# REMOVAL OF COD AND TURBIDITY TO IMPROVE WASTEWATER QUALITY USING ELECTROCOAGULATION TECHNIQUE

Moh Faiqun Ni'am\*<sup>1</sup>, Fadil Othman<sup>2</sup>, Johan Sohaili<sup>2</sup>, Zulfa Fauzia<sup>2</sup>

<sup>1</sup> Civil Engineering Department, Universitas Islam Sultan Agung, Semarang, Indonesia <sup>2</sup> Environmental Dept., Civil Engineering Faculty, Universiti Teknologi Malaysia, Johor, Malaysia

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#### Abstract

Electrocoagulation (EC) is becoming a popular process to be used for wastewater treatment. The removal of COD and turbidity from wastewater by EC using iron (Fe) electrode material was investigated in this paper. Several working parameters, such as pH, current density, and operating time were studied in an attempt to achieve a higher removal capacity. Wastewater sample was made from milk powder with initial COD of  $1140 \text{ mgL}^{-1}$  and turbidity of 491 NTU. Current density was varied from 3.51 to 5.62 mA cm<sup>-2</sup>, and operating time of between 30 and 50 minutes. The results show that the effluent wastewater was very clear and its quality exceeded the direct discharge standard. The removal efficiencies of COD and turbidity were high, being more than 65 % and 95 %. In addition, the experimental results also show that the electrocoagulation can neutralize pH of wastewater.

#### Abstrak

Elektrokoagulasi (EC) merupakan suatu proses yang populer untuk digunakan pada perawatan air sisa. Penyingkiran COD dan kekeruhan daripada air sisa oleh EC yang menggunakan material elektrod besi (Fe) telah diselidiki dalam kertas kerja ini. Beberapa parameter, seperti pH, kerapatan arus dan masa perawatan dikaji dalam percubaan untuk mendapatkan kadar penyingkiran lebih tinggi. Air sisa dibuat dari tepung susu dengan nilai awal COD 1140 mgL<sup>-1</sup> dan kekeruhan 491 NTU. Kerapatan arus divariasi dari 3.51 hingga 5.62 mA cm<sup>-2</sup>, dan masa perawatan antara 30 dan 50 minit. Keputusan kajian mendapati bahawa efluen air sisa sangat jernih dan sesuai dengan piawai pelepasan. Efisiensi penyingkiran COD dan kekeruhan adalah tinggi, menjadi lebih dari 65 % dan 95 %. Sebagai tambahan, keputusan kajian juga mendapati bahawa elektrokoagulasi dapat meneutralkan pH air sisa.

#### Introduction

The reuse of wastewater has become an absolute necessity. Demands to the cleaning industrial and domestic wastewater to avoid environmental pollution and especially contamination of pure water resources are becoming national and international issues. Innovative, cheap and effective methods of purifying and cleaning wastewater before discharging into any other water systems are needed. Electrocoagulation (EC) due to some advantages over chemical coagulation is becoming a popular process to be used for wastewater treatment.

Electrocoagulation is not a new technology [1]. Treatment of wastewater by EC has been practiced for most of the 20<sup>th</sup> century with limited success [2]. Using electricity to treat water was first proposed in UK in 1889, and the application of electrolysis in mineral beneficiation was patented by Elmore in 1904 [3]. The principle of EC was used to treat bilge water from ships was first patented in 1906 by A. E. Dietrich [4].

The electrocoagulation is simple and efficient method for the treatment of many water and wastewaters. It has not been widely accepted because of high initial capital costs as compared to other treatment technologies. In recent years, many investigations have been especially focused on the use of EC owing to the increase in environmental restrictions on effluent wastewater [5-8].

The use of electrocoagulation for the treatment of wastewater has been reported by various authors, and several differences were found in comparison to the chemical coagulation process. A literature survey indicates that EC is an efficient treatment process for different wastes, e.g. soluble oils, liquid from the food, textile industries, or cellulose and effluents from the paper industry [9-10]. Electrocoagulation is an effective process for the destabilisation of finely dispersed particles by removing hydrocarbons, greases, suspended solids and heavy metals from different types of wastewater [12-13]. Electrocoagulation has been proposed in recent years as an effective method to treat various wastewaters such as: landfill leachate, restaurant wastewater, saline

wastewater, tar sand and oil shale wastewater, urban wastewater, laundry wastewater, nitrate and arsenic bearing wastewater, and chemical mechanical polishing wastewater [14].

Alumunium or iron were usually used as electrodes and their cations are generated by dissolution of sacrificial anodes upon the application of a direct current [8,10,13,15-17]. Electrocoagulation technique for treatment of wastewater samples have been conducted on a laboratory scale and good removal of COD, color, turbidity, and dissolved solids at varying operating conditions have been obtained [8,9,14-18].

# Experimental

This study is to investigate the effect of electrocoagulation (EC) process. This research is mainly focused on the capability of EC technology to improve wastewater quality, such as to increase removal efficiencies of COD and turbidity.

## Description of EC Technique

EC is an electrochemical wastewater treatment technology that is currently experiencing both increased popularity and considerable technical improvements. EC is a complicated process involving many chemical and physical phenomena that use consumable electrodes to supply ions into the wastewater stream [19]. The mechanism of the Electrochemical process in aqueous systems is quite complex [6]. It is generally believed that there are three possible mechanisms involved in the process: electro-coagulation, electro-flotation and electro-oxidation.

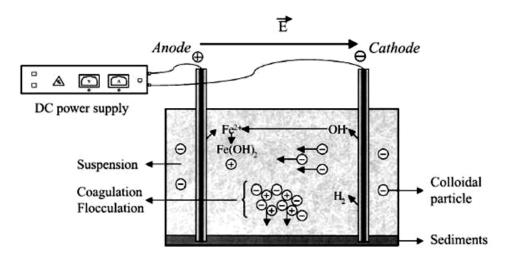


Figure 1. Principle of electrocoagulation process [15].

In EC with electrical current flowing between two electrodes, coagulant is generated in situ by electrolytic oxidation of the anode material. With an iron anode,  $Fe(OH)_n$  with n = 2 or 3 is formed at the anode. Simplified oxidation and reduction mechanisms at the anode and cathode of the iron electrodes are represented by [2,7,15,17,21]:

(a) Mechanism 1 :

Anode : Fe (s) 
$$\rightarrow$$
 Fe<sup>2+</sup> (aq) + 2 e<sup>-</sup>  
Fe<sup>2+</sup> (aq) + 2 OH<sup>-</sup> (aq)  $\rightarrow$  Fe(OH)<sub>2</sub> (s)  
Cathode : 2 H<sub>2</sub>O (1) + 2 e<sup>-</sup>  $\rightarrow$  H<sub>2</sub> (g) + 2 OH<sup>-</sup> (aq)  
Overall : Fe (s) + 2 H<sub>2</sub>O (1)  $\rightarrow$  Fe(OH)<sub>2</sub> (s) + H<sub>2</sub> (g)

(b) Mechanism 2 :

Anode : 4 Fe (s)  $\rightarrow$  4 Fe<sup>2+</sup> (aq) + 8 e<sup>-</sup> 4 Fe<sup>2+</sup> (aq) + 10 H<sub>2</sub>O (l) + O<sub>2</sub> (g)  $\rightarrow$  4 Fe(OH)<sub>3</sub> (s) + 8 H<sup>+</sup> (aq) Cathode : 8 H<sup>+</sup> (aq) + 8 e<sup>-</sup>  $\rightarrow$  4 H<sub>2</sub> (g) Overall : 4 Fe (s) + 10 H<sub>2</sub>O (l) + O<sub>2</sub> (g)  $\rightarrow$  4 Fe(OH)<sub>3</sub> (s) + 4 H<sub>2</sub> (g)

Principle of EC is as shown in Figure 1. Dissolving of metal anodes are used to continuously 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 [9,15,19]. Particles interact with the iron hydroxides and can be removed either by surface complexation or electrostatic attraction [2,7,15,17,21]. The electric field increases the probability of charges in suspension coming into contact with each other compared to chemical dosing and stirring. Therefore, the EC process improves coagulation [7,15,20].

# Artificial Wastewater Sample

The synthetic wastewater sample made from milk powder and treated by using HCl 1 M as pH adjusment and electrolyte. The composition of artificial wastewater is shown in Table 1. Concentration of HCl in this sample is 5 mL/L (0.5 %) and obtained maximal current 0.8 A at voltage 30 V. The current density was adjusted to a desired value before the coagulation process started.

Table 1. Characteristics of artificial wastewater used in all experiments

Parameter	Value
Chemical oxygen demand COD	1140 mgL <sup>-1</sup>
Total suspended solids (TSS)	$1400 \text{ mgL}^{-1}$
Turbidity	491 NTU
pH after adjusted by HCl	2.91

# Equipments and Procedures

The experiments carried out in this work were set in static methods. EC and wastewater batch tests were performed in a cylindrical glass cell (volume 2000 mL) with stirring at constant speed (Figure 2). Stirring was provided by a plate impeller from plastic material  $\emptyset$  3 cm) at a rotating velocity of 100 rpm (HEIDOLPH RZR-2101 Electronic).

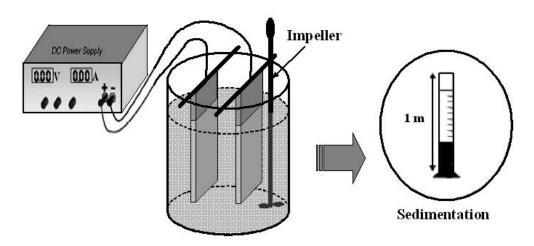


Figure 2: Schema of experimental apparatus

For EC tests, the monopolar iron (Fe) plate electrodes (130 mm x 50 mm x 4 mm) were used in this work. The total effective electrode area was  $142.40 \text{ cm}^2$  and the distance between both was 50 mm. Before each run, electrodes were washed with acetone solution to remove surface grease. At the end of run, the electrodes were washed thoroughly with water to remove any solid residues on the surfaces, and dried.

Electrodes were placed in two litres of fluid wastewater and connected to terminals of a DC Power Supply (LODESTAR 8107; 30V/10A) with potensiostatic or galvanostatic operational options. Bubbles generated from water electrolysis in EC could float flocs to the top of the suspension. After EC process, wastewater were degassed under low stirring speed with an impeller velocity 30 rpm.

The wastewater after EC treatment were placed in graduated sedimentation columns of 5 cm internal diameter and 1 m height (Figure 2). The effects of relevant wastewater characteristics such as pH, turbidity and COD removal efficiencies were studied during 30-240 minutes settling time.

#### Analytical Method

The pH of the solutions was measured by pH meter (Thermo Orion Model 420 A+). Thermal analysis was performed and equipped using HANNA Instrument Checktemp HI-98501. The turbidity removal was measured from wastewater samples by HACH DR/4000 (HACH Method 10047). COD measurments were determined according to the Standard Methods for Examination of Water and Wastewater (APHA, 1992). The COD samples were analysed using UV-Vis HACH DR/4000 spectrophotometer (HACH Method 8000).

The calculation of turbidity, COD and suspended solid removal efficiencies after electrocoagulation treatment were performed using this formula [17]:

$$CR(\%) = \frac{C_0 - C}{C_0} \times 100 \tag{1}$$

Where  $C_0$  and C are concentrations of wastewater before and after electrocoagulation in NTU or mgL<sup>-1</sup>, respectively.

The rate of change of wastewater concentration, such as turbidity, COD and suspended solid removal can be expressed as a first order kinetic model [20], as follows:

$$\frac{C}{C_{a}} = e^{-k_{2}t} \tag{2}$$

Where C, Co, and  $k_2$  are wastewater concentrations after EC, initial, and kinetic constant, respectively.

Hence, the loss of particles due to coagulation after treatment process [22], as follows:

$$\frac{C}{C_0} = a \, (b - e^{-kt}) \tag{3}$$

Where k is kinetic constant ; a and b are constant values.

## **Results and Discussion**

#### Effect of Operating Time

The effluent treated with iron electrode as anode, appeared greenish first and then turned yellow and turbid. This green and yellow colour must have resulted from  $Fe^{2+}$  and  $Fe^{3+}$  ions generated during the EC process.  $Fe^{2+}$  is the common ion generated in situ of electrolysis of iron electrode. It has relatively high solubility at acidic or neutral conditions and can be oxidised easily into  $Fe^{3+}$  by dissolved oxygen in water.

The effect of time was studied at constant current density of  $5.62 \text{ mA cm}^2$ . Figure 3 illustrates the removal of COD and turbidity as a function of operating time. It is clearly seen from Fig. 3 that, the operating time has a significant effect on the pollutant removal. When the operating time changed from 10 to 50 minute, the removal of COD from 967 to 267 mg/L and turbidity from 447 to 9 NTU were obtained. In this process, electrocoagulation involves two stages which are destabilization and aggregation. The first stage is usually short, whereas the second stage is relatively long.

It is explainable from Figure 3 that metal ions as destabilization agent are produced at the anode through electrochemical reactions. This matter will cause the charge loading is low, when the operating time is shortened. In this condition, the metal ion  $(Fe^{3+})$  dosage was not sufficient to destabilize all colloidal and finely suspended particles. Thus the COD and turbidity removal efficiencies were not high.

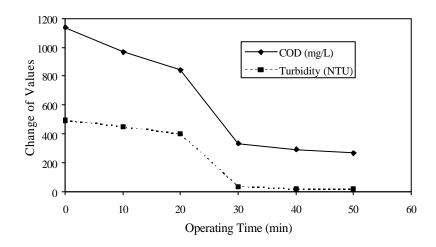


Figure 3. Effect of operating time on the removal of COD and Turbidity (current density: 5.62 mA cm<sup>-2</sup>)

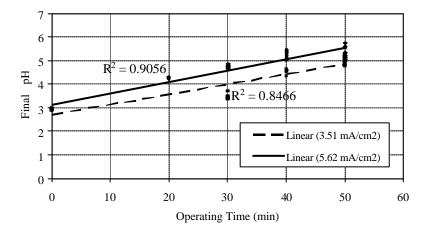


Figure 4. Effect of operating time on pH change

On the other hand, to explore the effect of the operating time on the pH value, the initial pH of solution was kept constant at 2.91. Figure 4 showed a relationship between operating time and pH value. As shown in Fig.4, the pH value increases as the time of EC process is increased. This happened because of the OH<sup>-</sup> ion accumulates in aqueous solution during the process. The increase of pH at acidic condition was attributed to hydrogen evolution at cathodes.

In the previous studies, such as were explained by Kobya et al. [16] and Daneshvar et al. [17], pH is an important factor influencing on the treatment performance of the electrocoagulation process. Generally, the pH of the medium changes during the process, as observed also by other investigators [5, 8].

#### Effect of Settling Time

The effect of settling time on the COD and turbidity removal as shown in Figure 5 and Figure 6, were investigated by varying the applied current of 3.51 and 5.62 mA cm<sup>2</sup> with operating time at 40 minutes. Figure 5(a) depicts the COD removal efficiency as a function of settling time. The COD removal efficiencies increase as the settling time is increased. For a current density of 3.51 mA cm<sup>2</sup>, the COD removal percentage was improved effectively from 8.16% to 32.81%. Whereas, an EC cell with current density of 5.62 mA cm<sup>2</sup> was

more effective in removing COD. Result showed that the efficiency of EC with current density of 5.62 mA cm<sup>-2</sup> was greater than 70 % when settling time was approximately 60 minutes.

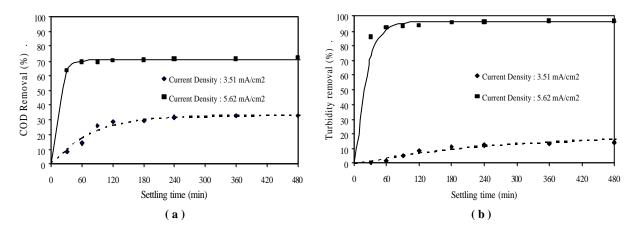


Figure 5a. Effect of settling time on COD and turbidity removal (treatment time = 40 min)

As depicted in Figure 5(b), the turbidity removal efficiency is increased to a value more than 95%, with current density of 5.62 mA cm<sup>2</sup>. On 480 minutes of settling time, turbidity removal is increased to 96.74% for 40 minutes treatment time. While for a current density of 3.51 mA cm<sup>2</sup>, the turbidity removal percentage was only improved from 1.22% to 18.73%.

Figure 6 shows the reduction of COD is in kinetics equation. The exponential curve of Fig. 6 is characteristic of a differential equation such as Eq. (3). The equation explain that the fraction of particles neutralized  $(b - e^{kt})$  is increasing with time, while the overall concentration, C, is decreasing. Initially, none of the particles are neutralized, and they must migrate toward the anode. After a certain time, a rather sharp drop in concentration occurs because coagulation has begun, and finally the rate begins to slow as most of the particles have become neutralized.

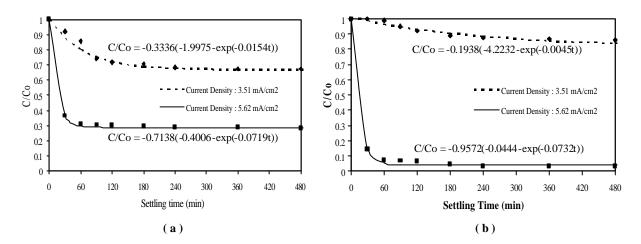


Figure 6. Effect of settling time on rate of change of COD and turbidity by EC process (t = 40 min).

The behaviour of those processes (Figure 5 and Figure 6) indicates that the EC technology can enhances the settling velocity of suspended particles.

# Conclusion

The removal efficiencies of COD and turbidity from wastewater were experimentally done by electrocoagulation technique. The processes were measured in the batch studies of a single stirred apparatus. The monopolar iron (Fe) plate electrodes were used in this work and were set at distance of 50 mm in the cylindrical glass cell (volume 2000 mL).

The results show that the COD removal efficiency is as high as 72.28% for EC process with current density of 5.62 mA cm<sup>2</sup>. Whereas, the turbidity removal in the process of wastewater with current density of 5.62 mA cm<sup>2</sup> was obtained 96.74%.

In general, 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. It mean that the EC technology can improve wastewater quality.

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