

# Batch electrocoagulation system using aluminum and stainless steel 316 plates for hospital wastewater treatment

*by* Ratnawati Ratnawati

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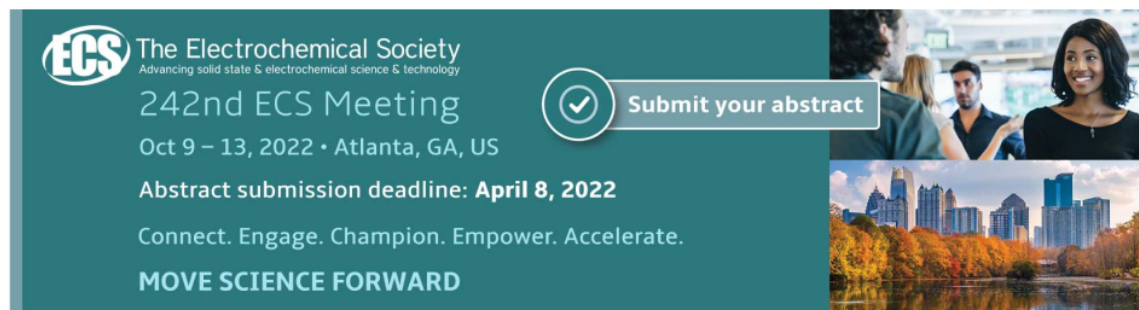
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## Batch electrocoagulation system using aluminum and stainless steel 316 plates for hospital wastewater treatment

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**Abstract.** Hospital wastewater is one of the most dangerous types of pollution that contaminates rivers due to the presence of pharmaceutical compounds and dyes. Antibiotic ciprofloxacin and methylene blue dyes, a hospital wastewater model, were investigated using the electrocoagulation method. This study aims to determine the optimum initial pH and time in electrolysis using the electrocoagulation method in a batch reactor system. The electrode that used is aluminum and stainless steel 316 plates. The DC power supply was set at 50 volts. Initial pH variations were 5, 7, 10, and contact time intervals were 1, 2, 3, and 4 hours. The results showed that the optimum conditions were obtained at the initial pH of 10 with a processing time of 4 hours. This condition has significant efficiency in pollutant removal from wastewater with the highest percentage of 84.60% and 68.19% for methylene blue and ciprofloxacin, respectively. A precipitated of  $Al(OH)_3$  coagulant was obtained as much as 4.77 grams. The findings in this research would be helpful for the removal of organic pollutants simultaneously in complex wastewater.

### 1. Introduction

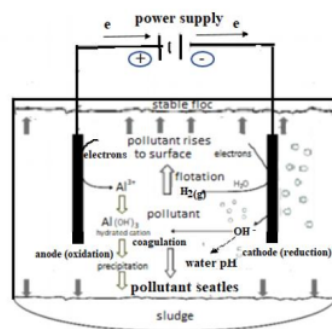
Hospital wastewater is one of the most dangerous types of pollution which contaminate river waterways. Pharmaceutical compounds and dyes are some of the pollutants that contaminate complex organic wastewater. High concentrations of the antibiotic ciprofloxacin have been reported in hospital effluents [1]. Ciprofloxacin is a fluoroquinolones (FQs) family of antibiotics that has a broad spectrum because it works to inhibit or even kill pathogens [2]. Besides, methylene blue is also widely used in hospital activities such as diagnosis, antiseptic for surgery, and methemoglobinemia [3]. The presence of antibiotics and dyes in the water is hazardous since it disrupts the balance of aquatic ecosystems and has an impact on damaging the effectiveness of bacterial colonies that can decompose various substances [4]. According to the regulation of the Minister of Environment and Forestry No P.68/MENLHK/Setjen/Kum.1/8/2016 about the organic waste quality standards, the maximum concentration of methylene blue active substances (MBAS) in hospital wastewater is 5 mg/L. Unfortunately, until now, there is no standard regulation regarding the content of ciprofloxacin in wastewater. As a consequence, not many research about wastewater treatment on ciprofloxacin antibiotics is performed in Indonesia. However, since it is categorized as a harmful pollutant, it should be treated effectively.

Many efforts have been performed to treat the hospital wastewater, especially those containing methylene blue [5] and antibiotic ciprofloxacin [6,7] either as a single waste by electrocoagulation or a mixture of both by photocatalysis [4]. Unfortunately, treatment for this pollutant is not easy. Antibiotic



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and dye-containing effluent are toxic to the environment since dyes are stable compounds with low biodegradability and can be carcinogenic mutagenic, or teratogenic [1,3]. Antibiotic and dye removal by conventional treatment methods (biological process) is limited and inadequate since the majority possess complex aromatic molecular structures, which resist biodegradation [8]. Recently, electrocoagulation is an alternative method widely used to treat wastewater with high effectiveness, simple equipment, easy to operate, and produces a coagulant that can adsorb pollutant which then forms sludge [9]. In this electrocoagulation process, the aluminum anode (Al) is oxidized to produce  $Al^{3+}$  ions, and it occurs on the anode surface with a certain area. Meanwhile, at the cathode, the reaction of  $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$  happens and the formation of  $H_2$  occurs at the cathode surface [9,10]. The mechanism of electrocoagulation is depicted in figure 1.



**Figure 1.** General mechanism of electrocoagulation.

The effectiveness of electrocoagulation is strongly influenced by operational parameters such as electric current density, initial pH, electrolysis time, voltage, plate thickness, and the distance between the electrodes [9,10]. In this study, the optimized parameters are initial pH and electrolysis time since pH adjustment is very important in the electrocoagulation process. Transfer of Al ions from the electrode (anode) to the solution causes the in-situ generation of coagulating agents by electro-oxidation of “sacrificial anodes” [11], and it is influenced by pH value. With the pH variation, the percentage efficiency of adsorption removal pollutant in solution is obtained. pH is very influential for aluminum species formed on the oxidation of aluminum at the anode. Electrolysis time will also determine the amount of  $Al^{3+}$  in the solution. In the current study, the application of the electrocoagulation process to treat the mixture of two types of pollutants (methylene blue dye and ciprofloxacin) simultaneously in wastewater has been rarely studied. In this study, the role of initial pH and electrolysis time in the electrocoagulation process was investigated.

## 2. Materials and method

### 2.1 Chemicals

Methylene blue dye with formula  $C_{16}H_{18}N_3SCl$  was obtained from the ROFA laboratory center, whereas antibiotic ciprofloxacin/CIP (generic tablets) were supplied by PT. Bernofarm. Meanwhile, nitric acid and sodium hydroxide purchased from Merck Company are used for pH adjustment.

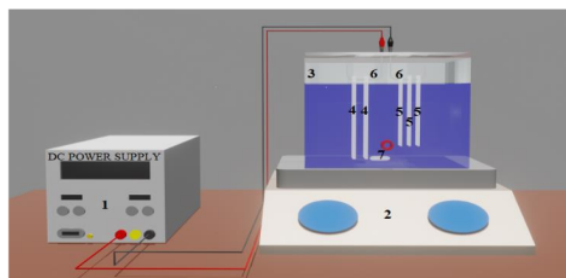
### 2.2 Experimental procedure

The electrocoagulation is carried out using a batch reactor system made of acrylic by the size of (16x8x11) cm. The electrocoagulation reactor with a volume of 1L mixed wastewater containing 10 mg/L ciprofloxacin and 10 mg/L methylene blue dye is equipped with a magnetic stirrer. Two aluminum plates sized (10x4x0.2) cm and three pieces of stainless steel 316 plates sized (8x2.5x0.1) cm act as an anode and cathode, respectively. The inter-electrodes distance was 1.5 cm. In this study, the electrodes were set in parallel connected to an electric current generating from a DC (Zhaoxin RXN-605D, 60V 5A) was set at a constant 50 V potential difference. At a time interval of 1, 2, 3, and 4 hours, the

concentrations of sample solutions were analyzed using a UV-Vis Spectrophotometer (Shimadzu UV 2450) with various initial pH of 5, 7, and 10. This equipment worked at the wavelength of 665 nm and 336 nm for absorbance measurement of methylene blue and ciprofloxacin, respectively. These wavelengths correspond to the maximum absorbance of the two pollutants. Percentage efficiency of adsorption removal is calculated with the formula as follows:

$$\% = \frac{C_0 - C_t}{C_0} \times 100\% \quad (1)$$

$C_0$  and  $C_t$  are the concentrations of methylene blue and ciprofloxacin solution at the beginning of the reaction and at a certain time, respectively. A schematic diagram of the reactor can be seen in figure 2 below:



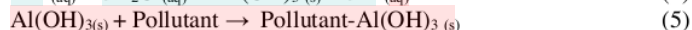
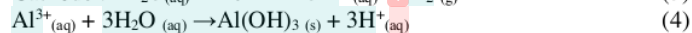
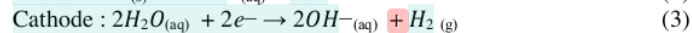
**Figure 2.** A Schematic diagram of the electrocoagulation reactor: (1) DC power supply, (2) magnetic stirrer, (3) acrylic reactor, (4) aluminum plates, (5) stainless steel 316 plates, (6) copper cable, (7) magnetic bars.

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### 3. Results and discussion

#### 3.1. Effect of initial pH and electrolysis time

In this study, Al electrodes are used with a larger size and more than one piece to produce coagulants in the form of  $Al^{3+}$  ions and much of  $OH^-$  ions which were able to bind pollutant ions more quickly. As a result, it would increase the efficiency of adsorption removal of methylene blue and ciprofloxacin with a longer contact time. According to (equation 2),  $Al^{3+}$  ions are produced from the aluminum plate, which is used as the anode [10]. This process is characterized by the appearance of pores on the aluminum plate after the process is completed. At the cathode, stainless steel 316 will produce hydroxide ions and hydrogen (equation 3). The presence of hydrogen at the cathode is indicated by the appearance of gas bubbles during the process so that the foam appears on the surface of the wastewater. The electron transfer affects the formation of flocs  $Al(OH)_3$ , which function to bind methylene blue and ciprofloxacin to become sludge. The process can be represented in the reactions as follows:



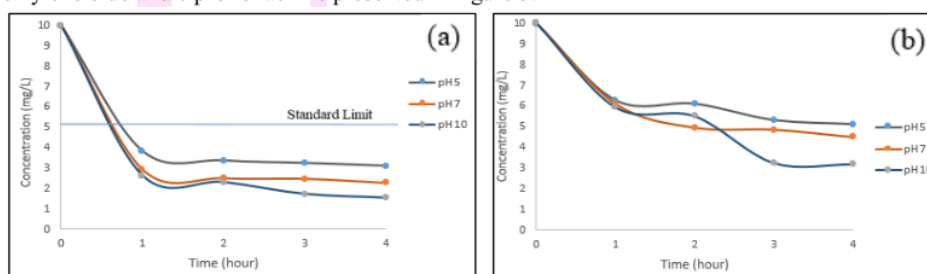
At the beginning of the electrocoagulation process, bubbles occur due to the formation of hydrogen, which is formed from the release of electrons from the cathode to the anode as a result of the electrocoagulation process. This condition causes the impurities formed to settle during the process, which precipitate the formed flocs [11,12]. While at the anode,  $Al^{3+}$  ions will be oxidized to form  $Al(OH)_3$ , which functions as a coagulant, and at the cathode, there is a reduction of cations (equation 4). The process takes 4 hours with pH variations at 5,7 and 10. The process of floc/sludge formation begins



with the release of electrons by the cathode so that  $\text{Al}(\text{OH})_3$  coagulant occurs, which will adsorb pollutants [13].

The  $\text{Al}(\text{OH})_3$  formed in the electrocoagulation process is a coagulant that functions to precipitate methylene blue and ciprofloxacin in the electrolyte solution. Coagulants that occur in the electrocoagulation process will cause the fast formation of precipitates, where the particles suspended in water have an electric charge on their surface due to the adsorption of ions ( $\text{OH}^-$ ) from the water. These ions surround the particle surface tightly and attract charged ions from the solution. The presence of attractive forces between particles (van der Waals forces) causes colloids to combine to form flocs [13].

At a high pH and length of time for electrolysis, the dissolution of aluminum at the anode increases, resulting in higher amounts of  $\text{Al}^{3+}$  and  $\text{Al}(\text{OH})_3$ . Increasing the concentration of  $\text{Al}^{3+}$  could raise the charge neutralization reaction of contaminants to form flocs. In addition, with increasing pH, the rate of formation of hydrogen bubbles at the cathode increases. The effect of initial pH in electrocoagulation of methylene blue and ciprofloxacin is presented in figure 3.



**Figure 3.** Effect of initial pH in electrocoagulation of (a) Methylene blue and (b) Ciprofloxacin with the initial concentration of 10 ppm.

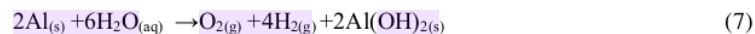
From figure 3, it can be seen that there is a significant effect of pH on the decreasing of methylene blue and ciprofloxacin concentration. At pH 10, there is better efficiency of methylene blue and ciprofloxacin removal in the electrocoagulation process than at pH 5 and 7. The higher the pH, the lower the pollutant concentration. From the electrolysis time point of view, there is little effect on the decreasing concentration of methyl blue after 1 hour of processing. It meant that after that time, the decrease in the methylene blue concentration had a little significant effect. In all pH and time variations, the final methylene blue concentration is in accordance with the quality standard (below 5 ppm). The same phenomenon also occurs in decreasing ciprofloxacin concentration. The higher the pH and the longer the processing time, the more the concentration of ciprofloxacin decreases. In this case, the magnitude of the pH value and the length of processing time have a significant effect on decreasing ciprofloxacin concentration.

According to Rakhi M.S. and Karoui et al. [11,14], pH solution can affect the reactions that occur during the electrocoagulation process. With increasing pH, the rate of pollutant removal increases since the effect of pH on the coagulant produced depends on the reactions as follows:

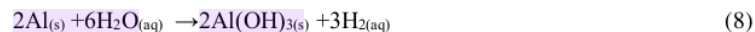
In neutral conditions:



In acid conditions:

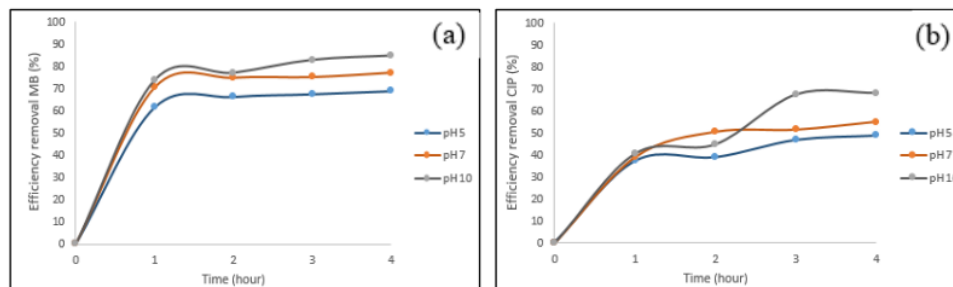


In alkali conditions:



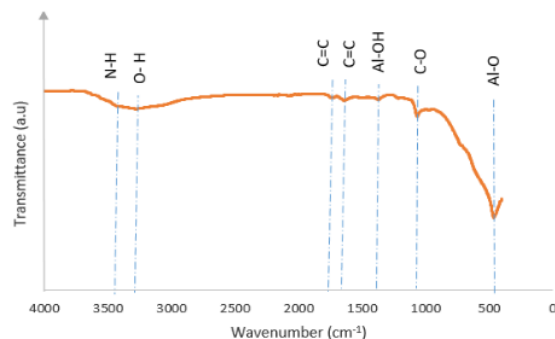
At pH 7, the reaction between Al and H<sub>2</sub>O ions will produce Al(OH)<sub>2</sub>, Al(OH)<sub>3</sub>, and H<sub>2</sub>. However, at pH 5, it produces Al(OH)<sub>2</sub>, H<sub>2</sub>, and O<sub>2</sub>, while at pH 10, it produces Al(OH)<sub>3</sub> and H<sub>2</sub>. As a result, the weight sludge of the precipitate Al(OH)<sub>n</sub> at pH 5, 7, and 10 were 0.001g, 0.0289g, and 4.77g, respectively. This result is directly proportional to the reduction in the pollutant concentration. Since Al(OH)<sub>3</sub> has a higher weight and density, therefore it settles faster with high efficiency of pollutant removal as it acts better in trapping pollutants to form a precipitate/sludge [14].

The removal efficiency of methylene blue and ciprofloxacin as a function of pH and time can be seen in figure 4. At 4 hours of electrolysis, the efficiency of adsorption removal is 84.60 % and 68.19% for methylene blue and ciprofloxacin, respectively.



**Figure 4.** Effect of pH and time on the removal efficiency of (a) Methylene blue (MB) and (b) Ciprofloxacin (CIP) with the initial concentration of 10 ppm.

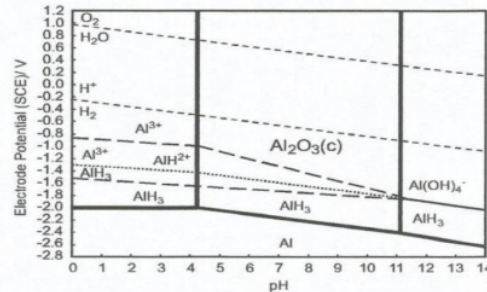
From figure 4, it can be stated that the longer the time, the higher the efficiency of pollutant removal until it reaches equilibrium or constant rate. At the beginning of the process, the removal efficiency of pollutant removal is very high due to the abundance of Al(OH)<sub>3</sub> coagulant. With increasing time, the removal efficiency of pollutant increases which is proportional to the availability of Al(OH)<sub>3</sub>, until it reaches an equilibrium condition.



**Figure 5.** Fourier transform infrared (FTIR) spectra of sludge.

The functional groups formed in the sludge that was identified by several peaks in the FTIR at the wavenumber of 490, 1350, and 3360 cm<sup>-1</sup> correspond to the stretching vibration of Al-O, Al-OH, and hydroxyl groups O-H, respectively [15]. These bonds indicate the presence of Al(OH)<sub>3</sub>. Peaks at around 1082 and 1700 – 1800 cm<sup>-1</sup> associated with the symmetric vibrations of C–O and C = C stretching of alkene, respectively, while FTIR spectra at around 3400 cm<sup>-1</sup> could confirm the symmetric vibration of N-H bonding. These peaks indicated the successful adsorption of methylene blue and ciprofloxacin onto the coagulant Al(OH)<sub>3</sub> [15].

According to Mochelhoff [16], aluminum is located in the zone of active dissolution at certain pH conditions. The role of initial pH is very influential for aluminum species formed on the oxidation of aluminum at the anode. The conditions that affect the formation of  $\text{Al}(\text{OH})_n$  are in accordance with the potential pH of  $\text{Al}-\text{H}_2\text{O}$  as shown in figure 6.



**Figure 6.** Aluminum E-pH (Pourbaix) diagram [16].

### 3.2 Overview of Kinetics on Electrocoagulation

The kinetic data obtained from this study were analyzed using a kinetic model for the first and second order. The general equation for the electrocoagulation kinetics of wastewater solution is as follows:

$$\frac{dC}{dt} = kC^n \quad (9)$$

First-order electrocoagulation kinetics can be calculated by integrating this equation with order  $n = 1$ , therefore equation (9) becomes:

$$\ln\left(\frac{C_t}{C_0}\right) = -k_1 t \quad (10)$$

Where  $C_0$  is the initial concentration (mg/L),  $C_t$  is the concentration remaining after processing for a time  $t$  (hours), and  $k_1$  is a kinetic constant for order 1 ( $\text{hours}^{-1}$ ). For second-order kinetics,  $k_2$  (L/mg.hours) is calculated by integrating equation (9) with  $n = 2$ , so that the result is as follows:

$$\left(\frac{1}{C_t} - \frac{1}{C_0}\right) = k_2 t \quad (11)$$

Plotting of equation 10 ( $-\ln C_t/C_0$  vs.  $t$ ) and equation 11 ( $1/C_t$  vs.  $t$ ) will get  $k_1$  dan  $k_2$  as slopes, and the result can be depicted in table 1. The  $k$  value is the adsorption rate constant and the  $R^2$  value is the correlation coefficient [17]. The greater the value of  $k$ , the faster the adsorption process so that the rate of pollutant removal is also greater. The high removal rate of pollutant result in high efficiency.

**Table 1.** Comparison of kinetic constants,  $k$  for the first and second-order reaction of methylene blue.

pH	first-order pseudo model		second-order pseudo model	
	$k_1$ ( $\text{hours}^{-1}$ )	$R^2$	$k_2$ (L/mg.hours)	$R^2$
5	0.2508	0,649	0.0493	0,730
7	0.3131	0,636	0.0743	0,733
10	0.4171	0,770	0.1304	0,928

**Table 2.** Comparison of kinetic constants,  $k$  for first and second-order reaction of ciprofloxacin.

pH	first-order pseudo model		second-order pseudo model	
	$k_1$ ( $\text{hours}^{-1}$ )	$R^2$	$k_2$ (L/mg.hours)	$R^2$
5	0.1499	0,791	0.0219	0,852
7	0.1829	0,793	0.0028	0,857
10	0.2895	0,921	0.0568	0,922



It can be concluded that at pH 10, the adsorption rate is highest compared to others due to the availability of a lot of  $Al^{3+}$  that reacts with  $OH^-$  to form a coagulant  $Al(OH)_3$  that can absorb many pollutants in wastewater. Based on the analysis of the adsorption kinetics data in table 1 and 2, it can be stated that the suitable model for adsorption kinetics is at pH 10 with second-order reaction ( $n=2$ ) since it gives the highest  $R^2$  which is close to 1 compared to adsorption kinetics model at initial pH 5 and 7.

#### 4. Conclusion

pH and electrocoagulation time affect the rate of methylene blue and ciprofloxacin removal from wastewater. At pH 10 and 4 hours process, the efficiency of adsorption removal of methylene blue and ciprofloxacin reach 84.60 % and 68.19%, respectively. For second-order pseudo model shows the highest correlation coefficient of nearly 1 at pH 10, and it is the best fit to the model. The initial pH of the solution can affect the amount of  $Al(OH)_3$  coagulant. The  $Al(OH)_3$  coagulant at pH 10 was obtained as much as 4.77 grams. In the future study, it is important to determine the addition of electrolyte solutions to obtain high effectiveness of the wastewater simultaneously.

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