Effect of operational parameters on dye removal of textile wastewater by electrocoagulation process using zinc electrode

Marwa Ali Elajel [1] (*), Ahmad Zuhairi Abdullah[2]

[1]Plasma Research Laboratory, Authority of Natural Science Research and Technology, Tripoli, Libya

[2] School of Chemical Engineering, Universiti Sains Malaysia Engineering Campus, Nibong Tebal, Penang, Malaysia

Abstract

Electrocoagulation is a promising technology that could effectively replace conventional wastewater treatment processes. Optimization of operating parameters might increase dye removal efficiency. Zinc electrode could help in improvement of treatment performance of textile

(*) Email: aldhirr@yahoo.com

wastewater. The aim of this study is to investigate the effect of operating parameters on the treatment of textile wastewater using zinc electrode. The performance of electrocoagulation process for treatment of a synthetic textile wastewater (prepared using reactive blue 261 dye) and a real textile wastewater was investigated. The operating parameters involved in this study were current density (2-8 mA/cm^2), initial dye concentration (20-100 mg/L) and initial pH (5-9). The preliminary study was conducted using synthetic textile wastewater to determine the optimum operating conditions that would lead to high dye removal. The optimal conditions were a current density of 4 mA/cm^2, an initial dye concentration of 100 mg/L and an initial pH of 9. The highest dye removal was recorded at 99.8% for treating of synthetic textile wastewater while a dye removal of 83.7% was achieved during treatment of real textile wastewater under the optimum operating conditions.

Keywords: Dye removal; Electrocougulation; Reactive blue dye; Textile wastewater; Zinc electrode

1. Introduction

Water pollution is a major environmental problem due to its effects on human life, plants, organisms and other life forms. Textile industry is one of the industries that generate large amount of wastewater that presents a significant environmental pollution problem due to high level of pollutants with high chemical oxygen demand (COD), high suspended solid, strong color, and high biotoxicity.Textile idustry consumes large amount of dyes and pigments compared with other industries [1]. Also, this industry uses chemicals and dyes in various fabrication processes (dyeing and finishing process) where large quantity of these dyes (up to 50%) are lost in wastewater. Wastewaters containing dyes constitute

substantial sources of pollution [2]. Dyes are colored compounds which absorb light in visible region and can strongly attach to the fiber due to physical and chemical bondings between the dye compound and fiber groups [3]. Various dyes are yearly produced worldwide and various industries use dyes in their manufacturing process. Different types of dyes used in textile manufacturing process are such as direct, vat, acidic, sulphur, metal complex and reactive dyes [4]. Reactive dyes are most important class of the dyes. They are used for dying of cellulosic and protein fibers such as wool and silk. They offer a wide range of bright colors and excellent color fastness to washing. This is due to the fact that the fixation of these dyes is conducted under alkaline conditions where strong covalent bonds between the fibers and dyes are formed.

Reactive dyes are sulphonated and produced in granular shape. The main characteristics of reactive dyes are their high water solubility and they can undergo hydrolysis into insoluble forms easily. This is why they are extensively used in textile industry. Reactive dyes are not absorbed onto biomass to any great level and generally pass through conventional biological wastewater systems. It has been reported that 20-50% of reactive dyes used in textile manufacturing can be released into wastewater [5]. In addition, dyes molecules are highly structured molecules that present a potential toxicity to organisms [6,7]. The loss of reactive dye in dyeing process is a major environmental problem in textile industry. Wastewaters containing reactive dyes are usually toxic and sometimes carcinogenic as well as causing aesthetic problem due to the color. Coloring materials are the main contaminants in textile wastewater and have to be treated and removed prior to discharge. Wastewaters from reactive dyeing process usually have high alkalinity (> 200mg $CaCO_3$/L),

inorganic (total dissolved solids (TDS) (>5000mg/L)) and organic matters content (COD >1000mg/L) [8].

Numerous methods used to treat textile wastewaters are such as biological process, chemical coagulation, activated carbon adsorption, ultrafiltration, ozonation, electro-coagulation and flotation process, and other physical processes [9]. Biological treatment of colored wastewater usually subjects low efficiency for dye removal due to toxicity of wastewater and the need for a continues of an aeration system [10]. Chemical coagulation process is still used for treating dyeing wastewater but a secondary water pollution might result [11]. Activated carbon is a common adsorbent but it is difficult to be regenerated and its performance is limited by equilibrium [12]. In ultrafiltration and reserve osmosis processes, the high cost and low production capacity are the main limitations [13,14]. Due to drawbacks of current treatment processes, developing a new and effective technology for treatment of textile wastewater is environmentally important. Electrocoagulation process has been proven to be an effective process to treat a variety of industrial wastewaters. These include paper mill wastewater [15], metal plating wastewater [16], oily wastewater, battery industry wastewater [17] and other wastewaters such as urban wastewater, restaurant wastewater, organic wastewater, heavy metals wastewater, textile wastewater [18,19] and landfill leachate wastewater [20].

Electrocoagulation process (EC) is an alternative process for wastewater treatment. It uses an electrochemical process in combination with chemical coagulation. In an electrocoagulation process, an applied potential releases the coagulant species into the solution by dissolving the metal anode such as aluminum or iron [21]. The produced metal coagulates are active and have higher effect on binding of particles.

Finally, metal coagulants are combined with negative charged particles to cause coagulation [22].

Generally, the main electrochemical reactions that occur at the electrodes are:

$M_{(s)} \rightarrow M_{(aq)}^{n+} + ne^-$ (at the anode) (1)

$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$ (at the anode) (2)

$n\ H_2O + ne^- \rightarrow \frac{n}{2}H_{2(g)} + n\ OH^-$ (at the cathode) (3)

$4H^+ + 4e^- \rightarrow 2H_{2(g)}$ (at the cathode) (4)

Where M = anode material, n = number of electrons involved in the oxidation and reduction reaction [23]. Ions of soluble metals such as Al and Fe are produced at the anode and react with the hydroxides ions formed at the cathode. Finally the metal hydroxides are generated and expressed as:

$M^{n+} + n\ OH^- \rightarrow M(OH)_n$ (5)

Theses insoluble metal hydroxides will react with the suspended and colloidal solids and precipitate [24]. The use of electrocoagulation process in the treatment of wastewater in recently favored due to its high performance in removal of pollutants. Several studies on electrocoagulation technology have been conducted by using aluminum and iron electrodes [25-30]. However, the search for a new material that could lead to high dye removal is still needed. To the best of our knowledge, zinc has not been used for treatment of textile wastewater using EC process and this is the first time the uses of this type of sacrificial electrode is reported. In this study, zinc was chosen as the electrode material and synthetic textile wastewater was prepared by using reactive blue 261 dye. The aim of this study was to investigate the effect of operating parameters on dye removal of synthetic textile wastewater in

order to obtain the best operating parameters such as current density, initial pH and initial dye concentration using EC process and zinc electrode. The best parameters were then used to treat a real textile wastewater.

2. Materials and methods

2.1. Synthetic textile wastewater

The dye used in the preparation of synthetic textile wastewater was Reactive Blue 261 dye provided by Penfabric textile factory in Penang, Malaysia. The synthetic textile wastewater was prepared by dissolving different concentrations of the dye (20-100mg) into one liter of distilled water.

2.2. Real textile wastewater (effluent)

The real textile wastewater used in this study was collected from the dying process at Penfabric textile factory. The main characteristics of real textile wastewater are presented in Table 1.

Table 1 : Characteristics of the real textile wastewater.

Basic property	Value
pH	9
Temperature (ºC)	28
Electrical conductivity (µS/cm)	28600
COD (mg/L)	1100
Turbidity (NTU)	90
λ_{max} (nm)	527
Absorbance at max wavelength	1.49

2.3. Experimental set-up

The electrocoagulation experimental set-up used in this study consisted of a reactor vessel, an electrode set, a DC power supply, a stirrer, a pH meter and an electroconductivity meter**.** The experimental reactor was made of double-walled Perspex with dimensions of 100mm x 100mm x 110mm and a capacity of 1.1 liter. Ten square electrodes i.e. five anodes and five cathodes of the same dimensions (70mm x 70mm x 1mm) were used in this study. The submerged effective surface area for each electrode was 55mm x 70mm. The distance between electrodes was maintained at 5mm while the gap between electrodes and the bottom of the reactor was set at 30mm in all experiments. Zinc electrode with a purity of 99.5% was used in this study. In all experiments, the anode and cathode materials were the same materials in order to avoid passivation of the anode surface or the formation of carbonate films on the surfaces of the cathode. The electrodes were connected to each other in parallel monopolar mode. Two outer electrodes were connected to a DC power supply (DAZHENG PS-305D; 32V, 5A) in order to supply and control the required current during the experimental run. A magnetic stirrer was used during the experiment and the solution was stirred at a constant desired speed to ensure uniformity and homogeneity of the solution. The experimental setup used in this study is schematically shown in Fig. 1.

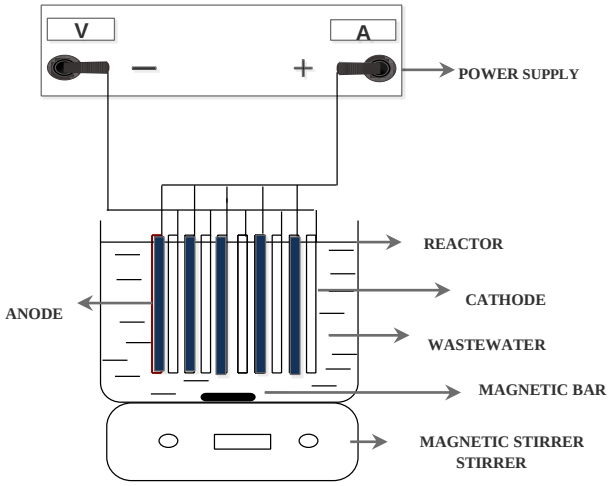

Fig. 1. The experimental set-up used for treating textile wastewater.

2.4. Experimental procedure

Before starting each run, the electrodes were washed with acetone in order to remove any contaminants such as oil, grease, scales and dust from the surface, then were dipped in a dilute HCl solution (10%) for 5 minutes for electrode activation and subsequently rinsed through with distilled water, dried and weighted. 1000 mL of wastewater solution was then fed into the reactor. The electroconductivity of the solution was adjusted by adding 5 g of NaCl to the solution and was measured by a conductivity meter while pH of the solution was adjusted to the required values by adding 0.1M NaOH or 0.1M H_2SO_4 solution. Then, the pH value was monitored using the pH meter. In order to get homogenous solution, the solution was continuously stirred at 250 rpm for all experiments. The electrode set was placed inside the reactor at a depth of 55 mm and connected to a DC power supply. The current density was adjusted to the desired values (2-8 mA/cm^2) and 60 minutes of operating time was set for each run. All experiments were conducted at room

temperature. The experimental run was started when the DC power supply was switched on. Samples of the treated wastewater were taken using a pipet from the middle of the reactor at 5, 10, 20, 30, 40, 50 and 60 minutes of operating time. The collected samples were filtered using a 0.45µm membrane filter paper and settled for 60 minutes for further analysis. At the end of the experiment, the DC power supply was switched off and the electrodes were disconnected and removed from the reactor, washed, rinsed dried and then reweighted.

Effects of various operating parameters (current density, initial dye concentration and initial pH) on dye removal were investigated in this study by varying one parameter at a time while the other parameters were kept constant. Four levels of operating parameters investigated were current density (2, 4, 6 and 8 mA/cm^2) pH of the sample (5, 6, 7, 8 and 9), dye concentrations (20, 30, 50 and 100 mg/l) and operating time upto 60 minutes.

2.5. Analysis

The dye removal was measured using a UV-vis spectrophotometer (Agilent 2010) based on the absorbance of the treated sample (absorbance before and after the treatment) carried out at the maximum wavelength of Reactive Blue 261 dye. In order to determine unknown concentration of the dye, the absorbances of several known dye concentrations were measured using a 1cm quartz cuvette put inside the chamber of UV-vis spectrophotometer. The calibration curve was created between the absorbance (A) and the dye concentration (C). The concentrations of dye before and after treatment are determined using the calibration curve and then the dye removal efficiency (ER) for synthetic textile wastewater is calculated using Equation 6 [31].

$$\% ER = (C_0 - C) / C_0 \times 100\% \quad (6)$$

Where, C_0 is the initial concentration of dye (mg/l) and C is the final concentration of dye (mg/l).

Meanwhile the dye removal of real textile wastewater was determined by measuring the absorbance of untreated and treated sample using UV-vis spectrophotometer. The efficiency of dye removal is calculated as follows [32,21].

$$\% ER = (Abs_0 - Abs) / Abs_0 \times 100 \quad (7)$$

Where, Abs_0 is the initial absorbance of wastewater and

Abs is the absorbance of wastewater after treatment at time t (min).

2.6. Electrode consumption (weight loss test)

The actual consumption of electrode was determined by using a weight loss method. The electrodes were weighted at the beginning and the end of each experiment. The weight loss of electrode was calculated by taking the difference between the initial and final electrode weight [33]. The electrode consumption is determined using the following equation [34]:

$$\Delta W = W1 - W2 \quad (8)$$

Where, ΔW is the weight loss of electrode material (g),

W1 is the weight of the electrode before EC treatment (g) and

W2 is the weight of the electrode after EC treatment (g)

2.7. Electrical energy consumption

The electrical consumption of energy during the electrocoagulation process for zinc electrode was calculated at optimum conditions using Equation 9 [35].

$$E = UIt / V \quad (9)$$

Where, E is the amount of energy consumed during EC process (kWh/m^3),

U is the operating voltage (Volt), I is the operating current (Ampere), t is the EC process time (hour) and V is the volume of wastewater (m^3).

2.8. Characterization of electrode surface using scanning electron microscope (SEM)

The characterization of electrode surface enables the understanding on the effect of wastewater treatment using EC process on the surface morphology, composition and microstructure of the electrode. Electrode surface characterization before and after wastewater treatment was conducted using a SEM (Philips model XL30S). Electrode sample with (dimensions of 10 x 10 x 1mm) was inserted into the sample chamber of SEM and observed at various magnifications.

3. Results and Discussion

3.1. Effect of current density on dye removal efficiency of synthetic textile wastewater

Current density plays an important role in the dye removal as it is the most important parameter that could determine the production rate of coagulant, adjust bubble production and affect the growth rate of the flocs [23,36]. The electrical current introduced to the electrocoagulation system determines the amount of metal ions such as Zn^{2+} ions that could be released from the electrodes materials. According to Faraday's law, increasing in the value of electrical current leads to an increase in the concentration of metal ions.

$$\Delta m = MQ/ FZ \qquad (10)$$

Where; Δm is the mass of the substance liberated at an electrode surface in gram; Q is the total electric charge passing through the electrode material; F= 96845Cmole^{-1} is the Faraday's constant; Z is the number of electrons transferred during the anodic reaction and M is the molar mass of the substance [37]. To determine the effect of current density on dye removal efficiency, four different values of current density i.e. 2, 4, 6 and 8 mA/cm^2 were studied at natural pH of synthetic textile wastewater (pH of 6.8), initial dye concentration of 50 mg/L and a maximum duration time of 60 minutes using zinc electrode.

As shown in Fig. 2, dye removal efficiency using zinc electrode was found to increase gradually as the current density was increased. The dye removal efficiency reached the highest values with current densities of 4, 6 and 8 mA/cm^2 in the first 10 minutes with the best removal of 99.4% recorded at 4 mA/cm^2. Meanwhile, the current density of 2 mA/cm^2 required more time (20 min) to reach the same value of dye removal (99.4%). This can be explained based on the conversion of dissolved Zn^{2+} ions into insoluble zinc hydroxide ($Zn(OH)_2$). With increasing current density, the generated $Zn(OH)_2$ also increased to have better ability to remove dye molecules by adsorption mechanism and this led to high dye removal [38]. Increasing process time beyond 10 minutes for the current densities of 4 to 6 and 8 mA/ cm^2 did not result in any noticeable changes in the dye removal efficiency. However, In order to avoid excessive evolution of hydrogen and to minimize the cost of operation, the current density should be as low as possible [39]. The lowest current density that led to higher dye removal in short treatment time was 4 mA/cm^2. As a current density of 4 mA/cm^2 showed the highest performance in terms of dye removal, it was then chosen for subsequent studies.

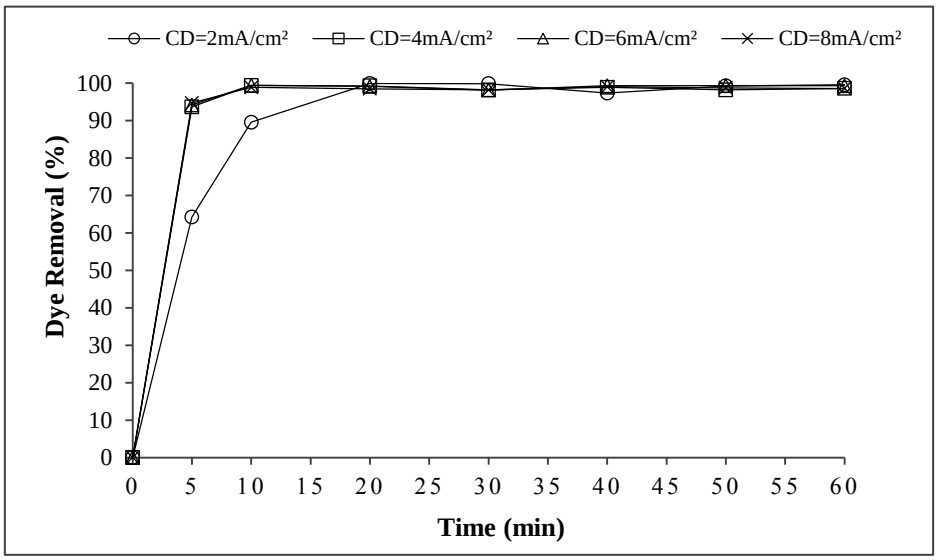

Fig. 2. Dye removal of synthetic textile wastewater at various current densities using

zinc electrode (pH = 6.8 and dye concentration = 50 mg/L).

3.2. Effect of initial pH on dye removal efficiency of synthetic textile wastewater

Initial pH of the solution is another important parameter that could affect the EC process performance. The maximum pollutant removal efficiency could be achieved at the optimum value of solution pH for that particular pollutant. The removal efficiency of pollutants may be decreased by either increasing or decreasing in the value of pH solution from the optimum pH [4]. The main reactions for zinc electrode are:

$$Zn \rightarrow Zn^{2+} + 2e^{-} \text{ (at the anode)} \quad (11)$$

$$2H_2O + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-} \text{ (at the cathode)} \quad (12)$$

$$Zn^{2+}_{(aq)} + + 2H_2O \rightarrow Zn(OH)_2\downarrow + 2H^{+} \quad (13)$$

$Zn(OH)_2$ flocs could adsorb the soluble organic particles then these flocs would be removed from the aqueous solution by sedimentation [38]. According to the Zn-H_2O purbaix diagram and thermodynamics point view of zinc material, the precipitation of $Zn(OH)_2$ would only be significant at pH (>8.6). However, the interfacial pH increase during the electrocoagulation process favored the zinc hydroxide formation [38].

The effect of initial pH on dye removal efficiency using zinc electrode is presented in Fig. 3. The removal of the dye was found to increase with increasing pH value. 99.7% of dye removal was achieved at an initial pH of 9. The dye removal reached the highest value in 10 minutes of the treatment and afterwards changes in the dye removal efficiency were very minimal. After 60 minutes of the treatment, the dye removal efficiency reached 99.1%. Zinc purbaix diagram suggests that the zinc hydroxides formation and the minimum solubility of zinc hydroxides $Zn(OH)_2$ occur in pH range of 7 to 9. Majority of zinc complexes and their precipitations for adsorption of dye were formed at pH 9 which might cause an increase in the dye removal.

At initial pH of 8 and 7, the dye removal efficiency reached 99.39%, and 99.35% respectively in the first 10 minutes. Meanwhile, at evidently lower pH value of 5, the dye removal efficiency decreased to 89.4%. Therefore, low pH value did not support the formation of hydroxides and hydroxyl ions and might indeed cause the inhibition of the EC process [40,33]. A pH value of 9 was therefore chosen to be used for further study.

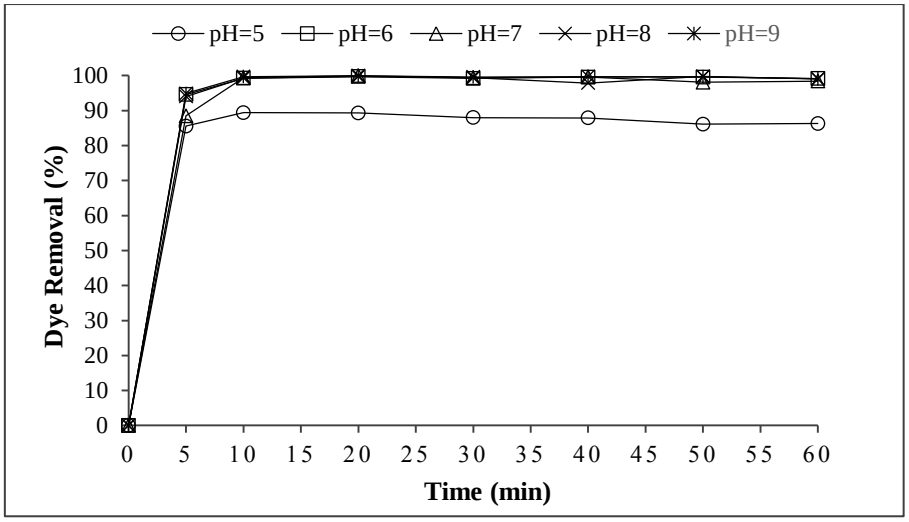

Fig. 3. Dye removal of synthetic textile wastewater at different pH using zinc electrode

(current density = 4 mA/cm^2 and dye concentration = 50 mg/L).

3.3. Effect of initial dye concentration on dye removal of synthetic textile wastewater

Fig. 4 illustrates dye removal efficiency as a function in initial dye concentration. The results show that the EC process gave satisfactory findings for all studied concentrations. More than 99% dye removals were obtained within 10 minutes of the treatment time for all studied dye concentrations.

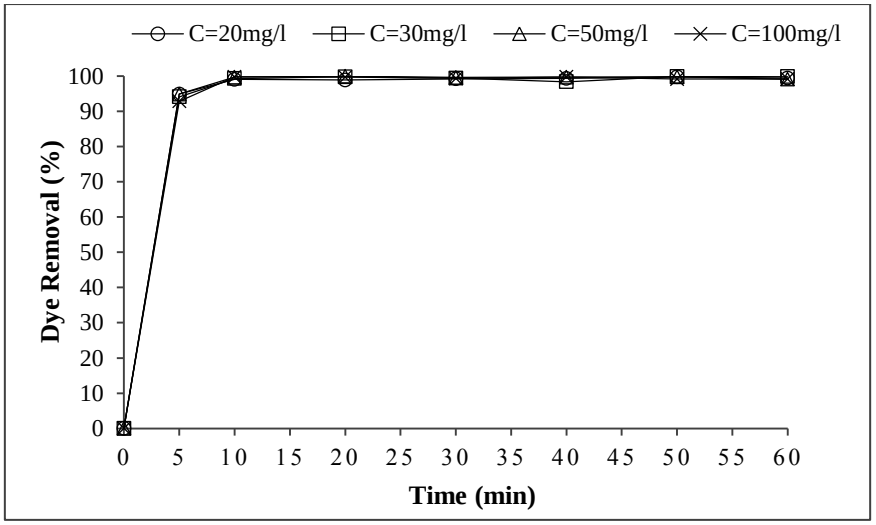

Fig. 4. Dye removals of synthetic textile wastewater at various concentrations using

zinc electrode (current density = 4 mA/cm^2 and initial pH = 9).

It was noted that the dye removal efficiency did not show noticeable changes with varying dye concentration from 20 to 100 mg/L using zinc electrode. This might be due to the flocs that were produced using zinc electrode that were able to adsorb nearly all dye molecules when the concentration was in the range of 20 -100 mg/L. In the meantime, dye removal remained at the highest value during the 60 minutes of the treatment. At initial dye concentration of 100 mg/L, the produced flocs were sufficient to adsorb all dye molecules and it led to high dye removal. Therefore, a concentration of 100 mg/L was selected to be the best dye concentration for zinc electrode.

3.4. Dye removal efficiency of synthetic and real textile wastewater using zinc electrode

Dye removal efficiency was investigated at an initial pH of 9, a current density of 4 mA/cm^2 and an initial dye concentration of 100 mg/L

for zinc electrode. The results of dye removal are shown in Fig. 5. High value of dye removal efficiency (99.8%) was obtained in 10 minutes of the EC treatment of synthetic textile wastewater. The change in color from originally blue color to colorless with more than 99% of dye removal during the first 10 minutes of the treatment was observed. By extending the treatment time up to 60 minutes, complete discolorization the blue solution was noted.

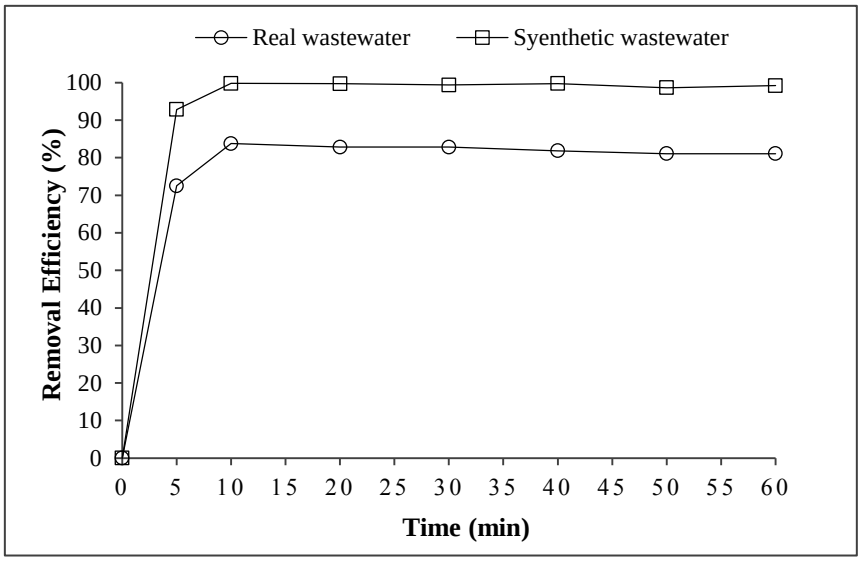

Fig. 5. Comparison between dye removal for synthetic and real textile wastewater using zinc electrode under operating conditions (current density = 4 mA/cm^2, initial pH = 9 and initial dye concentration = 100 mg/L).

Meanwhile treatment of real textile wastewater showed relatively lower dye removals where it reached only 83.7% in 10 minutes of the EC treatment using zinc electrode. A slight reduction in dye removal of treated real textile wastewater compared to synthetic textile wastewater was observed. This was due to the presence of other organic compounds such as sequestering, chemical and auxiliary agents in real textile

wastewater which could affect the performance of the EC process. Meanwhile, synthetic textile wastewater might contain only reactive dye which could be easily removed. It can be concluded that dye removal of synthetic wastewater was higher than that of the real textile wastewater.

3.5. Energy consumption during treatment of synth18etic and real textile wastewater

The operating cost of electrocoagulation process is associated with the cost of electrical energy consumed during the treatment of textile wastewater. The electrical energy required to treat textile wastewater needs to be estimated to avoid high operating cost and to evaluate the EC process from economic point of view. The calculation of energy consumption was done using equation 9 as reported in section 2.7. Energy consumption calculation was performed during the treatment of synthetic and real textile wastewater under optimum operating conditions. The current and cell voltage were measured at the optimum treatment time which was 10 minutes. The electrical consumption for treatment of real textile wastewater was found to be higher than that of the treatment of synthetic textile wastewater and the consumption was recorded at 0.69 kWh/m^3. However the power consumption reduced to 0.66 kWh/m^3 when zinc electrode was used in the treatment of synthetic textile wastewater. Thus, this type of electrode material had a considerable saving on electrical energy consumption during the EC process.

3.6. Electrode weight loss calculations

Weight loss of electrode during the EC process was calculated by measuring the initial electrode weight and eventual weight of the electrode after the treatment. The weight loss measurements were

performed under the optimum of operating conditions. The results are presented in Table 2.

Table 2 : Weight loss of electrodes during the EC treatment of synthetic and real textile wastewater for10 min.

Wastewater	Weight loss (kg/m^3 of wastewater)
Synthetic wastewater	0.240
Real wastewater	0.263

The lowest weight loss was recorded with zinc electrode which was 0.24 kg/m^3 during treatment of synthetic textile wastewater. Meanwhile, zinc electrode showed 0.26 kg/m^3 reduction in the weight of the electrode during the EC treatment of real textile wastewater.

3.7. Characterization of anode surface by using scanning electron microscopy (SEM)

SEM images of electrodes surface before and after the EC treatment are shown in Fig. 6. The images show the typical microstructure for zinc. The anodes surfaces were found to be smooth and uniform. No appearance of dents and dissolution species was detected on the surface of the electrodes.

After the EC treatment, SEM images of anodes surface show relatively rough surface, and the appearance of indention suggested the dissolution of certain species from the electrode surface. The formation of indention was attributed to anode material dissolution during the EC treatment where zinc ions were generated at anode surface.

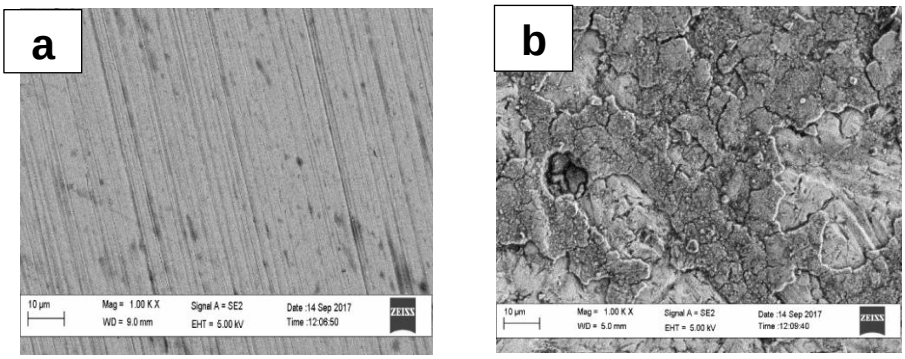

Fig. 6. SEM images for zinc anode (a) before and (b) after the EC treatment.

4. Conclusions

Various operating parameters such as current density, initial pH and, initial dye concentration were demonstrated to have significant effects on the dye removal efficiency of the synthetic textile wastewater. The treated synthetic textile wastewater under the optimum operating conditions showed that the highest dye removal (99.8%), lowest electrode weight loss (0.24 kg/m^3) and reasonable energy consumption (0.66 kWh/m^3) which were achieved at optimum operating time (10 min). Meanwhile, dye removal, power consumption and electrode weight loss were found at 83.7%, 0.69 kWh/m^3 and 0.26 kg/m^3 respectively for treatment of real textile wastewater. Comparison between the treatment of synthetic and real textile wastewater for removal of the dye showed a slight reduction in dye removal efficiency of real textile wastewater. It was due to the fact that the real textile wastewater might contain other components that could interfere with dye removal efficiency. Therefore the EC process demonstrated a good dye removal efficiency using zinc electrode material for treatment of synthetic and real textile wastewaters.

Thus, zinc electrode could be an effective electrode material for treatment of textile wastewater containing reactive dye.

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