IMPROVING LEACHATE TREATMENT BY USING LIMESTONE FILTRATION AND FREE WATER SURFACE CONSTRUCTED WETLAND

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ABSTRACT

Landfill is the most applicable method in reducing the solid waste. However, the landfill method causes generation of leachate. Landfill leachate contains high levels of organic and inorganic matters which mean it is very contaminated. Due to this problem, leachate cannot be directly discharge into water course. Therefore, it is necessary for leachate to undergo a treatment process before it can be discharged into environment. This study mainly focused on the method of landfill leachate treatment. In this study, combination of limestone filter and free water surface wetland are used to treat landfill leachate. 5 to 15 mm particle sized of limestone and *Eichhornia crassipes sp.* (water hyacinth) as wetland plant were used to treat leachate. Raw leachate sample was collected from Jerangau-Jabor Landfill. The treated leachate was analyzed for BOD, COD, A-N and total suspended solids improvement and heavy metal removal that consist of Fe, Cu, Cr, Mn, Zn and Pb. From the analysis, the combination of limestone filter and FWS constructed wetland was effective in treating landfill leachate. The result shows a great removal efficiency with 97.1% COD, 98.1% TSS, 97.6% A-N, 99.9% Fe, 99% Pb, 100% for Mn, Cu and Zn. Based on the study, it can be concluded that the combination of limestone filter and FWS constructed wetland has a high efficiency in removing leachate pollutants. The optimum contact time for the system is seven days.

ABSTRAK

Tempat pelupusan sampah (landfill) merupakan antara kaedah yang paling sesuai untuk mengurangkan sisa pepejal. Walaubagaimanapun, kaedah ini telah menyebabkan penghasilan air larut resap. Air larut resap daripada tapak pelupusan sampah mengandungi kepekatan bahan organik dan bahan tidak organik yang tinggi. Hal ini bermaksud, air larut resap sangat tercemar dan kontaminan. Sehubungan itu, air larut resap tidak boleh sewenangnya dilepaskan ke sumber air yang sedia ada. Jadi, air larut resap perlu melalui proses rawatan terlebih dahulu sebelum dilepaskan ke sumber air. Fokus kajian ini adalah kaedah rawatan air larut resap. Dalam kajian ini, kombinasi penapis batu kapur dan tanah bencah buatan digunaka untuk merawat air larut resap. Saiz batu kapur dari 5 ke 15 mm dan pokok *Eichhornia crassipes sp.* (keladi bunting) sebagai pokok untuk tanah bencah digunakan untuk proses rawatan air larut resap. Sampel air larut resap diambil dari tapak pelupusan sampah Jerangau-Jabor. Air larut resap yang dirawat telah dianalisa kadar penurunan BOD, COD, A-N, pepejal terampai dan juga penyingkiran logam berat yang terdiri daripada Fe, Cu, Cr, Mn, Zn dan Pb. Daripada keputusan analisis, kombinasi antara penapisan batu kapur dan tanah bencah buantan berkesan dalam merawat air larut resap. Keputusan menunjukkan kadar penyingkiran yang tinggi iaitu sebanyak 97.1% COD, 98.1% TSS, 97.6% A-N, 99.9% Fe, 99% Pb dan 100% untuk elemen Mn, Cu and Zn. Berdasarkan kajian yang telah dilakukan, ianya boleh dirumuskan bahawa kombinasi sistem antara penapisan batu kapur dan tanah bencah buatan sangat efisien untuk penyingkiran bahan pencemar dalam air larut resap. Kadar masa yang optimum untuk sistem ini berfungsi adalah tujuh hari.

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LIST OF ABBREVIATIONS AND SYMBOLS

Al	-	Aluminum
A-N	-	Ammonia Nitrogen
APHA	-	American Public Health Association
AAS	-	Atomic Absorption Spectrometer
BOD	-	Biological Oxygen Demand
BOD_5	-	5-day Biological Oxygen Demand
Ca	-	Calcium
CaO	-	Lime
$CaCO_3$	-	Calcium Carbonate
Cd	-	Cadmium
COD	-	Chemical Oxygen Demand
Cr	-	Chromium
Cu	-	Copper
DMS	-	Dimethyl Sulphide
DO	-	Dissolved Oxygen
DOC	-	Dissolved Organic Carbon
DOE	-	Department of Environmental
EPA	-	Environmental Protection Agency
FKASA	-	Fakulti Kejuruteraan Awam & Sumber Alam
Fe	-	Iron
FWS	-	Free Water Surface
HLR	-	Hydraulic Loading Rate
HRT	-	Hydraulic Retention Time
LOI	-	Loss on Ignition
Mg	-	Magnesium
Mn	-	Manganese
Ni	-	Nickel

NH	-	Ammonium
P	-	Phosphorus
Pb	-	Lead
PAH	-	Polycyclic Aromatic Hydrocarbon
PCB	-	Polychlorinated Biphenyls
QA/QC	-	Quality Assurance and Quality Control
RZM	-	Root Zone Method
S	-	Sulfur
SOP	-	Soluble Orthophosphate
SS	-	Suspended Solid
SSF	-	Sub Surface Flow
TKN	-	Total Kjeldahl Nitrogen
TN	-	Total Nitrogen
TOC	-	Total Organic Carbon
TP	-	Total Phosphorus
TSS	-	Total Suspended Solids
UMP	-	Universiti Malaysia Pahang
USEPA	-	United State Environmental Protection Agency
Zn	-	Zinc
%	-	Percent
°C	-	Degree Celcius

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CHAPTER I

INTRODUCTION

1.1 Research Background

Over the years, industrialization and urbanization with high growth rate has causes several environmental problem all over the world. Nowadays, solid waste management and wastewater treatment are among the important problems worldwide. Like most of the developing countries, Malaysia is facing an increase of the generation of waste and of accompanying problems with the disposal of this waste. The amount of solid wastes produced around the world is increasing at high rates. Landfill is one of the most widely employed methods for the disposal of municipal solid waste (MSW). Up to 95% total MSW collected worldwide is disposed of in landfills (El-Fadil *et al*, 1997). However the landfill causes generation of leachate. Landfill leachate will cause environmental problems without proper handling. Increase in landfill leachate creates challenges for cost effective treatment methods to process wastewater.

Leachate is characterized by high concentration of organic matter (biodegradable and non-biodegradable), ammonia nitrogen, heavy metals, and chlorinated organic and inorganic salts (S. Renou *et al*, 2008). The characteristics of leachate are highly variable (D. Kulikowska *et al*, 2008) depending on the waste composition (P. Palaniandy *et al*, 2009), amount of precipitation, site hydrology, waste compaction, cover design, sampling procedures, and interaction of leachate with the environment, landfill design and operation (D. R. Reinhart *et al*, 1998).

Organic content of leachate pollution is generally measured in terms of biological oxygen demand (BOD_5) and chemical oxygen demand (COD).

The concentration of leachate contaminants may range over several orders of magnitude (Y. Deng *et al*, 2006). A combination of pollutants (BOD_5, COD, ammonia, inorganic salts, etc.) in higher concentrations renders landfill leachate as a potential source of contamination both to ground and surface waters, hence necessitates its treatment prior to discharge to water resources (M. J. K. Bashir *et al*, 2010).

1.2 Problem Statement

Leachate can contaminate groundwater where landfills are not provided with liners. It may also contaminate if it is not collected and treated prior to its discharge. Most of the landfills in developing countries including Malaysia are not designed with proper leachate collection mechanism. There are 230 landfills in Malaysia recognized officially (J. Y. M. Alkassasbeh *et al*, 2009). Most of these landfills do not come under sanitary landfill classification because there are no facilities for collection and/or treatment of leachate and there is no infrastructure to collect landfill gas (J. Y. M. Alkassasbeh *et al*, 2009). Many of these landfill sites are located near rivers and streams which are a major source of agriculture and productivity, industrial and domestic water supply.

In addition, leachate contains mercury, iron, manganese and copper which is a heavy metal pollutant. Large doses of heavy metal can be detrimental to human health. For example, ingestion of inorganic mercury salts may cause acute effect in terms of gastrointestinal disorders such as abdominal pain, vomiting, diarrhea, and hemorrhage (ATSDR, 1989). Repeated and prolonged exposure of inorganic mercury will result in severe disturbances in the central nervous system, gastrointestinal tract, kidneys, and liver. Meanwhile, large doses of manganese cause apathy, irritability, headaches, insomnia, and weakness of the legs while the acute toxicity for ingested copper is characterized by abdominal pain, diarrhea, vomiting, tachycardia and a metallic taste in the mouth. Continued ingestion of copper compounds can cause cirrhosis and other debilitating liver conditions (Mueller-Hoecker *et al.*, 1989). Therefore, since leachate can affect aquatic ecosystems and human health, proper leachate treatment is needed before leachate is discharged into receiving water (Paredes, 2003).

Usually, a combination of two physical-chemical treatment or physico-chemical and biological treatment is required for optimum treatment of stabilized leachate (Kurniawan *et al.*, 2006, Kargi and Pamukoglu, 2003, Tatsi *et al.*, 2003). Thus, this research will focus on leachate treatment using combination of limestone and wetland in one system. Type of wetland that will be use in this research is free water surface (FWS).

1.3 Research Objective

There are three objectives for this research based on the problem statement. The objectives are as below:

i. To investigate the suitability of combination limestone filter and free water surface (FWS) constructed wetland for leachate treatment system. The parameters to be studied are ammoniacal nitrogen, biological oxygen demand (BOD), chemical oxygen demand (COD), total suspended solid (TSS) and heavy metals.

ii. To determine the optimum contact time for the system to remove pollutant from leachate.

iii. To determine the percentage removal of leachate in limestone filter and FWS constructed wetland.

1.4 Scope of Research

The scopes of this study are includes: set-up two stage of constructed wetland (FWS) and limestone filter to treat the landfill leachate. The experiments are carried out in the Environmental Laboratory, Faculty of Civil Engineering and Earth Resources, Universiti Malaysia Pahang (UMP).

The limestone will be obtained from Tinjau Makmur Sdn. Bhd. quarry located at Felda Bukit Sagu 4, Kuantan, Pahang. The limestone will be crushed at Faculty of Civil Engineering and Earth Resources laboratory to achieve the desired size of limestone.

Leachate sample will be taken from Jerangau - Jabor Landfill in Kuantan District with cooperation from Majlis Perbandaran Kuantan (MPK) and Terang Bersih Sdn. Bhd. The plants that will be used in this study is *Eichhornia crassipes* (water hyacinth) as floating plant for FWS wetland which can be obtained at the swamp nearby University Malaysia Pahang area.

Leachate characteristics are identified in terms of physical and chemical characteristics. A series of experiments will be conduct to determine the chemical characteristics based on the parameter of ammoniacal nitrogen, biological oxygen demand (BOD), chemical oxygen demand (COD), total suspended solid (TSS) and heavy metals. Thus, this research will focus on leachate treatment using combination of limestone and wetland in one system. Finally, lower cost of leachate treatment system using limestone and wetland proposed as a cost effective treatment compared to other conventional treatment.

1.5 Significant of the study

The study is conducted to evaluate the performance of limestone in combined constructed FWS wetland to treat the landfill leachate. It also an environmental friendly approach. Leachate poses a number of environmental problems. This is due to variable types of waste and its composition. Leachate can contain high concentration of organic matters, nutrients and heavy metals. The study of leachate treatment by using limestone and wetland are important in order to improve the efficiency and provide variation methods of leachate treatment. Leachate treatment by using limestone and wetland will become another alternative treatment which will reduce the overall cost of conventional leachate treatment. This treatment can be used and practice widely by municipal authorities in order to control and reduce pollution posed especially from transfer station by leachate.

CHAPTER II

LITERATURE REVIEW

2.1 Introduction

This chapter includes literature review on landfill leachate, constructed wetland and limestone removal that related to the scope of the study. Landfill leachate has become highly contaminated due to the high population growth rate, industrialization and urbanization. As leachate migrates away from a landfill, it may cause serious pollution to the adjacent surface waters as well as groundwater. Constructed wetland combined with limestone filter has become the promising method to treat a landfill leachate.

2.2 Leachate

Leachate is a liquid that has percolated through solid waste and has extracted, dissolved, and suspended materials that may include potentially harmful materials. The type of solid waste, physical, chemical, and biological activities that occur in the solid waste determines the quality of leachate. The quantity of leachate seeping from the landfill is proportional to the build up of leachate within the landfill, alternatively called leachate mound. Leachate can cause serious problems since it able to transport contaminating materials that may cause a contamination of soil, groundwater and surface water (Warith, 2003).

2.2.1 Leachate Composition

Leachate is the main issue for the landfill pollution, and it consists of different inorganic substances and organic matters. The composition of leachate will influence by types and age of waste deposited, the prevailing physico-chemical conditions and the microbiology and the water balance in the landfill (McBean, 1995). Leachate is the liquid percolation that drains through the waste in the landfill vary widely depends on waste type and the waste age (Christensen, 1994). The composition of leachate varies greatly from site to site, and can vary within a particular site. Some of the factors affecting composition include:

a. Age of landfill
b. Types of waste
c. Degree of decomposition that has taken place
d. Physical modification of the waste (e.g. shredding)

Leachate is highly variable and heterogeneous. Generally, leachate produced in younger landfills is characterized by the presence of substantial amounts of volatile acids, as a result of the acid phase of fermentation. In mature landfills, the great portions of organics in leachate are humic and fulvic-like fractions. From older literature view it can be concluded that in leachate from young landfills, the concentration of organics (as COD) is above 10000 mg/l, while in leachate from landfills older than 10 years COD is below 3000 mg/l (Chang, 1989).

Typically, the leachate can be characterized into three major groups as shown in **Table 2.1**. The three major groups are mainly organic matters, inorganic matters and xenobiotic organic compounds. Beside these, other compounds are also likely present in the leachate such as arsenate, barium, borate, cobalt, lithium, mercury, selenate and sulfide however in small quantity and of less significant level.

Table 2.1: Pollutant in leachate

Group of Pollutants In Leachate	**Components**
Organic matters	Acids, alcohols, aldehydres and others usually quantified as COD (Chemical Oxygen Demand), BOD (Biochemical Oxygen Demand), DOC (Dissolved Organic Carbon), Other Volatile fatty acid and refractory compound include fulvic-like and humic like compounds
Inorganic matters	Sulfate, chloride, ammonium, calcium, magnesium, sodium, potassium , hydrogen carbonate, iron and manganese and heavy metal like lead, nickel, copper, cadmium, chromium and zinc
Xenobiotic organic compounds	Aromatic hydrocarbon, phenols, chlorinated aliphatics, pesticides and plastizers include PCB, Dioxin, PAH, etc

2.2.2 Leachate Treatment Review

Conventional landfill leachate treatments can be classified into three major groups: (a) leachate transfer: recycling and combined treatment with domestic sewage, (b) biodegradation: aerobic and anaerobic processes and (c) chemical and physical methods: chemical oxidation, adsorption, chemical precipitation, coagulation/flocculation, sedimentation/flotation and air stripping (S. Renoua, 2007).

a. Leachate transfer

i. Combined treatment with domestic sewage

Few years ago, a common solution was to treat the leachate together with municipal sewage in the municipal sewage treatment plant. It was preferred for its easy maintenance and low operating costs (W.-Y. Ahn, 2002). However, this option has been increasingly questioned due to the presence in the leachate of organic inhibitory compounds with low biodegradability and heavy metals that may reduce treatment efficiency and increase the effluent concentrations (F. Cecen, 2004). Combined treatment was investigated by Diamadopoulos *et al.* using a sequencing batch reactor (SBR) consisting of filling, anoxic, oxic and settling phases. When the ratio of sewage to leachate was 9/1, nearly 95% BOD and 50% nitrogen removals were obtained at the end of the daily cycles.

ii. Recycling

Recycling leachate back through the tip has been largely used in the past decade because it was one of the least expensive options available (J.M. Lema, 1988). Bae *et al.* reported that leachate recirculation increased the moisture content in a controlled reactor system and provided the distribution of nutrients and enzymes between methanogens and solid/liquids. Significant lowering in methane production and COD was observed when the recirculated leachate volume was 30% of the initial waste bed volume (S. Chugh, 1998). Also, Rodriguez *et al.* reported a 63–70% CODlowering in an anaerobic pilot plant with recirculation. The leachate recycle not only improves the leachate quality, but also shortens the time required for stabilization from several decades to 2–3 years (D.R. Reinhart, 1996).

b. Biological treatment

Biological treatment methods are processes whereby microbes are used to destroy or at least reduce the toxicity of a waste stream. Normally biological treatment of predominantly aqueous wastes such as leachate is accomplished in specially designed bioreactors. A suitable culture of the micro-organisms or microbial association, either aerobic or anaerobic, is chosen (Roux). Biological treatment is firmly established as the standard method of waste treatment for some wastes. i.e.:

- Domestic sewage
- Waste from food processing
- Hazardous waste e.g. phenols, cyanide, oils
- Leachates

However, for leachate treatment, a large number of approaches to biological treatment are proposed, but many are unproven and have not yet been shown to be effective on site (Roux).

The most common biological treatment is activated sludge - a suspended-growth process that uses aerobic microorganisms to biodegrade organic contaminants in leachate. With conventional activated-sludge treatment, the leachate is aerated in an open tank with diffusers or mechanical aerators. After the aeration phase, the mixed liquor of microorganisms and leachate is pumped to a gravity clarifier. The rotating biological contactor (RBC) is an attached-growth, aerobic, biological treatment process in which a series of discs are partially submerged in a tank of leachate. The disks eventually develop a slime layer, and then rotational shear forces strip off the excess solids and carry them with the effluent to a clarifier, where they are settled and separated from the treated waste. The carbon technique removes dissolved organics from the leachate. Although carbon systems may be useful with some older leachates, the cost of the carbon in the regeneration stage can make the process one of the most expensive treatment options (Aarts, 1994).

c. Physical / Chemical methods

Physical and chemical processes include reduction of suspended solids, colloidal particles, floating material, color, and toxic compounds by flotation, coagulation/flocculation, adsorption, chemical oxidation and air stripping. Physical/ chemical treatments for the landfill leachate are used in addition at the treatment line (pre-treatment or last purification) or to treat a specific pollutant (stripping for ammonia) (S. Renoua, 2007).

Pre-treatment with physical technologies prior to biological treatment have been largely using sedimentation, coagulation and flocculation or filtration in order to remove suspended solids. After biological treatment, the presence of high concentrations of salts normally prevents direct discharge to the environment. Options for treatment include evaporation or reverse osmosis with the recovery of a brine or solid salt material that often has to be disposed back into the landfill. Clearly, unless this process is managed carefully it is essentially self-defeating, since the salt can re-enter the leachate and the treatment cycle has to be repeated (Roux).

Chemical oxidation has also been widely used in South Africa. Hydrogen peroxide is being used at most H sites in South Africa for the mitigation of odours produced by the leachate, since it readily reacts with any sulphide and mercaptan components that normally cause the odour. Hydrogen peroxide is expensive and large amounts would be required to have any significant impact on the concentration of organics in the leachate. In the UK, ozone has been used to oxidize recalcitrant organics such as human acids, in order to break the molecules and make them more susceptible to biological treatment (Roux).

2.3 Limestone

Limestone is a sedimentary rock composed primarily of calcium carbonate ($CaCO_3$) in the form of the mineral calcite. It most commonly forms in clear, warm, shallow marine waters. It is usually an organic sedimentary rock that forms from the accumulation of shell, coral, algal and fecal debris. It can also be a chemical sedimentary rock formed by the precipitation of calcium carbonate from lake or ocean water.

2.3.1 Properties of Limestone

The properties of limestone can be divided into two types which are physical properties and chemical properties. Physically, limestone are quite impervious, hard, compact, and fine to very fine grained calcareous rocks of sedimentary nature and chemically, they are calcareous rocks principally of calcic minerals with minor amounts of alumina, ferric & alkaline oxides. The summaries of physical and chemical properties of limestone are illustrated in the **Table 2.2**. The color varies according to composition.