

# WASTEWATER

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

One of the major challenges facing mankind today is to provide clean water to the vast majority of the population around the world. The need for clean water is particularly critical in the third-world countries. River canals, estuaries, and other water bodies are being constantly polluted due to indiscriminate discharge of industrial effluents as well as other activities, and natural processes (1).

Freshwater is a finite resource, essential for agriculture, industry, and human existence. Without adequate freshwater supplies of adequate quantity and quality, sustainable development will not be possible. Water pollution and wasteful use of freshwater threaten development projects and make water treatment essential in order to produce safe drinking water (2). Demands to treat industrial and domestic wastewater to avoid environmental pollution, and contamination of drinking water supplies are national and international issues (3).

The reuse of wastewater has become an absolute necessity. There is, therefore, an urgent need to develop innovative, more effective, and inexpensive techniques for treatment of wastewater.

### EC with Iron Electrodes

Wide ranges of wastewater treatment techniques are known that include bio-

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logical processes and physical-chemical processes. A host of very promising techniques based on electrochemical technology are being developed, and existing ones improved that do not require chemical additions (1). These include electrocoagulation (EC), electroflotation (EF), electrodeposition (ED), electrooxidation (EO), and others (4). Even though one of these, EC, has reached profitable commercialization, it has received very little scientific attention (1). Figure 1 shows the principles of EC.

EC has been used for the treatment of wastewater 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) (5-9).

EC is an effective process for the destabilization of finely dispersed particles by removing hydrocarbons, greases, suspended solids, and heavy metals from different types of wastewater (5, 6). According to Can, et al., EC has been proposed in recent years as an effective method to treat various wastewaters such as: landfill leachate, restaurant wastewater, salty wastewater, tar sand and oil shale wastewater,

urban wastewater, laundry wastewater, nitrate- and arsenic-bearing wastewater, and chemical mechanical polishing wastewater (10).

Aluminum or iron are usually used as electrodes and their cations are generated by dissolution of sacrificial anodes upon the application of a direct current (5). Kobya, et al., has been investigated EC technologies for treatment of textile wastewaters using iron and aluminum electrode materials. The results show that iron is superior to aluminum as sacrificial electrode material, from chemical oxygen demand (COD) removal efficiency and energy consumption points (11).

Aluminum (Al) is usually used for water treatment, and iron (Fe) for wastewater treatment. The advantages of EC include high particulate removal efficiency, a compact treatment facility, relatively low cost, and the possibility of complete automation.

An EC has electrical current flowing between two electrodes. A coagulant is generated *in situ* by electrolytic oxidation of the anode material. In the work described here, iron (Fe) electrodes were used for the treatment of synthetic wastewater. 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

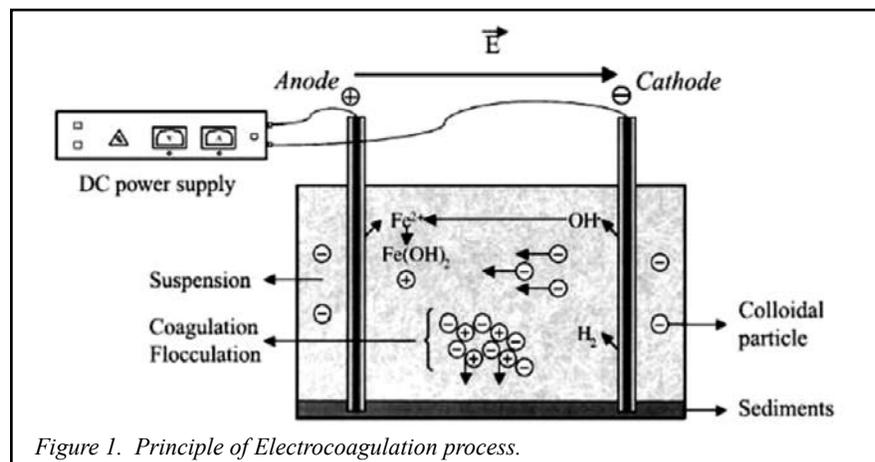


Figure 1. Principle of Electrocoagulation process.

the iron electrodes are represented by the equations represented in Mechanisms 1 and 2 shown in Table A (1, 8, 12-14):

According to Larue, et al., the generation of iron hydroxides (Fe(OH)<sub>n</sub>) is followed by an electrophoretic concentration of colloids (usually negatively charged) in the region close to the anode (8). Particles interact with the iron hydroxides and are removed either by surface complexation, or electrostatic attraction (1, 12-14).

### Materials and Methods

This research is mainly focused on the capability of EC technology to improve wastewater quality, such as to increase removal efficiencies of COD, turbidity, and suspended solids.

**Wastewater characteristics.** In this study, the synthetic wastewater sample made from milk powder and treated by using 1 molar (M) of hydrochloric acid (HCl) as pH adjustment and electrolyte. The concentration of HCl in this fluid is 5 milliliters per liter (mL/L) (0.5 %). The current density was adjusted to a desired value and the coagulation was started. The composition of artificial wastewater is shown in Table B.

**Experimental apparatus and procedures.** A laboratory batch EC reactor was designed and performed in a cylindrical glass cell (volume 2,000 mL) with stirring at constant speed. Stirring was provided by a plate impeller from plastic material (3 centimeters diameter [cm]) at a rotating velocity of 100 revolutions per minute (rpm) (HEIDOLPH RZR-2101 Electronic). The experiments carried out in this work were setup into static methods.

The monopolar iron (Fe) plate electrodes (dimension 130 millimeter [mm] x 50 mm x 4 mm) were used in this work. The total effective electrode area was 142.40 square centimeters (cm<sup>2</sup>) and the net spacing between the iron electrodes (d) was varied from 14 to 50 mm. Electrodes were placed in 2 L of fluid wastewater and connected to terminals of a DC power supply (LODESTAR 8107 ; 30V / 10A) with potentiostatic or galvanostatic operational options. Before each run, electrodes were washed with acetone solution to remove surface

**TABLE A**  
**EC Reduction Mechanisms at the Anode and Cathode of Iron Electrodes**

Mechanism 1:	
Anode :	$Fe (s) \rightarrow Fe^{2+} (aq) + 2 e^{-}$
	$Fe^{2+} (aq) + 2 OH^{-} (aq) \rightarrow Fe(OH)_2 (s)$
Cathode :	$2 H_2O (l) + 2 e^{-} \rightarrow H_2 (g) + 2 OH^{-} (aq)$
Overall :	$Fe (s) + 2 H_2O (l) \rightarrow Fe(OH)_2 (s) + H_2 (g)$
Mechanism 2:	
Anode :	$4 Fe (s) \rightarrow 4 Fe^{2+} (aq) + 8 e^{-}$
	$4 Fe^{2+} (aq) + 10 H_2O (l) + O_2 (g) \rightarrow 4 Fe(OH)_3 (s) + 8 H^{+} (aq)$
Cathode :	$8 H^{+} (aq) + 8 e^{-} \rightarrow 4 H_2 (g)$
Overall :	$4 Fe (s) + 10 H_2O (l) + O_2 (g) \rightarrow 4 Fe(OH)_3 (s) + 4 H_2 (g)$

**TABLE B**  
**Characteristics of Wastewater**

<i>Parameter</i>	<i>Value</i>
Chemical oxygen demand COD (mg/L)	1,140
TSS (mg/L)	1,400
Turbidity (NTU)	491
Initial pH	7.45
pH after adjusted by HCl	2.91

grease. At the end of run, the electrodes were washed thoroughly with water to remove any solid residues on the surfaces, and dried. The DC current was kept constant at 0.50 A, 0.65 A, 0.80 A, and 1.0 A. Bubbles generated from water electrolysis in EC could float flocs to the top of the suspension.

During each 10 minutes (min) of treatment time, the effects of relevant wastewater characteristics such as pH, temperature, turbidity, and COD removal efficiencies have been explored. Samples of treated wastewater were collected at 10 cm from the surface of wastewater using pipette, and then filtered before being analyzed.

After the EC process, the treated wastewater was degassed under a low-stirring speed with an impeller velocity 30 rpm. Afterwards, the treated wastewater was placed in graduated sedimentation columns of 5-cm internal diameter and 1-meter (m) height. Samples were collected at 5 cm from the surface of wastewater using a pipette, at 30, 60, and 90 min, and 2, 3, 4, 6, 8, and 12 hours (h) of settling time. The effects of relevant wastewater characteristics such as suspended solids (SS) removal

efficiency has been explored.

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

To measure total suspended solids (TSS), the wastewater samples were filtered through a standard GF/F glass fiber filter. The residual retained on the filter was dried in an oven at 1,030°C to 1,050°C until the weight of the filter showed no further changes. The increase in weight of the filter represents the TSS (APHA Method 2540 D).

The calculation of turbidity, COD, and suspended solid removal efficiencies after EC treatment were performed using the formula in Equation 1 (14). (*Editor's*

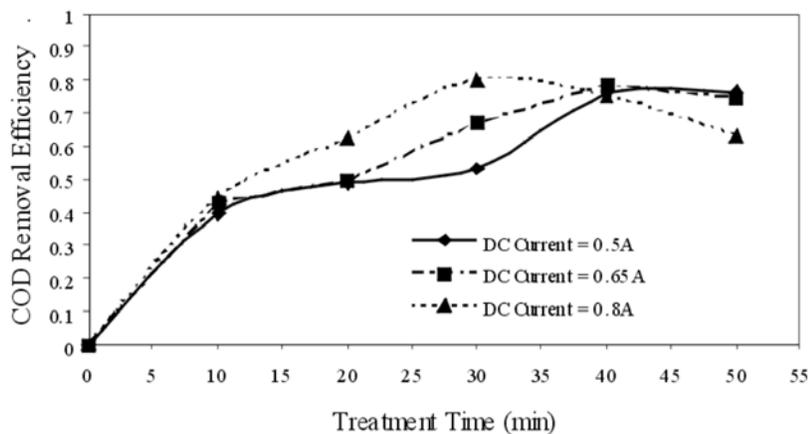


Figure 2. Effect of treatment time on the removal percentage of COD ( $d = 14$  mm).

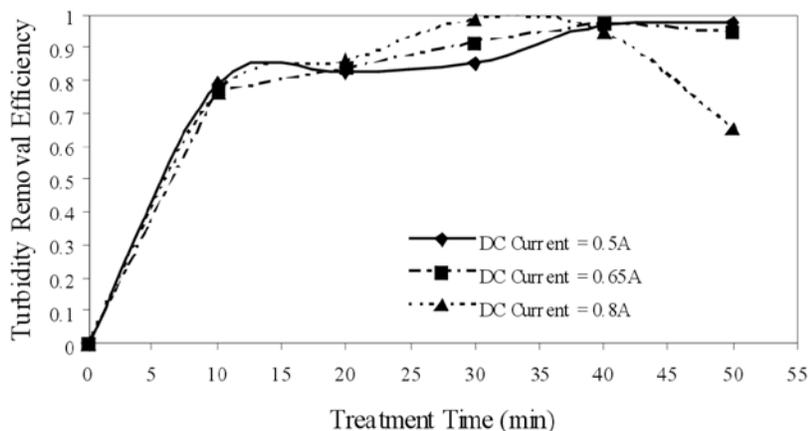


Figure 3. Effect of treatment time on the removal percentage of turbidity ( $d = 14$  mm).

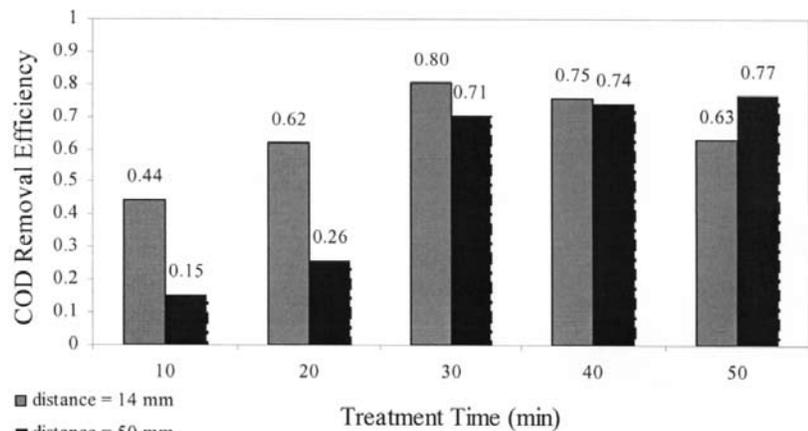


Figure 4. Effect of treatment time on the removal percentage of COD ( $I = 0.8A$ ).

note: Article equations appear together in an Equations Table.)

The rate of change of wastewater concentration, such as turbidity, COD and suspended solids removal can be expressed as a first order kinetic model (15), as follows in Equations 2. Hence, the loss of particles due to coagulation after treatment process (16) is illustrated through Equation 3.

## Results and Discussion

**Treatment time effect** at constant current between 0.50 to 1.0 A. Figures 2 and 3 illustrate the removal percentage of COD and turbidity as a function of treatment time. It is clearly seen from both figures that the treatment time has a significant effect on the pollutant removal. When the treatment time changed from 10 to 50 minute, the removal of COD moved from 691 to 226 mg/L, and turbidity from 117 to 9 nephelometric turbidity units (NTU) were obtained. In

this process, EC involves two stages that are destabilization and aggregation. The first stage is usually short, whereas the second stage is relatively long.

As seen from Figure 2, the optimal COD removal at DC current of 0.5A and 0.65 (> 75%) is reached after 40 min of wastewater processing. Whereas for the current of 0.8A, the optimal COD removal (80%) is reached at 30 min of EC process. And above of 30 min of treatment time, COD removal percentages decreased. This change probably occurs because the  $Fe^{3+}$  effects.

In this process, EC involves two stages, which are destabilization and aggregation. The first stage is usually short, whereas the second stage is relatively long. Figure 2 illustrates that metal ions as a destabilization agent are produced at the anode through electrochemical reactions.

The effluent appeared greenish first and then turned yellow and turbid. This green and yellow color 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 oxidized easily into  $Fe^{3+}$  by dissolved oxygen in water.

Analogous curves are observed for the turbidity. As seen clearly in Figure 3, the optimal percentage of turbidity removal at DC current of 0.5A and 0.65 (> 95%) is reached after 40 min of wastewater processing. And for the current of 0.8A, the optimal turbidity removal (98%) is reached at 30 min of EC process.

On the other hand, interelectrode distance ( $d$ ) have a significant effect on the efficiency of COD and turbidity removal. The effects of interelectrode distance depicted in the curves of relationship between treatment time and COD or turbidity removal in Figures 4 and 5, respectively.

In Figure 4, the optimum COD removal efficiency for the distance of 14 mm and 50 mm (80% and 77%) is reached at 30 min and 50 min of treatment time, respectively. While in Figure 5, the optimum turbidity removal efficiency (98%) is reached at 30 and 40 min treatment time, for  $d = 14$  mm and 50 mm, respectively.

**Cell current influence.** To determine the influence of DC current on the removal of COD and turbidity, experiments were conducted with treatment time at 10 to 30 min. The current was varied from 0.5A to 1.0A, corresponding to current densities in the range 35.10 to 70.22 A/m<sup>2</sup>.

Figure 6 illustrates the removal of COD as a function of cell current. It is clearly seen from Figure 6 that, the current has a significant effect on the COD removal. When the current changed from 0.5A to 1.0A, the optimum of COD removal percentage is reached till 76% (t=10 min) and more than 80% (t= 20 and 30 min). For the treatment time of 10 and 20 min, the optimum values are reached at 1.0A. While for the treatment time of 30 min, the optimum values are reached at 0.8A.

Somewhat the same results are found in the case of turbidity removal efficiency. The optimum of turbidity removal percentage is reached till 95% (t=10 min) and 98% (t= 20 and 30 min). For the treatment time of 10 and 20 min, the optimum values are reached at 1.0 A. While for the treatment time of 30 min, the optimum values is reached at 0.8A.

**Settling time effect and kinetic equation.**

From the batch studies involving various settling time presented for total suspended solids (TSS) removal experiment (Figure 7), the best efficiency started after 120 min of sedimentation. As seen in Figure 7, Suspended particles removal are increases as the settling time is increased. With an addition of treatment time from 30 min becomes 50 min, TSS removal is increased from 17.86% to 31.86% on 1,440 min (24 h) of settling time. Whereas, with 720 min (12 h) of settling time, TSS removal is increased between 15.71% to 31.29%.

Figure 8 shows the reduction of COD in kinetics equation. The exponential curve of Figure 6 is characteristic of a differential equation such as Equation 3. The equation explains 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

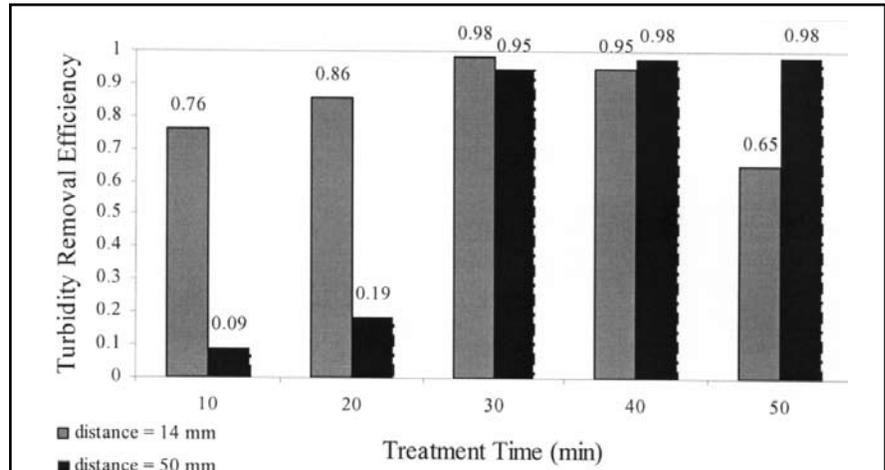


Figure 5. Effect of treatment time on the removal percentage of turbidity (I = 0.8A).

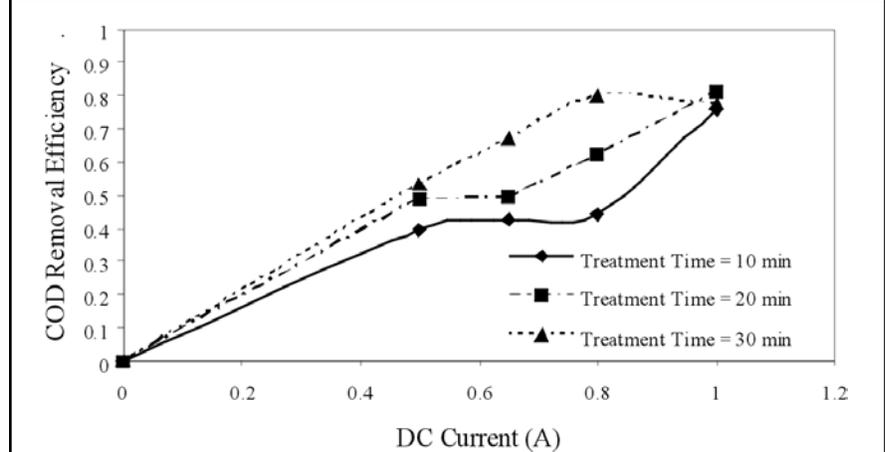


Figure 6. Influence of the current on the COD removal efficiency (d= 14 mm).

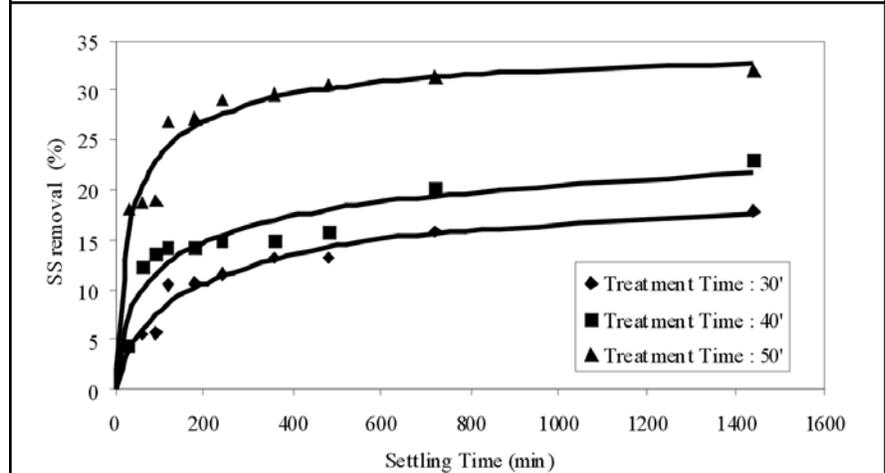


Figure 7. TSS removal efficiencies as a function of settling time (I = 0.8 A; d = 50 mm; t = 30 – 50 min).

to slow as most of the particles have become neutralized.

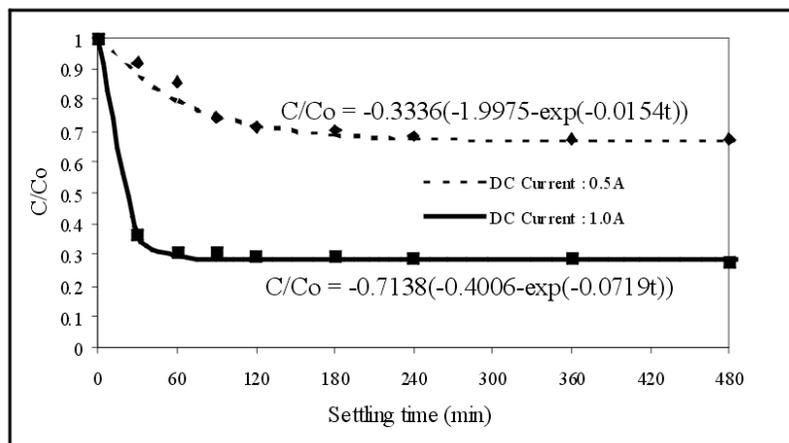
The kinetic rate law for describing the decrease in COD and turbidity concentration from wastewater with settling time is shown through Equation 3. A general reaction rate model (17) is illustrated through Equation 4.

According to Schnoor (18), a first order

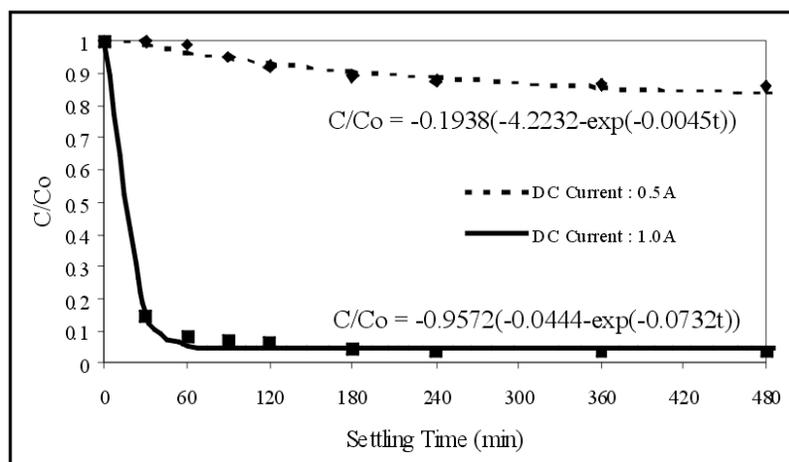
reaction is one in which the reaction rate is proportional to the concentration of the reactant to the first power. This is seen in Equations 5 and 6.

Equation 5 can be solved by separation of variables and integrated. It is a linear, ordinary, first order differential equation. This is shown in Equations 7 and 8.

Likewise, Equation 6 can be integrated



(a) COD removal



(b) turbidity removal

Figure 8. Effect of settling time on rate of change of COD and turbidity by EC process ( $t = 40$  min).

into these equations, resulting in Equation 9.

The assumption that the treatment process can enhance the amount of particles in B, so B represented of function of time (t) and treatment process (Tr) (16). This is progressively shown in Equations 10 through 13.

Substituting Equations 12 and 13 into Equation 10, we obtain Equation 14. From Equation 14, if  $t = \infty$ , and  $Tr = 0$ , and  $B = B_o$ ; the particle concentrations

in the initial of sedimentation, we obtain Equation 15.

If  $t = \infty$ ,  $Tr = Tr$ , and  $B = \infty$ , that is particles concentration in the final of sedimentation. We obtain equation 16. Substituting Equation 15 into Equation 16, we arrive at Equation 17. By substituting C and b from Equations 15 and 17 into Equation 14, we obtain Equation 18. The final concentration of particles is  $B_{\infty} = n$ ,  $CA_o$ , and  $n < 1$ , which is shown in Equation 19.

If  $CA = f(CA_o, Tr)$ , and  $B = f(CA_o,$

Tr), B always depends on CA, and can be expressed as seen in Equation 20. Substituting B from Equation 20 into Equation 19, we obtain Equation 21. Equation 21 can also be expressed as shown in Equation 22.

## Conclusion

This research is the preliminary stage from a study that combined magnetic fields and EC technique for improvement of wastewater quality. In this study, the effect of operational variables such as interelectrode distance, treatment time, current density, and settling time were examined.

The removal efficiencies of COD and turbidity were high, more than 65% and 95%, respectively. The suspended particles removal efficiency is as high as 31.86% on 24 h of settling time. The kinetics of the COD and turbidity removal were described by the empirical formulation, and fit to data.

In general, the results were obtained from the curves of treatment time and settling time, showing that the EC technique can enhance the settling velocity of suspended particles and removal of COD and turbidity. This means that the EC technique can remove the suspended particles, COD, and turbidity from wastewater and improve its quality.

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**Key words:** COD, ELECTROCOAGULATION, TSS, WASTEWATER

## EQUATIONS TABLE

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

Eq. 1

Where  $C_0$  and  $C$  = concentrations of wastewater before and after EC in NTU or mg/L, respectively.

$$\frac{C}{C_0} = e^{-k_2 t}$$

Eq. 2

Where  $C$ ,  $C_0$ , and  $k_2$  = wastewater concentrations after EC, initial, and kinetic constant, respectively.

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

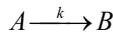
Eq. 3

Where  $k$  = kinetic constant  
 $a$  and  $b$  = constant values

$$\frac{dC_A}{dt} = \pm k C_A^a C_B^b \dots C_N^n$$

Eq. 4

Where  $k$  = the rate constant  
 $C_A$  = the concentration of substance A, etc.  
 $T$  = time



$$\frac{dC_A}{dt} = -k C_A$$

Eq. 5

$$\frac{dC_B}{dt} = k C_A$$

Eq. 6

$$\int_{C_{A0}}^{C_A} \frac{dC_A}{dt} = -k \int_0^t dt$$

$$\ln \frac{C_A}{C_{A0}} = -k t$$

Eq. 7

$$C_A = C_{A0} \cdot e^{-kt}$$

Eq. 8

$$\frac{dC_B}{dt} = k C_A = k C_{A0} \cdot e^{-kt}$$

Eq. 9

$$B = f(t) + f(Tr)$$

$$B = B_1 + B_2$$

Eq. 10

$$dB = \frac{dB}{dt} dt + \frac{dB}{dTr} dTr$$

Eq. 11

$$B_1 \rightarrow dB = \frac{dB}{dt} dt = k C_{A0} \cdot e^{-kt} dt$$

$$\int_0^{B_1} dB = \int_0^t k \cdot C_{A0} (e^{-kt}) dt = C_{A0} (1 - e^{-kt})$$

Eq. 12

$$B_2 \rightarrow dB = \frac{dB}{dTr} dTr = b dTr$$

$$\int_0^{B_2} dB = \int b dTr = b(Tr) + C$$

Eq. 13

$$B = C_{A0} (1 - e^{-kt}) + b \cdot Tr + C$$

Eq. 14

$$C = B_0 - C_{A0}$$

Eq. 15

$$B\infty = C_{A0} + b \cdot Tr + C$$

Eq. 16

$$B\infty = C_{A0} + b \cdot Tr + B_0 - C_{A0}$$

$$b = \frac{B\infty - B_0}{Tr}$$

Eq. 17

$$B = C_{A0} (1 - e^{-kt}) + \left[ \frac{B\infty - B_0}{Tr} \right] Tr + B_0 - C_{A0}$$

$$B = C_{A0} (1 - e^{-kt}) + B\infty - C_{A0}$$

Eq. 18

Where:  $B\infty = n$   
 $C_{A0}$ ;  $n < 1$

$$B = C_{A0} (n - e^{-kt})$$

Eq. 19

$$B = \frac{C_A}{a}$$

Eq. 20

$$\frac{C_A}{a} = C_{A0} (n - e^{-kt})$$

$$\frac{C_A}{C_{A0}} = a(n - e^{-kt})$$

Eq. 21

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

Eq. 22

Where  $C_0$  = initial concentration  
 $C$  = concentration after treatment/sedimentation  
 $a$  and  $b$  = constant values  
 $k$  = the rate constant  
 $t$  = time