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Electrocoagulation of Textile Wastewater with Fe Sacrificial Anode

Ajjam, S.K.[1] and Ghanim, A.N.[2]
Electrochemical Eng. Dep. / Eng. College/ Babylon University
(1) E-mail: satajam58@yahoo.com
(2) E-mail: ala_gh2003@yahoo.com

Abstract
Electrocoagulation is an electrochemical technique used to treat a high polluted effluent whereby sacrificial anodes corrode to release active coagulant precursors into the solution. This research focuses on the performance of EC technique to treat a high strength wastewater textile industry located in Al-Hilla-Iraq using a batchwise mode. Several working parameters such as current density, total suspension solid removal percent, chemical oxygen demand removal percent, turbidity removal percent and the operating time were studied. It was found that the application of 12mA/cm^2 provided 69.2%, 62.5% and 54.3% removal in turbidity, COD and TSS, respectively, while the application of 20mA/cm^2 current density provided 90.1%, 85.2% and 83.1% removal in turbidity, COD and TSS, respectively, within 60 min. of EC treatment and with an inter-electrode spacing of 5cm**.** Also it was found that the contaminants of treated textile wastewater such as BOD, COD, TDS, TSS, turbidity, nitrates, chloride, total hardness, sulfate, total phosphates, electrical conductivity, oil and grease and the total phenols were within allowable limits for wastewater reuse. The loss of particles due to electrocoagulation after treatment as a function of operation time was expressed as a first order kinetic model and the kinetic constant for each contaminant removal was predicted for each current density.

الخلاصة
التخثر الكهروكيمياوي هوتقنيةُ كهروكيمياويةُ تَستعملُ لمعالجة المياه الصناعية المتدفقه ذات المستويات العاليه من التلوّثَ بأستخدام أقطاب تضحية من الحديد أو الألمنيوم تَتأكل لإنتاج مادة مخثرة نشيطة ترتبط مع الملوثات لترسيبها. يُركّزُ هذا البحثِ على أداءِ هذه التقنيةِ في معالجة المياه الملوثه الناتجه من الشركة العامة للصناعات النسيجية في الحلة/العراق بأستخدام منظومة مختبرية تعمل بطريقة الوجبات. تم دراسة عدد من المتغيرات مثل كثافةِ التيارِ، نسبة الأزالة للمواد الصلبة العالقة الكلية، نسبة الأزالة لمتطلبِ الكيمياوي للأوكسجين ، ونسبة الأزالة للكدرة وكذلك زمن التشغيل الكهروكيمياوي . وُجِدَ أن تطبيقَ كثافة تيار 12mA/cm^2 يعطي نسب إزالة 69.2 %، 62.5% و54.3 % في الكدرةِ والمتطلب الكيمياوي للأوكسجين والمواد الصلبة العالقة الكلية على التوالي، بينما تطبيق كثافة تيار 20mA/cm^2 يعطي نسب إزالةَ 90.1 %، 85.2% و83.1 % إزالة في الكدرةِ والمتطلب الكيمياوي للأوكسجين و للمواد الصلبة العالقة الكلية على التوالي ، في زمن 60 دقيقة مِنْ العملية الكهروكيمياوية. وُجِدَ أيضاً أن نسبةجميع الملوثات بعد المعالجة هي ضمن الحد المسموح به عالميا لأعادة أستخدام هذه المياه . تم أقتراح موديل رياضي يمثل حركية التفاعل للجزيئاتِ الملوثة بسبب التخثر الكهروكيمياوي بدلالة وقتِ العمليةِ وكثافة تيار.

Keywords: Electrocoagulation, COD, TDS, Turbidity, operation time.

1. Introduction

Demands of clean industrial wastewater to avoid environmental pollution and especially contamination of pure water resources are become national and international issues for the reuse of wastewater. Innovative, cheap and effective methods of purifying and cleaning wastewater before discharging into any other water systems are needed. Electrocoagulation (EC) technique emerges a simple and efficient

process to be used for wastewater treatment. In recent years, many investigations have been especially focused on the use of EC owing to the increase in environmental restrictions on effluent wastewater (Mollah, M.Y.A et al. 2001, Calvo, L.S. et al. 2003, Carneiro, P.A et al. 2003). Electrocoagulation is an efficient treatment process for various type of wastes such as soluble oils, liquid from food, textile industries, cellulose and effluents from paper industry (Calvo et al., 2003). According to (Can et al., 2006), EC has been proposed in recent years as an effective method to treat various wastewaters such as landfill leachate effluent from restaurant, saline wastewater, tar sand and oil shale wastewater, textile wastewater, urban wastewater, laundry wastewater, nitrate and arsenic bearing wastewater and chemical-mechanical polishing wastewater. Apaydin, Ö. et al. 2009 pointed out that the removal efficiency of the pollutants is 10% higher in the electro-fenton (EF) process than the electrocoagulation (EC) process. In the first 5 min., significant COD removal is achieved in the EF process. Also, it was observed that energy consumption is 20% lower in the EF process than the EC process. Consequently, both the EF process and the EC process show a fast and efficient reaction in the treatment of tannery industry wastewaters. The rate of increase in the percentage COD removal of textile wastewater is high in the beginning of the EC process and approaches monotonical situation beyond 30 min. of process time (Chithra, K., et al., 2008). This can be explained that the more of coagulation process occurs within 30 min. of process time and the existence of excess colloids for the adsorption at high initial concentrations. Chithra, K, et al., 2008 found that the percentage COD removal efficiency increases with an increase in the applied current density. In addition, the rate of bubble generation also increases with increasing current density, which enhances the COD removal efficiency. The EC technique utilizing $Fe(OH)_3$ production through electrolysis with sacrificial iron electrodes can be used to remove a lot of pollutants included acid dyes very effectively via sorption onto the precipitated iron and via Fe(II) driven reduction of azo dyes to acrylamines (Philippe et al., 1998). The pollutants removal efficiency showed that iron was in competition with aluminum in treating the direct dye DR 23 (Phalakornkule et al., 2009). EC is found to be an effective method for the treatment of the slaughterhouse wastewaters (SWW) (Budiyono et al, 2010). Effluent temperature depends on suspended solid (SS) content and increases up to 98°C when SS content was 4000 mg/L. The EC technology can improve wastewater quality and the removal efficiencies of COD and turbidity from wastewater were experimentally done by electrocoagulation technique (Moh Faiqun Ni'am et al., (2007)). They observed that the results obtained from the curves of operating time and settling time indicates that the EC technology can enhances the settling velocity of suspended particles and removal of COD and turbidity. The removal of a reactive textile dye (C.I.Acid yellow 36) from the wastewater using the electrocoagulation method was studied (Kashefialasl, M., et al., 2006). They found that the released ions neutralize the particle charges and thereby initiate coagulation.

Mechanism of Electrocoagulation

The mechanism of the Electrochemical process in aqueous systems is well known. There are three possible mechanisms involved in the process, i.e. electrocoagulation, electroflotation and electro-oxidation. In EC, with electrical current flowing between two electrodes, the coagulant is generated in situ by electrolytic oxidation of the anode material. By using an iron anode the $Fe(OH)_n$ formation with n = 2 or 3 is released at the anode. Simplified oxidation and reduction mechanisms at the anode

and cathode of the iron electrodes are represented as follows (Daneshvar et al, 2003, Larue et al, 2003, Daneshvar et al., 2006, Chithra, K., et al, 2008):

(a) Mechanism 1:

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

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

Cathode: $2H_2O_{(l)} + 2e^{-} \rightarrow H_{2\ (g)} + 2OH^{-}_{(aq)}$

Overall: $Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)}$

(b) Mechanism 2:

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

$4Fe^{2+}_{(aq)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H^{+}_{(aq)}$

Cathode: $8H^{+}_{(aq)} + 8e^{-} \rightarrow 4H_{2(g)}$

Overall: $4Fe_{(s)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4\ Fe(OH)_{3(s)} + 4H_{2(g)}$

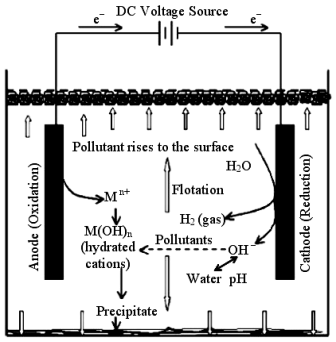

Figure 1: Principle of ectrocoagulation (Calvo et al., 2003; Mollah et al., 2004)

The principle of EC (Fig.1) involved the dissolving of metal anodes continuously to produce metal ions close to the anodic surface. The generation of iron hydroxides (Fe(OH)n) is followed by an electrophoretic concentration of colloids (usually negatively charged), which are swept by the electric field into the region close to the anode (Moh Faiqun Ni'am et al., 2007, Calvo, L.S. et al. 2003, Mollah, M.Y.A et al., 2004). Particles interact with the iron hydroxides and can be removed either by surface complexation or electrostatic attraction (Mollah, M. Y. A et al ,2001, Daneshvar, N., 2003, Larue and Vorobiev 2003). The electric field increases the probability of charges in suspension coming into contact with each other compared to chemical stirring (Mollah, M.Y.A et al.,2001, Larue and Vorobiev, 2003).

The aim of this work is to examine the capability of the EC to treat a high strength textile wastewater located in Al-Hilla-Iraq using a batchwise mode and to study the factors influencing the effectiveness of electrocoagulation process and the feasibility to reuse water after treatment.

2. Analytical method

The effects of textile wastewater characteristics such as total suspended solids, turbidity, and COD removal efficiencies were studied. The electrical potential was held constant for each run, being fixed at 30 volts. The pH, conductivity and turbidity of the solutions were measured by pH meter (pH meter-pHM84), conductrometer (HANNA HI-99301), turbidtimeter (HACH 2100P). COD and total suspended solids measurements were according to the Standard Methods for Examination of Water and Wastewater (APHA, American Public Health Association 2000). The calculation of turbidity, total suspended solids and COD removal efficiencies after electrocoagulation treatment were performed using the following formula (Carneiro, P.A. , 2003).

$$C_R\ (\%) = [(C_0 - C)/C_0] \times 100 \quad (1)$$

Where C_R is contaminant removal, Co and C are concentrations of wastewater before and after electrocoagulation. The rate of change of wastewater concentration, such as turbidity, COD and suspended solid removal can be expressed as a first order kinetic model (Matteson, M. J 1995):

$$C/C_0 = \exp(-kt) \quad (2)$$

3. Experimental Procedure

The electrocoagulator cell used was 1500 ml glass reactor with upper Teflon cover contain four glands for electrodes fixation and equipped with magnetic stirrer without temperature control. The electrolysis time was established of 60 minute using DC power supply of 30 V and a current controlled via rheostat. Two current densities of 12mA/cm^2 and 20mA/cm^2 were tested at ambient temperature between 25-27°C. Removal percents, were examined using two variables: Current density mA/cm^2, and EC operation time (min.). Fig.2 represent the schematic diagram for the experimental set up.

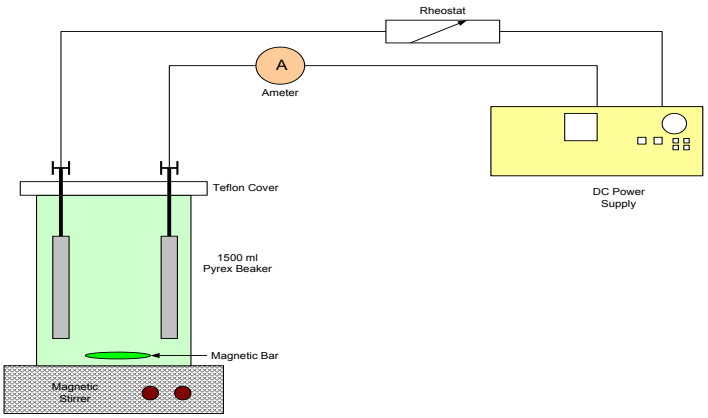

Fig.2 Scheme of experimental set up

In each run, 1 liter of sar gulation process. Two electrodes of iron with surface area of 20cm^2 were used. The separation between the anode and the cathode was kept at 5 cm (Fadil Othman et al., 2006). The solution in the reactor was stirred by a magnetic plate stirrer with Teflon bar at a rotating velocity of 500 rpm (HP-3000). Controlled direct current was supplied by a DC power supply (ATTEN DC power supply type APR 3002A of 0-30V). The current was kept invariant in each test by a rheostat (Wheatstone Type 2755-Japan) and measured by an ammeter (Aswar DT830D, China).

Several EC batch experimental runs were performed in the laboratory. Wastewater was rigorously stirred for 5 min. for homogenization of sample. Then a current density of 12mA/cm^2 was applied for various time periods (ranging from 5 to 60 min.). Treated wastewater samples were collected after settling for predetermined time to estimate the effect of EC treatment time on removal efficiency of TSS, COD and turbidity. In order to see the influence of current density another value of 20mA/cm^2 was used for the same operation time. The sample was allowed to settle for 60 min. after treatment so that the flocs and coagulates that were formed during electrocoagulation may settle. In each experimental run, after settling, about 50 ml supernatant sample was collected for laboratory analysis.

4. Results and Discussion

Characteristics of Raw Textile Wastewater

The sampling and analysis procedures were adopted from Standard Methods for the Examination of Water and Wastewater (APHA, 2000). Details of methods and instruments are presented in Table 1. As observed, the average BOD, COD and TSS concentrations are at the medium strength side as compared to the characteristics of

typical textile wastewater (Chithra, K. 2008). The high value of TDS or electrical conductivity is advantageous to the EC treatment of wastewater since it will eliminate the need to add an electrolyte that is necessary to enhance the current passage in the wastewater.

Table (1): Properties of raw textile wastewater before and after EC technique.

Contaminants	Units	Instruments/Method	Raw Wastewater Before EC	Raw Wastewater After EC	Allowable Limit EPA 1996*
Electrical conductivity	mS/cm	HANNA HI-99301	1324	440	ID
Turbidity	NTU	HACH 2100P	390	38	ID
Total suspension solid TSS	ppm	Gravimetric	1180	190	250-300
Total dissolved solid TDS	ppm	Gravimetric	1241	164	5-180
Ph	_	pH meter-pHM84	4.52	7.62	6-8
Total Hardness	ppm	Gravimetric	270	20	ID
Free Chlorine	mg/l	Titration	198.2	30	ID
Chlorides	mg/l	Titration	50	0.7	ID
Sulfate	mg/l	Spectrophotometer	672	190	ID
Phosphate	mg/l	Spectrophotometer	6.9	0.2.	ID
Nitrates	mg/l	Spectrophotometer	9	0	ID
Phenols	mg/l	Gravimetric	345	0.009	10
Oil and Grease	mg/l	Solvent Extraction	2	0.6	5-40
Biochemical oxygen Demand (BOD)	ppm	DO-meter	110.8	5	5-45.5
Chemical Oxygen Demand (COD)	ppm	Closed Reflex, Titrimetric Method	985	140	91-600

ID: Insufficient Data

* EPA/625/R-96/004 Sep-1996, VOL-1, Appendices

Also, the presence of chloride at relatively high concentrations helps in the production of chlorine as a result of the electrochemical process. Chlorine is an oxidizing agent that can participate in oxidizing soluble ferrous ions into insoluble ferric ions (Larue and Vorobiev, 2003). Furthermore, lower values of phenol shows the quality of textile wastewater is favorable for reuse purpose. Also, the low values of O&G is show that this textile wastewater can be treated easily by EC technique. A comparison of the treated textile wastewater quality with the international wastewater quality standard (EPA 1996 and Commission Decision 2002) allows to judge the effectiveness of EC

process for the treatment of the present textile wastewater to be reused or utilized for other purposes. The analysis shows that BOD, COD, TDS, TSS and turbidity are within the allowable limits of standards. However, the final pH value of treated wastewater is slightly basic (7.6 ± 0.2) which is within the allowable limits. Similarly conductivity, nitrate, total hardness, total phosphates, chloride, sulfate, oil and grease, and the total phenols are below allowable limits.

Effect of EC Operating Time

The textile wastewater effluent treated with iron electrode, appears greenish at the beginning and then turned yellow and turbid. This green and yellow color may be resulted from Fe^{2+} and Fe^{3+} ions generated during EC process. Fe^{2+} is the common ion generated in situ of electrolysis of iron electrode (Moh Faiqun et al. 2007). It has relatively high solubility at acidic or neutral conditions and can be oxidized easily into Fe^{3+} by dissolved oxygen in water (Babu et al., 2007, Chithra, K. et al. 2008). The effect of time was studied with current density of 12mA/cm^2 and 20mA/cm^2.

Figure (3) illustrates the percent removal of turbidity as a function of operating time. Curves show that EC time has an effect on the turbidity %removal. As the operating time increased from 10 to 30 minutes, the % turbidity removal increased from 38% to 67% at 12mA/cm^2 and from 52.4% to 83.8% at 20mA/cm^2. Results show that maximum efficiency of EC process was obtained at a treatment time of 30 minutes and further increase in treatment time has insignificant improvement in the removal efficiency with rate constant (k) of 0.10229 min^{-1} at 12mA/cm^2 and 0.20087 min^{-1} at 20mA/cm^2.

Figure (4) illustrates the percent COD removal as a function of operating time. It shows that EC time has a significant effect on the %COD removal. As the operating time increased from 10 to 30 minutes, the % COD removal increased from 36% to 60% at 12mA/cm^2 and from 48.5% to 80.2% at 20mA/cm^2. Results show that high efficiency of EC process was obtained at a treatment time of 30 minutes and further increase in treatment time has some significant improvement in the removal efficiency with rate constant (k) of 0.08519 min^{-1} at 12mA/cm^2 and 0.16595 min^{-1} at 20mA/cm^2.

Figure (5) illustrates the percent TSS removal as a function of operating time. The curve shows that EC time has a significant effect on the %TSS removal. As the operating time increased from 10 to 30 min., the %removal TSS increased from 25% to 52.5% at 12mA/cm^2 and from 41.3% to 75.5% at 20mA/cm^2. Results show that high efficiency of EC process was obtained at a treatment time of 30 minutes and further increase in treatment time has some significant improvement in the removal efficiency with rate constant (k) of 0.06802 min^{-1} at 12mA/cm^2 and 0.15442 min^{-1} at 20mA/cm^2.

In all processes for Turbidity, COD, and TSS percent removal the EC involves two stages which are destabilization and aggregation. The first stage is usually short, whereas the second stage is relatively long (Fadil Othman et al. 2006, Moh Faiqun Ni'am. Et al., 2007)

Effect of Applied Current Density

Effect of current density on the removal efficiency of EC process was investigated. From the results it appears that for a given time, the Turbidity, COD, TSS removal efficiencies increased significantly with the increase in current density. As the current density decreased, the time needed to achieve similar efficiencies increased. In this investigation it seems clearly from Fig's.3,4 and 5 that the treatment efficiency was mainly affected by charge loading expressed as current density. If current density increased the % removal of contaminants also increased. The increase of current density always increase the cost of treatment, so it is necessary to select an optimum value of current density for efficient treatment and minimum cost. The result shows that the current density of 20mA/cm^2 is reasonable for the present textile wastewater EC treatment because above this value significant power consumption and insignificant % removal is observed**.**

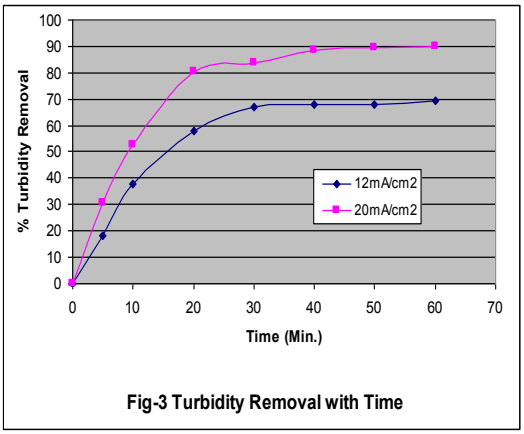

Fig-3 Turbidity Removal with Time

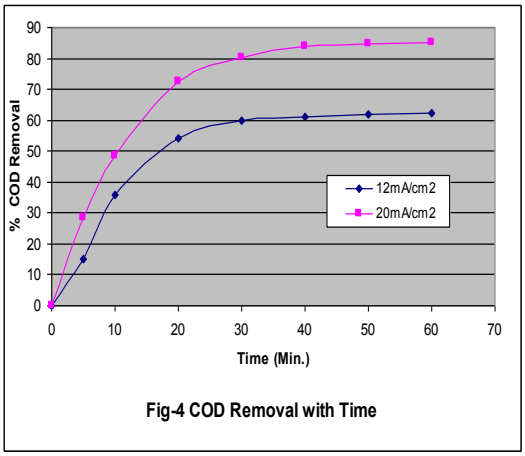

Fig-4 COD Removal with Time

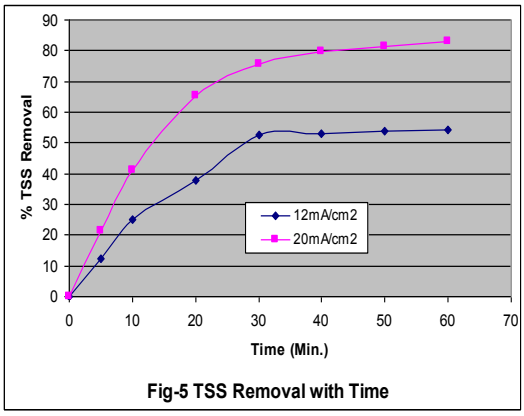

Fig-5 TSS Removal with Time

5. Conclusion

EC is an electrochemical process involving many chemical and physical phenomena that use consumable electrodes to supply ions into the treated wastewater. This research is mainly focused on the capability of EC technology to improve a textile wastewater quality located in Al-Hilla Iraq. Effect of parameters such as operating time, current density was evaluated. It was found that 69.2%, 62.5% and 54.3% removal in turbidity, COD and TSS, respectively, were achieved within 60 minutes at a current density of 12 mA/cm^2 while 90.1%, 85.2% and 83.1% removal in turbidity, COD and TSS, respectively, were achieved within 60 minutes of EC treatment at 20mA/cm^2 current density. It was found that the current density of 20mA/cm^2 is reasonable for the present textile wastewater by EC treatment. After EC treatment, the wastewater quality was compared with standards international wastewater quality. It was found that the studied parameters were within the allowable limit. According to kinetic model (2):
The rate constant for Turbidity removal is 0.10229 min^{-1} at 12 mA/cm^2 and 0.20087 min^{-1} at 20 mA/cm^2.
The rate constant for COD removal is 0.08519 min^{-1} at 12 mA/cm^2 and 0.16595 min^{-1} at 20 mA/cm^2.
The rate constant for Turbidity removal is 0.06802 min^{-1} at 12 mA/cm^2 and 0.15442 min^{-1} at 20 mA/cm^2.

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6. Notation

COD chemical oxygen demand (mg/l)
C_o initial concentration of contaminant (mg/l)
C final concentration of contaminant (mg/l)
C_R contaminant removal
k rate constant (min^{-1})
t electrocoagulation time (min.)
TSS total suspension solid (mg/l)