile carpets factory located in Alexandria-Egypt, Which produce synthetic and wool carpets. The composition of the wastewater is shown in table 1.

Table 1
Characteristics of the textile wastewater

Parameter	Value
Chemical Oxygen Demand (COD), mg/l	4120
Biological Oxygen Demand (BOD), mg/l	226
Total Dissolved Solids (TDS), mg/l	19160
Total Suspended Solids (TSS), mg/l	675
pH	9
Conductivity, mS/cm	14.6

Fig. 11, shows the effect of electrolysis time on the efficiency of color removal at optimum operating conditions. It is seen that the color removal reaches 92.6% after 45min and 100% after 55min, i.e the electrocoagulation process used in this study is an efficient method for the treatment of textile wastewater.

3.8. Absorbance spectra of the dye solutions and textile wastewater

Fig. 12 (a and b), illustrates the UV-vis adsorption spectra for the disperse dye solution and textile wastewater before and after electrocoagulation process. It can be seen that the decrease of absorbance peaks was directly proportional to the reduction of the dye concentration and almost a complete color removal was achieved for both of them.
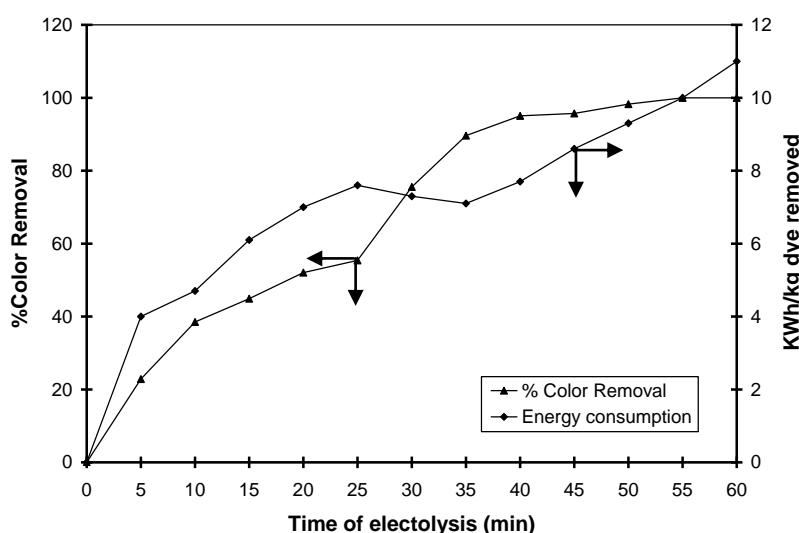

Fig. 8. Effect of electrolysis time on the efficiency of color removal and energy consumption (C_o= 300 ppm, c.d =5.76 mA/cm2, NaCl = 1.5 g/l, pH=8, Temp. =25oC).
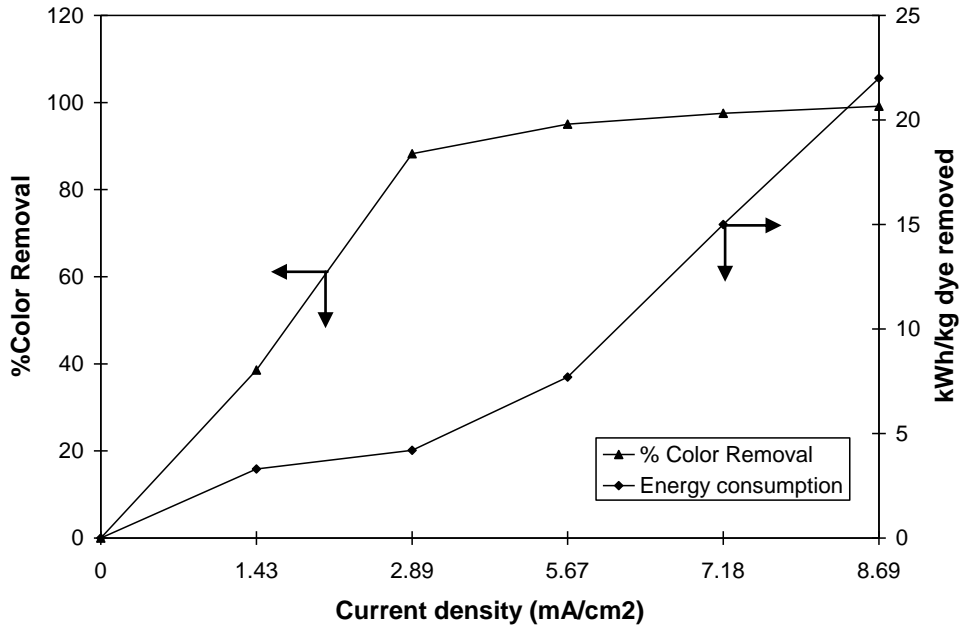
Fig. 9. Effect of current density on the efficiency of color removal and energy consumption
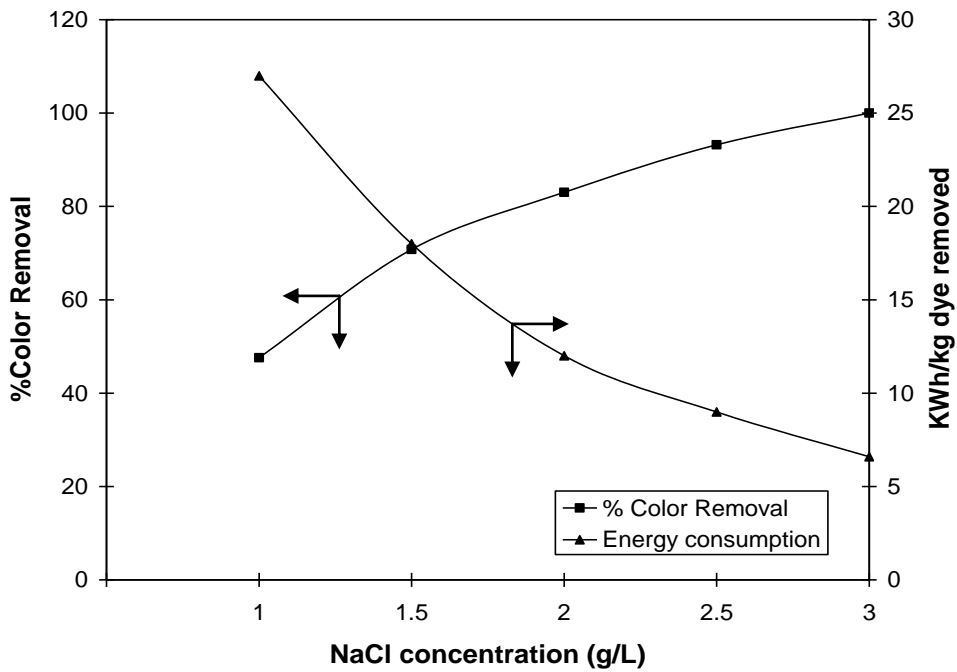(Time= 40 min, C_o= 300 ppm, NaCl = 1.5 g/l, pH=8, Temp. =25ºC).

Fig. 10. Effect of sodium chloride concentration on the efficiency of color removal and energy consumption
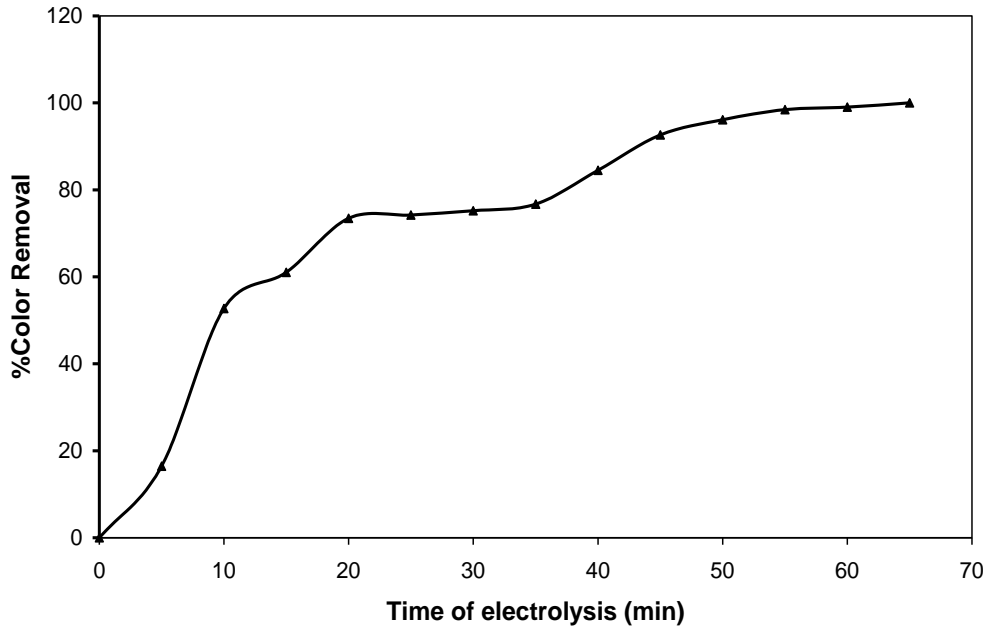(Time=40 min, C_o= 300 ppm, c.d =5.76 mA/cm^2, pH=8, Temp. =25ºC).

Fig. 11. Effect of electrolysis time on the efficiency of color removal for treatment of textile wastewater obtained from textile carpet factory (Alexandria, Egypt) at optimum operating conditions (c.d =5.76 mA/cm^2, NaCl = 1.5 g/l, pH=9, Temp. =25^{0}C).
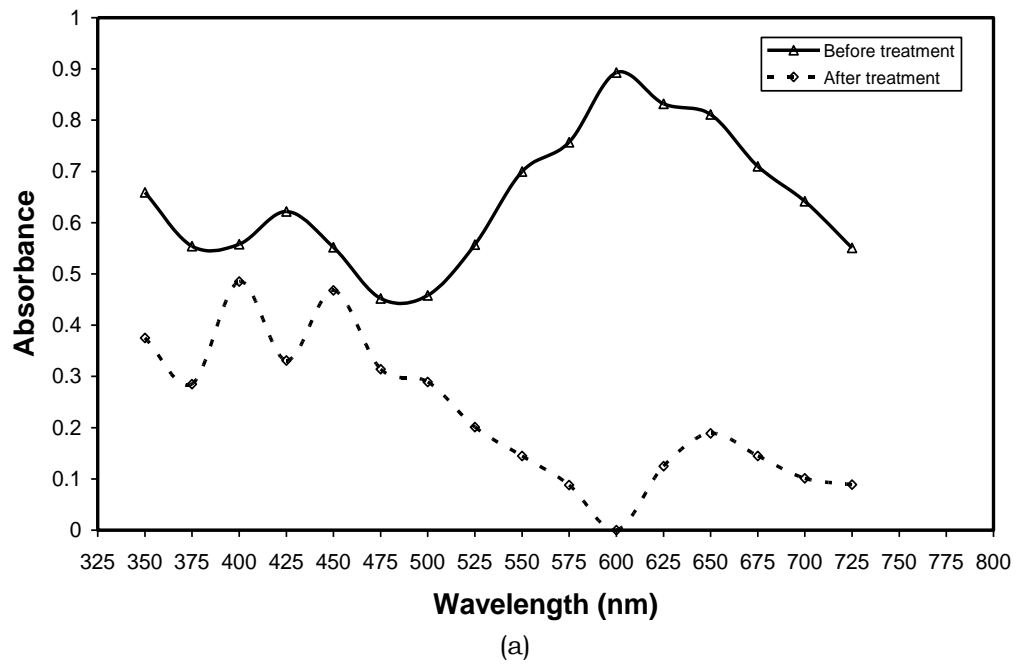

(a)

Fig. 12-a. UV-vis spectra of dispersed dye solution before and after electrocoagulation (Time=40 min, C_o= 300 ppm, c.d =5.76 mA/cm^2, pH=8, NaCl=1.5g/l, Temp. =25^{0}C).
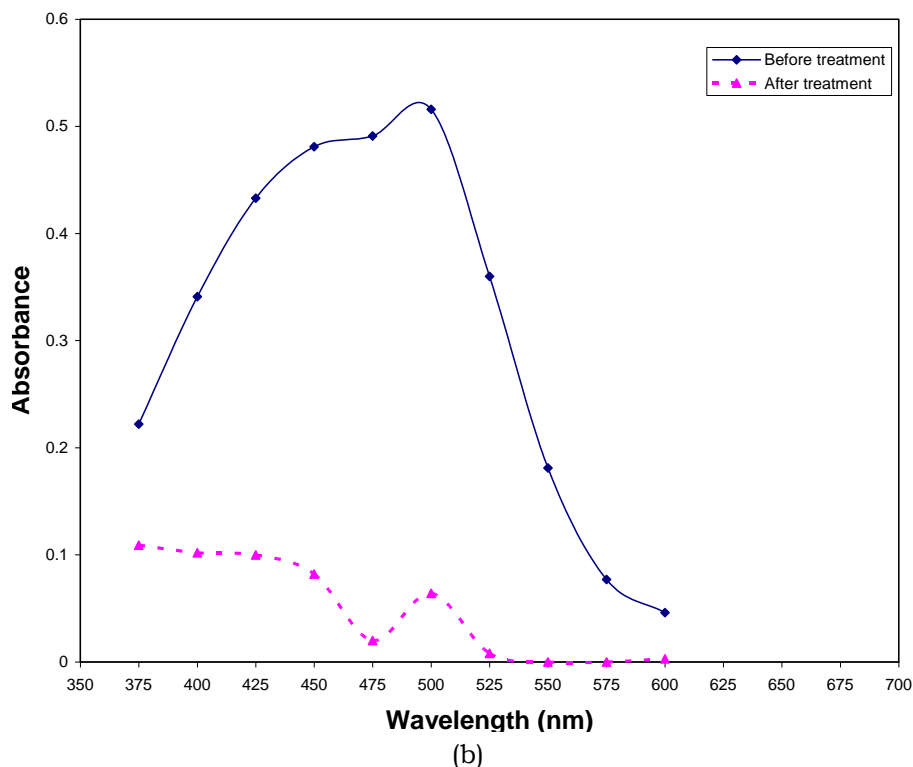
Fig. 12-b. UV-vis spectra of textile wastewater before and after electrocoagulation
(Time=40 min, c.d =5.76 mA/cm^2, pH=9,NaCl=1.5g/l, Temp. =25^{0}C).

4. Conclusions

The electrocoagulation process is successfully applied to remove disperse dye from aqueous solution. The effect of various operational parameters on the efficiency of color removal was investigated. The results showed that the color removal increases with the increase in current density, electrolysis time and NaCl concentration, while it was found to decrease with the increase in initial dye concentration. The increase in solution temperature was found to increase the color removal up to a certain temperature (40°C). The pH was found to be an important parameter for the electrocoagulation process, the maximum efficiency of color removal was in neutral and slightly alkaline medium (6< pH <9). To assist in the economic evaluation of the present technique compared to other competing techniques such as adsorption, anodic oxidation, wet oxidation, photocatalytic oxidation, etc., energy consumption was calculated. Energy consumption ranged from 3.3 to 27 kWh/ kg of dye removed depending on current density, electrolysis time, and NaCl concentration. An actual textile wastewater was treated in the present study by applying the optimum operating conditions. The electrocoagulation set-up described in this study is simple in design and operation and can be used as a convenient tool in the treatment of textile wastewater.

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