Electrocoagulation Treatment of Sanitary Landfill Leachate in Malaysia

M R Taib, B N Mook, M I H M Tahir, M A A Aziz*

School of Chemical and Energy Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

*Corresponding author: m.arif@utm.my

Abstract. The tropical climate condition and high moisture of solid waste characteristic in Malaysia led to the increase of leachate generation from landfill. Current leachate treatment showed less efficient to treat leachate due to various refractory pollutants in the leachate. This study employed electrocoagulation (EC) to investigate the effects of electrode material, current density and initial pH condition in reducing total suspended solid (TSS), chemical oxygen demand (COD), biological oxygen demand (BOD₅), ammonia-nitrogen and colour from the leachate. The result showed that aluminium electrode was able to remove all the parameters more effectively as compared to iron electrode. The optimum current density and pH condition were found to be 100 mA/cm² and at pH 7, respectively. The COD in the leachate was predominantly exist as particulate matter and it was removed in the form of suspended solids. Although all the pollutant parameters still exceeded the standard discharge limit after the treatment, the electrocoagulation is proposed to act as pretreatment prior to biological treatment in sanitary landfill. The electrocoagulation also can treat leachate from open dumping landfill sites in order to reduce the leachate contamination into the environment.

1. Introduction
Malaysia generated more than 38,000 tonne of solid waste per day in which 89% of the solid waste collected were ended up in landfill. Malaysia has 165 operating landfill with only 8 considered as sanitary landfill equipped with leachate treatment system. This resulted to the continuous generation of untreated leachate which represent a threat to the environment. In addition, current leachate treatments were still not efficient to treat leachate due to the promulgation of a new leachate regulation in year 2009. Landfill leachate is a very dark coloured liquid formed by the percolation of precipitation through the solid waste in landfills. The risk of untreated leachate generation in Malaysia is serious due to its tropical climate condition and unique waste characteristics such as high load of organic waste. Therefore, a suitable treatment is needed in order to reduce the leachate concentration from closed and non-sanitary landfill and also to improve the leachate treatment efficiency in sanitary landfill. Electrocoagulation is an electrochemical process which able to generate in-situ coagulants from the dissolution of sacrificial anode. Comparing to conventional coagulation-flocculation treatment, the operation of electrocoagulation is easier to operate which requires no continuous input of coagulant and competent operator. This paper discussed the effectiveness of electrocoagulation to treat leachate in Malaysia.
Leachate is a liquid that percolated through the solid waste in landfills. It is generated from liquids present in the waste and from outside water, including rainwater [1]. Leachates composition consisted of various types of chemicals. The composition of leachate is characterized by the age of landfill, moisture content of landfill and waste composition [2, 3]. The generation of leachates depends on the water balance on the landfill site. The water balance includes the precipitation (i.e rainfall), evaporation, surface runoff, moisture content of deposited waste, microbial activity in the landfill and leachate generation [4]. High rate of rainfall which infiltrates through the landfill will increase leachate generation.

Precipitation and evaporation are governed by climate factor. Malaysia is located in tropical climate that is being hot and humid throughout the year. This climate condition has encouraging to the high generation of leachate. Leachate contains contaminant with high value of chemical oxygen demand (COD), biological oxygen demand ($\text{BOD}_5$), ammonia-nitrogen, total suspended solids, heavy metals and color. The typical composition of leachate in Malaysia were mostly comes from organic waste. Around 50% of the total solid waste is organic matter [5]. This high load of organic matter increased the generation of leachate which caused environmental pollution if it were not treated properly. Source of leachates were largely generated from dumping sites. Although the dumping sites were closed, the generation of leachates were still active. The absence of proper treatment in the closed landfills were led to uncontrolled leachate generation which threat to the environment. In addition, current leachate treatment facilities in sanitary landfill were less efficient to treat leachate due to the promulgation of a new leachate regulation in year 2009.

Leachate treatment methods depend on the type of leachate characteristics. According to the landfill age, the leachates can be classified into three types: young, intermediate and old. Biological treatments such as aerobic, anaerobic, sequencing batch reactor, aerated lagoons, and activated sludge are effective in removing organic matters in the young leachate [6, 7]. On the contrary, biological treatment is inefficient when it comes to the treatment of old leachate. This is due to the presence of refractory organic compound such as non-biodegradable organic compound, substances toxic for the microorganism and the limited availability of necessary nutrients for microbial growth [8].

Prior to the biological treatment, a suitable pretreatment can be employed to reduce leachate concentration. Physico-chemical processes can be applied to treat refractory organic compounds in especially in old leachate [9]. Coagulation-flocculation is commonly used method as a pretreatment for leachate. However, coagulation-flocculation method has some drawbacks such as high maintenance, requires continuous input of chemical coagulants and requires a competent and full time operator. Compared with conventional coagulation-flocculation, electrocoagulation is not dependent on solid chemical coagulant.

Electrocoagulation does not require continuous input of coagulants. This technology is easy to operate, low maintenance and requires minimal supervision. Therefore, this method is more feasible to treat leachate compared to the conventional coagulation-flocculation method. This promising electrocoagulation method can improve the efficiency of leachate treatment by acting as pretreatment for current leachate treatment system in sanitary landfill. The method can also be used to treat leachate concentration from closed landfills. This reduces the impact of leachate contamination to the environment. The purpose of this research is to investigate the efficiency of electrocoagulation to treat leachate in Malaysia. The objectives are to determine the effect of electrode materials, initial pH condition and current density on the performance of leachate treatment.

2. Materials and methods
2.1. Description of landfill site
Leachate was sampled from a Sanitary Landfill which located in southern region of Malaysia. The landfill has been classified as level 4 which equipped with leachate treatment facility. The landfill has been operated since 2004 with lifespan of 20 years. The landfill area is 275 acres with 15 millions tonnes of landfill capacity. The landfill receives 1800 tonnes of solid waste daily and the capacity of leachate generation for treatment is 540 m$^3$ per day.
2.2. Leachate sampling and characterization

The leachate sampling for this study was carried out during normal day. During the sampling, a container with sealed cap was used for collecting the leachate sample using APHA Standard Methods. No bubbles were present in the sampling bottle. The pH of the sample was measured in situ using the pH probe, Hanna Model, No. 8033. The concentrations of chemical oxygen demand (COD), Total suspended solids (TSS) and ammonia-nitrogen were determined using the Standard Methods of APHA. The concentration of biological oxygen demand (BOD) was determined using BOD$_5$ test. The colour of sample was determined using HACH Method 10048. The concentration of particulate matter in the sample was obtained using filtration method. Table 1 showed the characteristics of raw leachate obtained with comparison to the Standard Emission Rate of the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation 2009 (PU(A) 433). The results clearly showed that the leachate sample had exceeded the standard discharge limit.

Table 1. The properties of leachate sample from a Malaysian Sanitary Landfill and Standard discharges limits.

| Analysis               | Value  | Standard Discharge Limits$^c$ |
|------------------------|--------|-----------------------------|
| pH                     | 3.9    | 6.0-9.0                     |
| TSS (mg/L)             | 1690   | 50                          |
| COD (mg/L)             | 10953, 1520$^a$ | 400                        |
| BOD$_5$ (mg/L)         | 3600   | 20                          |
| Ammonia-nitrogen (mg/L)| 384    | 5                           |
| Colour (ADMI)$^b$      | 2794   | 100                         |

$^a$ Determine using filtration method on raw leachate  
$^b$ American Dye Manufacturer’s Institute  
$^c$ Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation 2009 (PU(A) 433)

2.3. Electrocoagulation procedures

Electrocoagulation treatment of leachate was carried out in a batch mode. Figure 1 depicts the experimental setup of electrocoagulation in this study.

Figure 1. Experimental setup for electrocoagulation
The experimental setups for electrocoagulation were consisted of a DC power supply, magnetic stirrer and electrodes. 500 mL of leachate was placed in a glass jar with two electrodes dipped inside. The electrodes were made up of the same type of material with dimensions of 150 mm (height) x 50 mm (width) x 5 mm (thickness). The spacing between the electrodes was set at 50 mm and arranged in parallel. The electrodes were connected to the positive and negative pole of the DC power supply to serve as the cathode and anode. Anodes and cathodes were weighed before and after electrocoagulation treatment. The electrodes were also connected to voltmeter and ammeter. Aluminium and iron electrodes were used in this study. Slow mixing was provided using magnetic stirrer to ensure the homogeneous mixing of reactor content.

The electrocoagulation treatment was conducted for 30 minutes at room temperature for each trial. Initially, the study was performed by conducting the electrocoagulation using different electrodes, iron and aluminium electrode at initial pH 7 and current density of 20 mA/cm$^2$. Subsequently, electrocoagulation was carried at different current density ranging from 20, 40, 60, 80 and 100 mA/cm$^2$ using aluminium electrode and at initial pH of 7. Then, the electrocoagulation was conducted at different initial pH of 3, 5, 7 and 10. The initial pH was adjusted using hydrochloric acid and sodium hydroxide. After each run the electrodes were cleaned and rinsed with distilled water. At the end of each electrocoagulation treatment, a solution with flocks was allowed to settle for 60 mins in the container before chemical analysis.

3. Results and discussions
The electrocoagulation treatment was conducted using aluminium and iron electrode to remove the TSS, COD, BOD$_5$, ammonia-nitrogen and colour. The experiment was conducted for 30 minutes at 20 mA/cm$^2$ and initial pH of 7. Fig. 2 depicts the removal efficiency of pollutant parameters by aluminium and iron electrode. The result showed the superior removal of all pollutants by using aluminium electrode than iron electrode. Higher percentage of total suspended solids removal efficiency is observed for aluminium sacrificial anode (44.38%), while for iron sacrificial anode, it is merely 38.93%. The percentage removal efficiency of COD is 42.88 % and 40.82 % by aluminium and iron sacrificial anode, respectively. The percentage removal efficiency of BOD$_5$ by aluminium and iron sacrificial anode is 25.88 % and 23.89 % respectively. For ammonia-nitrogen, the removal efficiency by aluminium electrode is at 13.54% while the removal efficiency by iron electrode is only 11.72%. The percentage removal of colour by aluminium and iron electrode is 31.21% and 25.81%, respectively.

![Figure 2. The effect of Al- and Fe-electrode on the removal efficiency of the TSS, COD, BOD$_5$, NH$_3$-N and Colour. (Current density = 20 mA/cm$^2$, pH = 7). (A) before treatment, (B) After treatment.](image-url)
form metal hydroxide. The metal hydroxides were served as coagulant to coagulate the pollutants. The generation of metal cations showed an ability to neutralize negatively charged particles, which then encouraged to bind together to form aggregates of microflocs composed of a combination of colloidal particles and metal hydroxide. Through Van der Waals attraction, the microflocs were assembled or bind together to form a larger floc.

Iron sacrificial anode undergoes electrochemical dissolution to form $\text{Fe}^{2+}$ ions. The $\text{Fe}^{2+}$ species are oxidised into $\text{Fe}^{3+}$ in the presence of dissolved oxygen $[\text{10}]$. The $\text{Fe}^{3+}$ species formed is hydrolysed to form $\text{Fe(OH)}_3$. The anodic reactions for iron electrodes are shown as follows:

(1) At alkaline conditions:

$$\text{Fe}^0 - 2e^- \rightarrow \text{Fe}^{2+} \quad (5)$$
$$\text{Fe}^{2+} + \text{OH}^- \rightarrow \text{FeOH}^+ \quad (6)$$
$$\text{FeOH}^+ + \text{OH}^- \rightarrow \text{Fe(OH)}_2 \quad (7)$$

(2) At acidic conditions with the presence of dissolved oxygen:

$$4\text{Fe}^{2+} + \text{O}_2 + 4\text{H}^+ \rightarrow 4\text{Fe}^{3+} + 2\text{H}_2\text{O} \quad (8)$$
$$\text{Fe}^{3+} + 3\text{H}_2\text{O} \rightarrow \text{Fe(OH)}_3 + 3\text{H}^+ \quad (9)$$

The evolution of hydrogen takes place at the cathode electrode and the reaction is as shown below:

$$2\text{H}_2\text{O} + 2e^- \rightarrow \text{H}_2 + 2\text{OH}^- \quad (10)$$

In contrary to the iron sacrificial anode, the aluminium sacrificial anode dissolved to form $\text{Al}^{3+}$ directly according to the following reactions:

$$\text{Al}^0 - 3e^- \rightarrow \text{Al}^{3+} \quad (11)$$

The evolution of hydrogen and $\text{OH}^-$ ion takes place at the cathode electrode and the reaction is as followed:

$$2\text{H}_2\text{O} + 2e^- \rightarrow \text{H}_2 + 2\text{OH}^- \quad (12)$$

The aluminium ions released from the dissolution of sacrificial anodes were immediately reacted with $\text{OH}^-$ ions in water to form monomeric and polymeric hydrolyzed species.

$$\text{Al}^{3+} + \text{H}_2\text{O} \rightarrow \text{Al(OH)}_2^{2+} + \text{H}^+ \quad (13)$$
$$\text{Al(OH)}_2^{2+} + \text{H}_2\text{O} \rightarrow \text{Al(OH)}_3^{+} + \text{H}^+ \quad (14)$$
$$\text{Al(OH)}_3^{+} + \text{H}_2\text{O} \rightarrow \text{Al(OH)}_4^- + \text{H}^+ \quad (15)$$
$$\text{Al(OH)}_4^- + \text{H}_2\text{O} \rightarrow \text{Al(OH)}_5^- + \text{H}^+ \quad (16)$$

This direct dissolution of $\text{Al}^{3+}$ ions may be explained the superior removal of the pollutants by aluminium as sacrificial anode. The settleability of particle formed by $\text{Al(OH)}_3$ is seen better than that formed by $\text{Fe(OH)}_3$. Similar observation was also reported by Ilhan et al. in which the Al electrodes showed a higher treatment efficiency than Fe ones on the basis of COD removal $[\text{11}]$. In this study, the aluminium electrode is regarded as the optimum choice for leachate treatment.

As electrocoagulation removes pollutants in the form of particulate matters, the removal of dissolved matters of COD, BODs, $\text{NH}_3$-$\text{N}$ and colour were probably being removed by coprecipitation and/or adsorption by flocculants. For example, the humic substances which give a dark colour of leachate are probably being removed synergistically by coprecipitation and/or adsorption by flocculants ($\text{Al(OH)}_3$ or
Fe(OH)$_3$) using van der Waals forces [12]. However, in this study, the presence of COD was found mostly in the form of particulate matter. This was evidenced by filtration method on the raw leachate. The concentration of COD remained in the solution after filtration method was found to be 1520.28 mg/L (Table 1). The percentage of COD removed through filtration is 86.12%. Therefore, it can be deduced that the COD removed was predominantly exists in the form of particulate matter which can be retained as solid through filtration.

**Figure 3.** The effect of current density on the removal efficiency of TSS, COD, BOD$_5$, NH$_3$-N and Colour. (Al-electrode, pH = 7).

The current density is an important operating factor which determines the coagulant dosage [13]. In this study, the electrocoagulation was conducted at different current density of 20, 40, 60, 80 and 100 mA/cm$^2$ using aluminium electrode and at initial pH of 7. Fig. 3 depicts the removal efficiency of TSS, COD, BOD$_5$, ammonia-nitrogen and colour at different current density. From the result, it can be observed that the removal efficiencies of all pollutant parameters increased as the current density is increased. Among the pollutant parameters, BOD$_5$ has the most affected by the current density, while NH$_3$-N is the least affected. This is due to the generation of higher amount of metal cation at higher current density which lead to more coagulants formed from the dissolution of sacrificial anode. Subsequently, it can be observed that the production of floc increases. According to Faraday’s Law, the increment of current density leads to greater extent of anodic dissolution and the dosage of coagulants increase [14]. The weight loss for sacrificial anode is the highest when the current density is the greatest.

Figure 4 showed the effect of initial pH on the removal efficiency of TSS, COD, BOD$_5$, ammonia-nitrogen and colour through electrocoagulation. The initial pH was adjusted to 3, 5, 7 and 10 at constant current density of 20 mA/cm$^2$ and using aluminium as sacrificial anode. From the result, it shown that electrocoagulation process worked effectively at neutral pH in removal of all pollutants studied except ammonia-nitrogen. For example, the maximum removal of COD was observed 42.88 % at initial pH of 7. The drop of removal efficiency occurred when initial pH value tends towards acidic or basic values for all parameters studied except for ammonia-nitrogen which showed a slight increase in removal at initial pH of 10.

In general, electrocoagulation is highly pH dependant [15]. The pH influences the nature of produced polymeric metal species that will be formed as soon as the metal coagulants are dissolved in water. The monomeric species accommodates Al(OH)$_3$$^{2+}$, Al$_2$(OH)$_5$$^{4+}$ and Al(OH)$^{3+}$. The polymeric species includes Al$_6$(OH)$_{15}$$^{3+}$, Al$_1$(OH)$_7$$^{2+}$, Al$_8$(OH)$_{26}$$^{4+}$, Al$_3$(OH)$_{23}$$^{4+}$ and Al$_{13}$(OH)$_{34}$$^{5+}$ (Rebhun, 1993). The influence of pH on electrocoagulation may be considered as a balance of two competitive forces: (1) between H$^+$
and metal hydrolysis products for interaction with organic ligands, and (2) between hydroxide ions and organic anions for interaction with metal hydrolysis products [16].

![Figure 4](image-url)

**Figure 4.** The effect of initial pH (3, 5, 7 and 10) on the removal efficiency of TSS, COD, BOD$_5$, NH$_3$-N and Colour. (Al-electrode, Current density = 20 mA/cm$^2$).

In this study, at initial pH of 3, the removal efficiency of TSS, COD, BOD$_5$, ammonia-nitrogen and colours is low. At low pH, the aqueous complex, Al(H$_2$O)$_6$$_{3+}$ and Al$_3^+$ are more dominant and the concentration of insoluble precipitates are low [17]. This is due to the competitive forces between H$^+$ and the aqueous complex with organic ligand. Thus, the coagulation effect is become less insignificant. The removal efficiency of the parameters increases when the initial pH is closer to the neutral pH of 7. At basic initial pH of 10, the aqueous complex, Al(OH)$_4^-$ is dominant due to high concentration of OH$^-$ in the solution. The Al(OH)$_4^-$ complex does not coagulate with pollutants except ammonia nitrogen. When the concentration of OH$^-$ increases, the removal efficiency of ammonia-nitrogen increases. The removal efficiency of TSS, COD, BOD$_5$ and colours at initial pH of 10 is low except for ammonia-nitrogen. At initial pH of 7, the removal efficiency of TSS, COD, BOD$_5$ and colour is the highest compared to initial pH of 3, 5 and 10. At this pH condition, the Al metal cation hydrolyzed to form myriad monomeric species and polymeric species. This is due to the less competitive of H$^+$ (low pH) and hydroxide ions (high pH) during the formation of the monomeric species and polymeric species. Due to the acidic nature of leachate sampled in this study, it is needed to regulate the initial pH of raw leachate to neutral pH prior to the treatment. This is to increase the removal efficiency of the pollutants.

4. Conclusions

The pollutant parameters of TSS, COD, BOD$_5$, ammonia-nitrogen and colour in the leachate in Malaysia had exceeded the standard discharge limit of the Environmental Quality (Control of Pollution From Solid Waste Transfer Station and Landfill) Regulation 2009. Treatment is required for the leachate prior discharge to the surrounding. In this study, electrocoagulation was employed as a primary treatment to treat the leachate. Aluminium electrode used in the electrocoagulation process is able to remove the selected parameters of TSS, COD, BOD$_5$, ammonia-nitrogen and colour more effectively as compared to iron electrode. The removal efficiency of TSS, COD, BOD$_5$, ammonia-nitrogen and colour are greater when the current density is higher. At current density of 100 mA/cm$^2$, the removal efficiency of TSS, COD, BOD$_5$, ammonia-nitrogen and colour is the highest as compared to the lower current density. The optimum initial pH to conduct electrocoagulation is at pH 7 except for ammonia-nitrogen. The COD in the leachate was predominantly exist as particulate matter. Electrocoagulation removed the COD in the form of suspended solids from leachate. Although the pollutant parameters still exceeded the standard
discharge limit after the treatment, the electrocoagulation is proposed to act as pretreatment prior to biological treatment in sanitary landfill. The electrocoagulation also can treat leachate from open dumping landfill sites in order to reduce the leachate contamination into the environment.

Acknowledgments
This work was supported by the Universiti Teknologi Malaysia (Grant No. 16J64)

References
[1] Jayawardhana Y, Kumarathilaka P, Herath I and Vithanage M 2016 Environmental Materials and Waste Municipal Solid Waste Biochar for Prevention of Pollution From Landfill Leachate Academic Press chapter 6 pp 117-48
[2] Moody C M and Townsend T G 2017 Waste Manage. 63 267
[3] Naveen B P, Mahapatra D M, Sitharam T G, Sivapullaiah P V and Ramachandra T V 2017 Environ. Pollut. 220 1
[4] Bengtsson L, Bendz D, Hogland W, Rosqvist H and Åkesson M 1994 J. Hydrol. 158 203
[5] Jalil M A 2010 J. Sustain. Dev. 3 91
[6] Baiju A, Gandhimathi R, Ramesh S T and Nidheesh P V 2018 J. Environ. Manage. 210 328
[7] Miao L, Yang G, Tao T and Peng Y 2019 J. Environ. Manage. 235 178
[8] Iskander S M, Zhao R, Pathak A, Gupta A, Pruden A, Novak J T and He Z 2018 Water Res. 145 297
[9] Kamaruddin M A, Yusoff M S, Aziz H A and Hung Y T 2015 Appl. Water Sci. 5 113
[10] Li X, Song J, Guo J, Wang Z and Feng Q 2011 Procedia Environ. Sci. 10 1159
[11] Ilhan F, Kurt U, Apaydin O and Gonullu M T 2008 J. Hazard Mater. 154 381
[12] Moussavi G, Khosravi R and Farzadka M 2011 Desalination 278 288
[13] Holt P K, Barton G W and Mitchell C A 2005 Chemosphere 59 355
[14] Schneider M, Simunkova L, Manko M and Lohrenge M M 2019 J. Solid State Electrochem. 23 345
[15] Hashim K S, Khaddar R A, Jasim N, Shaw A, Phipps D, Kot P, Pedrola M O, Alattabi A W, Abdulredha M and Alawsh R 2019 Sep. Purif. Technol. 210 135
[16] Stephenson R J and Duff S J B 1996 Water Res. 30 781
[17] Un U T, Koparal A S and Ogutveren U B 2009 J. Environ. Manage. 90 428