Enhancement of an Electrocoagulation Process for the Treatment of Textile Wastewater under Combined Electrical Connections Using Titanium Plates

Ahmed Samir Naje¹, Shreeshivadasan Chelliapan^{1*}, Zuriati Zakaria², Saad A. Abbas¹

 ¹Department of Engineering, UTM Razak School of Engineering and Advanced Technology, Universiti Teknologi Malaysia, Jalan Semarak, 54100, Kuala Lumpur, Malaysia.
²Department of Environmental Engineering and Green Technology, Malaysia-Japan International Institute of Technology (MJIIT), Universiti Teknologi Malaysia, Jalan Semarak, 54100, Kuala Lumpur, Malaysia.

^{*}E-mail: <u>shreeshivadasan.kl@utm.my</u>

Received: 27 February 2015 / Accepted: 2 April 2015 / Published: 28 April 2015

The performance of an electrocoagulation (EC) process by adding titanium plates (electrooxidation, EO) was studied for the treatment of textile wastewater using combined electrical connections in a single reactor. Aluminum (and iron) and Titanium (Ti) electrodes were fixed in a bipolar (Bp), and monopolar (Mp) structure in the same electrolytic cell. The performance of the reactor was characterized in terms of electrolysis time (RT), current intensity (I), pH, chemical support, interelectrode distance (IED), and stirring speed (M_{rpm}). Furthermore, energy and electrode consumption, sludge compaction, operating expenses as well as comparison with the conventional EC process was also examined. The most suitable EC-EO performance was achieved by using Mp Ti-Bp Al plates. Preliminary results showed the following optimal operating conditions: I=0.6 A, pH=6, IED=1cm and M_{rpm} =500 rpm. The implementation of these parameters on textile wastewater revealed a relatively high removal efficiency of COD (93.5%), TSS (97%), color (97.5%), BOD₅ (90%), TDS (89%), turbidity (96%), phenols (99%) and phosphate (97%). The overall operating consumption, chemicals, and sludge disposal.

Keywords: Electrocoagulation; textile wastewater; aluminum electrode; titanium electrode

1. INTRODUCTION

Textile industry produces massive levels of wastewater that causes numerous environmental issues [1]. In the past, electrochemical technology has witnessed a great deal of attention for its efficacy in treating various types of wastewater [2]. Electrochemical treatment can be characterized as

a process that uses simple tools and equipment, short retention time, and simple operation. These characteristics help to decrease the costs of operation in scalable applications [3]. The electrocoagulation (EC) process has been the center of attention compared to other approaches that use electrochemical technology. This process has been applied for the removal of various types of pollutants, such as dyes [4], heavy metals [5, 6] and organic-based substances [7]. Many researchers have used EC and electrocoagulation- electroflotation (EC-EF) process for the treatment of textile wastewater [1, 8]. EC consists in situ generation of coagulants by electro-dissolution of aluminum or iron electrodes [9]. Metallic ions (Al⁺³ or Fe⁺²/Fe⁺³) can potentially react with different OH⁻ ions that are produced near cathode and can adsorb pollutants. Amalgamation of EF with EC boosts flock flotation that is formulated within EC system.

The electrooxidation (EO) process is an electrochemical mechanism that is commonly used to treat effluents that contain refractory organic compounds such as textile effluents [10], landfill leachates [11,12], olive oil wastewater, sewage sludge [13], as well as tannery effluent [14,15]. The intuition behind electrochemical oxidation is based on its ability to react with pollutants by direct and indirect effect of current. In direct oxidation, the pollutants are adsorbed by the anode and removed through transfer of the electron. In contrast, indirect oxidation takes place because of oxidants that are electrochemically mediated such as HClO, H₂O₂, Cl₂, Fenton reagent and O₃ [16 -18]. A number of experimental studies demonstrated that the metallic oxide anodes (Ti/IrO₂, Ti/SnO₂ and Ti/PbO₂) could be successfully utilized for pollutant removal [19-21]. The hydroxyl radicals (OH°) and Cl⁻ produces HClO, which is characterized among the powerful oxidants. It is formulated in the following equations [22, 23];

$$MO_x + H_2O \rightarrow MO_x(OH^\circ) + H^+ + e^-$$
 (1)

 $Cl^{-} + OH^{\circ} + H^{+} \rightarrow HClO + H_{2}O$ (2)

Where MO_x represents metal oxide electrodes. The formation of HClO occurs as an effect of Cl_2 hydrolyzation based on the equations below [24];

 $2\mathrm{Cl}^{-} \rightarrow \mathrm{Cl}_{2} + 2\mathrm{e}^{-} \qquad (3)$

 $Cl_2 + H_2O \rightarrow HClO + H^+ + Cl^-$ (4)

It may be worthy to integrate EC with iron or aluminum electrodes, and also EO with titanium electrodes in a single reactor. Accordingly, the primary goal of this study is to propose a hybrid process (EC-EO) integrating monopolar EO and bipolar EC in a single reactor for the treatment of textile wastewater. The performance of the reactor was characterized in terms of bipolar electrodes, current intensity, reaction time, pH, electrolysis support, distance between electrodes and stirring speed. In addition, the amount of sludge produced after treatment and comparison between EC-EO and EC alone was also evaluated.

2. MATERIALS AND METHODS

2.1. Description of the textile wastewater

The wastewater used in this work was collected from a major textile-based industry in Iraq (Babylon). This industry uses the Imperon Violet KB (CAS #: 6358-46-9) for the fabric dyeing process.

Tables 1 and 2 show the main characteristics of the textile wastewater and the properties of the Imperon Violet KB, respectively.

Table 1.	Characteristics	of textile	wastewater
----------	-----------------	------------	------------

Parameters	Values
Electrical conductivity (µS/cm)	245
Turbidity (NTU)	390
Total suspension solid, TSS (mg/l)	3280
Total dissolved solid, TDS (mg/l)	1241
Dissolved oxygen, DO (mg/l)	0.73
pH	4.52
Chlorides, Cl ⁻ (mg/l)	32
Sulfate (mg/l)	677
Phosphate (mg/l)	7.2
Nitrates (mg/l)	9
Phenols (mg/l)	345
Oil & Grease (mg/l)	3.5
BOD (mg/l)	110.8
COD (mg/l)	985

Table 2. Properties of Imperon Violet KB



*Absorbance of 0.34 at 533

2.2. Experimental setup

The treatment of textile wastewater was performed using a batch electrolytic cell that was made of Plexiglas having upper Teflon cover with a length, width and height of 18, 14 and 14 cm respectively. The sets of electrodes include five iron (or aluminum) electrode plates and six plates made of titanium, each with a surface area of 120 cm^2 ($12 \text{ cm} \times 10 \text{ cm}$). The inter-electrode distance was 1 cm. Iron (or aluminum) and titanium plates were continuously exchanged within the electrode pack, in which titanium plates were made up of titanium that was fully covered with iridium dioxide (Ti/IrO₂) plates for the anode, and titanium plates with no cover (Ti) for the cathode. The titanium plates were attached to the main power supply.



Figure 1. Schematic view of EC-EO experimental setup: (1) DC power supply, (2) Voltmeter, (3) Variable resistance, (4) Ammeter, (5) Monopolar set (Ti/IrO₂ anode -Ti cathode), (6) Bipolar set (aluminum or iron).

The iron (or aluminum) electrodes were positioned in between two electrodes made up of titanium, but not electrically connected, as depicted in Figure 1. A total of five inner iron (or aluminum) electrodes were used as bipolar form and sacrificial electrodes. The thickness of each electrode was 0.1 cm. The electrodes were placed in the reactor in a vertical manner and secured by plastic screws. A magnet stirrer and hot plate (SCILOGEX) with a 500 rpm rotating velocity were used to stir the contents of the cell. The cathode and anode groups were connected to the negative and positive parts of a DC power supply (YIZHAN, 0-6 A; 0-40 V). The main voltage has a value of 30 V for every round. The current level was controlled at a constant value in every run using a variable resistor, and measured with a multiple range ammeter. An effluent of 3 L working volume was applied for every run. After each run, the electrolytic cell was thoroughly cleaned using a 5% (v/v)hydrochloric acid solution. Experiments were conducted at ambient temperatures in the range of 25-27°C and pH was adjusted from 5 – 10 by adding 0.5 M NaOH. A supporting electrolytes (NaCl and Na₂SO₄) were added to the wastewater to examine the impact of conductivity. Sodium chloride, sodium sulphate, and sodium hydroxide are the analytical type reagents (Mat Laboratory). The EO is dependent upon chlorine generation, where it was estimated from CIO⁻, CI₂ and HCIO. Approximately 100 ml of supernatant was used for the analysis in replicate and 30 ml samples were taken to measure

the metal concentration residue for sludge compaction study. The optimal conditions were reused in triplicate to confirm the reproducibility and effectiveness of the EC-EO.

2.3. Sludge compaction

The textile wastewater sludge was given an hour for settle to increase its consolidation. The cationic polymer was examined using two different concentrations (10 and 40 mg/l). Space taken by solid (ml) was checked at regular time intervals. The solid portion (wet residue) was estimated and dried for 24 h at a temperature of 100°C in order to find out the total residual solids. The specific resistance to filtration (SRF) and also cake-dry solid were estimated using the following formula [25];

RF (SRF) = $(2 * Kb * P * A^2) / \mu * a$ (5)

Where Kb is the plot slope (V vs. t/V); A is the filter area; P is the pressure during filtration of sludge; μ is the viscosity of filtrate; and a is the weight of the solid per unit volume of the filtrate. The solid concentration was estimated using the following formula;

Sludge dryness (%) = 100 * [(m3-m1) / (m2-m1)] (6)

Where m2 and m1 are the mass of the cup before and after filtration; and m3 is the mass of the cup after drying at a temperature of 100°C for a period of 24h.

The SVI (ml/g) is the volume (in mm) used by 1 g of a suspension subsequent to 30 minutes of settling [26].

 $SVI = VD_{30} / TSS$ (7)

Where VD_{30} is the volume of settled sludge (ml/l); TSS is the concentration of suspension solids (g/l).

2.4. Chemical analysis

The sample analysis was conducted according to standard methods [26]. The COD was measured by a Closed Reflex-Titrimetric method. TSS, TDS, and phenols were measured by Gravimetric method and oil and grease (O & G) were measured by a Solvent Extraction unit. BOD and dissolved oxygen (DO) were measured using a DO meter (Eutech Instrument Cyberscan model 110). pH (pHM84), conductivity (HANNA HI-99301), and turbidity (HACH 2100P) were also investigated, all according to standard methods [26]. Color was measured via absorbance by a UV-V Spectrophotometer (Shimadzu UV 1700) with a wave length of 533 nm. Ion analysis was performed using an ionic chromatography (ICS-2000).

3. RESULTS AND DISCUSSION

3.1. Effect of the bipolar electrode materials and electrolysis time

In electrolysis process, the COD removal efficiency depends on the concentration of metal ions produced, electrode material, mode of electrical connection and reaction time [27–29]. The bipolar

configuration within the EC-EO of textile wastewater treatment was performed using an aluminum or iron electrodes with 0.8 A current intensity at a natural pH of 4.52.



Figure 2. Effect of bipolar electrode materials and electrolysis time on COD removal efficiency at current intensity of 0.8 A.

Table 3. Effect of bipolar materials using aluminum or iron electrodes on EC-EO performance treating textile wastewater (I=0.8 A, RT = 90 minutes)

Parameters	Raw effluent	MpTi/Bp Al	Mp Ti/Bp Fe
Bipolar electrode	-	Al	Fe
Mean voltage (v)	-	30	30
Conductivity (µS/cm)	245	445	452
Final pH	4.52	6.40	6.70
Actual Al or Fe dose (mg/l)	-	98	205
Theoretical Al or Fe (mg/l)	-	134.20	416.63
Current efficiency (%)	-	73.00	49.20
COD (mg/l)	985	236.4	305.3
COD removal (%)	-	76	69

Figure 2 illustrates the COD removal efficiency of aluminum and iron electrodes. An increase in electrolysis time from 20 to 90 minutes resulted in high COD removal efficiency for the bipolar electrodes (76%, aluminum and 69%, iron electrodes). The theoretical value of Al or Fe dissolution is calculated using Faraday's law [30, 31];

 $C_{theoretical} = M_i I RT / ZF V$ (8)

 $C.E = ZFC_i V/I RT M_i \qquad (9)$

Where $C_{\text{theoretical}}$ represents Faraday's theoretical amount of Fe or Al in solution (mg/l), C.E= current efficiency, $C_i = \text{iron/aluminum}$ or active chlorine (mg/l), $M_i = \text{molecular mass}$ (Al, 26.9 g/mol;

Fe, 55.8 g/mol; Cl, 35.4 g/mol), I = current, RT = reaction time (s), Z = electron moles (three for Al, two for Fe, and two for Cl), F = Faraday's constant (96500 C/mol), and V = wastewater volume.

Table 3 illustrates the effect of bipolar materials on EC-EO process performance treating textile wastewater. It is evident that total intake of iron dosage (205 mg/l) was greater than the aluminum (98 mg/l), and confirms the suitability of aluminum electrode for the EC case.

3.2. Effect of current intensity

It has been shown that current intensity influences the treatment efficiency of wastewater [32, 33]. Several current intensities (0.2 - 1 A) were selected to investigate its effect on COD, TSS and color removal performance. When current intensity was increased from 0.2 to 0.6 A, the COD, TSS and color removal efficiencies increased to a peak removal of 76%, 81%, and 83% respectively, which were achieved at a current of 0.6A (Figure 3 (a)).



Figure 3. (a) Effect of current intensity on COD, TSS and color removal efficiency.

A greater number of cations made of aluminum are generated at higher current, and, as a result, the formation rate of Al $(OH)_3$ was increased. There was a decrease in the effectiveness when the current was increased from 0.8 to 1 A. Figure 3 (b) illustrates the current efficiency for the EC-EO process. An optimum efficiency of 75% for oxidant and 87% for coagulant was achieved at a current intensity of 0.6 A.



Figure 3. (b) Effect of current intensity on current efficiency of EC-EO process

3.3. Effect of pH



Figure 4. Effect of initial pH on COD, TSS and color removal efficiency and final pH.

Several researchers have investigated the effect of pH in electrocoagulation process using various types of wastewater [30,34-36]. To investigate the effect of pH on COD, TSS, and color removal, multiple experiments were carried out at varying pH between 5 and 10 at optimal conditions (current= 0.6 A, Mp Ti-Bp Al and reaction time = 90 minutes). Figure 4 illustrates the effect of pH on EC-EO, and up to 90% COD, 92% TSS, and 94% color removal efficiencies were achieved at pH 6. A large reduction of treatment efficiencies was also observed at pH 5 and 10.

A minor increase in pH was noticed in the acidic solution (pH 5 and 6). This is probably due to the increase in hydroxide ion (OH⁻) near the cathode [37]. The Al (OH)_{3(s)} flocks have large surface areas which aided high adsorption of soluble compounds (organic) and colloidal particles. When pH levels were 4 to 9, aluminum monomeric and polymeric species transformed to insoluble amorphous Al (OH)_{3(s)} through polymerization or precipitation-based kinetics [38]. At pH 10, the concentration of soluble anion Al (OH)₄⁻ was increased due to decrease in Al (OH)_{3(s)}, and this reduces the treatment efficiency (72%, 79% and 83% for COD, TSS, and color respectively). A minor reduction in pH (8.5) occurs based on the following reaction [9]:

 $Al (OH)_{3(s)} + OH^{-} \rightarrow Al (OH)_{4}^{-}$ (10)

These results confirm that the average pH of 6 is the optimum condition for the treatment of textile wastewater.



3.4. Effect of the electrolyte support

Figure 5. The effect of electrolysis support on COD, TSS and color removal under optimal conditions of EC-EO process (MpTi /Bp Al, I = 0.6 A, RT = 90 minutes and pH = 6) at different concentrations: (a) NaCl (b) Na₂SO₄.

The addition of an electrolyte (Na₂SO₄ or NaCl) can improve wastewater conductivity, decrease cell voltage and reduce electrical energy [1, 39]. In this experiment, Na₂SO₄ and NaCl (0, 0.02, 0.05, and 0.10 kg/m³) were added to the textile wastewater to investigate the influence on COD, TSS, and color removal performance (Figure 5a and 5b). According to Chen et al. [40], addition of

 Na_2SO_4 and NaCl does not change the COD, TSS and color removal efficiency in EC-EO process. After the addition of Na_2SO_4 and NaCl (0.02, 0.05 and 0.10 kg/m³), the COD removal efficiency was 90%, 90.5%, and 91.20% for NaCl and 90.5%, 90.75% and 91% for Na_2SO_4 . The energy intake for the EC-EO process may be computed from the equation below [41]:

 $C_{energy} = U I RT/V$ (11)

Where U=voltage (v), I=electrical current (A), RT=EC-EO electrolysis time (s), V=volume of textile wastewater.

The maximum decrease in voltage was 30 to 28 volts, and from 30 to 28.5 volts with the addition of 0.1 kg/m³ NaCl and Na₂SO₄ respectively. Since the voltage is decreased, the energy intake shifted from 9 to 8.4 kW \cdot h/m³, and from 9 to 8.55 kW \cdot h/m³, when NaCl and Na₂SO₄ were introduced. This change in energy intake that was witnessed during the analysis in both removal and the addition of a supporting electrolyte was assumed of less importance [42].

3.5. Effect of the inter-electrodes distance



Figure 6. Effect of inter-electrodes distance on COD, TSS and color removal efficiency under optimal conditions of EC (Mp Ti /Bp Al, I = 0.6 A, RT = 90 minutes, and pH = 6).

Several scholars have investigated the impact of electrode distance on pollutant removal efficiency. Inter-electrode distance (IED) depends on the nature of pollutants, structure of electrodes and hydro-dynamic characteristics [33, 43-45]. In this study, the impact of IED was studied at three different distances; 0.5, 1 and 1.5 cm. The COD, TSS, and color removal efficiencies were increased to 90%, 92% and 94% respectively at an electrode distance of 1 cm (Figure 6). However, a minor

decrease was observed when the electrode gap was increased to 1.5 cm (COD, 87%; TSS, 89% and color, 92%). Similar results were also observed by Modirshahla et al. [43] during the treatment of 4-nitrophenol containing wastewater using EC. A decrease in the treatment efficiency was observed when the space between the electrodes was increased. Molecule interactions with both oxidants and coagulants become weak when the distance becomes more than 1 cm, which leads to a drop in the treatment efficiency.

3.6. Effect of stirring speed



Figure 7. Effect of stirring speed on COD, TSS and color removal efficiency under optimal conditions of EC (Mp Ti /Bp Al, I = 0.6 A, RT = 90 minutes, pH = 6, and IED=1cm).

Stirring speed is an important parameter in EC-EO process and may enhance the mass transfer kinetics. Proper oxidant and coagulant disperse in reactor could promote excellent homogenization. However, high stirring speed may also effect the chemical reactions of oxidants and coagulants. In this study, three stirring speed (M_{rpm}) was investigated (250, 500 and 750 rpm) and results showed a high removal efficiency of COD (90%), TSS (92%) and color (94%) when the reactor was operated at 500 rpm (Figure 7). However, when the stirring speed was increased to 750 rpm, the reactor performance decreased substantially (COD, 86%, TSS, 88% and color, 92%). At optimum stirring speed, Al (OH)₃ flocks combined with one another, and oxidants had the ability to break up the contaminants and increase the mass transfer, and the precipitation was easier. A number of researchers have investigated

the effect of stirring speed on EC process, and their findings are almost identical with the current investigation [33, 46, 47].

3.7. Sludge compaction

Sludge compaction of textile wastewater was studied using a flocculating agent (LPM 3135) at a concentration of 0.01 and 0.04 kg/m³ [48]. The evaluation was carried out at an optimal conditions (current = 0.6 A; Mp Ti-Bp Al ; pH=6 ; IED=1cm ; M_{rpm} =500 rpm). An introduction of 0.01 kg/m³ LPM 3135 resulted in a high COD removal efficiency (68.6% to 82.3%). Moreover, a great change in the turbidity level from 34 to 7 NTU, and TSS from 120 to 85 mg/l was also observed during this experimental period. The relationship between electrolysis time and metallic sludge production was investigated with and without the flocculant agent (Figure 8).



Figure 8. The effect of floculant agent on metallic residues of textile wastewater under optimal conditions of EC-EO (Mp Ti /Bp Al, I = 0.6 A, RT = 90 minutes, pH = 6, and IED=1cm and M_{rpm} =500).

It can be seen that the optimum metallic sludge was 2.8 and 2.9 kg/m³ at a concentration of 0.01 and 0.04 kg/m³ respectively. Sludge dryness, SRF, and SVI was also evaluated in this study. A 7% sludge dryness and 70 ml/g SVI was observed during the analysis. Olmez [49] studied the treatment of Cr (VI) contaminated industrial-based wastewater using EC process, and their results showed an SVI value of 80 ml/g. It should me mentioned here that the initial concentration of TSS applied in their study was 80 mg/l, and in the current study, the concentration was 3285 mg/l.

Furthermore, the SRF utilized in this study was lower (5.9 $*10^{12}$ m/kg) compared to their study (7.8 $\times 10^{12}$ m/kg).

3.8. Operational cost analysis

In EC-EO process, the most important costs are the material, chemical, sludge and electrical costs. As a result, these elements are incorporated in this study to investigate the operational cost. The following equation was used [41]:

Operating $cost = a C_{energy} + b C_{electrode} + d C_{sludge} + e C_{chemicals}$ (12)

Where C_{energy} , $C_{electrode}$, C_{sludge} , $C_{chemicals}$, a, b, d, and e represent the energy intake for every cubic meter of wastewater (kWh/m³), electrode intake for treating one cubic meter of wastewater (kg/m³), quantity of sludge for one cubic meter of wastewater (kg/m³), chemicals intake (kg/m³), electricity expenses 0.075US\$/kWh, fixed expenses of aluminum (2.5 US\$/kg), sludge removal expenses (not including the drying process) and transportation (0.06 US\$/kg), chemicals (NaOH: 0.5 US\$/kg, LPM3135: polymer 3 US\$/kg).

The current efficiency was 87% when operated at an aluminum dosage of 0.087 kg/m³, which is much lower than the theoretical value (0.1 kg/m^3 based on Faraday's law, Eq.8), with final energy intake of 9 kWh/m³ (Eq.11). This calculation was based on the amount sodium hydroxide (1.2 kg/m^3) and polymers (0.01kg/m^3) consumed during the process. The quantity of sludge produced was 2.88 kg/m³. Hence, the operating costs for the treatment of one cubic meter of textile wastewater at optimum operating conditions was approximately 1.69 US\$.

3.9. Efficiency and validation of the EC-EO process performance

The treatment of textile wastewater using EC-EO process was performed at optimal conditions (current=0.6 A, Mp Ti - Bp Al, reaction time = 90 minutes, pH=6, inter-electrode distance=1cm, mixing speed=500 rpm, and 0.01 kg/m³ of polymer LPM 3135). Table 4 shows the treatment profile of the textile wastewater, and it can be seen that a COD removal efficiency of 93.5 % was achieved in the treatment system. The Al (OH)₃ generated in the electrolytic cell neutralizes the colloids and the HClO generated by indirect oxidation destabilize the organic compounds. The HClO promotes hydrophilic particulates and provide a more suitable aggregation of colloidal and dehydrates the sludge. The color, TSS, TDS, BOD and O & G was reduced to 97%, 97.0%, 89%, 90% and 96% respectively. The hydrophobic volume of the O & G gives a suitable affinity with the H₂ bubbles created near the cathode where the (O & G)-H₂ complex can be skimmed easily at the liquid's surface [50]. The EC-EO also reduces the phosphate content of the textile wastewater from 6.9 to 0.21 mg/l. A number investigation was carried out for the treatment of various type of wastewater using EC-EO process. Hu and Li [51] demonstrated the integration of EC-EO process for the treatment of chemical thermo mechanical pulp (CTMP) wastewater that contains a COD of 33000 mg/l and color, 48000 CU.

Table 4. The validation of EC-EO performance treating textile wastewater at optimal conditions (Mp Ti / Bp Al, I = 0.6 A, RT = 90 minutes, pH=6, and LPM $3135 = 0.01 \text{ kg/m}^3$)

Parameter	Raw	Treated	Allowable	Pollutant
	effluent	effluent	Limit EPA	Removal
			1996*	(%)
Electrical conductivity (µS/cm)	250	550	ID	
Initial pH	4.60	6.00	-	
Final pH		7.00	6-8	
Energy consumption (kwh/m ³)		9.00		
Electrode consumption (kg/m ³)		0.087		
Sludge production (kg/m ³)		2.88		
Polymer consumption (kg/m ³)		0.01		
Hydroxide sodium consumption (kg/m ³)	—	1.20		
O&G (mg/l)	4	0.160	5-40	96.0
$BOD_5 (mg/l)$	118	11.80	5-45.5	90.0
COD (mg/l)	990	64.40	20-500	93.5
TSS (mg/l)	3290	98.70	60-300	97.0
Color observance at 533 NM	0.3400	0.0085	ID	97.5
TDS (mg/l)	1235	135.8	5-180	89.0
Turbidity (NTU)	402	16.1	15-50	96.0
DO (mg/l)	0.69	13.5	4.5-15	
Sulfate (mg/l)	662	13.24	ID	98.0
Phosphate (mg/l)	6.9	0.210	ID	97.0
Nitrates (mg/l)	8.5	9.880	ID	
Phenols(mg/l)	340	0.345	10	99.9
Aluminum (mg/l)	1.30	6.500		
Sludge dryness (%)		7.30		
SRF (m/kg) * 10^{12}		6.00		
SVI (ml/g)	150	70.60		
Electrical energy cost (US \$/m ³)		0.675		
Electrode consumption cost (US\$/m ³)	—	0.217	—	
Sludge disposition cost (US \$/m ³)		0.170		
Polymer cost (US \$/m ³)		0.030		
Hydroxide sodium cost (US\$/m ³)		0.600		
Total operating cost (US \$/m ³)		1.690		

ID: Insufficient Data

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

They utilized stainless-steel as the anode and graphite for the cathode. At a reaction time of 90 minutes and a current intensity of 0.6 A, 80% and 90% COD and color removal efficiency, was

observed in their investigation. Hernandez et al. [16] demonstrated a successful treatment of the electro-coagulated effluent by electro-oxidation. They used two stage treatment process integrating EC with iron and then EO with boron doped diamond (BDD). The COD, BOD, and turbidity was removed within 90 minutes. These results could be compared to the current study where 93.5% COD removal efficiency was noted during the textile wastewater treatment. The BDD electrode has proven to be one of the most effective anode materials for the production of hydroxyl radical (OH°); however it requires high potential voltage and considered expensive [52, 53]. A comparison with the international textile wastewater quality standards [54, 55], showed that the EC-EO process used in this study could be applied in practical for the treatment of textile wastewater. The TDS, BOD₅, DO, turbidity, COD, pH, O & G and phenol were all under the allowed range. The SRF, SVI, and sludge dryness were 6.0×10^{12} m/kg, and 70.6 ml/g, and 7.3 % respectively.

3.10. Comparison of the EC-EO performance with EC process

Table 5. Comparison the EC-EO process with EC alone for treatment of textile wastewater under the same conditions (I=0.6A, RT=90 minutes, and pH=6)

Parameters	EC-EO	EC process
	process	
Configuration of electrode	Mp Ti/Bp Al	Mp Al/Bp Al
Conductivity (µS/cm)	550	560
Final pH	7.00	6.92
COD%	93.5	92.3
TSS%	97.0	96.0
Color%	97.5	96.0
Electrode consumption (kg/m ³)	0.087	0.1
Energy consumption $(kW.h/m^3)$	9.00	8.49
Sludge quantity (kg/m^3)	2.88	3.50
NaCl consumption (kg/m^3)	-	0.1
NaOH consumption (kg/m^3)	1.20	1.26
Polymer consumption (kg/m^3)	0.01	0.01
Electrode consumption cost (US\$/m ³)	0.217	0.250
Energy consumption cost $(US\$/m^3)$	0.675	0.636
Sludge disposal cost (US \$/m ³)	0.17	0.21
NaCl consumption cost (US\$/m ³)	-	0.005
NaOH consumption cost (US\$/m ³)	0.60	0.63
Polymer consumption cost (US\$/m ³)	0.03	0.03
Total operational cost (US\$/m ³)	1.69	1.76

A comparative study was made between EC-EO and EC process alone at optimal conditions. The EC treatment was conducted using 11 aluminum electrodes. Six were in monopolar settings, and the remaining five were in bipolar settings. Table 5 shows the treatment profile using both methods. It can be seen that the COD, TSS, and the color removal efficiency during the EC process alone was 92%, 96%, and 96% respectively. The EC process can remove suspended solids and colloidal particles [9, 17, 56] while the EO process oxidizes the dissolved pollutants [17]. The traditional EC process requires a support of 0.1 kg/m³ NaCl to enhance conductivity and pollutant removal capability. On the other hand, the EC-EO process showed excellent treatment without using electrolysis support. The electrode intake and sludge production was lower than the EC process alone. The amount of aluminum electrodes utilized by the EC was greater than the EC-EO process. As for the operating costs, the EC-EO process was lower than the EC (EC=1.76 US\$/m³, while EC-EO=1.69 US\$/m³).

4. CONCLUSIONS

The combination of EC-EO process for the treatment of textile wastewater showed an excellent approach. The final results may be presented as follows:

• The Mp Ti-Bp Al electrodes were found to be more effective for the treatment of textile wastewater compared to Mp Ti-Bp Fe.

• At optimal conditions the following results were obtained; 93.5% (COD), 97% (TSS), 97.5% (color), 90% (BOD₅), 89% (TDS), 96% (turbidity), >99% (phenols) and 97% (phosphate). The final concentration of DO was improved from 0.69 to 13.5 mg/l.

• The polymer agent LPM 3135 enhances the settling process at a dosage of 0.01 kg/m³.

• The costs for the treatment of textile wastewater using EC-EO process was approximately $1.69 \text{ US}/\text{m}^3$.

ACKNOWLEDGEMENTS

The authors thank Babylon Textile Plant, Iraq, for supplying the textile wastewater. The authors also thank Universiti Teknologi Malaysia and the Ministry of Education Malaysia for funding this research under a Ph.D. scholarship fund.

References

- 1. B. Merzouk, K. Madani and A. Sekki, Desalination, 250 (2010) 573.
- X. D. Zhang, J.D. Hao, W. S. Li, H. J. Jin, Q. M. Huang, D. S. Lu and H. K. Xu, J. Hazard. Mater., 170 (2009) 883.
- 3. M. Jović, D. Stanković, D. Manojlović, I. Anđelković, A. Milić, B. Dojčinović and G. Roglić, Int. *J. Electrochem. Sci.*, 8 (2013) 168.
- 4. J.B. Parsa, H. R. Vahidian, A. R. Soleymani and M. Abbasi, *Desalination*, 278 (2011) 295.
- 5. B. Al Aji, Y. Yavuz and A. S. Koparal, Sep. Purif. Technol., 86 (2012) 248.
- I. Kabdasli, T. Arslan, T. Olmez-Hanci, I. Arslan-Alaton and O. Tünay, J. Hazard. Mater., 165 (2009) 838.
- 7. M. Vepsalienen, M.Pulliainen and M. Sillanpaa, Sep. Purif. Technol. 99 (2012) 20.
- 8. B. Merzouk, M. Yakoubi, I. Zongo, J. P. Leclerc, G. Paternotte, S. Pontvianne and F. Lapicque, *Desalination*, 275 (2011) 181.
- 9. G. Chen, Sep. Purif. Technol., 38 (2004) 11.
- 10. A. Wang, J. Qu, H. Liu and J. Ge, Chemosphere, 55 (2004) 1189.

- 11. C.A. Martinez-Huitle and S. Ferro, Chem. Soc. Rev., 35 (2006) 1324.
- 12. Y. Deng and J. D. Englehardt, Waste Manag., 27 (2007) 380.
- 13. A. Morao, A. Lopes, M.T.P. De Amorin and I.C. Goncalves, *Electrochimica Acta*, 49 (2004) 1587.
- 14. N.N. Rao, K. M. Somasekhar, S. N. Kaul and L. Szpyrkowicz. J. Chem. Technol. Biotechnol., 76 (2001) 1124.
- 15. M. Panizza and G. Cerisola. Environ. Sci. Technol., 38 (2004) 5470.
- 16. I. Linares-Hernàndez, C. Barrera-Diaz, B. Bilyeu, P. Juàrez-Garcia Rojas and E. Campos-Medina. *J. Hazard. Mater.*, 175 (2010) 688.
- 17. P. Drogui, J. F. Blais and G. Mercier. Recent Pat. Eng. 1 (2007) 257.
- D. Stergiopoulos, K. Dermentzis, P. Giannakoudakis and S. Sotiropoulos. *Global NEST J.*, 16 (2014) 499.
- 19. M. Panizza, C. Cristina and G. Cerisola. Water Res., 34 (2000) 2601.
- 20. M.H. Zhou, Q.Z. Dai, L.C. Lei, C.A. Ma and D.H. Wang, Environ. Sci. Technol., 39 (2005) 363.
- 21. Y.H. Wang, K.Y. Chan, X.Y. Li and S.K. So. Chemosphere, 65 (2006) 1087.
- 22. Y. Liu, H. Liu, J. Ma and X. Wang. Applied Catalysis B: Environ., 91 (2009) 284.
- 23. G.R.P. Malpass, D.W. Miwa, D.A. Mortari, S.A.S. Machado and A.J. Motheo. *Water Res.*, 41 (2007) 2969.
- 24. F. Zaviska, P. Drogui, J.F. Blais and G. Mercier. J. Appl. Electrochem., 39 (2009) 2397.
- 25. R. Daghrir, P. Drogui, J.F. Blais and G. Mercier. J. Environ. Eng., 138 (2012) 1146.
- 26. American Public Health Association (APHA), 20th ed., Washington, DC, USA 2000.
- 27. M. Kobya, M. Bayramoglu and M. Eyvaz. J. Hazard. Mater., 148 (2007) 311.
- 28. S. Bellebia, S. Kacha, A. Z. Bouyakoub and Z. derriche. *Env. Prog. & Sustain. Energ.*, 31 (2012) 361.
- 29. Z. Zaroual, M. Azzi, N. Saib and E. Chainet. J. Hazard. Mater., 131 (2006) 73.
- 30. K. Bensadok, S. Benanma, F. Lapicque and G. Nezzal. J. Hazard. Mater., 152 (2008) 423.
- 31. S. Song, Z. He, J. Qiu and X. L. Chen. Sep. Purif. Technol., 55 (2007) 238.
- 32. A.E. Yılmaz, R. Boncukcuoʻglu, M.M. Kocakerim and E. Kocadaʻgistan. *Desalination*, 230 (2008) 288.
- 33. N. Daneshvar, H. Ashassi-Sorkhabi and A. Tizpar. Sep. Purif. Technol., 31 (2003) 153.
- 34. A.H. Essadki, M. Bennajah, B. Gourich, Ch. Vial, M. Azzi and H. Delmas. *Chem. Eng. Process.*, 47 (2008) 1211.
- 35. A. S. Koparal, Y.S. Yıldız, B. Keskinler and N. Demircio-glu. Sep. Purif. Technol., 59 (2008)175.
- 36. S. Song, Z. He, J. Qiu and X. L. Chen. Sep. Purif. Technol., 55 (2007) 238.
- 37. P. Canizares, C. Jimenez, F. Martinez, M. A. Rodrigo and C. Saez. J. Hazard. Mater., 63 (2009) 158.
- 38. M. Bayramoglu, M. Kobya, O.T. Can and M. Sozbir. Sep. Purif. Technol., 37 (2004) 117.
- 39. N. Daneshvar, A. Oladegaragoze and N. Djafarzadeh. J. Hazard. Mater., 129 (2006) 116.
- 40. X. Chen, G. Chen and P.L. Yue. Sep. Purif. Technol., 19 (2000) 65.
- 41. A. Dalvand, M. Gholami, A. Joneidi and N. M. Mahmood. Clean Soil, Air, Water, 39 (2011) 665.
- 42. P. Drogui, M. Asselin, S. K. Brar, H. Benmoussa and J. F. Blais. Sep. Purif. Technol., 61 (2008) 301.
- 43. N. Modirshahla, M.A. Behnajady and S. Mohammadi-Aghdam. J.Hazard. Mater., 154 (2008) 778.
- 44. N. Daneshvar, H. Ashassi Sorkhabi and M.B. Kasiri. J. Hazard. Mater., 112 (2004) 55.
- 45. N. Modirshahla, M.A. Behnajady and S. Kooshaiian. Dyes Pigments, 74 (2007) 249.
- 46. O.T. Can, M. Bayramoğlu and M. Kobya. Ind. Eng. Chem. Res., 42 (2003) 3391.
- 47. A. Gürses, M. Yalçın and Ç. Doğar. Waste Manag., 22 (2002) 491.
- 48. M. Asselin, S. K. Brar, H. Benmoussa and J. F. Blais. Sep. Purif. Technol., 61 (2008) 301.
- 49. T. Olmez. J. Hazard. Mater., 162 (2009) 1371.
- 50. M. Asselin, P. Drogui, S. K. Brar, H. Benmoussa and J. F. Blais. J. Hazard. Mater., 151 (2008) 446.

- 51. Z.J. Hu and Y.M. Li. Adv. Mat. Res., 233-235 (2011) 619.
- 52. Y. Samet, L. Agengui, R. Abdelhédi. Chem. Eng. J., 161 (2010) 167.
- 53. M. Murugananthana, S.S. Lathaa, G. Bhaskar Rajua and S. Yoshihara. J. Hazard. Mater., 180 (2010) 753.
- 54. Environmental Protection Agency (EPA), Document No. EPA/625/R-96/004, Ohio, USA 1996.
- 55. Commission Decision, 371, EC, 2002, (http://europa.eu.int/ecolabel).
- 56. M. Khemis, G. Tanguy, J. P. Leclerc, G. Valentin and F. Lapicque. *Process Saf. Environ. Prot.*,83 (2005) 50.

© 2015 The Authors. Published by ESG (<u>www.electrochemsci.org</u>). This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution license (http://creativecommons.org/licenses/by/4.0/).