UNIVERSITI PUTRA MALAYSIA

ELECTROCOAGULATION FOR REMOVAL OF CHEMICAL OXYGEN DEMAND AND HEAVY METALS IN LEACHATE FROM JERAM SANITARY LANDFILL, MALAYSIA

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By

ROSIE JOTIN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

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June 2013

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The increasing amount of solid waste generation in Malaysia has become a problem in Solid Waste Management (SWM), since it creates serious impact to the environment especially on the landfill site. The generation of leachate during landfilling operation may impair environment by contaminating surface and ground water system either directly or indirectly from the site. Eventhough at a present most sanitary landfill is equipt with high level of leachate treatment facilities, leachate is still considered as a significant source of environmental pollution because their constituent that are difficult to be treated successfully. Therefore, innovative, cheap and effective techniques of purifying landfill leachate are needed before the leachate being discharged into water systems. The present study was conducted to investigate the efficiency of electrocoagulation processes in the removal of Chemical Oxygen Demand (COD) and heavy metals (Zn and Cu) from Jeram Sanitary Landfill leachate influenced by several parameters, thus proposing the optimum condition for pollutants removal. The electrocoagulation reaction was conducted in a batch reactor using aluminum as sacrificial electrodes. The effecting parameters such as applied

voltage, conductivity and initial pH were studied to determine the condition of optimum removal. The variation of conductivity, pH and temperature also were noted during the electrocoagulation process. In this process, samples of 25 mL were taken out from the beaker at 10, 20, 40, 60, 80 and 100 minutes of electrolysis time. From the results obtained, the optimum operation conditions of the electrocoagulation process for the removal of COD, Zn and Cu was found to be as follows: applied voltage of 10 V (j=0.0207 mA/cm^2), conductivity of 28.00 mS/cm or lower, initial pH of 6 and 100 minuntes of electrolysis time. At these conditions, approximately, 61.86 % COD, 74.41 % Zn and 97.47 % Cu removals can be reached. In this study, the electrocoagulation process has proved to be effective in the removal of COD and heavy metals from sanitary landfill leachate.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

ELEKTROKOAGULASI DALAM MENYIKIRKAN PERMINTAAN OKSIGEN KIMIA DAN LOGAM BERAT DI DALAM AIR LARUT RESAP DARIPADA TAPAK PELUPUSAN SANITARI JERAM, MALAYSIA

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Peningkatan jumlah penjanaan sisa pepejal di Malaysia telah menjadi satu masalah dalam Pengurusan Sisa Pepejal (SWM), kerana ia memberikan impak yang serius kepada alam sekitar terutamanya di tapak pelupusan sampah. Penghasilan air larut resap semasa tapak pelupusan beroperasi boleh menjejaskan alam sekitar dengan mencemarkan sistem air permukaan dan bawah tanah sama ada secara langsung atau tidak langsung daripada tapak pelupusan tersebut. Walaupun tapak pelupusan sanitari telah dilengkapkan dengan kemudahan rawatan air larut resap yang tinggi, namun air larut resap masih dianggap sebagai sumber penting bagi pencemaran alam sekitar kerana kandungan kimianya yang sukar untuk dirawat dengan berkesan. Oleh itu, teknik yang inovatif, murah dan berkesan dalam pembersihan air larut resap di tapak pelupusan sampah diperlukan sebelum larut resap tersebut dilepaskan ke dalam sistem air. Kajian ini telah dijalankan untuk mengkaji keberkesanan proses elektrokogulasi dalam penyingkiran Permintaan Oksigen Kimia (COD) dan logam berat (Zn dan Cu) dari air larut resap tapak pelupusan sampah sanitari Jeram serta mengkaji pengaruh beberapa parameter, sekaligus mencadangkan keadaan optimum

untuk penyingkiran bahan pencemar. Teknik elektrokoagulasi telah dijalankan dalam reaktor kelompok dengan menggunakan elektrod aluminium sebagai elektrod yang terkorban. Kesan beberapa parameter mempengaruhi seperti voltan digunakan, kekonduksian dan pH awal telah dikaji bagi menentukan pencapaian penyingkiran yang lebih tinggi. Perubahan kekonduksian, pH dan suhu juga telah diambil kira semasa proses elektrokoagulasi. Dalam proses ini, 25 mL sampel telah diambil daripada bikar pada tempoh 10, 20, 40, 60, 80 dan 100 minit selepas elektrolisis bermula. Daripada keputusan yang diperolehi, operasi optimum bagi proses elekrokoagulasi untuk penyingkiran COD, Zn dan Cu telah didapati seperti berikut: voltan antara elektrod 10V (j=0.0207 mA/cm^2), konduktiviti 28.00 mS/cm atau lebih rendah, pH awal 6 dan 100 minit masa elektrolisis. Pada keadaan ini, kira-kira, 61.86% COD, 74.41% Zn dan 97.47% Cu penyingkiran boleh dicapai. Dalam kajian ini, proses EC telah terbukti berkesan dan mampu dalam menurunkan COD dan logam berat dalam air larut resap.

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LIST OF ABBREVIATIONS

AAS	Atomic Absorption Spectrometer
Al	Aluminum
$Al(OH)_3$	Aluminum Hydroxide
Al^{3+}	Aluminum Ion
BOD	Biochemical Oxygen Demand
Cd	Cadmium
CI	Chlorine
CI^-	Chloride Ion
COD	Chemical Oxygen Demand
Cu	Copper
DO	Dissolve Oxygen
EC	Electrocoagulation
g	Gram
H_2O	Water
H_2	Hydrogen Gas
H^+	Hydrogen Ion
j	Current Density
mA/cm^2	MilliAmpere per square centimeter
mg/L	Milligram per litter
mS/cm	MilliSiemens per centimeter
MHLG	Ministry of Housing and Local Government
MSW	Municipal Solid Waste
NH_3-N	Ammonia-Nitrogen

Ni	Nickel
nm	Nanometer
O_2	Oxygen Gas
OH^-	Hydroxide Ion
Pb	Lead
ppm	Parts per million
SS	Suspended Solid
SWM	Solid Waste Management
TDS	Total Dissolved Solid
TKN	Total Kjeldahl Nitrogen
TOC	Total Organic Carbon
TSS	Total suspended solid
V	Volt
Zn	Zinc
%	Percentage
^0C	Degree Celsius

CHAPTER 1

INTRODUCTION

1.1 Overview

Since independence, Malaysia has developed on a huge scale to achieve high quality of life and well-being of their population. However, increasing demand in quality life and current rate of economic development had resulted in the increase production of solid waste per capita. Furthermore, as a developing country, more industrialization largely was built in Malaysia. This had caused a tremendous generation rate of municipal solid waste (MSW) across the Malaysia municipalities (Tarmudi *et al.*, 2009). These phenomena had resulted in a crisis in solid waste management (SWM) because the land for waste disposal is becoming scarcer. This problem will continue at the waste disposal sites which lead to more pollution that are difficult to control, thus effecting the ecosystem of the environment.

Sanitary landfilling is the most common methods for disposal of municipal solid wastes in many countries around the world including Malaysia. This method has been adopted replacing the old open dumpsite technique, which was practiced before. Besides integrated by resource recovery, the sanitary landfill site was also equipped with higher level of treatment facilities to stabilize waste in a more effective manner. However, in sanitary landfilling, leachate is still become a big problem in MSW since the leachate compounds are difficult to treat to reach the acceptable level before being discharged. In addition, the landfill leachate may cause serious

pollutions to the surroundings since the leachate is characterized as high-strength wastewater exhibiting acute and chronic toxicity (Deng and Englehardt, 2006).

Contreras *et al.* (2009) has define a landfill leachate as a liquid that were generated by the precipitation which percolates through the waste layers deposited in a landfill site and gains dissolved and suspended components from the biodegrading waste through several physical and chemical reactions. Furthermore, Moreas and Bertazzoli (2005) emphasized that leachate derived from landfill is a dark grey and foul smelling solution. Besides containing a large amount of biodegradable compound, landfill leachate also, contain resistant organic matter (*eg.* BOD, TOC, COD), heavy metal, ammonium, chlorinated compounds and inorganic salts (Christensen *et al.*, 2001; Aziz *et al.*, 2004; Renou *et al.*, 2008; Pi *et al.*, 2009). The elements of heavy metal that commonly found in landfill leachate which are in high concentration include iron, manganese, zinc, chromium, lead, copper and cadmium (Aziz *et al.*, 2004).

Usually, the treatment system for landfill leachate is provided after leachates passes through the leachate production control in the purpose to minimize the volumes of leachate as well as reduce the cost of treatment. Due to the different requirements in a different country, leachates containing high concentration of organic and inorganic compounds have been treated with different standard and guidelines. Abbas *et al.* (2009) had classified landfill leachate treatments into four major groups. There are (1) Leachate Channelling (Combined Treatment with domestic sewage and Recycling), (2) Biological Processing (Aerobic and Anaerobic), (3) Chemical/Physical Treatment (Flotation, Coagulation/Flocculation, Chemical

Precipitation, Adsorption, Ammonium stripping, Chemical Oxidation, Ion exchange and Electrochemical) and (4) Membrane filtration (Microfiltration, Ultra-filtration, Nano-filtration and Reverse osmosis). According to Chen *et al.* (2004), during the past two decades, electrochemical technologies have regained their importance worldwide as well as engineering deposition technology for metal recoveries and for treating various wastewaters that contain high concentrations of pollutants. This is in line with the increase standard of drinking water supply and the stringent environmental regulations on wastewater discharge. However, Aziz *et al.* (2004) has informed that the removal of metals element from landfill leachate is not well documented, especially in Malaysia.

According to Emamjomeh and Sivakumar (2009), electrocoagulation is an electrochemical wastewater treatment technology that has shown its efficacy for the removal of metals, suspended particles, clay minerals, organic dyes, and oil and greases from a variety of industrial effluents through the highly charged polymeric metal hydroxide species in solution. In the present work, a series of experiments were conducted to investigate the efficiency of EC processes for the removal of COD and heavy metals concentration from the landfill leachate influenced by several parameters, thus propose the optimal operating conditions of the EC method for the greatest pollutants removal. The parameters include the applied voltage, conductivity and initial pH of wastewater. The effect of EC on the variation of conductivity, initial pH and temperature value were also observed. The experiments were performed in batch reactor by using aluminium as a selective electrode.

1.2 Problem Statement

As discussed before, the economic advancement, urbanization, rapid population growth and increase in living standards are the factors that contribute to the increasing of municipal solid waste (MSW) generation in Malaysia. However, according to MHLG (2006), the population growth is identified as one of the main source of solid waste generation in Malaysia. Usually, the increasing in population is caused by the migration of population from rural to urban areas due to lack of resources in rural areas and employment opportunities in urban areas. According to Tarmudi *et al.* (2009), the annual total population in Malaysia increased from 23.49 million in 2000 to 26.75 million in 2005, which recorded about 5.9 million tonnes of municipal solid wastes generated in 2005. In addition, the MHLG (2006) estimated that the amount of waste generation would increase to 31,500 tonnes per day, which equivalent to about 11.5 million tonnes per year in the year 2020.

This circumstance is further worsened by the uncontrolled dumping of wastes in dumpsite area especially in the squatter areas and in slum areas, which happened mostly in open spaces, access roads and watercourses thus extends the problems to public health (Latifah *et al.*, 2009). The impacts of increasingly amount of solid waste in Malaysia also enhanced the problem in the landfill site where the leachate and gasses emissions will be generated to the surroundings. The situations may become even worse when facilities in the landfill not properly designed and well managed which not meet the landfill standard. Umar *et al.* (2010) stated that most of the landfills in developing countries including Malaysia are not integrated with proper leachate collection system. According to Suratman *et al.* (2011), in Selangor,

most of the landfills were built and being operated without proper monitoring facilities and pollution controls such as liner materials, leachate collection and treatment ponds, methane gas ventilation pipes and groundwater monitoring wells.

Hui and Shu (2010) reported that landfill leachate has been characterized as a complicated wastewater where it contains not only the concentrated organic substances but also large amount of harmful inorganic substances; heavy metal is one of the harmful substances and has been proven that may lead to secondary pollution. The composition of leachate in landfill was also studied by other researchers (Robinson, 2007; Kjeldsen *et al.*, 2002). Robinson (2007) had predicted the quality of leachate during acetogenic phase and methanogenic phase at very large landfill including Malaysia. According to Robinson (2007), the maximum of COD, Zn and Cu concentrations in acetogenic leachate can reach as higher as 50,000 mg/l, 20,000 µg/l and 100 µg/l, respectively, whereas, in methanogenic leachate, the concentration were predicted to be about 6000 mg/l, 50 µg/l and 2000 µg/l, respectively.

From the above studies, it shows that the quality of leachate produced in different landfill ages would cause high pollutions at the sites. It was also proved by Lee *et al.* (2010) that leachate leaching out from sanitary landfill site would pose various pollutants to the environment. Generally, the major potential environmental impacts that related to the landfill leachate are pollution of groundwater and surface waters (Kjeldsen *et al.*, 2002). Within the context of leachate pollution, researchers search for new treatment process to achieve zero discharge for the environment and also reduce the cost of treatment facilities. Usually, combinations of different treatment processes are required to enhance the effectiveness of pollutants removal from

landfill leachate such as biological and physico-chemical treatment processes. However, due to the increasingly restrictive limits for the wastewaters discharge, researchers today are still trying to find a better landfill leachate treatment method at lower cost for landfill leachate thus helps to preserve the environment for the future.

In the recent decade, the treatment of wastewater by electrocoagulation process has been increasingly practiced in South America and Europe for treatment of many types of wastewater effluent (Feng, 2007). In Malaysia, electrocoagulation technique is not well defined as it is considered new method for the treatment of wastewater especially for the treatment of landfill leachate. However, there have been several studies and attempts to investigate the efficiency of electrocoagulation technique for the wastewater treatment such as palm oil mill effluent (Zamawi *et al.*, 2013), landfill leachate (Masoumeh *et al.*, 2012), river water (Mohanty *et al.*, 2012) and synthetic wastewater (Ni'am *et al.*, 2007). Overall, electrocoagulation can be effectively removes a variety of unwanted dissolved particles and suspended matter from an aqueous solution only by electrolysis (Emamjomeh and Sivakumar, 2009). The study done by Zamawi *et al.* (2013) showed that the electrocoagulation treatment method potentially reduces COD, SS and color from palm oil mill effluent using aluminium up to 72.75 %, 96.93 % and 92.79 %, respectively with the optimum operating conditions of current density of 80 A/m^2, retention time of 15 minutes and initial pH of 4.

Another study done by (Chitra and Balasubramanian, 2010) revealed the efficiency of electrocoagulation on the removal of COD from Acid Blue 113 dyes effluent used in textile industry. By depending on several parameters such as applied current,

electrolyte pH and initial effluent concentration, the electrocoagulation process can remove the COD from dyes in textile effluent more than 90% of removal efficiency. The study of the effect of electrical potential, initial pH, pollutant concentration and contact time on electrocoagulation process for the removal of zinc and copper from synthetic wastewater was conducted by Nouri *et al.* (2010). From the result obtained, it shown that the most effective removal rates of study metals could be achieved at 40 V of electrical potential, which reduced more than 98 % of zinc and copper concentration. Nouri *et al.* (2010) also concluded that electrocoagulation technique is an efficient, reliable, safe, and cost effective technique for removal of zinc and copper from industrial effluent such as plating baths wastewater. Therefore, the focus of this study is to investigate the efficiency of electrocoagulation process for the removal of COD, Zn and Cu from sanitary landfill leachate using aluminium electrodes with different effecting parameters such as applied voltage, initial pH, conductivity and operating time.

1.3 Research Objectives

The general objective of this study is to investigate the efficiency of electrocoagulation for removal of COD and heavy metals in sanitary landfill leachate.

The specific objectives of this present work are:-

(i). to investigate characteristics of raw leachate generated from Jeram sanitary landfill site.

(ii). to investigate the efficiency of electrocoagulation processes in the removal of chemical oxygen demand (COD) and heavy metals (Zn and Cu) from sanitary landfill leachate effected by several parameters, thus proposing the optimum condition for pollutants removal.

1.4 Significance of Study

The present study focused on the removal of COD and heavy metals (Zn and Cu) concentrations from sanitary landfill leachate using electrocoagulation process by searching a possible optimum operational condition of experiment. An electrocoagulation processes have been employed as a water treatment technology and it is proven effective in removing an extremely wide range of pollutants (Holt *et al.*, 2005). Besides the requirement of simple equipments, electrocoagulation is also designable for virtually any size and easily operable. By applying electrocoagulation process on landfill leachate, it can improve water quality on its surrounding by reducing groundwater and surface water pollution, as well as the soil pollution. In addition, this study could also help other investigators to decide their effecting parameters and operational conditions of electrocoagulation in the future experiment.

1.5 Scope of the study

Landfill leachate is a major problem in waste landfilling. The leachate that flushed away from the landfill site are heavily contaminated and consist of complex compounds that is very difficult to deal with. The scope of the study was to investigate the efficiency of electrocoagulation process for the removal of COD, Zn

and Cu in leachate from Jeram sanitary landfill site through the experiment. The study also focused on several working parameters such as applied voltage, conductivity of wastewater, initial pH and operating time during electrocoagulation process to determine the optimum condition for the best removal of COD, Zn and Cu in leachate. The study also include determination of the characterisitcs of Jeram sanitary landfill leachate by in-situ measurement and laboratory analysis. The in-situ measurement of leachate sample were done at the site, while, the laboratory analysis were performed in the wet laboratory of the Department of Environmental Sciences, UPM. The in-situ measurement includes pH, Temperature, Conductivity, Dissolved Oxygen (DO), Turbidity and Salinity. For the laboratory analysis, the parameters includes Chemical Oxygen Demand (COD), Color, Total Suspended Solid (TSS) and heavy metals (Cd, Pb, Zn, Cu and Ni).

In the present work, the main focus of the study was on investigating the efficiency of electrocoagulation process in removal of COD, Zn and Cu in leachate. Therefore, the effecting parameters, namely, applied voltage, conducitivity, intial pH and operating time were the independent variables for the study; while the COD, Zn and Cu were the dependent variables. Hence, this study will be centred more on the electrocoagulation technique for the removal of selected pollutant in landfill leachate. From the results of the experiments, the effectiveness of electrocoagulation process in removing COD, Zn and Cu from landfill leachate can be determined. The variation of some parameters during the electrocoagulation process such as conductivity, pH and temperature was also determined to see their changes over operating time.

1.6 Limitation of Study

The subject of the study is regarding the removal of pollutants from sanitary landfill leachate through EC process. Although the research was carefully prepared, there are some limitations and shortcomings. The first is the study site. The study was actually first conducted at Sungai Sedu landfill site but it was closed during running half of the whole experiment. The study site was then shifted to Jeram sanitary landfill site where it took more time to set up the experiment again due to different concentration of landfill leachate obtained. The second problem is the location of the new study site. The landfill site is a quite far from the laboratory thus limiting the sampling frequency. The third is the financial support. It is difficult to get financial resources from the institution. Even though the costs that was required to carry out the EC experiments is low but the costs of conducting the entire research, from leachate sampling to analysis of pollutants are expensive. On paper, policy statements seem to indicate carrying out research like this is simple, but the actual process is quite to the contrary. As a result of this constrain, the study on leachate stabilization was only attempted for COD, Zn and Cu. The author need to change the size and scope of the research by reducing the number of samples analyzed and the duration of sampling to capture differences in leachate quality due to changing weather conditions. The result presented in this work is the maximum leachate concentration since sampling was done during the dry season.

1.7 Thesis Organization

The thesis begins with Chapter 1 where it introduces the background information about municipal solid waste generation, sanitary landfill management, leachate definitions, issues, and leachate treatment. The problem statement and objectives of the study are discussed in this chapter.

Chapter 2 reviews the basic information and background knowledge concerning sources of municipal solid waste, sanitary landfill management, landfill leachate generation, effect of leachate to human and environment, previous studies related to leachate treatment, the application of electrocoagulation in wastewater treatment and the parameter affecting the performance of electroagulation. This chapter also discusses the issues and leachate characteristics in Malaysia context.

Chapter 3 explains the procedure on how the research was conducted and what sample was collected and analyzed. The methodological approach selected used to achieve the research objectives. The details of the sampling, raw leachate collected and data analysis activities are discussed throughout this chapter.

Chapter 4 described the result from the analysis of raw leachate characteristics by in-situ and laboratory procedure. The discussion also included the efficiency of electrocoagulation in the removal of COD, Zn and Cu from landfill leachate. The details on electrocoagulation process and the effecting parameters that contribute higher removal of selected pollutant were discussed together in this chapter.

Finally, Chapter 5 concludes the thesis with the conclusions that answered the general objective of the study, as well as giving some recommendation for a possible setup in electrocoagulation experiment and for further research.

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BIODATA OF STUDENT

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PUBLICATIONS

1. Electro coagulation for Removal of Chemical Oxygen Demand in Sanitary Landfill Leachate. (Integrated Publishing Services – International Journal of Environmental Sciences)