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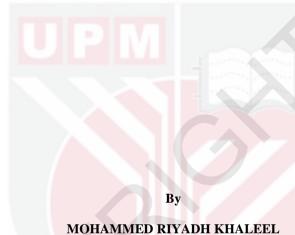
PERFORMANCE OF A COMBINED SYSTEM OF ELECTROLYSIS AND GRANULAR ACTIVATED CARBONS FOR LEACHATE TREATMENT OF JERAM SANITARY LANDFILL, MALAYSIA

MOHAMMED RIYADH KHALEEL

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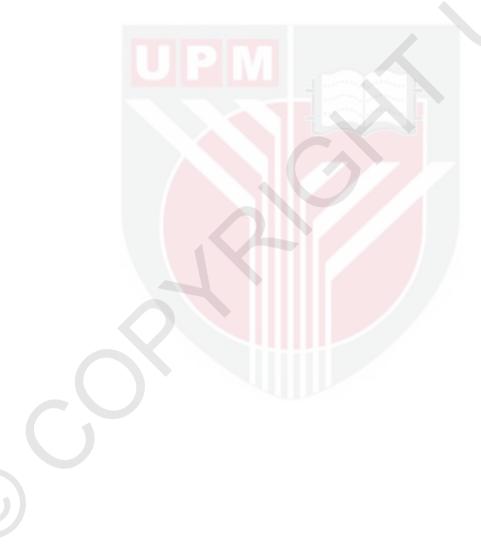
Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for the Degree of Master of Science

September 2015

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Abstract of the thesis presented to the senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science.

PERFORMANCE OF A COMBINED SYSTEM OF ELECTROLYSIS AND GRANULAR ACTIVATED CARBONS FOR LEACHATE TREATMENT OF JERAM SANITARY LANDFILL, MALAYSIA

By

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September 2015

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In this study, raw leachate collected from Jeram Sanitary Landfill (JSL) was characterized. The landfill leachate is a complex substance that contains toxic compounds, organic matter, ammonium, heavy metals and colloidal solids and a variety of pathogens potentially contaminate surface water and groundwater. The effluents are complicated to deal with and biological processes are totally inefficient for the toxic nature of stabilized leachate. Hence, there are coagulation-flocculation and adsorption process used to treat leachate. The coagulation-flocculation does by electrolysis process and adsorption by activated carbon. The raw leachate was treated using electrolysis treatment technique in which iron and stainless steel electrodes were utilized. In the electrolysis process, different voltages of 3, 6, 12, 18 and 24 volt and different retention times (RT) of 5, 10, 15, 20, 30, 40 and 50 min were used. The filtration process by quartz filter is subsequent treatment after electrolysis process. The adsorption process by using granular activated carbon (GAC) obtained from coconut shell (GACC) and oil palm shell (GACP) was final treatment after electrolysis and filtration processes. In the adsorption process, different AC dosages of 2, 4, 6, 8 and 10 g/l and different contact times (CT) of 1, 2, 3, 4, 5, 6, 8, 10 and 13 hr were used.

In electrolysis, the biochemical oxygen demand (BOD₅) removal efficiency was 68% and the chemical oxygen demand (COD) removal efficiency of 56% was achieved using the iron electrode. Total dissolved solids (TDS) removal efficiency of 55% was obtained at 20 min RT. Optimum total suspended solids (TSS) removal efficiencies of 69 and 75% were obtained using iron and stainless steel electrodes, respectively. Salinity removal efficiency was 53% and turbidity removal efficiency was 96%. The pH value was 9.4 at 40 min RT using iron electrode. The lowest electrical conductivity (EC) value was recorded as 156μ s/cm using iron electrode.

In adsorption process, the BOD₅ removal efficiency was 95%, while the COD removal efficiency was 88%. Total nitrogen (TN) removal efficiency was recorded as 98.7%, while phosphate (PO₄) removal efficiencies of 84 and 82% were obtained at CT of 4 (GACC) and 2 hr, (GACP) respectively. TDS removal efficiency was obtained as of 66 and 75% at 4 hr CT of GACC and GACP, respectively. Optimum TSS removal

efficiency was 90%. Salinity removal efficiencies using GACC and GACP were 81 and 74%, respectively. Turbidity removal efficiency of 95% was the highest removal efficiency recorded at 6 hr. The pH was 8.93 for both GACC and GACP. Using GACC and GACP, EC values were recorded as 102 and 83µs/cm, respectively.

After several combinations of voltage were used for the electrolysis process, where, 40 min RT and 24 volt were selected as the best combination for the highest removal efficiency. Also, GAC dosage of 10 g/l at 6 hr CT yielded the highest removal efficiency.

Generally, iron electrode is the cheaper and more resistant to corrosion than stainless steel. The results obtained from the iron electrode were close to stainless steel results. On the other hand, GACP is the cheaper than GACC. Also, GACP is abundantly produced in Malaysia as a biomass waste generated from agricultural activities. In conclusion, GACP can be considered a promising environmental-friendly adsorbent for the treatment of landfill leachate.

Abstrak tesis yang dikemukakan kepada senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Master Sains

PRESTASI SISTEM GABUNGAN ELEKTROLISIS DAN BERBUTIR DIAKTIFKAN KARBON BAGI LEACHATE RAWATAN JERAM SANITARY TAPAK PELUPUSAN SAMPAHOLEH, MALAYSIA

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September 2015

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Dalam kajian ini, larutan resap mentah yang dikutip dari Jeram Sanitary Landfill (JSL) telah digubal. Sampel larutan resap tapak pelupusan adalah bahan kompleks yang mengandungi sebatian toksik, bahan organik, ammonium, logam berat dan pepejal koloid dan pelbagai patogen yang berpotensi mencemarkan air permukaan dan air bawah tanah. Efluen yang rumit untuk menangani dan proses biologi adalah betul-betul tidak cekap untuk sifat toksik larutan resap stabil. Oleh itu, terdapat pembekuanpemberbukuan dan proses penjerapan digunakan untuk merawat larutan resap. Pembekuan-pemberbukuan tidak melalui proses elektrolisis dan penjerapan oleh karbon diaktifkan. Larutan resap mentah telah dirawat dengan menggunakan teknik rawatan elektrolisis di mana besi dan keluli tahan karat elektrod yang digunakan. Dalam proses elektrolisis, voltan yang berbeza 3, 6, 12, 18 dan 24 volt dan masa tahanan yang berbeza (RT) sebanyak 5, 10, 15, 20, 30, 40 dan 50 min telah digunakan. Proses penapisan oleh penapis kuarza adalah rawatan berikutnya selepas proses elektrolisis. Proses penjerapan dengan menggunakan karbon berbutir diaktifkan (GAC) yang diperolehi daripada tempurung kelapa (GACC) dan tempurung kelapa sawit (GACP) adalah rawatan akhir selepas elektrolisis dan penapisan proses. Dalam proses penjerapan, AC dos yang berbeza 2, 4, 6, 8 dan 10 g / l dan masa hubungan yang berbeza (CT) 1, 2, 3, 4, 5, 6, 8, 10 dan 13 jam digunakan.

Dalam elektrolisis, permintaan oksigen biokimia (BOD_5) kecekapan penyingkiran adalah 68% dan kecekapan keperluan oksigen kimia (COD) penyingkiran 56% telah dicapai dengan menggunakan elektrod besi. Jumlah kecekapan pepejal terlarut (TDS) penyingkiran 55% telah diperolehi pada 20 min RT. Jumlah pepejal terampai Optimum (TSS) kecekapan penyingkiran 69 dan 75% telah diperolehi dengan menggunakan besi dan keluli tahan karat elektrod, masing-masing. Kecekapan penyingkiran kemasinan adalah 53% dan kecekapan penyingkiran kekeruhan adalah 96%. Nilai pH adalah 9.4 pada 40 min RT menggunakan elektrod besi. Kekonduksian elektrik (EC) Nilai terendah yang dicatatkan sebagai 156 μ s / cm menggunakan elektrod besi.

Dalam proses penjerapan, kecekapan penyingkiran BOD₅ adalah 95%, manakala kecekapan penyingkiran COD adalah 88%. Jumlah nitrogen (TN) kecekapan



penyingkiran dicatatkan sebagai 98.7%, manakala kecekapan fosfat (PO₄) penyingkiran 84 dan 82% telah diperolehi di CT 4 masing-masing (GACC) dan 2 jam, (GACP). TDS kecekapan penyingkiran telah diperolehi pada 66 dan 75% pada 4 jam CT GACC dan GACP, masing-masing. TSS Optimum kecekapan penyingkiran adalah 90%. Kecekapan penyingkiran kemasinan menggunakan GACC dan GACP adalah 81 dan 74% masing-masing. Kekeruhan kecekapan penyingkiran sebanyak 95% adalah penyingkiran tertinggi kecekapan direkodkan pada 6 jam. PH adalah 8.93 untuk kedua-dua GACC dan GACP. Menggunakan GACC dan GACP, nilai SPR telah direkodkan sebagai 102 dan 83µs / cm, masing-masing.

Selepas beberapa kombinasi voltan digunakan untuk proses elektrolisis, di mana, 40 min RT dan 24 volt telah dipilih sebagai kombinasi yang terbaik untuk penyingkiran kecekapan tertinggi. Juga, GAC dos 10 g / l pada 6 hr CT menghasilkan penyingkiran kecekapan tertinggi.

Secara umumnya, elektrod besi adalah lebih murah dan lebih tahan kakisan daripada keluli tahan karat. Keputusan yang diperolehi daripada elektrod besi yang rapat dengan keputusan keluli tahan karat. Sebaliknya, GACP adalah lebih murah daripada GACC. Juga, GACP banyaknya dihasilkan di Malaysia sebagai sisa biojisim yang dihasilkan daripada aktiviti pertanian. Kesimpulannya, GACP boleh dianggap sebagai penjerap mesra alam menjanjikan untuk rawatan leachate tapak pelupusan.

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LIST OF ABBREVIATIONS

AC	Activated Carbon
AOP	advanced oxidation processes
BOD5	Biological Oxygen Demand
CAS	Conventional activated sludge systems
COD	Chemical Oxygen Demand
СТ	Contact Time
DC	Direct Electric Current
DOC	Dissolved Organic Carbon
DOM	Dissolved Organic Matter
EC	Electrical Conductivity
GAC	Granular Activated Carbon
GACC	Granular Activated Carbon based on coconut shell
GACP	Granular Activated Carbon based on palm shell
GHG	Green House Gas
HA	Humic Acid
HRT	Hydraulic Retention Time
IUPAC	International Union of Pure and Applied Chemistry
JSL	Jeram Sanitary Landfill
LR	Loading Rate
MAP	magnesium ammonium phosphate
MF	Microfiltratio
MSW	Municipal Solid Waste
Ν	Nitrogen
NF	nan filtration
NTU	Nephelometric Turbidity Units
PAC	Powder Activated Carbon
RO	Reversed osmosis
rpm	Revolution per minute
RT	Retention Time
SBR	Sequential Batch Reactor
SS	Suspended Solids

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- TCOD Total Chemical Oxygen Demand
- TDS Total Dissolved Solids
- TKN Total Kjeldahl Nitrogen
- TOC Total Organic Carbon
- TSS Total Suspended Solids
- UASB Upflow Anaerobic Sludge Blanket
- UF ultrafiltration

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- VFA Volatile Fatty Acids
- WWTP Waste Water Treatment Plant

CHAPTER 1

INTRODUCTION

1.1 Background

Sanitary landfills are defined as a place where the solid waste has been isolated from an exact environment till the mentioned solid waste is totally safe. It degrades biologically, chemically as well as physically. Solid waste in the landfill is a type of solid waste generated from community, commercial and agricultural operations. This includes wastes from households, offices, stores and other non-manufacturing activities. A site is a subject to be regarded as sanitary landfill after four basic conditions should be met, longer term aim should be introduced in order to meet them finally in full. Basic requirements are: partial or full hydrogeological isolation, Permanent control, planned waste emplacement and covering as well as formal engineering presentation.

The location of landfills for the deposition of domestic and industrial solid waste in remote areas is for health reasons. This is because of the emission of green house gas GHG (methane and carbon dioxide) from decomposing waste within the landfills that can be harmful to health and also pose major environmental problems. Additionally, there is the production of a liquid known as leachate when precipitation infiltrates the solid waste. Due to the high content of organic compounds and ammonium ions, leachate is highly polluted (Welander et al., 1997).

Leachate is generated from the garbage decomposition as well as precipitation which infiltrates and percolates throughout the waste material volume and settles down to the bottom of the landfill and generates chemical reaction as well as physical mixing together with ingredients that found in the subjected waste. Leachate commonly has high level of toxic compounds concentration together with matter of organic, heavy metals and ammonium. Inappropriate geological material under the landfill is the main cause of risk of leachate leakage to the groundwater. A long term humans health issues may be caused by heavy metals and toxic materials in leachate (Thörneby et al., 2003).

Leachate in landfill frequently exceeds standard for surface water and municipal waste water, often for several decades. Landfill leachate has the high possibility to pollute surface water and groundwater caused by pathway for leachate to the bottom of the landfill through the unsaturated soil layers to the groundwater, then by groundwater through hydraulic connections to surface water. Nevertheless, pollution may also outcome from the discharge of leachate through direct discharge of untreated leachate or by treatment plants. The main factors influencing the pollution chance from landfill leachate are the flux of the leachate and concentration. The landfill sitting such as the hydro geological setting and the degree of protection provided and the basic quality, volume, sensitivity of the receiving groundwater and surface water (Ghafari et al., 2009).



One of the methods to control leachate generation is to control the water infiltration in the sanitary landfill by solid waste compaction. This method reduces the infiltration rate while growing plants on the soil covers of the solid waste can also have the same effect. The sanitary landfill leachate properties are controlled by temperature, pH, solid waste properties, moisture content, redox potential, etc. Temperature has a significant effect on the decomposition process in a sanitary landfill. Besides, moisture is needed for the biological conversion and stabilization within the sanitary landfill. The redox and pH potential set the conditions for the different phases of decomposition and biological processes within the sanitary landfill. Thus, the microbial composition within the sanitary landfill effectively contributes to the sanitary landfill stabilization (Pokhrel et al., 2005).

Ground-water becomes contaminated due to buried solid waste that is above the level of water table. Ground-water gets contaminated likewise if leachate moves downwards from the sanitary landfill into the ground-water table as a result of precipitation infiltration (Madu, 2008).

The survey by O'Leary et al.(1995) investigates the objectives and factors that need to be considered in the design of a sanitary landfill that is related to biological, physical and chemical reactions at municipal solid waste landfills that occur simultaneously and result in waste decomposition leachate and gases (O'Leary et al., 1995).

The design procedure entails alternating layers of compacted municipality solid waste (MSW) with cover material when waste is disposed. This can be compost, soil, or any other approved material, where wastes are compacted after dumping by special bulldozers and the fresh layer of MSW is laid over with cover material to start another layer. This method helps to reduce odor problems, and prevents exposure to health hazards. All sanitary landfills are supplied leachate collection systems. A typical liner is composed of layers of synthetic material, plastic, gravel, and clay to prevent leachate from escaping as shown in Figure 1.1. A lined landfill is also fitted with a pipeline network to collect and drain the leachate. Leachate recirculation is practiced at a solid waste landfill, or it is treated and discharged (Nora, 2006).

The design of the sanitary landfill location would prevent, or it also might reduce any undesirable outcomes on the environment and the effect on human health. It is very important to adopt methods, standards and operational systems based on current best practices in the design, which reflects progress in management techniques and containment standards. Protecting the environment and health should be the main aims when designing a landfill. The findings of the environmental assessment, risk assessment and the conceptual design proposals are interactive process in landfill design. The main aim behind waste management is sustainability (Manandhar, 2009).

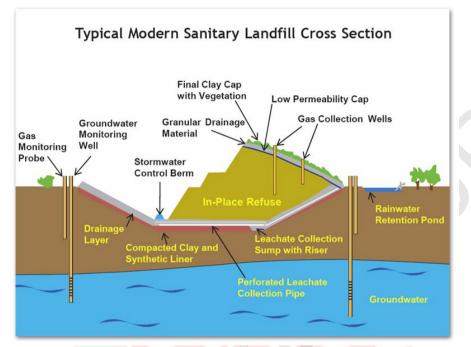


Figure 1.1: Cross-section of a Sanitary landfill showing the composition of layers (http://runcoenv.com/landfill.htm)

The central objective of waste policy is to reduce the harmful health and environmental impacts of waste. In order to meet this objective, it is particularly important to:

- prevent the generation of waste
- promote reuse of waste
- promote biological recovery of waste and recycling of materials
- promote energy use of waste not suited for recycling
- ensure that the treatment and disposal of waste does not cause any harmful impacts

The main climate-related objective of waste policy is to reduce the greenhouse gas emissions generated by waste, particularly by reducing the methane emissions resulting from treatment at landfills. In order to reach the objective, the amount of landfilled biodegradable waste will be substantially reduced, while at the same time measures will be taken to increase the recovery rates of methane generated at landfills (Graveland et al., 2003).

It is a known fact that all living plants and beings need nutrients which are essential for development. However, excessive use can cause adverse effects. As an example, aquatic life is affected by excess nutrient discharge in natural water bodies, as it increases oxygen demand and eutrophication, while human beings will suffer various health problems from excess nutrients. Human daily activities produce a high concentration of phosphorous and nitrogen and due to the discharge of wastewater that causes eutrophication in water bodies. Therefore, there is an urgent need to improve wastewater treatment technology to a level when it can efficiently remove organic

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matter, nutrients and other harmful constituents. These problems have led to the realization that there is an urgent need to think of solutions and alternative methods and available materials in the process of wastewater treatment. One of the natural methods and substances available to treat leachate are electrolysis treatment and activated carbon (AC).

The process of circulating direct current (DC) through an ionic substance is known as electrolysis; the user substance in the electrolysis process is either molten or dissolved in a suitable solvent, which produces a chemical reaction at the electrodes and separates the materials (Morimitsu, 2000).

In 1990, the world production of AC to meet demand was estimated to be 375,000 tons, except Eastern Europe and China. In the United States the demand for activated carbon reached 200,000 tons per year. In 2002, demand further increased and market growth for these materials for various applications was estimated at 4.6% per year (Mozammel et al., 2002). AC performance in water applications showed low cost compared to the use of other possible competitive inorganic materials such as zeolites and this has an effect on the positive market position.

1.2 Problem Statement

The JSL produces huge amount of the update every day. The landfill leachate is a complex substance which generated when water is absorbed into the solid waste disposal site that contains toxic compounds, organic matter, ammonium, heavy metals and colloidal solids and a variety of pathogens potentially contaminate surface water and groundwater. The landfill leachate properties are different and these differences are caused by several factors such as availability of oxygen and moisture content, design and life expectancy of the solid waste and operational of the sanitary landfill (Tzoupanos et al., 2010).

The important potential pollution source of surface and ground water is landfill leachate. Leachate are not correctly collected, treated and safely disposed, causing extensive contamination of water wells, creeks and streams (Li et al., 2010). The effluents are complicated to deal with and biological processes are totally inefficient for the toxic nature of stabilized leachate. Hence, there are requirement to physical, chemical and biological treatment and alternative technology. Coagulation-flocculation and adsorption process are widely used in wastewater treatment plants because of implementation and operation simplicity (Rivas et al., 2004).

The electrolysis is applied for landfill leachate treatment (Peng 2013; Tsai et al.1997). It had higher performance than classical chemical coagulation process and it can be applied as a step of a joint treatment. Kabuk et al. (2013) investigated on leachate treatment with electrolysis and optimization by response surface methodology. At optimum working conditions, 60.5 % COD removal, 92.4 % total suspended solids (TSSs) removal, 60.8 % total organic carbon (TOC) removal, 28.3 % total Kjeldahl nitrogen (TKN) removal, 99 % PO4-P removal, and 28.9 % NH3-N removal results were obtained.

Therefore, in this study, a novel low-cost process integrating electrocoagulation with an activated carbon (AC) contactor is developed for the first time to improve the treatment of the increasing volume of leachate. The optimum pollutant removal efficiencies (for BOD5, COD, TDS, TSS, and pH) are identified by extensive laboratory analysis. The proposed process is an ecofriendly, sustainable technique for leachate treatment, which reduces treatment cost and saves energy, and which also helps in protecting the environment.

The optimal conditions of electrolysis for landfill leachate treatment have not been investigated in Malaysia. In addition the optimal conditions of AC dosage for landfill leachate treatment have not been studied in Malaysia. Moreover, a combined system of electrolysis and AC have not been studied until now. It is hope that by applying the combined system; we can get high removal efficiency for various parameters (BOD₅, COD, TN, PO₄, TDS, TSS, Turbidity, pH, Salinity and electrical conductivity).

1.3 Research Objectives

The main objective of the current study is to examine the performance of a combined system of electrolysis and granular activated carbon adsorption to treat landfill leachate collection from Jeram Sanitary Landfill.

The specific objectives of this study can summarize as bellow:

- 1. To evaluate the performance of iron and stainless steel electrodes to treat landfill leachate by electrolysis.
- 2. To evaluate the pollutants removal efficiency from landfill leachate by using the granular activated carbons based on coconut and oil palm shells.
- 3. To determine the optimum conditions of hydraulic retention time, voltage and AC dosage for electrolysis process followed by AC adsorption.

1.4 The scope of the study

In this study, leachate samples were collected from JSL, followed by laboratory testing procedures in order to evaluate and determine the levels biochemical oxygen demand (BOD), chemical oxygen demand (COD), total nitrogen (TN), phosphate (PO₄), total suspended solid (TSS), total dissolved solids (TDS), turbidity, pH, salinity and electrical conductivity (EC) in these samples. In January 2007, the landfill started operations. The landfill leachate was collected without any pre-treatment performed. To keep the properties of the wastewater unchanged, the leachate of the collected landfill was stored in refrigerator at 4°C. The type of leachate landfill is medium leachate. The treatment process is divided into three stages: Firstly; the electrolysis process for leachate treatment was conducted using iron and stainless steel electrodes at various retention time (5, 10, 15, 20, 30, 40 and 50 min) and different voltages (3, 6, 12, 18 and 24 volt). Secondly; quartz filters were employed to remove the particles. Finally, GAC commercial coconut shells were used in leachate treatment with different contact time (1, 2, 3, 4, 5, 6, 8, 10 and 13 hr) and dosage (2, 4, 6, 8 and 10 g/l) to



determine the treatment efficiency of leachate. Furthermore a commercial GAC based on palm shells was also used in leachate treatment with different contact times in order to compare the treating efficiency outputs of leachate for each GAC type using laboratory testing techniques; BOD, COD, TN, PO4, TSS, TDS, turbidity, pH, salinity and EC, to find the optimal one with preeminent quality and determine the HRT, voltage, CT, AC dosage, etc.



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