TREATMENT OF STABILIZED LANDFILL LEACHATE USING BATCH ELECTROCOAGULATION WITH VIBRATION-INDUCED ELECTRODE PLATES

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by

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

Page

LIST OF PLATES

LIST OF SYMBOLS

A	Unit of current, Ampere
CO_2	Carbon dioxide gas
L	Unit of volume, Liter
mV	milivolts
pH	Unit measurement of acidity/alkalinity of solution
°C	Unit for temperature

LIST OF ABBREVIATIONS

Al $(OH)_3$	Aluminum hydroxide
BOD_5	Biological oxygen demand
$CaCO_3$	Calcium carbonate
COD	Chemical oxygen demand
DAF	Dissolved air flotation
DC	Direct current
DOE	Department of Environment
EC	Electrocoagulation
EC-DAF	Electrocoagulation-dissolved air flotation
EDX	Energy dispersive X-ray spectroscopy
FI	Flocculation index
IR	Internal resistance
MPPP	Penang Island City Council
MPSP	Seberang Perai City Council
MRF	Material Recovery Facility
MSW	Municipal solid waste
NaCl	Sodium chloride
NH_3-N	Ammoniacal nitrogen
PBSL	Pulau Burung Sanitary Landfill
PIV	Particle image velocimetry
PTFE	Polytetrafluoroethylene
ROI	Region of interest
RPM	Rotation per minute
TOC	Total Organic Carbon
TDS	Total dissolved solids
TN	Total nitrogen
TSS	Total suspended solids
UNDP	United Nations Development Program
VOA	Volatile organic acid
XRD	X-ray powder diffraction

LIST OF APPENDICES

RAWATAN LARUT RESAP STABIL MENGGUNAKAN KUMPULAN ELEKTROKOAGULASI MENGANDUNGI PLAT ELEKTROD DENGAN GETARAN TERINDUKSI

ABSTRAK

Di dalam proses elektrokoagulasi, peranan gelembung gas adalah penting kerana ia akan menentukan kecekapan pemindahan ion dan membantu dalam proses flokulasi dan pengapungan. Hidrodinamik gelembung gas dikatakan sebagai faktor yang penting untuk kecekapan penyingkiran yang tinggi dalam elektrokoagulasi, namun, kelemahan pada pengumpulan dan kemelekatan gelembung gas pada permukaan elektrod semasa proses rawatan belum dikaji secara menyeluruh dan peranannya tidak menjadi fokus utama terhadap pemnambahbaikan dan pengubahsuaian yang dibuat pada elektrokoagulasi dalam kajian sebelum ini. Kajian ini mengkaji prestasi elektrokoagulasi berkelompok yang dipasangkan dengan plat elektrod dengan getaran terinduksi dalam rawatan larut resap tapak stabil untuk meningkatkan penyingkiran warna, COD dan NH_3-N. Keamatan gelembung gas telah diperkenalkan di dalam reaktor melalui getaran elektrod menggunakan penggetar motor dan kesan intensiti getaran, pH awal, masa rawatan dan intensiti arus ke atas prestasi elektrokoagulasi telah dikaji. Kajian hidrodinamik seperti saiz gelembung gas, halaju kenaikan dan corak aliran dinilai menggunakan perisian PIVLab dan kamera berkelajuan tinggi digunakan untuk mengambil imej gelembung gas. Keputusan menunjukkan bahawa penyingkiran bahan pencemar tertinggi dicapai pada intensiti getaran optimum pada 2.8 V. Dengan plat yang bergetar, rintangan plat adalah 4%-20% lebih rendah daripada elektrokoagulasi dengan plat statik, oleh itu berkesan menyingkirkan 90% warna, (dengan kepekatan disingkirkan sebanyak 2462 mg/L),

37% COD (821 mg/L) dan 575 NH_3-N (154 mg/L). Saiz gelembung gas yang lebih kecil dengan luas permukaan yang tinggi dapat dibentuk dan dilepaskan sepenuhnya daripada permukaan plat pada intensiti getaran 1.8 V dan seterusnya. Halaju kenaikan gelembung didapati berkurang apabila plat bergetar dan halaju ini mencapai aliran mencapai aliran sekata pada intensiti arus tinggi dan dipengaruhi oleh kedua-dua komponen v- dan u-, yang bertentangan dengan hanya satu arah ke atas dalam elektrokoagulasi dengan plat statik. Daripada keputusan potensi zeta, elektrokoagulasi dengan plat getaran terinduksi menghampiri kadar koagulasi-flokulasi yang lebih pantas dengan data kinetik penyebaran Brownian adalah mengikut tindak balas urutan kedua dengan pemalar kadar 0.0001 $L.mg^{-1}s^{-1}$, dengan demikian, mekanisma flokulasi adalah dominan melalui peneutralan cas. Julat penggunaan tenaga elektrik yang dikira adalah dalam anggaran 0.47 kWh/m^3 ke 21.66 kWh/m^3 di mana ianya adalah 4% - 20% lebih rendah yang dapat dicapai oleh elektrokoagulasi dengan plat statik. Oleh itu, penggunaan plat dengan getaran terinduksi dapat dicadangkan sebagai alternatif kepada mekanisma pengadunan dalam elektrokoagulasi.

TREATMENT OF STABILIZED LANDFILL LEACHATE USING BATCH ELECTROCOAGULATION WITH VIBRATION-INDUCED ELECTRODE PLATES

ABSTRACT

In electrocoagulation, the role of gas bubbles is significant as it will determine the efficient mass transfer of the coagulant ions and help in the flocculation-flotation process. The hydrodynamics of gas bubbles are said to be the important factor of high removal efficiency in electrocoagulation, yet, the drawbacks on the accumulation and adherence of gas bubbles on the electrode surface during the treatment have not been thoroughly studied and their behaviour have not been the main focused in the previous enhancement and modification made on the electrocoagulation. This study investigated the performance of batch-electrocoagulation with vibration-induced electrode plates in the treatment of stabilized landfill leachate to enhance the colour, COD and NH_3-N removal. The intensification of gas bubbles was introduced in the reactor through the vibration of electrodes using motor vibrators and the effect of vibration intensities, initial pH, operating time and current intensities on the performance of electrocoagulation were investigated. The hydrodynamics study such as bubbles size, rise velocities and flow pattern were evaluated using the PIVLab software and a high-speed camera was used to capture the bubble's image. The results showed that the highest removal of pollutants was achieved at the optimum vibration intensity of 2.8V. With vibration-induced plates, the plates resistance was 4% - 20% lower than electrocoagulation with stationary plates, hence effectively removed more than 90% of color (with concentration removed was 2464 mg/L), 37% of COD (821 mg/L) and 57% of NH_3-N (154 mg/L). Smaller bubbles sizes with high surface area

were able to be formed and completely detached from the plate surface at a vibration intensity of 1.8 V onwards. The bubble rise velocities were found to be reduced when the plate vibrated and these velocities achieved a homogenous flow at high current intensities and were affected by both v- and u-component, as opposed to only single upward direction in electrocoagulation with stationary plates. From the zeta potential results, electrocoagulation with vibration-induced plates approached a faster rate of coagulation-flocculation with the Brownian diffusion kinetic data followed the second order reaction and a rate constant of 0.0001 $L.mg^{-1}s^{-1}$, thus, the flocculation mechanism of the electrocoagulation was dominantly through charge neutralization. The electrical energy consumption was calculated within 0.47 kWh/m^3 to 21.66 kWh/m^3 which was 4% to 20% lower than the one achieved in electrocoagulation with stationary plates. Therefore, the use of vibration-induced plates can be proposed as an alternative of agitation mechanism in electrocoagulation.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Sanitary landfill is a method of disposing municipal solid waste (MSW) on land without creating nuisance or hazards to the public health or the environment. The solid waste is confined to the smallest practical area, with reduced volume, and daily covered with a layer of earth (Arockiam JeyaSundar et al., 2020). Sanitary landfilling has been widely recognised as a practical waste disposal system and a common solution for developing countries compared to incineration, gasification and composting (Kamaruddin et al., 2017; Shadi et al., 2020).

Despite the advantage of MSW disposal by sanitary landfill this method however, generates fluid leakage due to the physicochemical and biological changes of solid waste over a period of time which is known as leachate. Because of the increased in the number of populations in Malaysia, it is expected that the MSW generated would increase to more than 30,000 tons per day which indirectly increased the amount of leachate generation (Abdul Aziz & Ramli, 2018). Leachate is harmful to the ecosystem as it consists of toxic compounds such as heavy metals, suspended solids and ammoniacal nitrogen (NH_3-N). The leachate quality discharged from sanitary landfill system are bound by environmental regulations and laws such as Environmental Quality Act 1974, Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 and other laws and regulations adopted by the local authorities. In addition, the leachate physico-chemical properties change over time. Hence, an effective treatment method comparable with the leachate characteristic is required to comply with the requirement of the relevant laws and regulations.

The conventional treatment methods for treating landfill leachate can be either by biological or physicochemical. Of all the wastewater treatment methods in use today, the electrocoagulation process recently sparked renewed interest due to its potential to remove wastewater pollutants. This technology has been proven to be able to effectively remove harmful substances from various wastewater sources as found in previous literatures, for example, the chemical mechanical polishing of wastewater from semiconductor industry (Liu et al., 2021a; Moersidik et al., 2020), the treatment of printing ink and dye (Núñez et al., 2019; Papadopoulos et al., 2019), crude oil from petroleum wastewater (Keramati & Ayati, 2019), palm oil mill effluent (Mohamad et al., 2022) as well as leachate from municipal solid wastes (Gautam et al., 2022). In treating landfill leachate, electrocoagulation has shown the effectiveness in reducing chemical oxygen demand (COD), colour and suspended solids (Dia et al., 2017; Kasmuri & Ahmad Tarmizi, 2018; Mohamad Zailani et al., 2018; Rusdianasari et al., 2017; Sitorus et al., 2018).

Electrocoagulation is an electrochemical method, involving the release of active coagulant ions (usually aluminium or iron cations) from the corroded sacrificial anodes in the solution and the simultaneous formation of hydroxyl ions and hydrogen gas at the cathode. When an electric current is applied, the anode will undergo oxidation, while the cathode will experience a reductive deposition of elemental metals. The formation of solid hydroxides and oxyhydroxides during the process will provide active surface for the adsorption of pollutants, followed by the agglomeration of particles.

Although electrocoagulation is an energy-intensive process due to its high use of electricity, it has advantages over other approaches such as its capacity to operate at ambient temperature and pressure, and its robust performance and its capacity to adjust to variations in the influent compositions and flow rate (Ghimire et al., 2019). This method also produces a small amount of sludge at the end of treatment and consumes fewer chemicals, which can be regarded as environmentally friendly compared to the conventional coagulation process.

Since water is also electrolysed as part of the parallel reaction in electrocoagulation, this will induce a simultaneous production of oxygen and hydrogen bubbles at the corresponding anode and cathode, respectively. Without any flow or agitation mechanism in electrocoagulation treatment, the oxygen and hydrogen gas generated in-situ can produce a soft mixing which helps to attract the colloids, destabilise to flocculate and will then carry the flocculated pollutants up to the surface through natural buoyancy (Mousazadeh et al., 2021). This later process which can help to further enhance the pollutant removal efficiency is named as electro-flotation, another main mechanism in electrocoagulation.

In most of electrocoagulation processes, the efficiency in removing pollutant is not just dependent on anode oxidation but is also determined by the action of gas bubbles produced during the treatment. The bubbles formation and bubbles rise velocities are important especially in electrochemical treatment for which generated gas bubbles improved the ionic transfer and dispersion of particles. Nevertheless, the drawback of this process is the accumulation of gas bubbles surrounding the electrode plate surface, which results in high internal resistance between the electrode plates that obstruct optimum ionic transfer, subsequently reducing the electrocoagulation efficiency (Angulo et al., 2020)

Recent trends of electrocoagulation research have been made to focus on the intensification methods in wastewater treatment through a series of physical enhancement (Fan, Deng, Feng, et al., 2020; Goren & Kobya, 2021; Nippatla & Philip, 2020; Panikulam, Yasri, & Roberts, 2018; Sandoval, Fuentes, Nava, et al., 2019). The aims were to improve the ionic transfer in electrocoagulation as a way of increasing the efficiency of pollutant removal. However, most of the research deal mostly with mechanical enhancement on the electrode plates, with still less or no attention being paid in investigating the role of gas bubbles in electrocoagulation treatment. Using the Particle Image Velocimetry (PIV) as the tool for measuring time-varying bubble hydrodynamics and characteristics, the present study aims to explore bubble characteristics in terms of size, rise velocity and flow behaviour in a batch electrocoagulation with vibration-induced electrode plates for the removal of colour, COD and NH_3-N in stabilized landfill leachate.

1.2 Problem Statement

Electrocoagulation is one of the efficient treatment methods for old or stabilized leachate. In electrocoagulation, other than coagulant ions, the role of gas bubbles is important during the treatment as the bubbles capture the pollutants and bring the flocs to the top surface of electrocoagulation reactor by flotation. During the treatment, the gas bubbles develop as bubble nuclei, grow in size, break off from the electrode surface and rise in the liquid. Unfortunately, over time and with or without any enforced agitation, the nucleated bubbles tend to adhere and accumulate on the electrode surface, which deactivate the parts of the electrode surface and the bubbles growth could reduce the mass transfer in the electrode boundary layer (Naje, Chelliapan, Zakaria, & Abbas, 2016). Adhering bubbles could also exert a substantial effect on the plate ohmic resistance, preventing current to reach a portion of the electrocatalytic area, and induce a non-uniform current density distribution in the area adjacent to the bubbles, affecting further ionic transfer in this region (Angulo et al., 2020).

To overcome the bubbles adherence on the electrode plates, the implementation of flowing electrolyte has shown to mitigate the potential loss due to adhered bubbles. Furthermore, the enhancement is more significant when bubbles detach from the electrode plates introducing turbulence (Zhao et al., 2019). Within the last five years, various efforts have been done in enhancing the mass transfer of ions in electrocoagulation with the prominent enhancement were made on the modifications of electrode plates such as electrode plate coated with surface active materials (Fan et al., 2020; Liu et al., 2021a), utilizing 3D aluminium electrodes (Goren & Kobya, 2021), rotating-disc electrodes (Nippatla & Philip, 2020), oscillating and rotating plates (Naje et al., 2019; Panikulam et al., 2018). Since gas bubbles are also the key-point in the success of electrocoagulation, there were several studies which enhanced flocculation during the treatment through coupling electrocoagulation with external forces of air supply systems to prevent nucleation or induce early attachment, mitigating the impact of bubbles in the overpotential of the system. The examples include electrocoagulation with dissolved ozone flotation (Li et al., 2021), sono-assisted electrocoagulation using ultrasonic bath (Prajapati, 2021) and electrocoagulation with microbubble diffuser (Abdulrazzaq et al., 2021).

Even though the results showed that the modifications made on the electrode plates able to achieve the turbulence flow, indirectly improve the mass transfer of the ionic species and increase the removal efficiency of the electrocoagulation, the previous studies however focused only on the physical enhancement of the electrocoagulation, with still lacked focus on the gas bubbles roles and behaviour in electrocoagulation which are known to induce convection and mass transfer rates. On the other hand, the combinations of electrocoagulation with air supply system allow the generation of microbubbles which could help to increase the treatment efficiency. Nevertheless, these methods are dependent on an external source of air supply, which require further pressurization and air injection system, leading to another consumption of power densities and high operating costs. Previous application of high frequency vibration on the anode contributed to the enhanced transport of ions (Dubrovski et al., 2018), yet, the study utilized piezoelectric transducer and amplifier to induce vibration for electropolishing of copper in the electrochemical cell. Still, there were no attempts made on applying a simple vibration technique of using vibration motor for electrocoagulation cell enhancement.

This study proposed to develop a novel batch electrocoagulation with vibration-induced electrode plates in treating stabilized landfill leachate from Pulau Burung Sanitary Landfill. Without requiring any external force to supply gas bubbles, the electrode plates were induced with vibration using motor vibrators which were expected to intensify the in-situ generation of microbubbles. This is the novelty of the study in which the vibration-induced plates helped to enhance the parallel flotation-flocculation process in electrocoagulation by the gas bubbles action. Other than its performance, this study contributed to the comprehensive understanding on the role and the impact of gas bubbles intensification during the vibration-induced electrocoagulation treatment, and investigation of its flocculation mechanism through relevant electrophoretic mobilities and electrokinetic modelling were also investigated.

1.3 Hypothesis

The main hypotheses of this research are as the followings:

i) It was hypothesized that by inducing the electrode plates to vibrate at a certain range of intensity, the vibrations could stimulate the detachment and distribution of bubbles and pollutants in the water, indirectly enhance the electrocoagulation process.

ii) The vibration-induced electrode plates were able to enhance the gas bubbles sizes and bubbles rise velocity which help for the flocculation and floatation process.

iii) The vibration-induced electrode plates were able to enhance the flow and mixing of particles during the electrocoagulation.

1.4 Research Objectives

This research aimed to achieve the following objectives:

i) To evaluate and compare the performance between batch electrocoagulation with vibration-induced and stationary plates in terms of the overall resistance and removal of color, COD and NH_3-N.

ii) To analyze the hydrodynamic of gas bubbles in electrocoagulation with vibration-induced and stationary plates in terms of bubbles size, bubbles rise velocity, and flow behavior using the PIV tool.

iii) To evaluate the flocculation mechanism and the electrical energy consumption between the electrocoagulation treatment induced with vibration and electrocoagulation with stationary plates.

1.5 Scope of Research

This research mainly focused on the laboratory works as the followings:

i) The working variables used for the performance study include vibration intensity (0.8, 1.3, 1.8, 2.3, 2.8 and 3.3 Volt), initial pH (5, 6, 8, 9 and 10), operating time (10, 23, 35, 48 and 60 minutes) and current intensity (0.5, 1.5, 2.5, 3.5, and 4.5 A). A batch electrocoagulation with aluminium plates attached with vibration motor was set-up for this purpose.

ii) Since the study on the batch electrocoagulation with vibration-induced plates were at its preliminary stage, the study on the performance of the batch electrocoagulation with vibration-induced electrode plates were not focused on optimizing the variables, instead it focused on the feasibility of this electrocoagulation in treating leachate through comparison of performance between vibration-induced and stationary electrode plates at the selected variables.

iii) The hydrodynamic study mainly focused on the comparison of the bubble size, bubble rise velocity and the bubbles flow regime and pattern, between vibration-induced and stationary electrode plates.

iv) All experimental runs as in (i) and (ii) were conducted at constant volume of raw leachate which is 2 L at room temperature.

1.6 Organization of Thesis

There are five chapter in this thesis and each chapter provides specific information about the research.

Chapter 1 provides the research background. This chapter starts with the past and current scenario on the MSW generation in Malaysia. It also gives a brief information about the current disposal method of MSW which is the sanitary landfilling and also the discussion about the leachate pollution from landfilling method. The problem statement, research objectives and research scopes are also stated clearly in this chapter.

Chapter 2 explains about the related literature review on the research. The chapter provides further discussion on the MSW process cycles from its generation to landfilling, the comparison between young and old leachate, and provide information about Pulau Burung Sanitary Landfill (PBSL), located in Southern Penang. The chapter also discuss the types of leachate treatment which then lead to a deep discussion about electrocoagulation. This chapter also highlights the recent trend on the modifications in electrocoagulation system. Other topics covered in this chapter include the description about the hydrodynamic of gas bubbles, PIVLab tools and the kinetic models in electrocoagulation

Chapter 3 describes the materials and methodology used in this research. The first part explains about the research experimental flow chart, followed by the list of material and reagents and chemicals used in this research. The subsequent topics explain about the electrocoagulation experimental set up, sampling and characterization procedures of landfill leachate, followed by the procedures on the electrocoagulation treatment and the hydrodynamic study of the gas bubbles. The final part of the chapter discusses on the flow and kinetic behavior of the electrocoagulation treatment.

Chapter 4 highlights the results and discussion obtained from the research. The first part of the results consists of the physicochemical properties of the landfill leachate. This is followed by the results and discussion on the removal efficiency of electrocoagulation with vibration-induced plates and stationary plates as comparison. The third part of the chapter includes the data analysis on the hydrodynamic study. The final part of the chapter presents the zeta potential and the kinetic modelling of the electrocoagulation treatment for determination of its particle distribution mechanism. The final part of the results and discussion also include the comparison of electrical energy consumption between electrocoagulation with vibration-induced plates and stationary plates.

Chapter 5 concludes the research following its research objectives. Recommendations for future work related to this research are also given.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter gives the overall overview of MSW problems and its management, leachate characteristics and its treatment methods. This is followed by the overview of electrocoagulation treatment mainly its theory and factors affecting the process. The recent enhancement in electrocoagulation, particularly on its mechanical intensification was further discussed. Further discussion was made on the hydrodynamic study in electrocoagulation and finally the kinetic study of electrocoagulation explained at the end of the chapter.

2.2 Municipal Solid Waste Generation in Malaysia

Municipal solid waste (MSW) can be defined as any scrap, unwanted or rejected materials which are highly nonhomogeneous mixture of residential, commercial and industrial sectors such as paper, boxes, clothing, plastics and food. Although the composition of MSW are highly variable, it is generally accepted that organic materials are the largest component of MSW (Adhikari et al., 2018). With the rapid growth of population and industrialization, the increase in household and industrial activities have resulted in a tremendous production of MSW in Malaysia. From Table 2.1, it can be seen that the population growth in Malaysia is increasing each year, with a 19% of increment of population number in 2020 as compared to year 2012. Apparently, the estimated daily solid waste generation for household, industrial, commercial and institutional increased proportionally with the number of populations (JSPN, 2022).

Table 2.1 Number of populations in Malaysia and estimated solid waste generation

Year	Number of populations in Malaysia (Malaysian and non-Malaysian	Household: Estimated daily solid waste generation (tons/day)	Industrial, commercial & institutional: Estimated daily solid waste generation (tons/day)
2012	28,334,135	21,533.94	11,617.00
2013	28,844,149	21,921.55	11,826.10
2014	29,363,343	22,316.14	12,038.97
2015	29,891,883	22,717.83	12,255.67
2016	30,429,936	23,126.75	12,476.27
2017	30,977,674	23,543.03	12,700.84
2018	31,535,272	23,966.80	12,929.46
2019	32,102,907	24,398.21	13,162.19
2020	33,782,300	25,674.55	13,850.74

MSW requires a systematic management to minimize undesirable impacts on the ecosystem. Due to its economical and eco-friendly method, Malaysia is still dependent on landfill for the MSW disposal with approximately 89% of waste collected ending up in landfills, while the remaining waste is sent for treatment by small incineration, recycling or reprocessing or illegal dumping (JPSPN, 2015).

2.3 MSW Management in Penang

The solid waste management practices in Penang mainly involves recycling and sanitary landfill. Based on the reports of domestic waste generation from year 2005 to 2012, it shows that the amount of disposed domestic waste to landfill gradual reduction with the increased recycling effort in Penang. In 2012, it was estimated that a total of 816, 652 tonnes of waste has been generated. Of the total waste generated in the year, about 29.35% of the waste has been recycled compared to 18.3 % as recorded in year 2005 (Majlis Perbandaran Pulau Pinang & Majlis Perbandaran Seberang Perai, 2013). The recent trend of waste generation reported a further reduction in which a total of 74,749 tonnes of waste was generated in April 2019 and the amount reduced to 49,596 tonnes in April 2020 (PLB Terang Sdn. Bhd., 2020).

The Penang State Government together with two local authorities : Penang Island City Council (MPPP) and Seberang Perai City Council (MPSP) are the administers who are responsible in solid waste management in Penang. The solid wastes collected would be first transferred to two existing dumpsites which are the Batu Maung Transfer Station in the Penang Island, while waste collected in the mainland would be transferred to the Ampang Jajar Transfer Station located at Seberang Perai. All collected waste from all entire Penang will be compacted and will be sent on to the Pulau Burung landfill for final disposal.

2.4 Pulau Burung Sanitary Landfill

In Penang, Pulau Burung Sanitary Landfill (PBSL) is the only approved municipal solid waste landfill. The PBSL is located in the southern region of Penang and has three phases as shown in satellite image of Figure 2.1, with total area of 127.4 hectares land combining with the existing phase 1 and 2 (Green Earth, 2017c).

Figure 2.1 Satellite image of Pulau Burung Sanitary Landfill: A – Phase 3, B – Phase 1 and 2

The Phase 1 began to operate in 2001 and has been closed in 2008, while the Phase 2 started its operation in 2008 and the operation has transferred to the new landfill of Phase 3, which started its operation in early 2021 with the total daily waste handling capacity of 1,800 tonnes per day (Green Earth, 2017a, 2017c). During the initial phase of operations, a semi-aerobic system was introduced with established a controlled tipping technique, complying with Level II of the sanitary landfill standards (Mohd Azme & Murshed, 2018). The landfill was then upgraded to Level III employing controlled tipping with leachate recirculation (Kamaruddin et al., 2016). The next landfill phase will establish a Level IV standard (the highest standard) for landfill by incorporating a modern Material Recovery Facility (MRF) to further increase the total recycling rate in Penang (Green Earth, 2017b).

The semi-aerobic system known as Fukuoka method which is still been applied by the PBSL, is considered as the most desirable landfill design for Malaysia because of its simple structure and is cost effective due to the utilization of readily available materials and methods, and less intensive leachate treatment system (Kamaruddin et al., 2016)(Department of Local Government, Ministry of Housing and Local Government (MHLG), 2006; Kamaruddin et al., 2016; Matsufuji, 2007). This method enable ambient air flows into the waste body through the leachate collection pipes by passive ventilation to promote rapid landfill stabilization and environmental preservation (Kamaruddin et al., 2017; Matsufuji, 2007). As shown in Figure 2.2, a collection and discharge pipe with a large cross section in the bottom of the landfill is built to rapidly collect and discharge leachate away from the landfill site. In addition, gas ventilation facilities are also constructed, hence maintaining the aerobic environment in the landfill (Matsufuji, 2007).

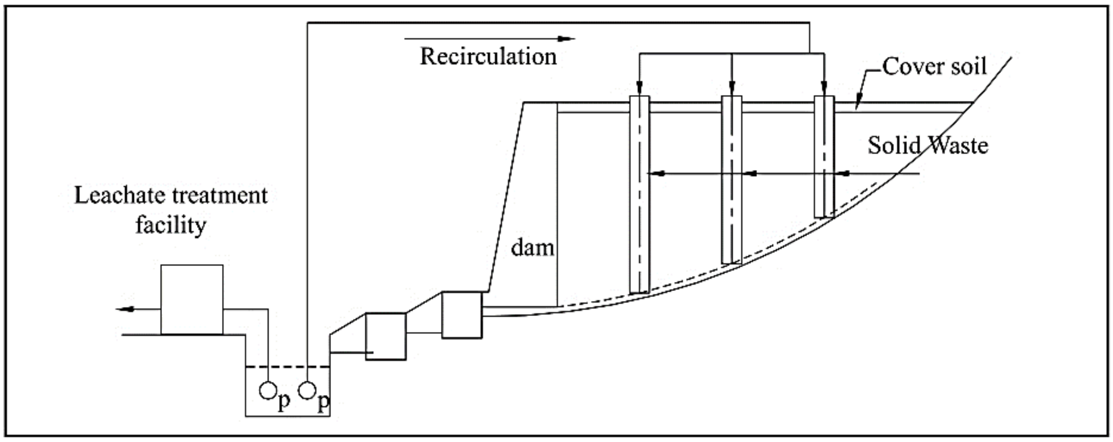

Figure 2.2 Semi-aerobic Landfill Type

The waste decomposition in landfill involves decomposition by physical, chemical, and biological process, in which the progress rates are dependent on the waste characteristics and also several factors such as the structure of solid waste, climate change, age of landfill, moisture content and pH (Kamaruddin et al., 2017). Among the three processes involved, the biological degradation is the utmost concern since it produces highly contaminated hazardous leachate and gases through solubilization and gasification, which are governed by both aerobic and anaerobic decomposition. (Matsufuji, 2007). The solubilization process produces ammonia, organic acids, and carbon dioxide, which consequently increases the BOD and reduce the pH level in leachate. Meanwhile, the gasification process produces gas from organic acid, hence reduce the BOD level and increase the pH (Matsufuji, 2007).

The main problem associated with landfill is the leachate produced, other than the free emission of landfill gas. From Figure 2.2, it can be seen that leachate is collected through collection pipe and diverted to the leachate retention pond for further treatment. Prior to the retention pond, the leachate may be recirculated back to the waste layer to be reprocessed and further decompose to improve leachate quality. The leachate flowing from waste dumps and disposal sites can cause serious pollution of groundwater and waterways if not treated in a proper way. PBSL has two leachate retention ponds and one leachate treatment plant which has been developed and was in test and commissioning phase (Green Earth, 2017c).

2.5 Characteristics of Young and Old Leachate

The solid waste dumped in the landfill will produce toxic liquid overflow which is commonly referred as leachate. Without treatment, the leachate can contaminate drains, rivers, groundwater, and soil with chemical, organic, and inorganic pollutants. Figure 2.3 further breakdowns the components in leachate. The organic materials may contain biodegradable and non-biodegradable materials, and the example of heavy metals include cadmium (Cd^{2+}), chromium (Cr^{3+}), lead (Pb^{2+}), nickel (Ni^{2+}) and zinc (Zn^{2+}), while the example of non-organic salts are sodium (Na^{+}), calcium (Ca^{2+}) and ammonia (NH^{4+}).

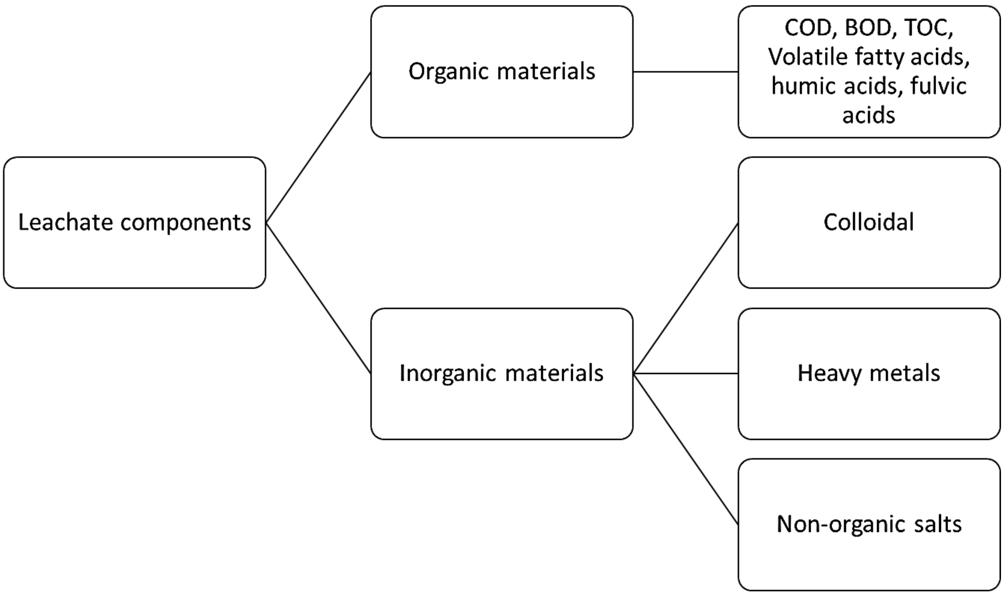

Figure 2.3 Components of leachate

The leachate quality is normally characterized by parameters such as its temperature and pH, colour, COD, BOD_5, NH_3-N, suspended solids, turbidity, conductivity, and heavy metals. COD measures the oxygen equivalent of the organic matter content that is susceptible to oxidation by a strong chemical oxidant (Kamaruddin et al., 2018a). High COD value represent high oxygen demand for the degradation of organic matters in solid wastes which contribute to the leachate formation (Kamaruddin et al., 2016). The COD values in leachate also can be varied depending whether it is categorized as biodegradable and non-biodegradable COD. It has been reported from a study that based on the leachate characteristics such as the ratio of BOD_5/COD, PBSL contains more non-biodegradable COD (Kamaruddin et al., 2018b).

The decomposition rates of the leachate pollutants are dependent on the physical, chemical and biological of waste biodegradation, hence the leachate quality may vary over time depending on the waste biodegradation phases (Kamaruddin et al., 2017). Figure 2.4 shows the bio-chemical decomposition of the organic matter present in MSW which follows a sequence of the four phases: i) hydrolysis, ii) acidogenesis iii) acetogenesis and iv) methanogenesis by the action of hydrolytic-, acidogenic-, acetogenic- and methanogenic microorganism, respectively.

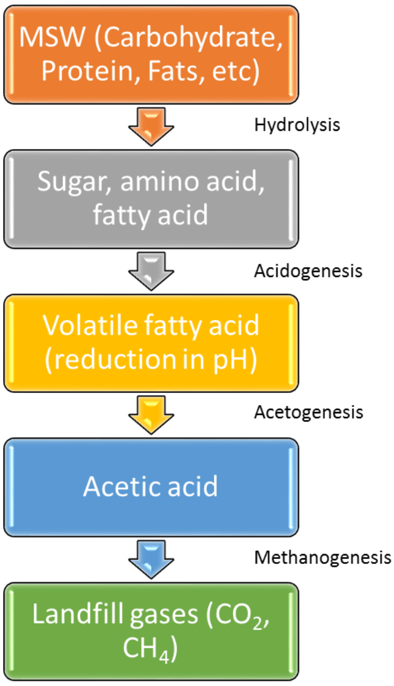

Figure 2.4 Leachate decomposition phases

From this figure, it can be seen that during the decomposition process, initially carbohydrates, proteins and fats get hydrolysed into soluble sugars (glucose, fructose and sucrose), amino acids and fatty acids, which further are mineralized into the long-chain and short-chain volatile fatty acids (VFAs) (i.e acetic acid, propionic, butyric acid, valeric acid, etc) and alcohol. Subsequently, in the methanogenesis phase, products of acetogenesis are mineralized to methane (CH_4) and carbon dioxide (CO_2) by either acetate cleavage or by the action of the hydrogen scavenging bacteria (Mohammad et al., 2022)`Table 2.2 provides a detailed summary of the leachate decomposition process at landfill site (Mohammad et al., 2022).

Table 2.2 Descriptions of leachate phases

Phases	Descriptions
i) Hydrolysis	• Hydrolysis of organic matter generates several acids • pH of leachate becomes acidic, which inhibits microbial activities • As time progress, the pH value starts increasing (pH > 7) due to the consumption of these acids by microorganism and the formation of ammonia during ammonification process (Luo et al., 2020)
ii) Acidogenesis	• Products of hydrolysis are converted into various volatile organic acids • The higher concentration of volatile fatty acids inhibits decomposition of organic matter due to the acidic pH (pH < 5)

Table 2.2 Continued

Phases	Descriptions
iii) Acetogenesis	• Decomposition of MSW generates acetic acid due to favourable conditions of acid formation
iv) Methanogenesis	• Leachate reaches a neutral or slightly alkaline state at this phase. • Gas present in landfill comprises of 55% - 60% of methane, and 40% - 45% of CO_2 when the phase stabilized • Established leachate of pH level between 7 and 8

Even though the biochemical decomposition process would be the important factor which affect the leachate quality, nevertheless lack of information about the leachate or landfill gas characteristics make the determination of landfill stabilisation stage nearly impossible (Kamaruddin et al., 2017). Hence, works in relating the landfill age with the prediction of leachate quality instead of biodegradation phases are the feasible alternative factor. As shown in Table 2.3, the landfill stabilisation status is categorized based on the landfill age, that is landfill of < 2 years is considered as young, landfill of age between 2 – 10 years is considered as intermediate and landfill of > 10 years is considered mature or stabilised landfill (Alvarez-Vazquez et al., 2004).

Table 2.3 Landfill constituent concentration as a function of landfill age

Parameter (mg/L) ***No unit for pH, BOD_5/COD and TOC/COD**	**Landfill age (years)**		
	< 2	**2 – 10**	**>10**
Stabilisation status	Young (fresh)	Intermediate	Mature (stabilised)
BOD_5	2000 – 30,000	N. A	100 – 200
COD	3000 – 60,000	3000 – 15,000	100 – 2800
TOC	1500 – 20,000	N. A	80 – 160
BOD_5/COD	0.5 – 1.0	0.06 – 0.5	<0.1
TOC/COD	<0.3	0.3 – 0.5	>0.5
Total Kjeldahl nitrogen	100 – 2000	N. A	N. A
Ammoniacal-nitrogen	10 – 800	30 – 1800	20 – 900
Organic nitrogen	10 – 800	N. A	80 – 120
Nitrate	5 – 40	N. A	5 – 10
pH	4.5 – 7.5	6.5 – 7.5	6.6 – 7.5
Alkalinity as $CaCO_3$	1000 – 10,000	N. A	200 – 1000
Total hardness as $CaCO_3$	300 – 10,000	N. A	200 – 500
Total suspended solids	200 - 2000	N. A	100 – 400
Heavy metals	>2.0	<2.0	<2.0
Total phosphorus	5 – 100	N. A	5 – 10
Orthophosphorus	4 – 80	N. A	4 – 8
Calcium	200 – 3000	N. A	100 – 400
Magnesium	50 – 1500	N. A	50 – 200

Table 2.3 Continued

Parameter (mg/L) ***No unit for pH, BOD_5/COD and TOC/COD**	**Landfill age (years)**		
	< 2	**2 – 10**	**>10**
Stabilisation status	Young (fresh)	Intermediate	Mature (stabilised)
Potassium	200 – 1000	N. A	50 – 400
Sodium	200 – 2500	N. A	100 – 200
Chloride	200 – 3000	N. A	100 – 400
Sulphate	50 – 1000	N. A	20 – 50
Total iron	50 – 1200	N. A	20 – 200

N. A – not available

Young landfills generally produce leachate with high concentration of COD, and BOD_5, and low concentration of NH_3-N, with high BOD_5/COD ratio and pH value of < 6.5. They are also characterized by a bad odour and strong colour. Meanwhile, stabilised landfills usually produce leachates that is highly contaminated with in NH_3-N and heavy metals concentration, moderately high in COD content but lower BOD_5/COD ratio (usually < 0.1) (Zainol et al., 2012). The BOD_5/COD ratio is another important indicator which represent the proportion of biodegradable organics in landfill leachate. A high ratio of BOD_5/COD indicates that the leachate is relatively biodegradable, whereas a low ratio shows that the leachate biodegrades more slowly (Kamaruddin et al., 2018a).

High organic pollutants in leachate could deplete the dissolved oxygen level and affect the aquatic life. Usually, low BOD implies good water quality. Similarly, inorganic pollutants are toxic to aquatic lifeforms in the river, hence must be properly treated before discharge. Meanwhile, the presence of a high level of NH_3-N in leachate may lead to algal growths and accelerate eutrophication (Kamaruddin et al., 2013). Since PBSL has been in operation for more than 20 years, the leachate currently produced by the landfill is considered as old or stabilised leachate. Identification of the status of the landfill age is essential as it is one of the factors that may influence the suitability of landfill leachate treatment methods.

2.6 Landfill Leachate Treatment Methods

The landfill leachate can be treated either using biological or physicochemical treatment. The effectiveness of each treatment method is varied depending on the landfill age as shown in Table 2.4 (Abbas et al., 2009). It was reported that young leachates with high levels of organic materials (COD of > 10,000 mg/L, ratio of BOD_5/COD between 0.4 and 0.8) and low concentrations of ammoniacal nitrogen are best treated using biological methods. However leachates that contain a high ammoniacal nitrogen concentration and a low level of biological compounds are suitable to be treated using physicochemical process and/or a combined treatment with biological process (Shadi et al., 2020). Thus, biological methods such as aerobic and anaerobic process are effective for treating young leachate but it is a poor treatment method for old or stabilized landfill leachate due to its unstable organic materials and the difficulty in getting it to biodegrade (Mohd Azme & Murshed, 2018).