REMOVAL OF COLOUR, COD AND NH_3-N FROM SEMI-AEROBIC SANITARY LANDFILL LEACHATE USING SULFONIC ACID AND QUATERNARY AMINE FUNCTIONAL GROUP RESINS

by

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DEDICATION

This thesis is dedicated to my beloved parents for their prayers and tremendous sacrifices. They are always a constant source of inspiration and motivation in my life. Their support and love have pulled me throughout my difficult times.

Mohammed J. K. Bashir

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LIST OF ABBREVIATION

AF	Anaerobic filters
ANOVA	Analysis of variance
AOPs	Advanced oxidation processes
AP	Adequate precision
APS	Activated sludge process
ASBR	Anaerobic sequencing batch reactor
BET	Brunauer-Emmett-Teller
BOD	Biochemical oxygen demand
CCD	Central composite design
CEC	Cation exchange capacity
COD	Chemical oxygen demand
CV	Coefficient of variance
DAF	Dissolved air flotation
DoE	Design of experiment
EBCT	Empty bed contact time
EC	Electrochemical oxidation
EL	Electrostatics interactions
FO	Fenton oxidation
FTIR	Fourier transform infrared
GAC	Granular Activated Carbon
HRT	Hydraulic retention time
MAP	Magnesium ammonium phosphate
MBBR	Moving-bed biofilm reactor
MBR	Membrane bioreactor
MF	Microfiltration

MSW	Municipal solid waste
MTZ	Mass transfer zone
NF	Nanofiltration
NH_3-N	Ammoniacal nitrogen
NPM	Non-polar part of organic
OH•	Hydroxyl radicals
PAC	Poly-aluminum chloride
PBLS	Pulau Burung Landfill Site
pH	Hydrogen ions
PRESS	Predicted Residual Sum of Squares
R^2	Coefficient of determination
R^2_{Adj}	Adjusted Coefficient of determination
R_L	Dimensionless equilibrium parameter
RO	Reverse osmosis
RSM	Response surface methodology
SBR	Sequencing batch reactor
SD	Standard deviation
SEM	Scanning electron microscopes
SG	Specific gravity
SS	Suspended solids
t_b	Breakthrough time
UASB	Upflow anaerobic sludge blanket
UF	Ultrafiltration

PENYINGKIRAN WARNA, COD DAN NH_3-N DARI LARUT LESAPAN KAMBUS TANAH SANITARI SEMI-AEROBIK MENGGUNAKAN RESIN KUMPULAN BERFUNGSI ASID SULFONIK DAN KUARTENARI AMINA

ABSTRAK

Aplikasi proses penukaran ion dalam olahan larut lesapan kambus tanah masih belum banyak dibincangkan dalam literatur. Keadaan-keadaan optimum pengoperasian dan interaksi di antara pemboleh ubah dalam proses olahan ini merupakan di antara jurang pengetahuan yang masih belum dikenal pasti. Dalam kajian ini, olahan larut lesapan dari tapak kambus tanah stabil menggunakan resin– kation, anion, kation diikuti dengan anion (kation-anion), dan anion diikuti dengan kation (anion-kation) telah dijalankan dan didokumentasi buat pertama kalinya. Kaedah permukaan respon (RSM) secara rekabentuk pusat komposit (CCD) digunakan bagi mendapatkan proses olahan yang optimum dan juga menilai kesan-kesan individu dan interaksi pemboleh ubah operasi terhadap keberkesanan penyingkiran setiap aplikasi dari segi warna, keperluan oksigen kimia (COD) dan nitrogen ammonia (NH_3-N). Model yang sesuai bagi menerangkan isoterma dan kinetik oleh kedua-dua resin kation dan anion juga ditentukan. Sampel larut lesapan diambil dari Tapak Pelupusan Pulau Burung (PBLS), Pulau Pinang, Malaysia dan dicirikan. Ujikaji dijalankan pada skala makmal. Keputusan ujikaji menunjukkan, resin kation adalah media berkesan untuk penyingkiran NH_3-N. Dos optimum kation, masa sentuhan optimum dan kelajuan goncangan optiumum dicapai masing-masing pada 24.0cm^3, 10 min dan 150 rpm, di mana 68.9% warna, 38% COD dan 91.8% NH_3-N berjaya disingkirkan. Keputusan ini menunjukkan resin kationik sahaja tidak mampu untuk menyingkirkan warna dan COD dengan berkesan. Sebaliknya, resin anion sahaja adalah media yang berkesan untuk menyingkirkan warna dan COD. Keadaan optimum berlaku pada 35.0 cm^3 dos anionik, 74 min masa sentuhan, 150 rpm kelajuan goncangan dan pH 3.3. Keadaan ini menghasilkan penyingkiran masing-masing 91.7%, 70.7% dan 11.8% untuk warna, COD dan NH_3-N. keputusan menunjukkan, penggunaan resin anion sahaja mampu untuk menyingkirkan warna dan COD dengan berkesan. Walau bagaimanapun, resin anion kurang berkesan untuk penyingkiran NH_3-N. Data penyingkiran keseimbangan untuk NH_3-N oleh resin kationik selain dari warna dan COD menggunakan penukaran anion amat berpadanan dengan isoterma penjerapan lurus Langmuir dan Freundlich. Namun begitu, data yang didapati bagi kedua-dua model untuk penjerapan NH_3-N ke atas resin anion dan warna dan COD keatas resin kation tidak berpadanan. Keputusan yang didapati bagi setiap model kinetik mematuhi persamaan pseudo aturan-kedua, menandakan kadar reaksi serapan dikawal oleh mekanisma aturan-kedua (serapan kimia). Untuk olahan kationik-anionik, keputusan ujikaji menunjukkan penyingkiran optimum untuk warna, COD dan NH_3-N adalah masing-masing 96.8%, 87.9% dan 93.8%. Sebaliknya, aplikasi olahan anionik-kationik masing-masing menghasilkan 91.6%, 72.3% dan 92.5% penyingkiran. Nilai R^2 adalah 0.8727, 0.9487 dan 0.9987 untuk model penyingkiran warna, COD dan NH_3-N. Keputusan kajian ini menunjukkan bahawa aplikasi turutan kationik-anionik untuk olahan larut lesapan semi aerobik tapak kambus tanah stabil adalah lebih berkesan berbanding turutan anionik-kationik. Masa bolos (t_b) yang dijangkakan di tapak pelupusan sebenar menggunakan penuras kation dan anion tanpa proses pra-olahan adalah masing-masing 5.2 dan 124 hari. Walau bagaimanapun, penggunaan proses enap cemar teraktif diikuti pengoksidaan elektrokimia sebagai proses pra-olahan sebelum penapisan anion-kation boleh meningkatkan t_b penapisan kation dan anion kepada masing-masing 30 dan 650 hari.

REMOVAL OF COLOUR, COD AND NH_3-N FROM SEMI-AEROBIC SANITARY LANDFILL LEACHATE USING SULFONIC ACID AND QUATERNARY AMINE FUNCTIONAL GROUP RESINS

ABSTRACT

The application of ion exchange process in landfill leachate treatment was not well established in literature. Optimized operational conditions and the interaction among process variables for this treatment process were unidentified, leaving a substantial gap in landfill leachate treatment knowledge. In the present study, the treatment of stabilized landfill leachate using resin- cationic, anionic, cationic followed by anionic (cationic-anionic), and anionic followed by cationic (anionic-cationic) were established and documented for the first time. Response surface methodology (RSM) concerning central composite design (CCD) was used to optimize each treatment process and to evaluate the individual and interactive effects of operational variables on the effectiveness of each application in terms of colour, chemical oxygen demand (COD) and NH_3-N removal efficiencies. Suitable model describing the isotherms and kinetics for both cationic and anionic resins were determined. The stabilized landfill leachate samples were collected from Pulau Burung Landfill Site (PBLS), Penang, Malaysia and characterized. Experiments were performed at laboratory scale. According to the results, cationic resin was an effective media for NH_3-N removal. The optimum cationic dosage, contact time and shaking speed were found to be 24.0 cm^3, 10min and 150rpm, respectively at which 68.9% colour, 38% COD and 91.8% NH_3-N removals were achieved. The results showed that cationic resin alone was not able to remove colour and COD effectively. Anion resin alone was an effective media for colour and COD removals. The optimized conditions occurred at 35.0cm^3 anionic dosage, 74 min contact time, 150 rpm shaking speed and pH 3.3. These conditions resulted in 91.7, 70.7 and 11.8% removal of colour, COD and NH_3-N, respectively. The results indicated that utilizing anion resin alone was effective for colour and COD reduction. However, anion resin was inadequate for NH_3-N removal. Equilibrium removal data for NH_3-N by cationic resin besides colour and COD by anion exchanger fitted well with Langmuir and Freundlich linear adsorption isotherms. However, the data obtained from both models for NH_3-N adsorption on anion resin and colour and COD on cationic resin was incompatible. The results obtained from each kinetic model showed good compliance with the pseudo-second-order equation indicating that the rate of the sorption reaction was controlled by the second-order mechanism (chemical sorption). For cationic-anionic treatment, the experimentally achieved optimum removal of colour, COD and NH_3-N were 96.8, 87.9 and 93.8%, respectively. However, the application of anionic-cationic treatment resulted in 91.6, 72.3 and 92.5% removal, respectively. The values of R^2 were 0.8727, 0.9487 and 0.9987 for colour, COD, NH_3-N removal models, respectively. Consequently, the results imply that the application of the cationic-anionic sequence for the treatment of semi-aerobic stabilized landfill leachate was more effective than the anionic-cationic sequence. The predicted breakthrough times (t_b) to be used at the active landfill site for cation and anion filters, without pretreatment process, were 5.2 and 124 days, respectively. However, using activated sludge process followed by electrochemical oxidation prior to anion-cation filtrations can increase the t_b of cation and anion filters to 30 and 650 days, respectively.

CHAPTER 1

INTRODUCTION

1.1 Background of Study

The population, material consumption and various developmental activities have resulted in a concomitant increase in the amount of municipal solid waste. At least a portion of the solid waste eventually reaches landfills. In urban areas of Southeast Asian Nations (ASEAN), solid waste is one of the most noticeable environmental problems. Since the late 1980s, the ASEAN region has experienced rapid urban growth which has resulted in significant increase in the overall MSW generation. Since many cities are not capable to manage increasing quantities of MSW due to institutional, economical, technological, regulatory, knowledge, and public participation shortcomings, increased waste production has created severe environmental issues in this region. Inadequate management and disposal of waste ultimately results in environmental degradation and other associated problems (Ngoc and Schnitzer, 2009).

Abdul Rahman et al. (2009) reported that Malaysia generates about 6.2 million tons of solid waste per year, which amounts approximately 17000 tons per day. This amount is expected to increase to more than 30000 tons per day by 2020 due to the increase in population and per capita waste generation. Therefore, solid waste management is one of the major challenges for Malaysia to address in the light of 2020, when the country plans to become a fully developed nation (Abdul Rahman et al., 2009). In 2003, the average amount of MSW generated in Malaysia was 0.5-0.8 kg/person/day while in major cities it was as high as 1.7 kg/person/day

(Kathirvale et al., 2003). About 95 percent of the collected solid wastes in Malaysia are disposed in more than 230 landfills.

Sanitary landfilling remains the most prevalent method for the disposal of MSW as compared to other disposal techniques. Such prevalence can be attributed to its several advantages, such as ease of disposal, economically feasible and landscape-restoring effect on the holes from mineral workings (Williams, 2005). Additionally, landfilling is the only method that can deal with all kinds of solid wastes generated. However, the contamination of surface and groundwater through leachate, soil contamination through direct waste contact or leachate, air pollution through burning of wastes, spreading of diseases by different vectors such as birds, insects and rodents, odor and uncontrolled release of methane by anaerobic decomposition of waste are among the major disadvantages associated with this method of MSW disposal (Ngoc and Schnitzer, 2009). Unfortunately, the generation of landfill leachate is rapid in tropical countries such as Malaysia because the rainfall generally exceeds the amount that can be evaporated during the rainy season (Lema et al., 1988).

1.2 Problem Statement

Landfill leachate is defined as a liquid that become polluted by percolating through the waste within the landfill site (Kurniawan et al., 2006). The resulting liquid contains high concentration of pollutants which can have adverse effects on the environment (Renou et al., 2008; Tchobanoglous and Kreith, 2002). These pollutants include biochemical oxygen demand (BOD), chemical oxygen demand (COD), NH_3-N, suspended solids (SS), heavy metals and inorganic substances. If not properly treated and safely disposed, landfill leachate could be a potential source of surface and groundwater contamination as it may percolate through soils and sub-soils. Hence treatment of landfill leachate is considered an essential step prior to its discharge (Tatsi et al., 2003; Aziz et al., 2004a).

Typically, leachates from stabilized landfills contain lower levels of pollutants compared to young leachate (age< 5yr.). Typically, young leachates are characterized by high BOD_5 (4000–40,000 mg/L), high COD (6000–60,000 mg/L), NH_3-N (<400), BOD_5:COD ratio typically ≤ 1.0, and pH range from 4.5 to 7.5 (Alvarez-Vazquez et al., 2004; Ehrig, 1983). Studies have shown that landfills older than 10 years produce stabilized leachates with low biodegradability. In this stage, leachates produce large amounts of non-biodegradable organic compounds such as humic and fulvic substances. Stabilized landfill leachates are normally characterized by moderately high strengths of COD (500–4500 mg/L), low BOD (20–550 mg/L), high NH_3–N (>400), a pH range of 7.5–9.0, and a BOD_5:COD ratio of <0.1 (Alvarez-Vazquez et al., 2004; Ehrig, 1983). Due to its characteristics, a stabilized leachate is difficult to treat using biological processes (Rivas et al., 2004; Renou et al., 2008).

This study focuses on the treatment of stabilized landfill leachate generated from the Pulau Burung semi-aerobic landfill site. Pulau Burung landfill site (PBLS) is situated within Byram Forest Reserve at 5º 24' North Latitude, 100º 24' East Longitude in Penang, Malaysia, which is around 20 km southeast from the Penang Island. This landfill produces a dark colour liquid with pH level more than 7.0 and is classified as stabilized leachate with high concentration of COD, NH_3-N, and low BOD_5/COD ratio (Aziz et al., 2007).

Although the leachate was treated, the effluent characteristics still did not comply with Malaysian Standard (Aziz et al., 2010). Various characteristics of PBLS leachate in particular colour, COD, BOD and NH_3-N have been reported in many studies (Aziz et al., 2004a, 2007, 2009; Ghafari et al., 2009; Mohajeri et al ., 2010 a,b; Palaniandy et al., 2010). Previous studies have reported that the concentrations of colour, COD and NH_3-N in leachate from this landfill site are high. The BOD_5/COD values between 0.04-0.17 have also been reported in literature (Aghamohammadi et al., 2007). Because of its low BOD_5/COD ratio and high concentration of NH_3-N, raw PBLS leachate is recognized as highly stabilized with low biodegradability. The high concentration of colour, COD, and NH_3-N can be classified as a typical problems associated with stabilized landfill leachate. The toxicological effects of these parameters onto the ecosystem are well established (Jokela et al., 2002; Kurniawan et al., 2006; Karadag et al., 2008). In Malaysia, The leachate should be treated properly for the above mentioned parameters to achieve compliance with the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, under the Laws of Malaysia-Malaysia Environmental Quality Act 1974.

The choice of a treatment technique is strictly based on the type of leachate (young or old) and hence the characteristics. The biological treatment is effective in removing organic matters from young leachate when the BOD_5/COD ratio> 0.4 (Kurniawan et al., 2006). In contrast, biological treatment has several limitations when it comes to the treatment of stabilized leachate due to the narrow range of biodegradable components in leachate, the presence of substances toxic for the microorganism (Li et al., 1999) and the limited availability of necessary nutrients for microbial growth (Li et al., 1999; Amokrane et al., 1997). Therefore, physico-chemical processes are recommended for the treatment of stabilized landfill leachate. Several physico-chemical treatment processes have been applied to treat landfill leachate including chemical oxidation, chemical precipitation, coagulationflocculation; activated carbon adsorption and membrane filtration, and ion exchange (Kurniawan et al., 2006).

Ion exchange resins have been widely used in water and wastewater treatment for extraction, separation, and purification of organic substances (Jorgensen, 2002; Cavaco et al., 2007; Kiefer et al., 2007). Ion exchange technology was investigated as a polishing step in the treatment of landfill leachate (Primo et al., 2009). However, a literature review reveals that the studies on the removal of colour, COD, and NH_3-N from stabilized landfill leachate using ion exchange resin as pretreatment process remain limited. The same is true for studies concerning the optimization of ion exchange treatment process for the studied parameters removal from leachate using design- expert software that could statistically design experiments and analyze related data.

In the present study, the treatability of stabilized landfill leachate via anion and cation ion exchange resins was investigated. Furthermore, the performance of anion and cation ion exchangers was investigated based on different sequences of treatment systems.

1.3 Research Objectives

The research focused on colour, COD, and NH_3-N removal from stabilized landfill leachate.

The main objectives of the present study included the followings:

1. To investigate appropriateness of different ion exchange resins with different mobile ion forms and different treatment sequences for the treatment of semi-aerobic stabilized landfill leachate.

2. To develop the equation of colour, COD and NH_3-N removal efficiency (as a response) from stabilized leachate with respect to process conditions (i.e., dosage, contact time, shaking speed, and pH) using response surface methodology RSM and central composite design (CCD) and to determine the optimum operational conditions of studied applications via RSM.

3. To determine the adsorption capacity using the best fit isotherm model and to find out the adsorption kinetics. Moreover, to evaluate suitable models describing the isotherms and kinetics for both cationic and anionic resins

4. To investigate the effectiveness of the exhausted media regeneration. Also, to examine the performance of the media in column study, individually and in sequence.

1.4 Layout of the Thesis

The rest of the thesis is divided into the following chapters:

Chapter 2 **Literature review:** A comprehensive review of literature surrounding the sciences of solid waste management, landfill, landfill leachate, landfill leachate treatments, ion exchange materials, process theory, applications and influencing factors, and implementations of RSM for parameters optimization is presented in this chapter.

Chapter 3 **Materials and Methods**: This chapter presents the experimental programs and procedures of both batch and column studies. In addition, to the site location and characteristics, materials properties and samples preparation are presented in this chapter. This chapter also describes the main methods used to determine leachate properties, operational variables, process optimization using RSM, and the physical and chemical characteristics of cationic and anionic rein used in this study.

Chapter 4 **Results and Discussions:** This chapter provides characterization of leachate and extensively discussed the optimum removal efficiency of colour, COD, and ammoniacal nitrogen from leachate as obtained from batch and column experiments using cationic and anionic exchange resins both individually and in various treatment sequences. The equations of removal for colour, COD, and ammoniacal nitrogen removals in terms of individual process parameters and their interactions are presented and discussed. Further, Freundlich and Langmuir isotherms, adsorption kinetics and regeneration results obtained from the experiments are presented and elaborated.

Chapter 5 **Conclusions and Recommendations:** The conclusions and recommendations based on the study finding are discussed and directions for future work are suggested.

CHAPTER 2

LITERATURE REVIEW

This chapter consists of six sections. The first section provides general overview of MSW sources and management. The second section describes a landfill and its types while the third section gives an overview of leachate characteristics. The fourth section reviews different methods of leachate treatment including physico-chemical treatment. The fifth section focuses on the ion exchange resins in terms of their types, properties, process theory and application in wastewater treatment while the last section gives a general outline of experimental design and analysis using RSM.

2.1 Solid Waste

Solid waste generally refers to unwanted solid materials produced as a result of human and animal activities. With respect to this definition, many items can be considered as waste such as domestic rubbish, sewage sludge, wastes from manufacturing activities, packaging items, discarded cars, discarded electronic devices, garden waste, old paint containers etc. Accordingly, all our daily activities can give rise to a large variety of different wastes arising from different sources (Ngoc and Schnitzer, 2009). Based on their sources, solid wastes can be classified into various types which include MSW, hazardous waste, agricultural waste and industrial waste (Tchobanoglous et al., 1993).

The present study focuses on the treatment of leachate generated from MSW landfills. MSW represents waste collected from households, in addition to the commercial waste collected by a municipality but it generally excludes hazardous wastes. Because urbanization and modernization have rapidly increased the rate of MSW production and disposal in many cities of the world, the management of MSW has emerged as a major concern around the world particularly the rapidly developing countries. According to Tanaka (2006), the generation of solid waste is expected to increase steadily along with economic growth (Figure 2.1).

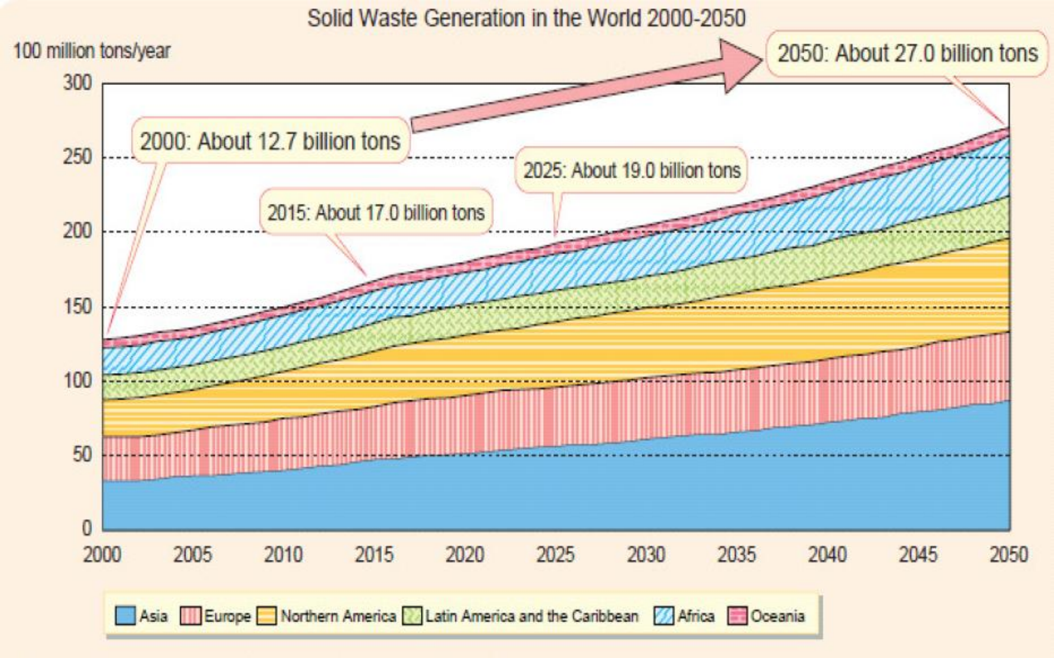

Figure 2.1: Predicted solid waste generation (Tanaka, 2006)

As illustrated in Figure 2.1, the amount of global waste generation is increasing all around the world. This is mainly because of the rapid economic growth in developing countries, especially in Asia, and the associated generation of huge amounts of wastes. Therefore, waste management problems become more serious (Tanaka, 2006). In Malaysia, Chong et al. (2005) reported that solid waste is one of the biggest environmental problems in Malaysia and the generation rate of solid waste is expected to increase tremendously due to the rapid increase in population and economic growth in the country.

2.1.1 Composition of MSW

Idris et al. (2004) reported that the data on composition of wastes is critical for the formulation of new waste management plans. Waste minimization can not be carried out effectively without having reliable waste composition data. Sufficient waste composition data is required to evaluate the impacts of certain types of wastes and to estimate the life of landfills. However, reliable data on solid waste composition is difficult to obtain and even if available, it is often not updated (Idris et al., 2004). The composition of wastes varies from time to time and place to place. Table 2.1 shows the MSW waste compositions from Southern Asian Nations.

Asian countries with higher rural population produce more organic waste and a smaller amount of recyclables such as paper, plastics and metals as shown in Table 2.1.

Table 2.1: MSW waste compositions in Southern Asian Nations

Country	**Waste composition (%)**					
	Organic waste	Paper cardboard	Plastic	Glass	Metal	Others
Brunei	44	22	12	4	5	13
Cambodia	55	3	10	8	7	17
Indonesia	62	6	10	9	8	4
Laos	46	6	10	8	12	21
Malaysia	62	7	12	3	6	10
Myanmar	54	8	16	7	8	7
Philippines	41	19	14	3	5	18
Singapore	44	28	12	4	5	7
Thailand	48	15	14	5	4	14
Vietnam	60	2	16	7	6	9

Source: Ngoc and Schnitzer., 2009.

2.1.2 Integrated MSW Management and Waste Management Hierarchy

MSW management is strongly required in order to deal with the huge daily amounts of solid waste. Solid waste management involves the overall management of activities associated with waste such as generation, storage, collection, transportation, processing, reuse, recycling and disposal (Hamer, 2003; Khatib and Al-Khateeb, 2009). Normally, an integrated waste management approach is used to cover all these activities. Several types of solid waste management systems have been implemented in different countries. Most of the countries including Malaysia employ landfill as the most common method of MSW management. However, some countries have adopted different waste management schemes such as incineration. In Japan, incineration is the most common method of MSW management (Beychok, 1987). Solid waste management hierarchy classifies waste management strategies according to their desirability. As shown in Figure 2.2, the most common methods used for MSW management are waste prevention, minimization, reuse, recycle, energy recovery and landfill disposal (UNDP, 2008).

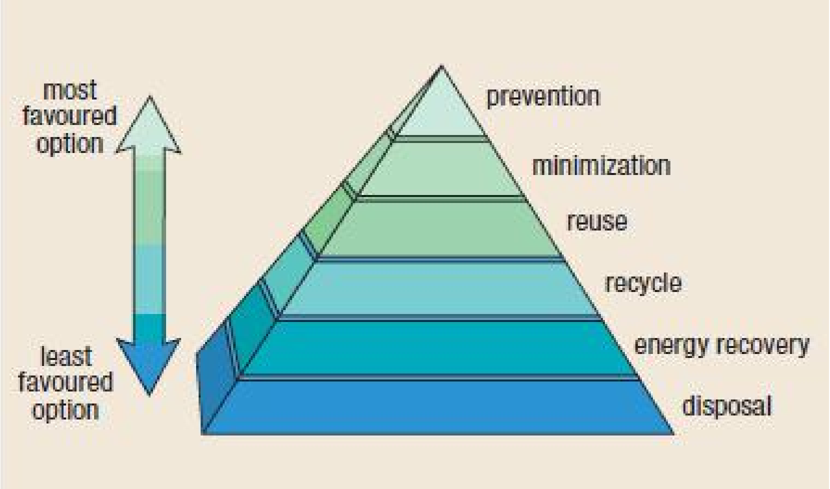

Figure 2.2: Solid waste management hierarchy (UNDP, 2008)

Sustainable waste management simply means managing waste by prioritizing as per the waste hierarchy (Butt et al., 2008). A proper waste source separation is expected to improve the overall recycling process. The waste hierarchy prioritizes prevention and source reduction of waste followed by reuse, recycling and the optimization of ultimate disposal (UNDP, 2008).

2.2 Landfill

2.2.1 General Overview

A landfill is like a big rubbish bin (JICA, 2005). In the last few decades, the disposal of waste to land has become the most favored means of waste management (Chong et al., 2005). Among others management alternatives, landfilling is the only management technique that is both necessary and sufficient. Several wastes are basically not recyclable and they finally reach a point where their intrinsic value is dissipated completely. Recycling also generates residuals that require ultimate disposal in a landfill (Messineo and Panno, 2008). The challenge is to ensure that all operating landfills are properly designed and monitored after their closure. Currently, operating landfills have gas control systems, liners, leachate collection and extensive groundwater monitoring systems (Theisen, 2002)

In Southeast Asian countries, open landfill sites are the most popular solid waste disposal method which is primarily due to their ability to deal with high quantities of solid waste generated each day (Ngoc and Schnitzer, 2009). Further, landfilling provides economical MSW disposal and it is also suitable for the type of wastes which constitute higher percentage of organic matter. However, landfills in many places in ASEAN are classically unsanitary open disposal sites without a leachate management system. Figure 2.3 illustrates the waste management methods used in ASEAN.

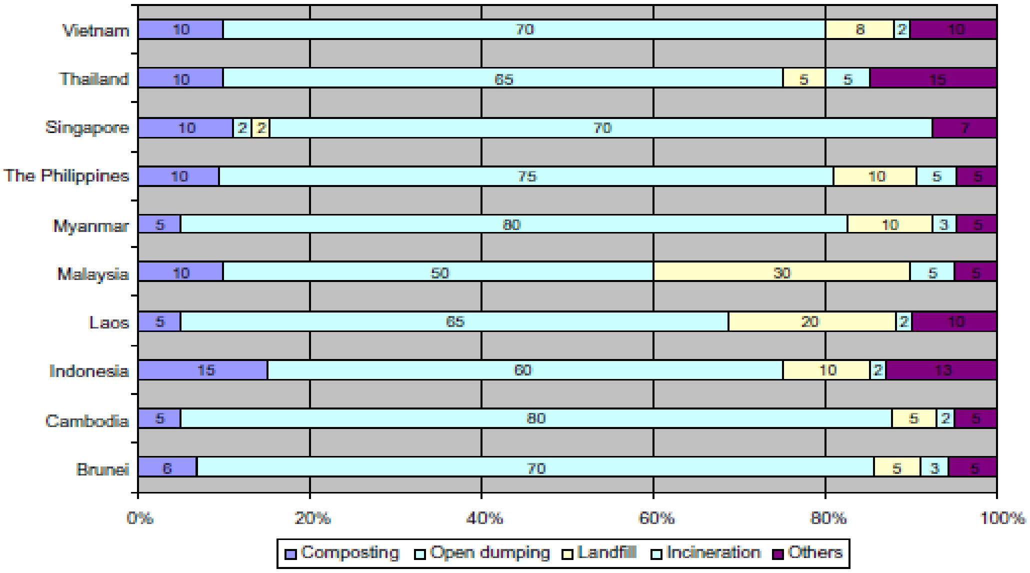

Figure 2.3: Solid waste management methods in ASEAN (Ngoc and Schnitzer, 2009)

Ngoc and Schnitzer (2009) mentioned that only 80% of solid waste in Malaysia disposed in landfill. In contrast, according to the information recorded by UNDP (2008) and Oh et al. (2010), MSW in Malaysia involves the disposal of approximately 95-98% of the total MSW to landfills. Oh et al. (2010) reported that there are more than 261 landfill sites in Malaysia of which approximately 150 sites are still operating

2.2.2 Sanitary Landfill

Sanitary landfilling is the principal method for MSW disposal in most countries (Wiszniowski et al., 2006). According to JICA (2005), sanitary landfill is a method of disposing refuse on land without creating nuisances or hazards to public health or safety.

Careful preparation of the fill area including the use of clay and/or synthetic liners and control of water drainage is required to ensure proper landfilling. Heavy equipment is used to spread, compact and cover the daily waste with at least 6 inches of compacted soil to confine the solid waste to the smallest practical area and reduce it to the smallest practical volume (Tchobanoglous et al., 1993). The soil cover application is very effective in the prevention of environmental pollution. It is important to maintain a landfill site clean and sanitary and maximize its capacity by good operation (Tchobanoglous et al., 1993). Sanitary landfills have leachate collection systems, methane gas controls and environmental monitoring systems (Diaz et al., 2002).

2.2.3 Classification Systems of Landfill Sites

In Malaysia, two classification systems for landfill sites are used which are introduced in the following Sections (2.2.3.1 and 2.2.3.2).

2.2.3.1 Landfill Sites Classification System Based on the Operational Purposes

Unfortunately, most of Malaysian landfills are not classified as sanitary landfills because they lack facilities to collect or treat the leachate and there is no infrastructure to exploit the landfill gas. According to Agamuthu (2001) the desired level of improvement of sanitary landfill system can be achieved in 4 stages:

Level 1: controlled tipping

Level 2: sanitary landfill with a bund (embankment) and daily soil covering

Level 3: sanitary landfill with a leachate recirculation system

Level 4: sanitary landfill with leachate treatment facilities

The latest assessment of the landfill sites in Malaysia was carried out in 2002 and the results demonstrated that there were 77 open dumps, 49 controlled tipping landfills (level 1) and only 35 levels 2, 3 and 4 landfill sites. The results also showed that the largest number of open dumps was in Sarawak followed by Johor, Sabah and Kelantan (Idris et al., 2004).

2.2.3.2 Landfill Sites Classification System based on the Landfill Structure

The second classification system is based on the landfill structure. Yamamoto, (2002) and Matsufuji et al. (1993) reported that landfill sites are classified according to their structure into 5 types as shown in Table 2.2.

Table 2.2: Classification of landfill structure

No	Type	Characteristics
i	Anaerobic landfill	Solid wastes are filled in excavated area of plane field or valley. Wastes are filled with water under anaerobic conditions.
ii	Anaerobic sanitary landfill	Anaerobic landfill with sandwich like cover shape. Conditions of solid waste are same as the anaerobic landfill
iii	Improved anaerobic sanitary landfill	This has leachate collection system at the bottom of the landfill site. The condition is still anaerobic and the moisture content is much less than the aerobic sanitary landfill
iv	Semi-aerobic landfill	Leachate collection duct is bigger than the improved sanitary landfill. The opening of the duct is surrounded by air and the duct is covered with small crushed stones. Moisture content in solid waste is lower. Oxygen is supplied to solid waste from leachate collection duct.
v	Aerobic landfill	In addition to the leachate collection pipe, air supply pipes are attached and air is enforced to enter the solid waste by which condition becomes more aerobic than semi-aerobic.

Source: Environmental Pollution Control Center, Osaka Prefecture, 2009).

2.2.4 Semi-aerobic Landfill (Fukuoka method)

This section focuses on the semi-aerobic landfill type (Fukuoka method) and its suitability to be implemented in the tropical countries. The first semi-aerobic landfill was developed by Fukuoka university Japan in 1975 (Matsufuji, 2004). According to Hanashima (1999), semi-aerobic landfills are suitable for Asian tropical weather conditions where leachate treatment and management are significant issues. This method has been practically tested in many countries such as Japan, Malaysia, Iran and China (Chong et al., 2005). Shimaoka et al. (2000) reported that creating aerobic atmosphere in an anaerobic landfill makes it possible to control the generation of methane gas and to reduce the amount of pollutants in leachate. Thus, a semi-aerobic landfill is graded between anaerobic and aerobic landfill. The mechanism of Fukuoka method for semi-aerobic landfill is shown in Figure 2.4. In Fukuoka method, the air is allowed to inflow through leachate collection pipes which lay at the bottom of the landfill leading to extended aerobic conditions. The aerobic conditions improve the quality of leachate by lowering the level of leachate concentration and reducing the generation of hazardous gases, all of which lead to faster stabilization of the landfill as shown in Figure 2.5.

The advantages of the semi-aerobic landfill type as listed by the Environmental Pollution Control Center, Osaka Prefecture and JICA, (2005) include the followings:

i. Leachate is discharged after collection which leads to reduced seepage of leachate
ii. The introduction of fresh air through the pipes lead to faster stabilization of waste, improves leachate quality and reduces the cost of final treatment of leachate.

iii. The release of gas from gas ventilation pipes reduces gas pressure and the chance of gas explosion

iv. The waste stabilization is enhanced, hence reduces the time for the reuse of completed landfills (for vegetation, open space, parks, recreation etc.)

v. It helps reducing the global warming by lowering the generation of CH_4 while increasing the concentration of CO_2. Global warming potential of CH_4 is about 25 times more than that of CO_2.

vi. The initial investment and maintenance cost of semi-aerobic landfill is lower than that of aerobic type of landfill which makes semi-aerobic landfill economically more feasible.

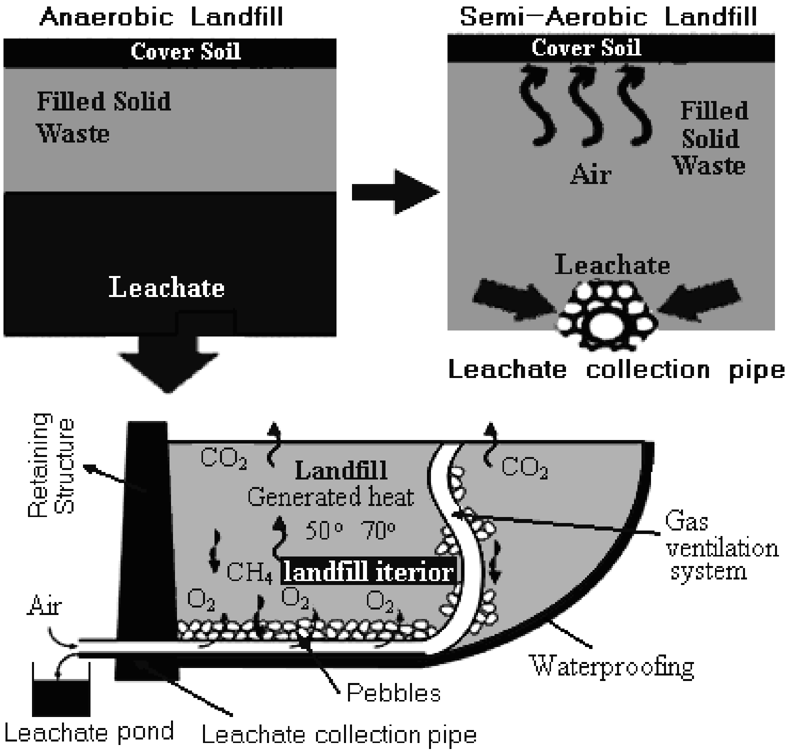

Figure 2.4: Mechanism of semi-aerobic landfill, Fukuoka method (JICA, 2005; Environmental Pollution Control Center, Osaka Prefecture, 2009)

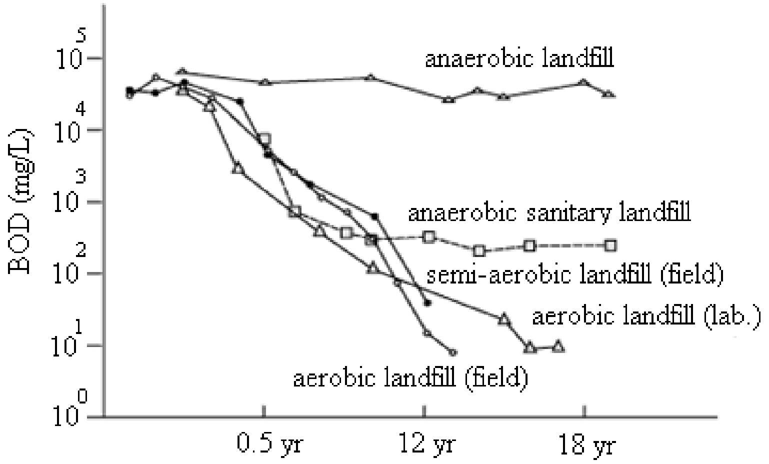

Figure 2.5: Change in the BOD concentration of leachate by landfill type (JICA, 2005; Shimaoka et al., 2000)

2.2.5 Principals of Decomposition in Landfill

Solid waste in a sanitary landfill undergoes various physical, chemical and biological degradation processes immediately after the landfill is covered. The processes initiate directly after the solid waste is placed in a landfill and continue until the end of landfill life. The decomposition period of solid waste normally depends on the waste characteristics while solids, liquids and gases are produced. These products should be considered in the landfill management system.

Physical decomposition of solid waste occurs in the operation solid waste management systems include, component separation, mechanical volume reduction and mechanical size reduction. Physical decomposition does not involve a change in phase (solid to gas), unlike chemical and biological decomposition processes (Tchobanoglous et al., 1993). Chemical decomposition of solid waste classically involves a change of phase. The chemical decomposition processes achieve a reduction in volume and convert wastes to new products. The principle chemical processes consist of combustion, pyrolysis and gasification (Tchobanoglous et al., 1993). Biological degradation processes include the aerobic and anaerobic degradation. According to Crawford and Smith (1985) the decomposition of landfilled waste is carried out by bacteria in five phases (Figure 2.6).

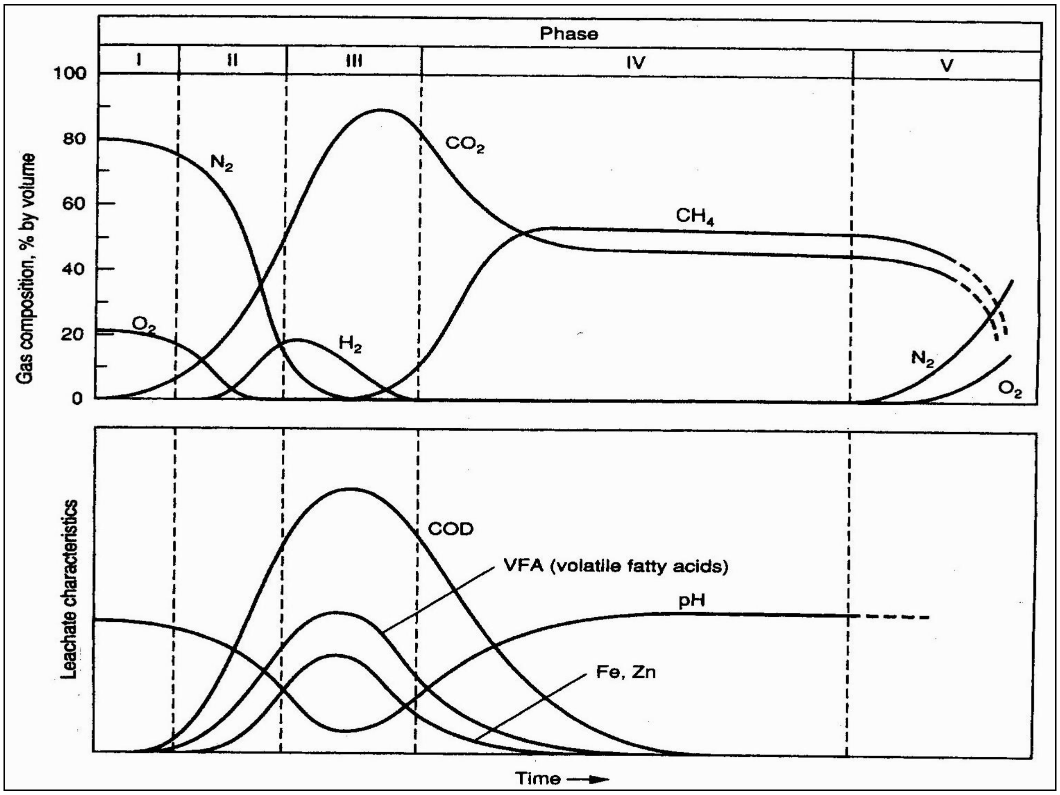

Figure 2.6: Phases of landfill gas composition and leachate characteristics (Tchobanoglous et al., 1993)

According to Pohland et al. (1985), the rate and characteristics of waste and biogas generated from a landfill vary from one phase to another and reflect the microbially mediated processes taking place inside the landfill. The rate of progress during these stages is dependent on the physical, chemical and microbiological conditions developed within a landfill over time.

2.3 Landfill Leachate

2.3.1 Leachate Definition

Normally MSW landfills release various types of contaminants in the surrounding environments including gas emissions, liquid leachate and nondegradable solid materials. Rainfall is the main contributor to the generation of leachate. Other contributors to leachate generation include groundwater inflow, surface water runoff and biological decomposition. A widespread example of leachate formation is presented in Figure 2.7.

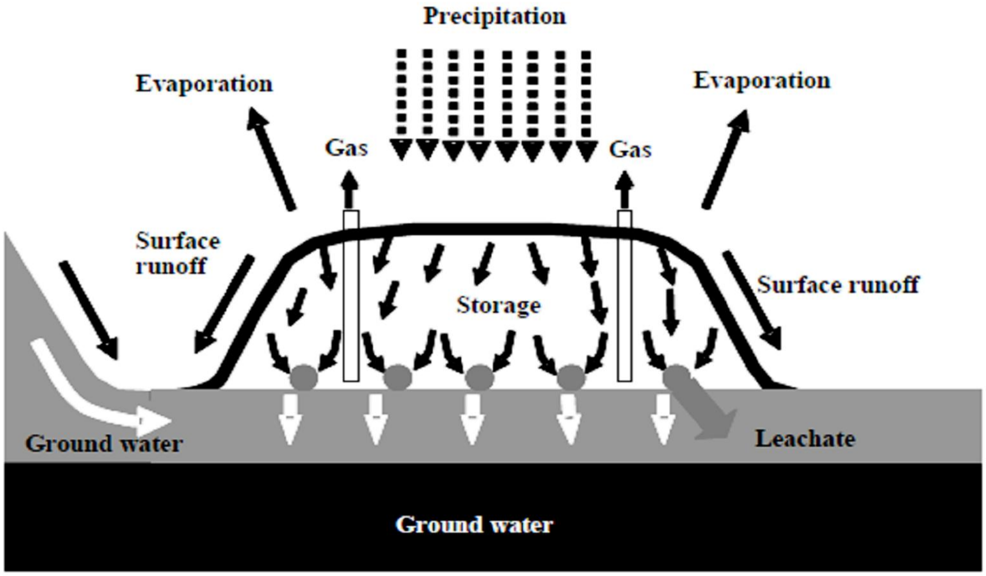

Figure 2.7: Water movements in the landfill (Wichitsathian, 2004)

In general, leachate represents the water that passes through the waste and water generated within the landfill site. The resulting liquid contains SS, soluble components of the waste and products from the degradation of the waste by various microorganisms (Williams, 2005; Tchobanoglous and Kreith, 2002). According to Bagchi (1990), leachate is defined as a liquid that has become polluted or toxic by percolating through the rubbish. Leachate contains many substances depending upon the types of waste disposed into a landfill (Gotvajn et al., 2009). If not properly treated and safely disposed, landfill leachate could be a potential source of surface and groundwater contamination as it may percolate through soils and sub-soils (Tatsi et al., 2003).

2.3.2 Leachate Composition and Characteristics

In general, leachate is highly contaminated with a mixture of organic substances (biodegradable and non-biodegradable carbon) and inorganic materials (heavy metals, sodium, calcium, sulphate and ammonia nitrogen) (Kang et al., 2002; Wang et al., 2002; Aziz et al., 2004a). During the first few years (< 6.5 years), the landfill is in acidogenic phase and the leachate produced is commonly known as "young leachate". Landfills older than 10 years are normally in the methanogenic phase and the leachate produced is referred to as "old leachate". Moreover, Tatsi et al. (2003) reported that leachate with high concentration of COD, nitrogen, turbidity and colour intensity are produced in methanogenic phase because of the biochemical reactions that occur during the percolation process. Christensen et al. (2001) claim that many parameters change dramatically as the landfill stabilizes.

The quality of landfill leachate can be demonstrated in terms of COD, BOD_5, total organic carbon (TOC), BOD_5/COD ratio, pH, SS, turbidity, NH_3-N, total Kjeldahl nitrogen (TKN), and heavy metals content (Gotvajn et al., 2009). The likely concentration of these parameters provides a precondition insight into the prediction of potential tendency of leachate quality, the design and operation (Foo and Hameed, 2009).

2.3.3 Seriousness of colour, COD, and NH_3-N at PBLS

The current study focuses on the leachate generated from Pulau Burung Landfill Site (PBLS). Although the leachate was treated, the effluent characteristics still did not comply with Malaysian Standard for such kind of wastewater (Aziz et al., 2010). Various characteristics of PBLS leachate in particular colour, COD, and NH_3-N have been reported in many studies (Aziz et al., 2004a, 2007, 2009; Aghamohammadi et al., 2007; Ghafari et al., 2009; Mohajeri et al ., 2010 a,b; Palaniandy et al., 2010).

Previous studies have reported that the concentrations of colour, COD and NH_3-N in leachate from this landfill site are high. A colour intensity between 2430-8180 Pt-Co, COD strength between 1533-3600 mg/L and NH_3-N concentration between 983-2117 mg/L has been reported by various authors (Aziz et al., 2007; Aghamohammadi et al., 2007; Ghafari et al., 2009; Mohajeri et al., 2010a; Palaniandy et al., 2010). The BOD_5/COD values between 0.04-0.17 have also been reported in literature (Aziz et al., 2004a; Aghamohammadi et al., 2007). Because of its low BOD_5/COD ratio and high concentration of NH_3-N, raw PBLS leachate is recognized as highly stabilized with low biodegradability. The high concentration of colour, COD, and NH_3-N can be classified as typical problems associated with landfill leachate. The toxicological effects of these parameters onto the ecosystem are well established (Jokela et al., 2002; Kurniawan et al., 2006; Karadag et al., 2008). In Malaysia, The leachate should be treated properly for the above mentioned parameters to achieve compliance with the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, under the Laws of Malaysia-Malaysia Environmental Quality Act 1974.

2.3.4 Factors Affecting Leachate Composition

Typically, leachate quantity and composition vary depending on the nature and partitioning of the landfill ages, climatic conditions, solid waste composition, moisture content, temperature, geological characteristics, groundwater inflow, surface water runoff, pH, availability of oxygen, microbial activity, local precipitation patterns, chemical equilibrium solubility, landfill design (size, depth top cover, bottom linear) and operation, vegetation and separate points of samplings (Christensen et al., 2001; Kjeldsen et al., 2002; Tatsi et al., 2003; Kargi and Pamukoglu, 2004). The most significant factors are discussed in more details (Section, 2.3.4.1-2.3.4.6).

2.3.4.1 Landfill Age

According to Aziz et al. (2007), age of a landfill is one of the main factors that affect leachate characteristics. As a landfill becomes older , the biological decomposition of the deposited wastes shifts from a relatively shorter initial period to longer decomposition period which has two distinct sub-phases, an acidic phase and a methanogenic phase. Leachates from these distinct stages contain different constituents and young leachates tend to be acidic due to the presence of volatile fatty acids. In UK, Alvarez-Vazquez et al. (2004) summarized the characteristics of different types of landfill leachate based on age (Table 2.3). In Malaysia, as a tropical country, the recorded characteristics of stabilized sanitary landfill leachate by Aziz et al. (2010), Ghafari et al. (2010), and Palaniandy et al. (2010) agree with the values in Table 2.3.

Generally, during the early settlement phase (acidogenic phase) of the disposed MSW young landfill leachate contains large amount of biodegradable and non-biodegradable materials, particularly in terms of volatile fatty acids (Renou et al., 2008; Kurniawan et al., 2006). At this stage, leachate is characterized by high concentrations of BOD_5 (4000-40000 mg/L), COD (6000- 60000 mg/L) and NH_3-N (500-2000 mg/L) while the BOD_5/COD ratio is typically ≤1.0 and pH range from 4.5 to 7.5 (Christensen et al., 2001; Alvarez-Vazquez et al., 2004).

Table 2.3: Characterization of different types of landfill leachate

Type of leachate	**Young**	**Intermediate**	**Stabilized**
Age of landfill (years)	<2	2-6.5	>6.5
pH	<6.5	6.5-7.5	>7.5
BOD/COD	0.5-1.0	0.1-0.5	<0.1
COD (mg/L)	>15000	3000-15000	<3000
NH_3-N (mg/L)	<400	NA	>400
TOC/COD	<0.3	0.3-0.5	>0.5
Kjehdal nitrogen (g/L)	0.1-2	NA	NA
Heavy metals (mg/L)	>2	<2	<2

Source: Alvarez-Vazquez et al., 2004.

At later stages (methanogenic phase), the biodegradable fraction of organic compounds are decomposed over a long period of time resulting in the production of stabilized leachate which has large amount of non-biodegradable organic compounds with high molecular weights such as humic and fulvic substances. Therefore, stabilized leachate cannot be treated using biological processes (Renou et al., 2008; Aziz et al., 2007). Stabilized landfill leachate is normally characterized by high concentrations of COD (5000-20000 mg/L), low BOD_5 (20-550 mg/L), pH range 7.5 to 9.0 and BOD_5/COD ratio of less than 0.1 (Christensen et al., 2001; Alvarez-Vazquez et al., 2004; Rivas et al., 2004).

2.3.4.2 Climatic Condition

Seasonal variations can considerably influence the nature of solid waste. For instance, in the rainy periods waste retains much moisture and is denser (Trankler et al., 2006; Kulikowska and Klimiuk, 2008). According to Trankler et al. (2001), in hot and humid weathers, leachate production is much higher and varies more than in hot and arid regions due to intensive microbial activity. Usually, rainfall gives the required moisture content for methane production and biological activity. During dry season, the leachate generation is very low because of the evaporation whereas in wet period, the leachate generation is associated to quantity of rainfall intensity. Furthermore, high rainfall leads to increased leachate production and reduced leachate strength. The influence of seasonal differences on the amount and composition of leachate varies from place to place and is also influenced by other factors. It is extremely essential to consider the hydrological and leachate quality data when suggesting a treatment plan in order to avoid environmental troubles caused by the direct discharge.

2.3.4.3 Solid Waste Composition

Kulikowska and Klimiuk (2008) stated that the quality of leachate varies widely due to the great variations in MSW composition and characteristics. Organic matters present in the waste mainly comprise of domestic waste while the inorganic components consist of plastic, glass, metal etc. The ratio of organic and inorganic components in the landfilled waste can highly influence the composition of leachate (Lu et al., 1985). As a result of hydrolysis and degradation of higher molecular weight organic compounds by microorganisms present in the waste, the organic content leaches into the leachate. Weerasekara et al. (2007) noticed that the leachate