LANDFILL LEACHATE TREATMENT BY COMBINATION OF ELECTRO - FENTON AND SEQUENCING BATCH REACTOR METHOD

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ABSTRACT

Landfill leachate contains a large amount of organic, inorganic and heavy metal contents. Untreated leachate is a potential source to effect of soil, surface and groundwater. The combined treatment offers an alternative technique in dealing to leachate treatment. This research is to determine the effectiveness of combined electro-Fenton and sequencing batch reactor (SBR) method on the removal of SS, colour, COD and NH_3-N. The experimental involved three major parts were coagulation-flocculation, electro-Fenton and SBR method. This process could be operated independently in a batch mode and optimum conditions for each treatment were identified. In the combined process, leachate was first fed to coagulation-flocculation for pre-treatment. Then, the effluent from that process was oxidized in electro-Fenton process. The final process is the effluent of leachate was fed to a SBR method. The combined treatment was operated under the optimum conditions for all the processes. The result of coagulation-flocculation shown PAC is more effective at 2500 mg/L of optimum dosage. After coagulation-flocculation process, the removal of SS, colour, COD and NH_3-N were 80%, 77%, 61% and 35% respectively. The result of electro-Fenton shown Al-Al is more effective at 200 A/m^2 of optimum current density, 25 minutes of optimum reaction time, 4 of optimum pH, 800 mg/L of optimum H_2O_2 dosage and 1000 mg/L of optimum $FeSO_4{\bullet}7H_2O$ dosage. After electro-Fenton process, the removal of SS, colour, COD and NH_3-N were 87%, 95%, 82% and 65% respectively. The final process of SBR effluent was approaching neutral pH at 6.90 at 2800 mg/L of optimum MLSS and 6 h of optimum reaction time. The overall performance of combined treatment on the removal of SS, colour, COD and NH_3-N were 84%, 82%, 87% and 78% respectively. Thus, this combined treatment offers as an alternative technique for landfill leachate treatment on the removal of pollutants.

ABSTRAK

Larut lesapan mengandungi bahan organik, bukan organik dan logam berat yang tinggi kandungannya. Larut lesapan yang tidak diolah adalah berpotensi sebagai sumber pencemaran kepada tanah, permukaan dan air bawah tanah. Olahan gabungan larut lesapan adalah sebagai satu kaedah alternatif dalam olahan larut lesapan. Kajian ini menentukan keberkesanan olahan gabungan elektro-Fenton dan kaedah reaktor kelompok berjujukan (SBR) keatas penyingkiran SS, warna, COD dan NH_3-N. Experiment ini melibatkan tiga bahagian utama iaitu pengumpalan dan pengelompokan, elektro-Fenton dan SBR. Ketiga- tiga olahan ini beroperasi secara individu di dalam reactor masing-masing dan keadaan optimum bagi setiap proses ditentukan. Dalam olahan gabungan larut lesapan dimulakan dengan proses pengumpalan dan pengelompokan sebagai pra-olahan. Kemudiannya, effluen dari pengumpalan dan pengelompokan dioksidakan dalam proses elektro-Fenton. Akhir sekali, effluen larut lesapan diolah dalam SBR proses. Olahan gabungan dijalankan dengan menggunakan keadaan optimum untuk semua proses. Keputusan oleh proses pengumpalan dan pengelompokan menunjukan PAC adalah lebih berkesan pada 2500 mg/L dos optimum. Selepas proses pengumpalan dan pengelompokan, penyingkiran SS, warna, COD dan NH_3-N adalah masing-masing 80%, 77%, 61% dan 35%. Keputusan oleh proses elektro-Fenton menunjukan elektrod Al-Al adalah lebih berkesan pada 200 A/m^2 ketumpatan arus optimum, 25 min masa tindakbalas optimum, 4 pH optimum, 800 mg/L dos optimum H_2O_2 dan 1000 mg/L dos optimum $FeSO_4 \cdot 7H_2O$. Selepas proses elektro-Fenton, penyingkiran SS, warna, COD dan NH_3-N adalah masing-masing 87%, 95%, 82% and 65%. Efluen larut lesapan dalam proses akhir iaitu SBR telah mencapai neutral pH 6.90 pada 2800 mg/L MLSS optimum dan 6 jam masa tindak balas optimum. Keseluruhan olahan gabungan ke atas penyingkiran SS, warna, COD dan NH_3-N adalah masing-masing 84%, 82%, 87% and 78%. Oleh itu, olahan gabungan ini dapat dijadikan sebagai kaedah alternatif bagi olahan larut lesapan dalam menyingkirkan pencemaran.

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LIST OF SYMBOLS AND ABBREVIATIONS

A_0	-	Initial Concentration
Al	-	Aluminium
APHA	-	American Public Health Association
AWWA	-	American Water Works Association
BOD	-	Biochemical Oxygen Demand
cm	-	centimeter
COD	-	Chemical Oxygen Demand
DC	-	Direct Current
$FeCl_3$	-	Ferric Chloride
H_2SO_4	-	Sulphuric acid
H_2O_2	-	Hydrogen Peroxide
HRT	-	Hydraulic retention time
mg /L	-	Milligram per liter
MLSS	-	Mixed liquour suspended solid
NaOH	-	Sodium Hydroxide
NH_3-N	-	Ammonia Nitrogen
^0C	-	Degree Celcius

PtCo	-	Platinum Cobalt
PAC	-	Polyaluminium Chloride
SBR	-	Sequencing batch reactor
SRT	-	Solid retention time
SS	-	Suspended Solid
St	-	Stainless steel
UTHM	-	Universiti Tun Hussein Onn Malaysia
WEF	-	Water Environment Federal

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CHAPTER 1

INTRODUCTION

1.1 Introduction

Malaysia is experiencing a rapidly in population mobility due to economic changes and urbanization. Most of the population living in urban areas has a good access to a variety of social services and economic opportunities. The rapid growth of industrial and commercial around the world leads effects in increases of both municipal and industrial solid waste generations. Generally, landfill is a site for management of solid waste which provides a part of an economically and environmentally satisfactory method for the disposal of solid waste throughout the world. Up to 95% of solid waste generated worldwide is currently disposed in landfill (Bohdziewicz and Kwarciak, 2008). According to 10th Malaysia Plan 2011-2015, Malaysia has a population of 28.9 million people in 2012 and expected to increase to 29.8 million people in 2015. Malaysian produced 15000 -18000 tonnes of waste per day and statistic shows waste produced are increasing every year and total estimation of waste 7,772,402 tonnes per year in 2015 (Department of Statistics, 2011). According to 9th Malaysia Plan (2006-2010), in Malaysia the components of municipal solid waste (MSW) are categorized for several components such as food waste (45%), plastic (24%), paper (7%), iron (6%), glass and others (3%) (Tarmudi *et al.*, 2009). Percentage composition of solid waste is different between areas because it is influenced by several factors such as age of the landfill, hydrogeology of the site, quality and quantity of solid waste, site climate, season, biological and chemical processes occurring in the landfill and the amount of precipitation and percolation of rainwater, landfill morphology, waste depth, landfill condition and operation of facilities (Li *et al*., 2010; Bohdziewicz and Kwarciak, 2008). Recently, the problem of solid waste at the landfill site is an important issue for the environmental protection. In the landfill processing, the solid waste will undergo physical-chemical and biological changes. During decomposition of landfill processes, municipal solid waste (MSW) will produce leachate and gas emissions. Leachate is known as high strength wastewater formed as a result of percolation of rainwater and moisture through solid waste (Bohdziewicz and Kwarciak, 2008; Amuda and Os, 2006) and may cause significant pollution such as dissolved oxygen depletion, increasing toxicity of water bodies and eutrophication. Landfill leachate lead environmental problem when it comes polluted of surrounding soil, groundwater and surface water. Landfill leachate contains a high load of organic matter, high content of nitrogen (mainly ammonia or nitrogen), solids, halogenated hydrocarbon, inorganic salt and heavy metal (Tatsi *et al*., 2003). In fact, ammonium ions in landfill leachate will be toxic to aquatic life when it appears as an ammonia species at high pH value.

Landfill leachate can be generated in otherwise dry or arid environment where groundwater is located far below the landfill site. Variations in leachate composition occur for a wide range of reasons. The information of landfill leachate characteristic is required for the control of landfill function and design and operation of leachate treatment facilities.

Collection and treatment of landfill leachate before discharge must be implemented in order to meet the required effluent standard. It is because, the discharge of landfill leachate can affect the environment as they may percolate through soils and sub soils, causing extensive pollution of ground and also surface water if untreated properly and safely disposed. Consequently, selection and design of landfill leachate treatment depend on leachate quality, final discharge requirement and economic aspects. To minimize toxicity and hazardous material content of landfill leachate before it discharged into nature environment, there are various methods used to treat leachate such as biological treatment, chemical and physical treatment, advanced oxidation process (AOP) and combined treatment. The most popular treatment of landfill leachate was biological treatment which is anaerobic digestion or aerobic activated sludge method (Lin and Chang, 2000).

Among AOPs, oxidation using Fenton's reagent is an attractive and effective technology for the degradation of a large number of hazardous and organic pollutants. This technology has promising environmentally friendly. Combined treatment is considered as modular or multistage units skills in the leachate treatment and usually used advanced biological treatment and chemical or physical. The combined treatment was offered a good performance in the removal of pollutants in leachate.

Hence, this research was studying the effectiveness landfill leachate treatment by combination of electro-Fenton and sequencing batch reactor (SBR) method on the removal SS, colour, COD and NH_3-N. This treatment could be as an alternative method to treat the landfill leachate properly and safely disposed to nature environment.

1.2 Problem statement

Groundwater pollution identified as a major problem in most countries in the world that exists in a sanitary landfill (Pujari *et al*., 2007). Decomposing waste at the landfill site create major environmental problems, such as production of liquid known as leachate. Landfill leachate categorized as high strength wastewater with contains large variables of organic, inorganic compounds and heavy metal (Wang *et al*., 2010; Tyrell *et al*., 2002). The water quality affected when leachate move downwards from landfill through storm water runoff, evapotranspiration or infiltration into groundwater and surface water. The high concentration of ammonia and heavy metals in leachate can inhibit the biological microorganism process because they can survive in the long term in leachate. This problem affected on the water quality and surface water. The quality of landfill leachate is different depends on several factors such as composition and depth of waste, moisture and oxygen content, design and operation of landfill sites and lifespan of solid waste. According to Nasir and Chong, (1999) found that 71.4% of local authorities facing serious groundwater pollution and 57.2% comes from dealing of leachate management. Due this problem, Malaysia was facing a difficult to obtain a satisfactory for landfill leachate treatment. The type of landfill leachate treatment depends on characteristics of a landfill site. The technology used for landfill leachate treatment can classified as biological, physical and chemical methods. Thus, landfill leachate treatment desires selection and design of treatment depends on leachate quality, final discharge requirement and economical aspects. Landfill leachate treatment requires a high cost and the composition of leachate need to identify, especially for high concentration of landfill leachate pollution (Ozturk *et al.*, 2003). Young leachate is easily to treat compared to stabilized leachate (Kargi and Pamukoglu, 2003). Biological treatment is suitable for landfill leachate containing high of organic content and the high concentration of volatile fatty acid (Trebouet *et al*., 2001). Meanwhile, physical and chemical treatment is suitable for treat landfill leachate containing low of volatile fatty acid because the presence of heavy metals in leachate inhibited the biological process whereas the effectiveness of microorganisms affected. Thus, the physical techniques as a pre-treatment have been found to be suitable not only for the removal of refractory substances from stabilized locate but also as a refining step for biological treatment. In the past decades, the most popular treatment of the landfill leachate was biological treatment which is anaerobic digestion or aerobic activated sludge method (Lin and Chang, 2000). Year after year, the alternative treatment has been studied such as advanced biological, chemical or physical treatment method and the processes are usually used in combination for better treatment efficiency. In the recent years, advanced oxidation process (AOPs) for landfill leachate treatment has been widely applied by Guo *et al*., (2010); Atmaca, (2009); Tizaoui *et al*., (2007); Deng and Englehardt, (2006). This application is able in degrades of pollutants in landfill leachate and most AOPs using a combination of strong oxidants with catalyst.

Rapid economic development and population increases, Malaysia faced with the challenge in managing the domestic and industrial solid waste. The local corporations have been applied vary methods for waste disposal such as open dumps, landfills, sanitary landfills as well as incineration plants. However, landfill methods faced with the problem of leachate management. In Malaysia, landfill leachate pollution is a major problem that must be addressed immediately. To reduce the effects of leachate pollution, the solid waste management, sustainable and effective landfill leachate treatment should be given serious attention. Leachate treatment is very complicated, expensive and generally requires multiple processes. The processes currently used often require combined techniques which are designed as modular or multistage units skilled in the treatment of contaminants which vary in concentration over the years. But, in Malaysia, the number of currently available information and studies on leachate treatment is quite limited and under review.

Therefore, the selection of the appropriate treatment method is very important to reduce operating cost, simple and safely dispose to the natural environment as well as consideration for environmental and public health aspects. Hence, this study was mainly focused on the determination of the effectiveness combined electro-Fenton and sequencing batch reactor method in landfill leachate treatment in order removal of SS, colour, COD and NH_3-N. Coagulation-flocculation widely used as a pre-treatment and relatively simple technique that may be employed successfully for the landfill leachate treatment. Electro-Fenton is one of an economical treatment. Also, it is the simple method which there is no mass transfer limitation due to its homogenous catalytic nature, both hydrogen peroxide and iron are cheap and non-toxic and there is no form energy involved as a catalyst (Guo *et al*., 2010; Li *et al*., 2010). Due to reliability and high cost effectiveness, biological treatment is commonly used for a reduce leachate pollutant that containing high concentration of BOD and COD and yields reasonable treatment performance on the removal of pollutants. Sequencing batch reactor (SBR) method has lower initial capital cost since less civil works during construction which means no clarifies and other requirements (Al-Rekabi *et al*., 2007).

1.3 Objective of study

The main objective of this research was to determine the effectiveness of landfill leachate treatment by the combination of electro-Fenton and sequencing batch reactor (SBR) method for the removal of suspended solid (SS), color, chemical oxygen demand (COD) and ammonia nitrogen (NH_3-N). To achieve this objective, the study, through several stages includes the following objectives:

a. To characterize the landfill leachate from Simpang Renggam Municipal Landfill Site.
b. To optimize the coagulant dose of polyaluminum chloride (PAC) and ferric chloride ($FeCl_3$) as a pre-treatment on the removal of SS, colour, COD and NH_3-N.
c. To investigate optimum conditions of electro-Fenton such as current density, reaction time, pH, hydrogen peroxide (H_2O_2) dose and ferrous sulphate heptahydrate ($FeSO_4 \bullet 7H_2O$) on the removal of SS, colour, COD and NH_3-N.
d. To investigate optimum conditions of SBR method such as mixed liquor suspended solid (MLSS) and reaction time on the removal of SS, colour, COD and NH_3-N.
e. To compare the final effluent with leachate discharge standard (EQA 2009).

1.4 Significant of study

Currently, Malaysia has an increasing number of population and incredible urbanization influence directly the municipal solid waste (MSW) generation. Thus, all local authorities were facing discriminating problems in the collection and disposal of MSW generation. The problems included shortage of adequate funds, manpower problem, lack of disposal sites, not properly in the management system and lack of expertise (Tarmudi *et al.*, 2009). Due to financial constraints, landfill usually lacks of environmental abatement measure such as leachate collection system and lining materials (Ismail and Manaf, 2013). Leachate is produced when water percolates through waste and containing organic, inorganic, high nitrogen content and heavy metal (Tatsi *et al.*, 2003). Landfill leachate is considered as the greatest environmental concern in solid waste management. In sanitary landfill, the leachate treatment should to treat with proper to avoid affected by water body, surface water and underground water (Daud, 2008). Currently, several of leachate treatment were studied by some researcher and shown the effectiveness of the removal of leachate pollutant. But, in Malaysia, the number of currently available information and studies on leachate treatment is quite limited and under review. Due the leachate treatment is complicated and requires a high cost there need to study combined treatment, including many processes such as aerobic- anaerobic decomposition, chemical oxidation, coagulation-flocculation and adsorption are applied as an alternative treatment offers a good performance in removal of leachate pollutants. This study was determined the effective combination of physicochemical and biological processes which involved coagulation-flocculation process as pre-treatment, electro-Fenton process sequencing batch reactor (SBR) method. Hence, the results obtained from this study may be used as references for students or researchers who enthusiastic to develop new methods in landfill leachate treatment in advanced.

1.5 Scope of study

This study was to determine the effectiveness of landfill leachate treatment by combination of electro-Fenton and sequencing batch reactor (SBR) method. The landfill leachate used in this study was taken from Simpang Renggam Municipal Landfill Site in Johor. The test parameter was investigated on the removal of suspended solid (SS), colour, chemical oxygen demand (COD) and ammonia nitrogen (NH_3-N). The characteristic of landfill site was identified, such as BOD, COD, SS, colour, NH_3-N, pH, total phosphate and turbidity. In the coagulation-flocculation process as a pre-treatment, polyaluminum chloride (PAC) and ferric chloride ($FeCl_3$) was used as coagulant. Coagulation–flocculation studies performed in a standard Jar- test apparatus by Phipps & Birds model PB-900. In the optimize of PAC and $FeCl_3$ dose, the range was selected for 0, 500, 1000, 1500, 2000, 2500, 3000, 3500, 4000, 4500 and 5000 mg/L.

In the electro-Fenton process was used two types of electrode made of aluminium (Al) and stainless steel (St) in bipolar (Al-Al and St-St) electrode modes. Electro-Fenton studies performed in a 1000 ml NICE glass beaker with the diameter of 11 cm by vertically positioned Al-Al and St-St electrode spaced by 5 cm which carried out in batch mode. The electrode dimension of each electrode was 20 cm x 5 cm x 1 mm. Total working area of electrodes is 0.010 m^2 when immerse 10 cm into leachate sample. The distance between electrodes and settling time was kept constant at 5 cm and 30 minutes respectively. Electro-Fenton process was investigated for the optimum conditions of current density, reaction time, pH, hydrogen peroxide (H_2O_2) dose and ferrous sulphate heptahydrate ($FeSO_4 \bullet 7H_2O$). The range determination of current density was selected for 50, 100, 150, m^2, 250 and 300 A/ m^2. The range determination of reaction time was selected for 5, 10, 15, 20, 25, 30, 40, 45 and 60 minutes. The range determination of pH was selected for 2, 3, 4, 5, 6, 7, 8, 9, 10, 11 and 12. The range determination of H_2O_2 dose was selected for 200, 400, 600, 800, 1000, 1200, 1500, 2000 and 2500 mg/L while Fe^{2+} dose was kept constant at 1200 mg/L. The range determination of Fe^{2+} dose was selected for 200, 400, 600, 800, 1000, 1200, 1500, 2000 and 2500 mg/L while H_2O_2 dose was kept constant at 600 mg/L.

In sequencing batch reactor (SBR) method was applied in aerobic condition. SBR studies performed in the SBR reactor are designed for height is 31.5 cm, inside diameter is 15 cm, depth is 28.5 cm and working volume is 3 L. The range determination of the MLSS was selected for 1000-7000 mg/L and the range determination of reaction time was selected for 2, 4, 6, 8 and 10 h. The dissolved oxygen (DO) was kept constant above 2 mg/L and the five cycles was at filled 0.5 h, react 2, 4, 6, 8 and 10 h, settle 2 h and decants 0.5 h. The procedure for all test parameters was repeated triplicate to get an average value.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Due the increasing population, various developments, industrial and material consumption around the world, have been resulted in an associated increase in the quantity of industrial and municipal solid waste generation (Bashir *et al*., 2009; Renou *et al*, 2008). Commonly, municipal solid waste (MSW) refers to waste collected and treated by municipalities which includes waste from households, commerce and trade, office buildings, institutions and small businesses, yard and garden waste, street sweeping and industrial cleansing waste.

Hence, the proper treatment of industrial and municipal solid wastes needs in considering constituting to environmental protection (Hur *et al*, 2000). Landfills create a major environmental problem as well as the production of a liquid from decomposes waste known as a leachate when precipitation infiltrates. (Ifeanyichukwu, 2008) which contained a large amount of contaminant. Landfill leachate can categorize as a high strength wastewater to consist of many chemicals which is a potential risk to human health (Abdul *et al*., 2009; Ifeanyichukwu, 2008). The leachate generated from municipal solid waste (MSW) are often disposed without any appropriate control facility, although the possibility that they may cause contamination of soil, surface and ground water (Abdul *et al*, 2009; Gupta and Singh, 2007; Heng *et al*., 2012; Rivas *et al*., 2004).

Nowadays, various option an alternative of technology for landfill leachate are used to treat leachate properly and efficiently. There are four major groups for leachate treatment technologies are leachate transfer including recycling, lagooning and combined treatment with domestic sewage, biodegradation including aerobic and anaerobic process, physico-chemical process including chemical oxidation, adsorption, chemical precipitation, coagulation/flocculation and air stripping and lastly membrane filtration including microfiltration, ultrafiltration, nanofiltration and reverse osmosis (Renou *et al*., 2008).

2.2 Leachate

Leachate defined as liquid from rain water that has percolated through solid waste. Leachate from landfill usually contains extracted, dissolved and suspended materials, some of which may be harmful. Thus, landfill leachate can be toxic, acidic and rich in organic group such as chemical oxygen demand (COD), biological oxygen demand (BOD) and ammonium (Renou *et al*., 2008). As a leachate is one of the environmental issues since it is potential health effect to human and biodiversity, landfill have to be designed to minimize the formation of leachate and amount of leachate. Therefore, landfill should design, planning and operation that involved the application of a variety of scientific, engineering and economic principles in control and minimize the leachate migration into natural environment.

2.3 Composition and characteristic of landfill leachate

When water percolates through solid wastes that are undergoing decomposition, both biological materials and chemical constituents are leached into solution. Leachate is composed of high concentration organic content (humic acid and fulvic acid) and inorganic compound (suspended solid, heavy metal, ammonia nitrogen and inorganic salts) substances. Consequently, these contaminants need to be removed due to their toxicity or become hazardous to the environment (Wiszniowski *et al*., 2006). Besides that, the chemical compositions of leachate vary due to factors such as age of the landfill, solid waste composition, hydrogeology of the site, site climate or season, biological and chemical processes occurring in the landfill and the amount of precipitation and percolation of rainwater, landfill morphology, waste depth, landfill condition and operation of facilities (Li *et al*., 2010; Bohdziewicz and Kwarciak, 2008).

Age of landfill is one of the main factors that will effects leachate characteristic (Goi *et al*., 2010; Renou *et al*., 2008). Table 2.1 shows the characteristic of landfill leachate versus the age of the landfill was reported by Li *et al*., (2010). In young landfill contains a large amount of biodegradable organic matter which means a rapid anaerobic fermentation occurs. This process is resulting in volatile fatty acids (VFA) as the major fermentation products. A high moisture content or water content in the solid waste depends on acid fermentation is enhanced (Wang *et al*., 2003). This early stage is known as acidogenic phase and release above 80 % of the organic contents (Li *et al*., 2010). The age landfill tends the biological decomposition of the deposited waste change from a shorter initial period to longer decomposition period with acidogenic and methanogenic phase (Li *et al*., 2010; Bashir *et al*., 2009). As a landfill becomes older, the methanogenic phase occurs. At this stage, microorganisms will expand in the waste and the volatile fatty acid (VFA) is changed to biogas (CH_4, CO_2). Then, the compound such as humic acid and fulvic acid resulted as a product. Consequently, leachate from the differing stages contain different their constituents. In general, young locate more polluted than old leachate which it tends to be acidic due to presence of volatile fatty acid with the pH normal at range about 6-7 or lower. Old leachate produces low COD and low biodegradability BOD_5/ COD at ratio < 0.1 (Bashir *et al*., 2009; Rivas *et al*., 2004).

The characteristics of the landfill leachate usually be characterized by the basic parameter is chemical oxygen demand (COD), five-day biochemical oxygen demand (BOD_5), BOD_5/ COD (biodegradability), total Kjeldahl nitrogen (TKN), nitrogen- ammonium ($N\text{-}NH_4$), pH, total dissolved solids (TDS), turbidity, alkalinity, colour, conductivity, salts, xenobiotic organic substances and heavy metal (Renou *et al*.,2008). Even though, leachate composition may differ broadly in aerobic, acetogenic, methanogenic and stabilization stages of the waste evolution, which are classified leachate into three major parts is young, intermediate and old (Li *et al*., 2010). Also, biological treatment is more effective for young leachate with a high

BOD_5/COD ratio and less effective in treating leachate from intermediate landfill with a low BOD_5/COD or high concentrations of toxic constituents (Abdul *et al.*, 2009). Collection and treatment of landfill leachate before discharge must be implemented in order to meet the required effluent standard. It is because, the discharge of landfill leachate can affect the environment as they may percolate through soils and sub soils, causing extensive pollution of ground and also surface water if untreated properly and safely disposed.

Table 2.1: Characteristics of landfill leachate versus the age of landfill (Li *et al.*, 2010).

Types of leachate	Young	Intermediate	Old
Age	<5	5-10	>10
pH	<6.5	6.5-7.5	>7.5
COD (mg/L)	>10,000	4000-10,000	<4000
BOD_5/ COD	0.5-1.0	0.1-0.5	<0.1
Organic Compounds	80% volatile fatty acid (VFA)	5%-30% VFA + Humic and fulvic acid	Humic and fulvic acid
Ammonia nitrogen (mg/L)	<400	N.A	>400
TOC/COD	<0.3	0.3-0.5	>0.5
Kjeldahl nitrogen (g/L)	0.1-0.2	N.A	N.A
Heavy metal (mg/L)	Low to medium	Low	Low
Biodegradability	Important	Medium	Low

N.A = Not available

2.4 Environmental pollution

Municipal solid waste (MSW) is a category of diverse waste which is generated from different sources such as residential, commercial, institutional facilities, construction and demolition activities, municipal services and agriculture. Landfills are today the most commonly used methods for waste disposal by far. In most landfills leachate is composed of the liquid that has entered the landfill from external sources such as surface drainage, rainfall, groundwater and water from underground springs and the liquid produced from the decomposition of waste. Landfill leachate became toxicity with containing a large amount of organic matter, inorganic compound, as well as heavy metal, high content of ammonium ions and inorganic salts (Isidori *et al*., 2003). Nowadays, many countries around the world is facing the challenge in the landfilling practice due to environmental pollution from leachate (Ismail and Manaf, 2013; Adeolu *et al*., 2011; McDougall *et al*., 2001). Landfill leachate often exceeds standard for drinking water and surface water. Commonly, landfill leachate has significant impending to polluted soil, surface water and groundwater. The frequent pathway for leachate to the environment is from the bottom of the landfill through the unsaturated soil layers to the groundwater and from groundwater through hydraulic connections to surface water. The significant potential of environmental pollution may also come from the discharge of leachate through treatment plant or from untreated leachate to nature environment. The most important factors influencing the pollution due to landfill leachate are the concentration and flux, the landfill sitting include the hydrogeological setting and the basic quality volume and sensitivity of the receiving groundwater and surface water (Ismail and Manaf, 2013). Landfill leachate frequently retained in the surrounding area of the landfill unless it reaches the surface and then leading to runoff. Thereby, groundwater moves down and pollutant migration from the landfill site increased (Singh *et al*., 2008). The major concern with the movement of leachate into the subsurface aquifer below unlined and lined landfills is the fate of the constituents found in leachate. Mechanisms that are operative in the attenuation of the constituents found in leachate as the leachate migrates through the subsurface soil include mechanical filtration, precipitation and co- precipitation, sorption, gaseous exchange, dilution and dispersion and microbial activity. The migration of leachate through compression and compaction thereby cause of refuse to reach field capacity. Field capacity is referring to the quantity of water that can be held against the pull of gravity. The potential quantity of leachate is the amount of moisture within the landfill in excess of the landfill filed capacity that leads contaminates underground water sources below (Ismail and Manaf, 2013). The climate is one factor that may also produce by leachate volume. According to Visvanathan *et al*., (2004) leachate production in warm climates is higher than in colder climates. Hence, the public should give awareness the environmental pollution due to leachate and need to control of leachate in landfills.

2.5 Landfill leachate management

The management of landfill leachate has become one of the main focus of the environmental management of landfill. When landfill site become older, there are no leachate or gas management facilities. This problem leads the land was contaminated and the problem can solved with a proper design of landfill site to control of leachate production. A number of alternatives have been used to manage the leachate collected from landfill, including leachate recycling, leachate evaporation, leachate treatment and discharge to municipal wastewater collection systems.

Leachate recycling an effective method for the treatment of leachate is to collect and recirculate the leachate through the landfill. During the early stages of landfill operation the leachate will contain significant amounts of organic, inorganic, ammonia content and heavy metal. When the leachate is recirculated, the constituents are attenuated by the biological activity and other chemical and physical reactions occurring within the landfill. Leachate recycling is the recovery of landfill gas that contains methane gas (CH_4). To avoid the uncontrolled release of gases when leachate is recycled for treatment, the landfill should be equipped with a gas recovery system (Tchobanoglous *et al*., 1993).

One of the simplest leachate management systems involves the use of lines leachate evaporation ponds. Leachate that is not evaporated is sprayed on the completed portions of the landfill. The lined leachate storage facility is covered with a geo membrane during the winter season to exclude rainfall. The accumulated leachate is disposed by evaporation during the warm summer months by uncovering the storage facility and by spraying the leachate on the surface of the operating and complete landfill (Tchobanoglous *et al*., 1993).

Leachate treatment is used when leachate recycling and evaporation is not used. Some pre-treatment or complete treatment will be required due to characteristics of leachate can vary significantly. The leachate treatment has been categorized as biological treatment and physical and chemical treatment. The treatment process selected will depend to a large extent on the contaminants to be removed. The type of the treatment facilities used depends primarily on the characteristic of the leachate and secondarily on the graphic and the physical location of the landfill. In the integrated leachate management system the leachate that moves down through the solid waste is first filtered as it passes the sand layer in the landfill and transported to lagoon which to reduce organic content and to control odors. The excess leachate is filtered as it passes through the shredded waste and the sand filter under drain system. The collected leachate is piped to a series of construction wetlands which to remove organic material, nutrients, heavy metals and other trace organic of leachate. The effluent from the wetlands is passed through a slow sand filter and then used for spray irrigation on the grass covered landscape at the landfill (Tchobanoglous *et al*., 1993).

Discharge to wastewater treatment plant such as biological process and physical and chemical process is one of the leachate management. This process may be required to reduce the organic content before the leachate can be discharged to the nature environment (Tchobanoglous *et al*., 1993).

2.6 Treatment of landfill leachate

Landfill leachate can be hazardous with a high contain of many chemicals. This contaminant is a potential effect to human health and the environment. Landfill has to design to minimize both the formation of leachate and the amount of leachate. Over the past years, many new methods have been applied such as biological treatment, physical and chemical treatment and also combined treatment.

In the past decades, the most popular treatment of the landfill leachate was biological treatment which is anaerobic digestion or aerobic activated sludge method (Lin and Chang, 2000). In biological treatment involved aerobic, anaerobic or anoxic path. Biological treatment was effective in removal of biodegradable compounds in leachate (Kargi and Pamukoglu, 2003). This process includes of activated sludge, rotating biological contractor, sequencing batch reactor (SBR), biologically aerated filter (BAF), lagoons, upflow anaerobic sludge blanket (UASB), anaerobic filter, moving bed biofilm reactor (MBBR) and membrane bioreactors (MBR). Many researchers were studies of physical and chemical treatment. Physical and chemical treatment is also used along with the biological method to improve treatment efficiency. This process includes coagulation and flocculation, precipitation, adsorption, floatation, chemical oxidation and ammonia stripping. In the recent years, advanced oxidation process (AOPs) for landfill leachate treatment has been widely applied by Guo *et al*., (2010); Atmaca, (2009); Tizaoui *et al*., (2007); Deng and Englehardt, (2006). Most AOPs use a combination of strong oxidants like ozone, oxygen or hydrogen peroxide with catalyst like transition metals, iron, semiconductor powders, radiation or ultrasound, for example O_3/UV, H_2O_2/UV, O_3/ H_2O_2/UV, H_2O_2/Fe (II), TiO_2/UV, and TiO_2/ H_2O_2/UV. Year after year, many researchers were studied in combination of biological and physical process are being considered as the greatest technologies for management of high strength effluents of leachate. Table 2.2 shows the treatment process of landfill leachate.

Table 2.2: Treatment process of landfill leachate

Treatment process		Application
Biological treatment	Activated sludge	Removal of organics
	Aerated lagoon	Removal of organics
	Rotating biological contractor (RBC)	Removal of organics
	Sequencing batch reactor (SBR)	Removal of organics
	Upflow activated sludge bed reactor (UASB)	Removal of organics
	Anaerobic filter	Removal of organics
Physical and Chemical treatment	Flotation	Removal of suspended matter
	Coagulation-flocculation	Removal of organics
	Adsorption	Removal of organics
	Chemical oxidation/ Advanced oxidation process	Removal of organics; detoxification of some inorganic species
	Air stripping	Removal of ammonia or volatile organics

2.6.1 Biological treatment

Biological treatment is commonly used for the removal of bulk of leachate encompassing high concentrations of BOD (Uygur and Kargi, 2004). The biological treatment process involved the optimum growth of microorganisms which are used to convert the colloidal, dissolved carbonaceous organic matter and inorganic element, such as nitrogen, phosphorus, sulfur, carbon and magnesium into cell tissue or into various gases (Wiszniowski *et al*., 2006). Biological treatment is very effective in removing organic and nitrogenous matter from young leachate which contains high of BOD/COD ratio at a value more than 0.5. Decomposition of microorganisms can degrade organic compounds to carbon dioxide and sludge under aerobic conditions and to biogas under anaerobic conditions (Renou *et al*., 2008). The parameters of biological process such as sludge retention time (sludge age), food microorganism ratio (F/M), and hydraulic retention time (HRT) and sludge volume index (SVI). These parameters allocate to evaluate the operating conditions of biological process (Wiszniowski *et al*., 2006). Biological treatment is allocated into aerobic and anaerobic process. Aerobic process involved of activated sludge, sequencing batch reactor (SBR), aerated lagoon and rotating biological contractor (RBC) while anaerobic process involved of, upflow activated sludge bed reactor (UASB), anaerobic filter and anaerobic fixed film reactor.

2.6.1.1 Activated sludge

Activated sludge is broadly applied in landfill leachate due the ability for removal of organic carbon, nutrients and ammonia content effectively. The process of activated sludge is suitable for treatment of young leachate. According to Ifeanyichukwu, (2008) the removal of COD were 87-97% and BOD were over 90%. The study by Aghamohammadi *et al.*, (2007) has been to investigate aerobic biodegradation of semi-aerobic leachate with and without powdered activated carbon (PAC) addition and conduct in laboratory-scale activated sludge reactors. The results obtained the removal of COD and colour were 49% and 50% respectively at the higher hydraulic retention time (HRT) of 22.2 d. Another study of activated sludge were reported by Kheradmand *et al*., (2010) showed low removal efficiency due of high organic load rate and require further treatment prior to the unit. From the results shown the system performance effectively in treating landfill leachate was achieved 94% removal efficiency of COD. Nevertheless, in the other recent decades, this treatment has been shown insufficient for landfill leachate treatment (Renou *et al*., 2008). The disadvantages included high capital cost, high energy consumptions, required skilled personnel, need for a longer aeration time, not easy to control the settling property of sludge as well as excess sludge production and microbial inhibition due to high ammonium nitrogen (Ifeanyichukwu, 2008; Renou *et al*., 2008; Wiszniowski *et al*., 2006).

2.6.1.2 Aerated lagoon

In the past decades, aerated lagoon is widely applied to treatment of wastewater as well as leachate due its low cost method and efficient in removal of pathogens, organic and inorganic matters. Aerated lagoon treatment is ability to reduce organic matter between 93 to 97% (Daud, 2008). The study of aerated lagoon by Robinson *et al*., (1992) has been found that the removal of COD, BOD and ammonia were achieved 98%, 99% and 90% respectively. However, the temperature dependence of laggoning is a significant limitation because it affected by microbial activity (Reno *et al*., 2008).

2.6.1.3 Rotating biological contractor (RBC)

The rotating biological contractor (RBC) also known as a biorotor is an attached growth technology. In the recent years, the rotating biological contractor (RBC) for landfill leachate has been applied because this process produces low energy consumption, simplicity, required less maintenance and monitoring (Ifeanyichukwu, 2008; Wiszniowski *et al*., 2006). According to Castillo *et al*., (2006) the removal of COD was obtained 52% at a HRT of 24 h and rotational speed of 6 rpm. A study by Spengel and Dzombek, (1991) reported a COD, BOD and ammonia removal of 38%, 80% and 98% respectively. Nevertheless, this process is not suitable for acetogenic leachate due the excess sludge growth that may be clogging of interstices within rotors.

2.6.1.4 Upflow activated sludge bed reactor (UASB)

The process UASB is most widely investigated in different scales and form of anaerobic digester since it used in landfill leachate treatment and wastewater treatment. This process is a modern anaerobic treatment that can have high treatment efficiency and a short hydraulic retention time (Lin *et al*., 2000). This process involves an upward passage of leachate through an anaerobic sludge bed in a tank. The study of UASB process by Chen and Christensen, (2010) has indicated the removal efficiency achieved more than 75%. In the findings by Neena *et al*., (2007) have been used a UASB reactor in laboratory scale to treat landfill leachate from coconut husk has indicated 82 % of total COD/kg coconut husk could be converted to biogas. However, UASB may be slow down the digestion of heavy metal and also high ammonium remains in the effluent may result ammonia toxicity.

2.6.1.5 Anaerobic filter

Anaerobic filter is an additional form of anaerobic digester. This process is able to elimination of secondary clarifiers removing all the related cost and operational problem. The growth of anaerobic bacteria can establish upon due to filter medium and then degrade organic substances in leachate moves slowly across that medium. A study by Wu *et al*., (2004) has been obtained 98% removal of BOD and 91% removal of COD at high concentration of organic matter. Wang and Banks, (2007) were study on anaerobic filter for leachate treatment have been obtained the removal of COD was between 75 to 90% and over 88% removal of sulphate.

2.6.2 Physical and chemical treatment

Physical and chemical treatment is applied along with the biological process to improve treatment more efficient. In physical and chemical process are used for removing non-biodegradable such as humic, fulvic acid and heavy metal from the leachate (Renou *et al.*, 2008). Also ability to reduce of suspended solids, colloidal particles, floating material, colour and toxic components. This process included of flotation, coagulation-flocculation, adsorption, chemical oxidation/advance oxidation process (AOP) and air stripping. In the treatment of leachate, physical and chemical processes are used in pre-treatment or last purification or to treat a specific pollutant such as stripping for ammonia. Currently, a study of advanced oxidation process (AOP) have been reported by researchers such as conventional Fenton, photo-Fenton, and electro-Fenton are powerful used in order to increase efficiency of electrochemical methods that effectively degrades refractory organics and persistent organic pollutants (POPs), which are difficult to degrade by another process such as bio-degradation (Abdul *et al*., 2009).

2.6.2.1 Floatation

Floatation process is able to reduce of colloids, ions, macromolecules, microorganisms and fibers (Rubio *et al*., 2002). This process has a good performance to remove humic acid after biological treatment which is the removal over 99% (Ifeanyichukwu, 2008). A study by Zaoboulis *et al*., (2003) indicated about 60% humic acid removal on the study of floatation in the column as a post-treatment step for removing residual humic acids.

2.6.2.2 Adsorption

Adsorption is one of the physical and chemical processes for landfill leachate treatment and sometimes used in biological process (Kargi and Pamukoglu, 2003). Generally, the most widely used as adsorbent is granular (columns) or powdered activated carbon (powder) will perform an effective removal efficiency of COD than the use of chemical method (Kargi and Pamukoglu, 2003; Zamora *et al*., 2000). The function of activated carbon adsorption is to ensure the final polishing level by removing toxic heavy metals or organic content and to support microorganisms. Moreover, some of the researchers were studying adsorption by using zeolite, vermiculite, illite, keolinite, activated alumina and municipal waste incinerator bottom ash. The study of adsorption by Kargi and Pamukoglu, (2003) has been investigated pre-treated leachate used coagulation-flocculation and air-stripping of ammonia. They have been used adsorbent of PAC and powdered zeolite. The results showed, the COD removal efficiency has achieved 87% and 77% with PAC and zeolite respectively. A study by Harvey, (2003) about the adsorption method by using peat as adsorbent obtained 100% removal efficiency of BOD and 69% of COD removal.

2.6.2.3 Chemical oxidation/advance oxidation process (AOP)

Chemical oxidation is suitable for leachate containing refractory organic compound such as soluble organics that cannot be removed by physical separation and biological oxidation. This process involves the presence of oxidants which directly react and mineral contaminants. The types of oxidants are chlorine, potassium permanganate, calcium hydrochloride, ozone with hydroxyl radicals generated when hydrogen peroxide is added to leachate in the presence of ferrous salts as is the case in the Fenton process (Wiszniowski *et al*., 2006). Year after year, advanced oxidation process (AOP) is widely applied for landfill leachate treatment due to its ability to remove organic compounds efficiently. The removal efficiency of COD obtained between 45 to 85% (Ifeanyichukwu, 2008).

2.7 Treatment with the coagulation-flocculation process

2.7.1 Mechanism of coagulation-flocculation

Coagulation-flocculation has been used as a pre-treatment for landfill leachate before the application of biological processes to remove non-biodegradable organic components (Aguilar *et al*., 2011). Coagulation and flocculation is a simple technique that may be applied effectively for the treatment of older landfill leachate (Yilmaz *et al*., 2010). The coagulation-flocculation process is used to destabilize colloidal materials in water with the addition of chemical agent. The process of coagulation and flocculation occurs in successive steps intended to overcome the forces stabilizing the suspended particles allowing particle collision and growth of flock. Coagulation indicates the process through which colloidal particles and very fine solid suspensions are destabilized and begin to agglomerate if the conditions are appropriate. Coagulation targets the colloid particles of size 10^{-7} to 10^{-14} cm in diameter. The colloid particles exhibit Brownian movement through the water, their surface is negatively charged so they repel one another and they form a stable dispersed suspension. If colloid particles or ions of positive electric charge are added it neutralizes the electric negative charge. Flocculation is the formation of aggregates of the destabilized colloids and need gentle mixing to allow effective collisions between particles to form heavy flocks which can be moved from water by settlement (Baghvand *et al*., 2010; Norulaini *et al*., 2001). The agglomerates formed by coagulation are compact and loosely bound, while the flocks formed by flocculation process are larger size, strongly bound and porous (Tripathy and De, 2006). It agglomerates of a few colloids then quickly bridge together to form microflocs which is turned into visible floc masses (Sahu and Chaudhari, 2013; Gregory, 2006). The effectiveness of the process depends on coagulants agent, dosage of coagulants, pH, rapid mixing requirement and ionic strength (Zazouli and Yousefi, 2008; Norulaini et al., 2001). The mechanism of coagulation is shown in Figure 2.1.

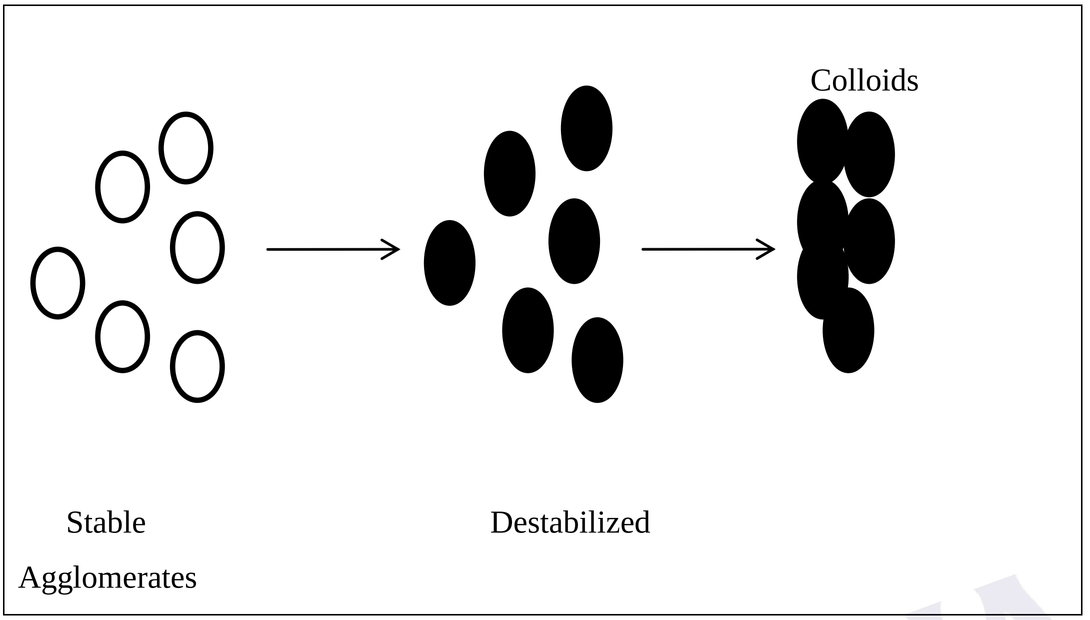

Figure 2.1: Mechanism of Coagulation (Sahu and Chaudhari, 2013; Gregory, 2006)

The removal of organics is associated with the removal mechanism of humic substances by the application of coagulation- flocculation. It is well known that humic substance can be effectively removed from the aqueous solution by adding hydrolyzing coagulants. There involved two main mechanisms regarding the removal of humic substances from coagulation- flocculation (Duan and Gregory, 2003). The first is binding of cationic metal species to anionic sites, results the neutralization of humic substances and the reduction of their solubility. The second mechanism is adsorption of humic substances onto the produced amorphous metal hydroxide precipitates. While, there is difficult to distinguish between the two mechanisms, it appears strongly pH dependent as for the solution pH values approximately 5.5-6.5 the humic substances are negatively charged whereas Al hydroxide is positively charged and resulting to strongly adsorption and charge neutralization.

2.7.2 Coagulants

The common coagulants used in wastewater treatment can be classified into inorganic coagulants (aluminium sulphate, polyaluminum chloride and ferric chloride), synthetic organic polymers (polyacrylamidde derivatives and polyethylene imine) or naturally occurring coagulants (plant extracts and chitosan) (Zainol et al., (2011). Mostly, coagulant of aluminium salts and iron salts are widely used for wastewater treatment. Besides that, the used of coagulants such as aluminium sulphate (alum), ferrous sulphate, ferric chloride are widely used in treating of wastewater (Silva *et al*., 2004; Wang *et al*., 2002). In the recent years, polyaluminum chloride (PAC) as a coagulant is broadly used in treating of wastewater (Rui *et al*., 2012; Lofrano *et al*., 2005). Frequently, aluminium sulphate (alum) is more widely used to treat water and wastewater because low of costly, stable of total dissolved solids and easily to handle compared with iron salts (Daud, 2008). When aluminium sulphate (alum) is added into wastewater that has alkalinity the reaction occurs as shown in equation 2.1.

$$Al_2(SO_4)_3 \cdot 14H_2O + 3Ca(HCO_3)_2 \rightarrow 2Al(OH)_3 + 3Ca(SO_4) + 14H2O + 6CO_2 \quad (2.1)$$

Aluminium sulphate (alum), ferrous sulphate, ferric chloride is effective in a pH between of 4-11 (Daud, 2008; Bratby, 2006). According to Daud, (2008) the pH of iron salts are effective in between of 5-8.5 and normally at pH 7.5. When ferric chloride and ferric sulphate is added into wastewater that has alkalinity, the reactions occur as shown in equation 2.2 and 2.3.

$$2FeCl_3 + 3Ca(HCO_3)_2 \rightarrow 2Fe(OH)_3 + 3Ca(SO_4) + 6CO_2 \quad (2.2)$$

$$Fe_2(SO_4)_3 + 3Ca(HCO_3)_2 \rightarrow 2Fe(OH)_3 + 3Ca(SO_4) + 6CO_2 \quad (2.3)$$

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