INTEGRATED ELECTROCOAGULATION-ULTRASONIC METHOD FOR ORGANIC MATTER REMOVAL FROM LANDFILL LEACHATE

SITI SUHAILA BINTI MOHD TARMIZI

SCHOOL OF CIVIL ENGINEERING
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by

SITI SUHAILA MOHD TARMIZI

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Name of Student: Siti Suhaila binti Mohd Tarmizi

I hereby declare that all corrections and comments made by the supervisor(s)and examiner have been taken into consideration and rectified accordingly.

Signature:

Date : 4/8/2021

Endorsed by:

PROF DR MOHD SUFFIAN BIN YUSOFF
Pusat Pengajian Kejuruteraan Awam
Universiti Sains Malaysia, Kampus Kejuruteraan
14300, Nibong Tebal, Pulau Pinang
Tel: 04-5996225, Fax: 04-5996906
Emel: suffian@usm.my

(Signature of Supervisor)

Name of Supervisor:

Date: 4.8.2021

Approved by:

(Signature of Examiner)

Name of Examiner: NAR

Date: 04/08/2021

(Important Note: This form can only be forwarded to examiners for his/her approval after endorsement has been obtained from supervisor)

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ABSTRAK

Penggunaan teknologi rawatan hibrid telah diakui sebagai salah satu kaedah rawatan yang dilakukan untuk meningkatkan kecekapan rawatan larut lesap kerana pelbagai cabaran seperti kepekatan tinggi bahan pencemar dan kemampuan terbatas proses tunggal. Larut lesap dari tapak pelupusan Pulau Burung (TPPB) telah dikategorikan sebagai larut biodegradabiliti yang lemah dan ia terdiri daripada bahan organik terlarut tinggi, komponen inorganik, dan bahan toksik. Dalam kajian ini, rawatan penggabungan antara elektropenggumpalan (EC) dan ultrasonik (US) diperkenalkan untuk menilai keberkesanan proses rawatan hibrid dalam rawatan larut lesap dari tapak pelupusan sampah dari penyingkiran bahan organik dalam bentuk parameter COD berbanding dengan satu rawatan dengan keadaan operasi yang berbeza iaitu voltan, masa rawatan, dan jarak antara elektrod. Selain daripada itu, pengaruh faktor kawalan terhadap penurunan berat elektrod sebanyak juga telah dikaji. Pengurangan COD yang maksimum dari kedua-dua kaedah EC tunggal dan gabungan EC-US telah mencapai keadaan eksperimen terbaik yang sama iaitu voltan 10 V, masa rawatan 25 min, dan jarak antara elektrod 2 cm tetapi peratusan penyingkiran COD melalui kaedah gabungan rawatan EC-US (61.13%) menyediakan penyingkiran COD yang lebih baik berbanding dengan proses rawatan kaedah EC tunggal (37.51%). Selain itu, penurunan berat elektrod paling tinggi diperhatikan di anod (Al-21.60%) dengan keadaan operasi yang sama dengan kecekapan penyingkiran terbaik dalam proses EC-US. Didapati bahawa katod (Fe) mengalami sedikit peningkatan berat akhir elektrod disebabkan oleh tindak balas hidrolisis. Oleh itu, kerja penyelidikan ini menunjukkan bahawa penggunaan rawatan gabungan EC-US secara signifikan telah meningkatkan penyingkiran COD dari larut lesap berbanding kaedah EC tunggal.

INTEGRATED ELECTROCOAGULATION-ULTRASONIC METHOD FOR ORGANIC MATTER REMOVAL FROM LANDFILL LEACHATE

ABSTRACT

The utilization of hybrid treatment technologies has been recognized as one of the treatment methods conducted to enhance the efficiency of leachate treatment due to the various challenges such as the high concentration of pollutants and the limited ability of a single treatment process. Leachate from Pulau Burung Sanitary Landfill (PBSL) has been categorized as poor biodegradability leachate and it is composed of high dissolved organic matter, inorganic components, and toxic substances. In this study, an integrated treatment between electrocoagulation (EC) and ultrasonic (US) was introduced to evaluate the effectiveness of the hybrid treatment process in landfill leachate treatment of organic matter removal in terms of COD compared to a single treatment with different operating conditions of voltage, treatment time, and distance of electrode. Other than that, the effect of the control factors on the weight loss of electrodes was also investigated. The maximum COD reduction from both of single EC and combined EC-US methods have achieved the same best operating condition which are potential of 10 V, treatment time of 25 min, and electrode distance of 2 cm but the combined EC-US treatment (61.13%) provide better removal of COD compared to the treatment process of a single EC method (37.51%). On the other hand, the highest weight loss of electrode was observed at the anode (Al-21.60%) with the same operating conditions of the best removal efficiency in the EC-US process. It is found that the cathode (Fe) experienced a slight increase in the final weight of the electrode due to the hydrolyzation reaction. Hence, this research work demonstrated that

the utilization of combined EC-US treatment significantly enhanced the removal of COD from leachate compared to the single EC method.

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LIST OF ABBREVIATIONS

AOP	Advanced Oxidation Process
APHA	American Public Health Association
BOD	Biological Oxygen Demand
COD	Chemical Oxygen Demand
DC	Direct Current
DO	Dissolved Oxygen
DOM	Dissolved Organic Matter
EC	Electrocoagulation
MSW	Municipal Solid Waste
PBSL	Pulau Burung Sanitary Landfill
SBR	Sequencing Batch Reactor
TDS	Total Dissolved Solids
TKN	Total Kjeldahl Nitrogen
TP	Total Phosphorus
TSS	Total Suspended Solid
US	Ultrasonic
USM	Universiti Sains Malaysia

LIST OF SYMBOLS

Al	Aluminum
$AgNO_3$	Silver Nitrate
Fe	Iron
CO_2	Carbon Dioxide
CH_4	Methane
Cr (III)	Chromium (III) ion
e^-	Electron
H^+	Hydrogen ion
H_2	Hydrogen
H_2O	Water
$HgSO_4$	Mercuric Sulfate
$K_2Cr_2O_7$	Potassium Dichromate
NH_3-N	Ammoniacal Nitrogen
NO_3^-	Nitrate
O_2	Oxygen
OH^-	Hydroxide ion
•OH	Hydroxyl Radicals
SO_4^{2-}	Sulfate

CHAPTER 1

INTRODUCTION

1.1 Research Background

Solid waste is the most crucial thing that can be an environmental challenge if there is a failure in managing solid waste. Globally, both developed and developing countries were experienced increasing population growth and urbanization include Malaysia which led to an increase in waste generation and high daily solid waste consumption. Many effective and efficient disposal practices are used worldwide which are landfill, incineration, waste compaction, composting, and recycling. The landfill is the most common practice and conventional method used for waste disposal due to low capital costs and it can minimize environmental impacts. However, the waste disposed of in the landfill will rot and decompose which produces harmful gases, liquid (leachate), and inert solids.

The generation of a liquid which is called leachate that present in the waste and due to humidity also rainfall (Baderna et al., 2019) is one of the problems and main issues at landfill sites. Landfill leachate is often defined as heavily polluted wastewaters that may be extremely hazardous to the groundwater and surface water if it is not properly treated and disposed of safely. The hazard result of leachate is because of its characteristics which it is contains high organic concentrations and inorganic contaminants including ammonia nitrogen, heavy metals, xenobiotics, and inorganic salts (Nurul, 2013). Treatment of wastewater from organic matter has become one of the important issues in the treatment industry as it consists of high variable compounds and it can pose threat to the environment and health (Zanki et al., 2020). Therefore, various methods of treatment that have been developed to treat leachate wastewater especially in the removal of organic matter from leachate including biological, physical/chemical and combination of treatment method to avoid the leachate from entering the environment.

As the current treatment technology for leachate is site-specific and costly, a low-method and efficient method is proposed. Various physio-chemical are often applied in the treatment of leachate for removal of organic matter such as electrocoagulation, photocatalyst, ultrasonic, etc. In this study, electrocoagulation (EC) and ultrasonic (US) techniques are used to treat the leachate. From literature, it is stated that a single process of treatment is not efficient compared to an integrated method which it is more efficiently in extract pollutant from wastewater (Asaithambi et al., 2020). Based on several studies, EC has been effectively used in combined treatment processes and it is evaluated to be effective in leachate treatment due to its wide adaptability towards various types of pollutants especially heavy metals without adding external chemicals but it couldn’t remove high concentration of organic matter from leachate due to its limited ability (Al-Qodah and Al-Shannag, 2019; Ding et al., 2021; Zanki et al., 2020). In order to remove organic matter from leachate efficiently, a US method is proposed in this research project as it has been used widely to degrade organic components from wastewater and it is one of the advanced oxidation processes that able to oxidize almost all organic pollutants (Mahvi et al., 2012).

1.2 Problem Statement

Landfill leachate is categorized as the main pollutant in wastewater as it has a complex structure and high pollutant load (Rusdianasari et al., 2017). It is one type of wastewater that consists high concentration of organic matter and nitrogen. Leachate also has high COD and toxic matter which are the most important problems in leachate landfills. The dissolved organic matter is a major component of complex leachate and it can cause the increase of antioxidant, bacteria, disinfection by-product generation, and membrane contamination in the treatment system (Liu et al., 2020). The presence of organic matter in leachate can pose a high environmental risk and a threat to human health as it easily interacts with metals and hydrophobic organic contaminants (Liu et al., 2020). Other than that, a high concentration of organic matter also can increase sludge volume which will affect the cost of the treatment process (Zanki et al., 2020). Therefore, various treatment has been developed to remove organic matter from leachate wastewater and the type of leachate treatment method should depend on its composition.

Many studies show that EC is one of the methods that have higher efficiency and more economic in treating wastewater include leachate. By using EC methods, a large variety of pollutants can be removed through simple equipment and easy operation. This treatment method is also known as an eco-friendly method because it doesn't require any addition of external chemicals to run the treatment (Zanki et al., 2020). However, the limited ability to remove a high concentration of organic matter is one of the main weaknesses of EC. A growing of passive layers at the electrode surface and limitation of mass transport passivation of the electrode also will affect the efficiency of this technique. Other than that, the production of excess sludge and desirable toxic chlorinated by-products also are the disadvantages of the EC process. Hence, an improvement treatment method can be made such as combined with other methods that can degrade the organic components in leachate such as the US.

The present study is conducted to concentrates on leachate treatment produced from a semi-aerobic landfill which is Pulau Burung Sanitary Landfill site (PBSL) located in Seberang Perai. The study is focused on COD removal using the integrated method between US and EC and a single method of EC in the treatment of leachate also the effect of the control factors on the weight loss of electrode.

1.3 Objectives

i. To investigate the effectiveness of integrated methods of ultrasonic (US) and electrocoagulation (EC) in comparison to single electrocoagulation (EC) method in organic matter removal (in terms of COD) from leachate wastewater.

ii. To determine the effect of control factors in combined EC-US and single EC method on weight loss of electrode.

1.4 Scope of work

This study is conducted to compare the efficiency of organic matter removal using individual EC methods and combined methods of EC-US method to treat landfill leachate wastewater. The selection of type for wastewater treatment will depend on its characteristics. As the leachate wastewater may contain large amounts of organic pollutants, it can be measured as COD and it is one of the most problematic parameters in treating leachate.

The leachate sample is collected from Pulau Burung Sanitary Landfill (PBSL) and will analyze for pH, temperature, conductivity, BOD, and COD. The study is performed in two phases which are leachate characteristics and treatability assessment by using combined EC-US and individual EC methods. The percentage of organic matter removal will evaluate in terms of COD analysis. Other than that, various operational parameters that might affect the process of treatment also will be considered such as voltage, treatment time, and distance of electrode. The effect of these control factors in combined EC-US and the single EC methods is investigated on the weight loss of the electrode for each set of experiments.

1.5 Dissertation Outline

Chapter 1, **Introduction**: Provides a brief background of this research project and issues related to landfill leachate. This chapter also comprehensively explain the objectives, and scope of work involved in the treatment of landfill leachate

Chapter 2, Literature Review: Discuss the previous studies which highlighted information on landfill leachate and the treatment methods that have been used by previous researchers. Other than that, fundamental and factors affecting the efficiency of electrocoagulation and ultrasonic treatment are also reviewed in this chapter.

Chapter 3, Methodology: Describe the flow of research work and procedure involved in the experimental work that involved in leachate characterization and treatment process. All of the laboratory apparatus and set-up of single and combined treatments are also described in detail in this chapter.

Chapter 4, Results and Discussion: Presents the results and discussion based on results obtained from the experiments of combined EC-US and single EC treatment process. This chapter consists of three main sections which discussed the leachate characterization, individual treatment of EC, and hybrid treatment of EC-US respectively. The third section reports the effect of the various operational parameters (voltage, distance of electrode, and treatment time) on the removal efficiency of COD and the electrode weight losses.

Chapter 5, **Conclusions and Recommendations**: Concludes from all the research work and provide suggestion to improve current work for future study.

References, followed with appendices with raw data are arranged in the final section of this thesis.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

In this chapter, general overview on the problems of municipal solid waste in Malaysia, waste disposal at the sanitary landfill, generation of landfill leachate, and the suitable methods based on its characteristics. The main concern of this research study is basically on electrocoagulation and ultrasonic method in organic matter removal from leachate.

2.2 Sanitary Landfill Leachate

Over recent years, Malaysia has been experienced an increase in population and urbanization which has affected the production of municipal solid waste (MSW). The population in Malaysia is expected to reach up to 33.4 million by the year 2020 and 37.4 million by the year 2030 due to the growth of fast pace urbanization (Chien et al., 2017). The increase in population growth has resulted in a high production rate of solid waste especially MSW. MSW is defined by the Malaysian Solid Waste and Public Management Act of 2007 (Act 672) as any substance that needs to dispose of because physically spoiled or contaminated, broken or worn out. Currently, the average amount of solid waste generated each day in Malaysia is about 1.1 kg/day and over 26,500 tonnes of waste is disposed of every day (Kamaruddin et al., 2017). The increase in waste generation has become a critical issue in solid waste management as it is one of the major problems of environmental in Malaysia. Therefore, various methods have been introduced and considered by the government for solid waste management such as inert landfill, incineration, composting, recycling non-organic waste, and unsanitary landfill (Kamaruddin et al., 2017).

Sanitary landfill is one of the conventional and most common methods for solid waste disposal all over the world due to its simple procedure and low cost (Abd El-Salam and Abu-Zuid, 2015). It is defined as a solid waste disposing of method on land and buried in layers of the earth with the knowledge of engineering without causing hazards to the public. It also disposed of solid waste by minimizing the environmental impacts and allow safe decomposition compared to other methods such as open-pit dumping and open-air burning sites (Abd El-Salam and Abu-Zuid, 2015). However, sanitary landfills also may result in environmental pollution such as unpleasant odors, groundwater pollution, landfill settlement, air pollution, global warming, and vegetation damage (Abd El-Salam and Abu-Zuid, 2015). These impacts might occur if improper waste management at sanitary landfills is not practiced which can cause groundwater contamination (leachate formation) and gas methane production (Hossain et al., 2011)

The types of landfills can be classified into anaerobic, anaerobic sanitary, improved anaerobic sanitary, semi-aerobic and aerobic landfills (Aziz and Ramli, 2018; Awang, 2011). Anaerobic and semi-aerobic landfills are the most common among these types and semi-aerobic landfills are more preferred as they have more benefits than other systems. In Malaysia, the semi-aerobic system has adopted in the landfill such as PBSL and remarkable improvements in leachate quality have been observed. The system of semi-aerobic landfill leachate with the collection pipe can provide various advantages such as prevent fouling of leachate in waste materials (Aziz and Ramli, 2018). The concept of a semi-aerobic landfill leachate collection pipe system is shown in Figure 2.1 and Figure 2.2.

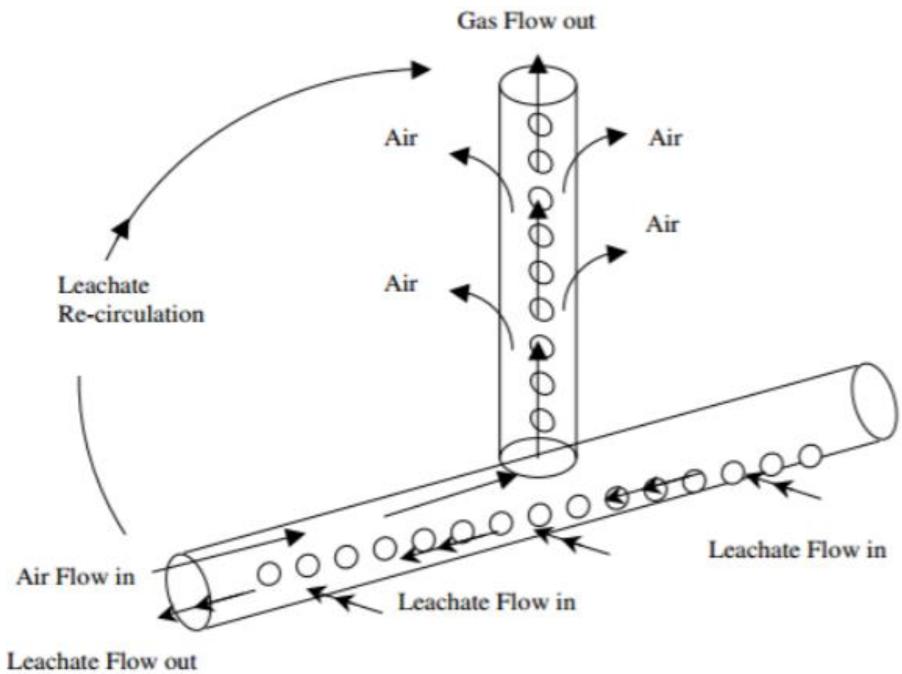

Figure 2.1: Concept of semi-aerobic landfill leachate collection pipes
(Source: Aziz and Ramli, 2018)

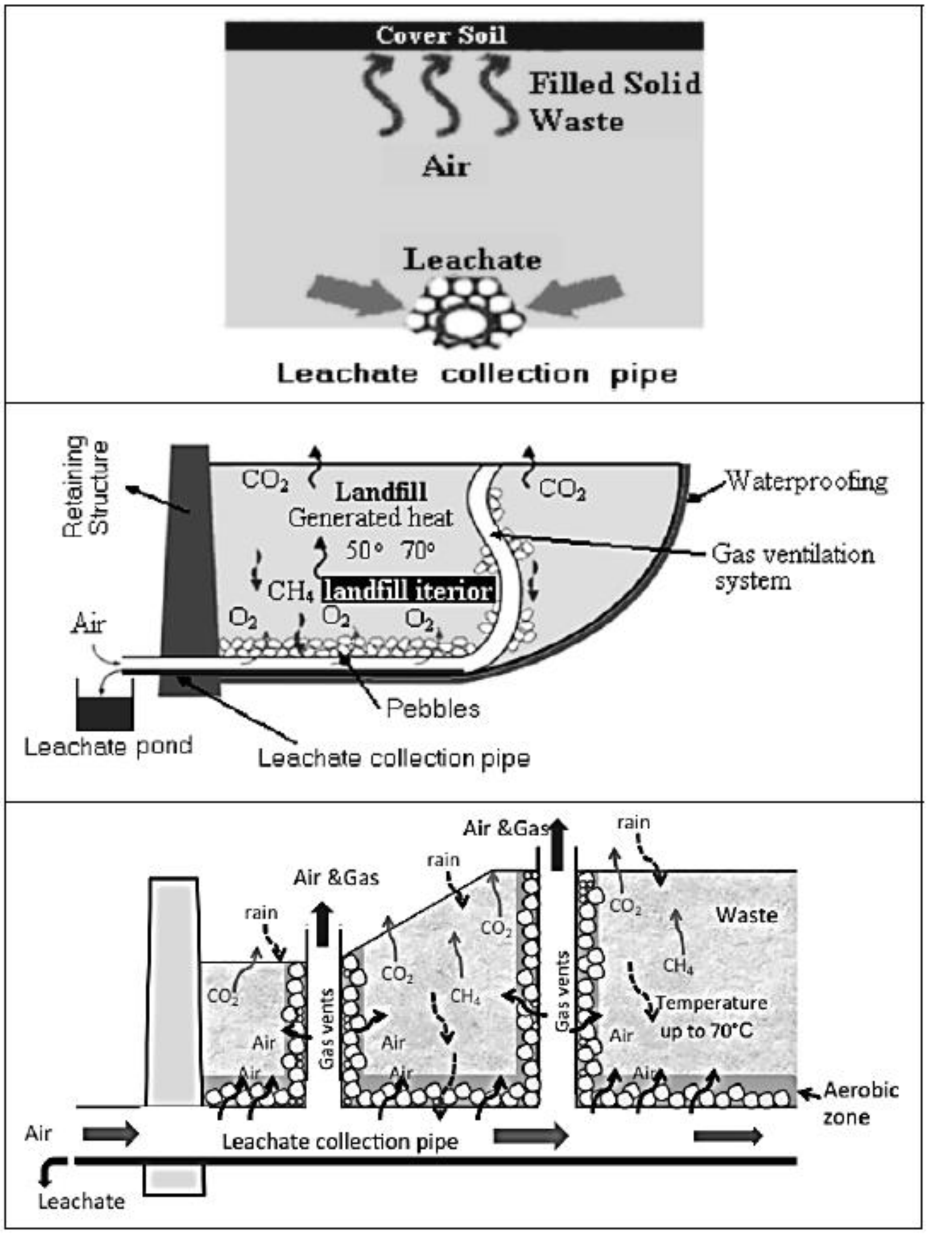

Figure 2.2: Concept of the semi-aerobic landfill
(Source: Aziz and Ramli, 2018)

2.3 Leachate Generation

Leachate is defined as a contaminated liquid generated from the percolation of water that passes through the MSW landfill that contains both soluble organic and inorganic compounds also suspended particles and it is considered a serious pollutant to the environment as well as affecting human health (Naveen et al., 2017). It is usually produced from the water generated from inside the waste, rainwater, surface water, drainage, and humidity (Zainol, 2015). The generation of landfill leachate is greatly influenced by climate factors, the age of solid waste, humidity, the composition of MSW, particle size, degree of compaction, and hydrology of the site (Aziz and Ramli, 2018; Zakaria and Aziz, 2018). According to Aziz and Ramli, (2018), it mentioned that leachate production is mainly contributed by rainfall. The rainwater that passes through the bottom of the landfill will infiltrate into the groundwater table which will cause extraction of a series of contaminants such as organic and inorganic matter, impurities, heavy metals, and other polluted harmful substances to the groundwater. The landfill degree of compaction also will influence the quantity of leachate where the less compacted landfill tends to produce more amount of leachate (Aziz and Ramli, 2018).

Generally, the process of waste decomposition will result in leachate production when water percolates through the waste and both biological and chemical materials are filtrated into the solution. Zainol, (2015) founds that there are five phases of decomposition at the landfill which are (1) initial adjustment phase, (2) transition phase, (3) acidogenic/acetogenic phase, (4) methane fermentation phase, and (5) maturation phase (Zainol, 2015). Meanwhile, another study by Zakaria and Aziz, (2018) describes the degradation process involved three stages which are acid fermentation, intermediate anaerobic phase, and anaerobic degradation. The first stage is the acid fermentation phase which commonly occurs in a short period and presents in the young landfill. Then, the degradation process is followed by the intermediate anaerobic phase where the ammonia is released. Lastly is the anaerobic degradation phase which is known as stabilized leachate. The different phases of the biological process will result in different ranges of several leachate parameters as shown in Table 2.1.

Table 2.1: Ranges of leachate parameters at different phases of biological process (Source: Zainol, 2015)

Parameter	Transition	Acid Formation	Methane Fermentation	Maturation
COD (mg/L)	480-18,000	1500-71,000	580-9760	31-900
Total volatile acids (mg/L as acetic acid)	100-3000	3000-18,800	250-4000	0
NH_3N (mg/L)	120-125	2-1030	6-430	6-430
pH	6.7	4.7-7.7	6.3-8.8	7.1-8.8
EC (µS/cm)	2450-3310	1600-17,100	2900-7700	1400-4500

2.4 Leachate Composition and Characteristics

Generally, the pollutant in leachate can be divided into four groups which are dissolved organic matter, inorganic components, heavy metal, and a xenobiotic organic compound. It is characterized based on its appearance, pH, odor, DO, total suspended solids (TSS), total dissolved solids (TDS), turbidity, biological oxygen demand (BOD_5), COD, BOD_5/COD ratio, chloride content (Cl^{-1}), nitrate (NO_3^{-1}), ammonium nitrogen ($NH_{3\_}N$), total Kjeldahl nitrogen (TKN, total phosphorus (P), and sulfate (SO_4^{-2}) loading (Saleem et al., 2017). The color of the landfill leachate can be yellow, brown, or black due to the process of decomposition of organic matter. These characteristics make leachate difficult to manage due to its complexity. According to Renou et al., (2008), leachate can be classified as a soluble organic and mineral compound that is generated when water that passing and infiltrates into the refuse layers, extraction of series of contaminants and triggered a complex interplay between the biogeochemical and hydrological reactions. Other than that, landfill leachate also can be classified based on the decomposition process of the waste and its characteristics where it is influenced by factors such as climatic condition, precipitation, types of landfill waste, landfill age, and operation mode (Aziz and Ramli, 2018; Zainol, 2015). Hence, the characteristics and quality of leachate are changed over time depends on the different waste biodegradation phases (Kamaruddin et al., 2017).

The most two important factors in characterizing the landfill leachate are volumetric flow rate and its composition (Mukherjee et al., 2015). Figure 2.3 below illustrates the water cycle in a sanitary landfill where the flow rate of leachate is closely linked to precipitation (P), surface run-off (R), and infiltration or intrusion of groundwater that percolation through the landfill (Kamaruddin et al., 2017). The climate also has a great influence on leachate production as precipitation (P) and evapotranspiration (E) are involved in the process of the water cycle in the landfill. The leachate flows are changing literally with the weather such as increase during rainy day periods.

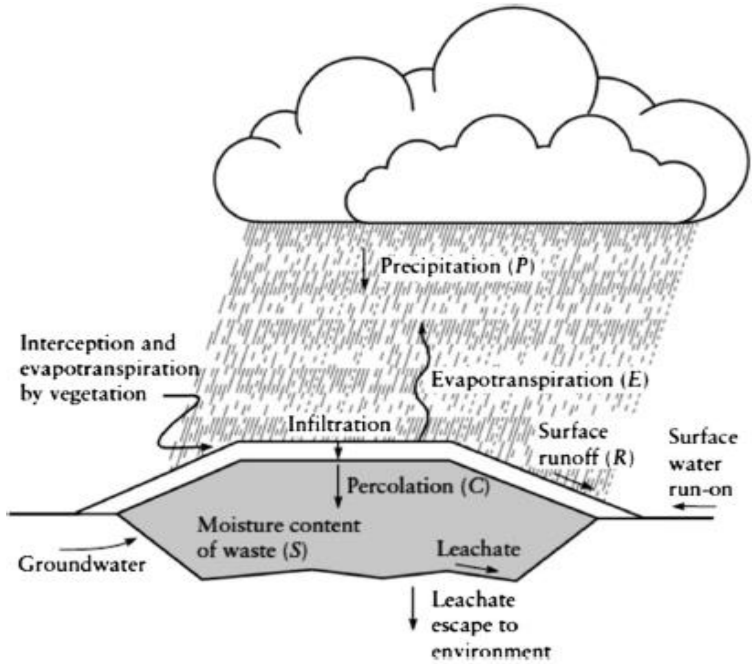

Figure 2.3: Water cycle in a sanitary landfill
(Source: Kamaruddin et al., 2017)

The composition of the landfill leachate depends on the age of the landfill (Zainol, 2015). The types of leachate can be classified according to landfill age which is young, intermediate, stabilized, and old. The composition of leachate may vary within the successive aerobic, acetogenic methanogenic, stabilization stages of the waste evolution also landfill age as shown in Table 2.2 (Mukherjee et al., 2015). The concentration and biodegradability of leachate usually decrease with its age and addition of time as the leachate strength will decline over consecutive years due to biological breakdown of compounds and precipitation of soluble elements (Aziz & Ramli, 2018; Mukherjee et al., 2015). Young leachate has a low molecular weight of organic compounds compared to old leachate that has organic compounds with a wide range of molecular weight (Mukherjee et al., 2015). Old landfill leachate is considerably more challenging as it is persistent and biologically difficult to cure (Zakaria and Aziz, 2018). Hence, the selection of suitable treatment methods should be based on the characteristics and composition of leachate to meet the standard of leachate emission.

Table 2.2: Physicochemical parameters of leachate according to different age

No.	*Parameter*	*Unit*	*Type of landfill leachate*		
			Young (<5 years)	*Intermediate (5–10 years)*	*Stabilised (>10 years) and semi-aerobic*
1	pH		<6.5	6.5–7.5	>7.5
2	COD	mg/L	>10,000	4,000–10,000	<4,000
3	BOD_5/COD		0.5–1.0	0.1–0.5	<0.1
4	Organic compound		80% VFA[a]	5–30% VFA[a] + HFA[b] HFA[b]	HFA[b]
5	NH_3-N	mg/L	<400	NA[c]	>400
6	TOC/COD		<0.3	0.3 –0.5	>0.5
7	Kjeldahl nitrogen	g/L	0.1–0.2	NA[c]	NA[c]
8	Heavy metals	mg/L	Low to medium	Low	Low
9	Biodegradability		Important	Medium	Low

(Source: Aziz & Ramli, 2018)

2.4.1 Dissolved Organic Matter

Dissolved organic matters (DOM) are the main components in the landfill leachate and their composition may provide useful criteria to choose a suitable treatment process (Liu et al., 2020; Awang, 2011). DOM usually contains a complex group and have active functional groups (carboxyl group, phenolic hydroxyl group, amino group, aldehydes, chlorinated benzenes, benzothiazolone, ketones, aliphatic compounds, nitrogen compounds, chlorinated dioxins and furans, phosphates, aromatic polyaromatic hydrocarbons, and pesticides) which it makes itself easy to interact with hydrophobic organic pollutant and metal (Liu et al., 2020; Scandelai et al., 2019). Generally, DOM can be classified into three fractions which are single compounds that can identify by specific analyses, biodegradable and low molecular weight; humic acid-like fraction characteristics; fulvic acid-like compounds (Liu et al., 2015). DOM plays an important role as a leachate pathway in the migration of heavy metals from MSW to environments due to its kind of heterogeneous mixture (Liu et al., 2015). Other than that, DOM also important in pollutant degradation as it is a bulk parameter that covering various organic degradation products and humic-like compounds and it contains both aromatic and aliphatic components. However, the reaction between DOM with others pollutants and chemicals can produce toxic contaminates and cause the problem of organic pollutants such as an increase of antioxidant and bacteria proliferation (Liu et al., 2020; Zanki et al., 2020).

2.4.1(a) Chemical Oxygen Demand (COD)

According to (Kamaruddin et al., 2017), the parameters that are commonly used to determine the content of dissolved organic matter in leachate are Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD), and Total Organic Carbon (TOC). COD, BOD, and TOC are determined by oxidizing organic matter using chemical agents, bacteria, and process of thermal respectively. COD is one of the important parameters in leachate treatment as it measured the oxygen required to break down the pollutants especially organic substances in leachate. It is commonly used for discharge consent and various type of organic matter can be measured using COD compared to BOD (Kamaruddin et al., 2017). Potassium Dichromate is one of the strong oxidizing agents that usually used to oxidize organic matter through the reaction of chemical oxidation

($K_2Cr_2O_7$ method). In this method, the presence of silver sulfate as catalyst and $K_2Cr_2O_7$ are reacting in oxidize the organic substances at 150°C for 2 h (Taromsary et al., 2019). The high concentration of COD in leachate will indicate a low concentration of volatile fatty acids, large amounts of fulvic and humic substances in leachate also it is considered stable in methanogenic conditions (Zainol, 2015). However, Kylefors et al., (2003) state that not only organic substances can be measured by COD but other substances such as inorganic substances also may affect the COD value. Taromsary et al., (2019) reported that the presence of salinity in landfill leachate could cause several challenges in treatment and interference in measurement of COD due to the presence of the chloride. According to a study by Geerdink et al., (2017), the presence of ammonia and the organic amine is also identified as the others substances that could affect the measurement of COD and cause a continuous reduction of dichromate occurred. Therefore, in order to reduce the interference of chloride and other substance, the addictive such as $HgSO_4$, $AgNO_3$ and Cr(III) or other modified methods can be used (Geerdink et al., 2017).

According to Standard Methods, APHA 2017, there are three methods in the determination of COD which are the open reflux method, the closed reflux (titrimetric method), and the closed reflux (colorimetric method). Over the last decades, more efficient analytical methods and instruments for the determination of COD have been developed and the spectrophotometer is one of them (Dimitrova et al., 2013; Li et al., 2018). Many studies used the spectrophotometric method in the determination of COD of landfill leachate due to fast reaction time, less time consuming, good precision, a lower chemical reagent used and sample volumes, and less waste produced (Ghanbari et al., 2020; Shadi et al., 2020; Li et al., 2018; Wang et al., 2019). Other than that, the spectrophotometric method also has been applied in environmental laboratories as it is rapid digestion time, minor environmental issues, and low analytical cost (Li et al., 2018). Therefore, in this research, the determination of removal of organic matter could be identified through the determination of COD using closed reflux which is the colorimetric method by using spectrophotometer instrument (DR 2800).

2.5 Treatment of Leachate

Sanitary landfill leachate has been recognized as high polluted industrial wastewater due to its high content of refractory materials. The composition of landfill leachate such as organic matter that can contribute to the toxicity of leachate which can cause serious environmental problems. The discharge of landfill leachate should be avoided and treat properly as it may percolate through soils and cause the contamination of groundwater (Awang, 2011). Therefore, special treatments and management of leachate are required before discharge to the environment and need to comply with the effluent standard limit (Environmental Quality Act 1974). Various methods and leachate treatment systems have been applied based on the characteristics of leachate. The selection of treatment process should consider the cost of treatment, treatment process residuals, etc. There are three categories of methods that are commonly adapted for wastewater treatment which are biological treatment, physical-chemical treatment, and a combination of both techniques (Luo et al., 2020). The efficient treatment should be applied based on the actual characteristics of landfill leachate such as the concentration of ammoniacal nitrogen and the biodegradability of the leachate. The selection of suitable treatment for landfill leachate is shown in Figure 2.4 below.

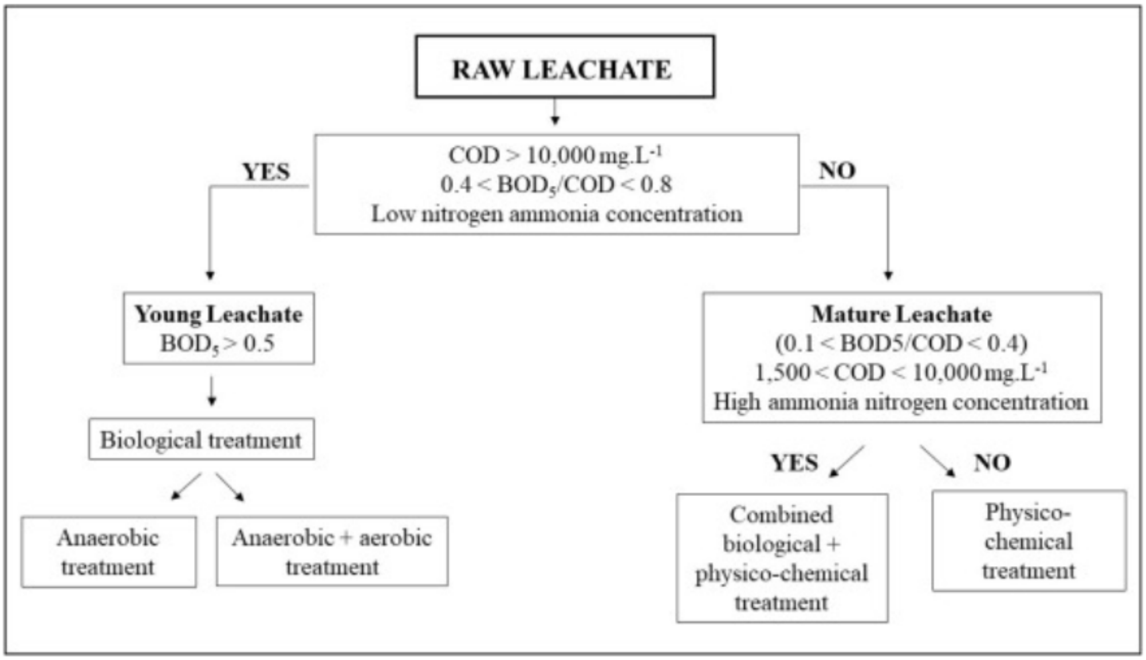

Figure 2.4: Parameters that considered in the selection of suitable treatment for leachate (Source: Costa et al., 2019)

2.5.1 Biological Treatment

Biological treatment in leachate usually is the most common practice as it is more economical compared to other methods such as physicochemical treatment (Zainol, 2015). Two processes are involved in biological treatment which are aerobic and anaerobic. The process of biological treatments involves the microorganism community degrading the organic compounds to CO_2 and sludge. This process also is very efficient for the treatment of leachates from young landfills (<5 years) as it contains high concentrations of biodegradable material such as volatile fatty acid (Costa et al., 2019; Zainol, 2015). However, for the leachates from mature landfills (>10 years), the treatment is not applicable as the leachate contains compounds that are not easily degraded such as humic acids (Costa et al., 2019).

Aerobic treatment is very effective in treating young leachate as it usually has a high concentration of COD and BOD. Through this method, the content of COD can be reduced up to 50%, and most organic components are transformed into CO_2 and sludge during the early stages of waste composition (Zainol, 2015). The type of most common aerobic treatments that have been adopted widely is aerated lagoons, conventional activated sludge processes, sequencing batch reactor (SBR), trickling filters, and moving bed biofilm reactors (MBBR) (Ahmad et al., 2018).

Other than that, the anaerobic treatment also one of the biological processes that involve the decomposition of organic matters and generates biogas which is CH_4 (Chelliapan et al., 2020). This treatment will be a more effective treatment solution for leachate compared to aerobic treatments due to its ability of bacteria to break down the complex organics (Chelliapan et al., 2020; Zainol, 2015). The anaerobic treatment usually involves two stages which are acid fermentation and methanogenic. The examples of the anaerobic process that adopted are anaerobic activated sludge, conventional anaerobic digester, and anaerobic filter. However, although the biological treatment method is known as very effective in leachate treatment, the removal of organic compounds through this technique is not easy (Ahmad et al., 2018). Thus, the development of a combination of the biological method with the physical/chemical method can enhance the effectiveness in the treatment of leachate.

2.5.2 Physical/Chemical Treatment

Numerous research studies in the treatment of leachate show that physical-chemical treatment is more applicable for older leachate compared to biological treatment

(Gautam et al., 2019; Luo et al., 2020). This is due to the characteristics of stabilized leachate which has low biodegradable organic fraction (low concentration of COD), refractory substances such as humic acids, and ammonia. According to Luo et al., (2020), physical-chemical treatment of leachate is usually used along with the biological method such as in pre-treatment of leachate or last purification to improve the effectiveness of leachate treatment. Other than that, this method is also used to treat specific pollutants such as ammonia or nitrate removal (Aziz and Ramli, 2018). Various type of physical and chemical treatment has been used successfully in treating stabilized and old landfill leachate which is coagulation and flocculation, flotation, ion exchange, adsorption, chemical oxidation/advanced oxidation process (AOP), electrochemical oxidation, and membrane filtration (Luo et al., 2020). Ullah et al., (2020) exposed that the individual physical-chemical treatments have limitations of its treatment and it is required to combine with other processes of treatment to achieve the guidelines of effluent.

Advanced Oxidation Process (AOP) has been discovered as a greener and innovative technology in the effective treatment of various types of wastewater (Babu et al., 2019; Gautam et al., 2019). The AOP mainly involves the generation of hydroxyl radicals (•OH) in treating wastewater by the process of purification (Gautam et al., 2019). This process also has been found to be effective in remove organic and inorganic pollutants and degrade highly toxic substances (Luo et al., 2020). There are five processes that involve the AOP process which are ozone, chemical, electricity process, photoassisted process, and miscellaneous as shown in Figure 2.5. Other than that, a study by Luo et al., (2020) founds that the AOP process also can be integrated with other processes to degrade the contaminants into harmless products. Thus, this AOP process can be applied in this research study to treat the landfill leachate as it has many benefits such as it discourage the use of chemicals, user-friendly, and can operate to purify the wastewater at ambient temperature, and pressure (Gautam et al., 2019). The electrocoagulation process in leachate treatment which is categorized under the AOP process of electricity is mainly focused in this research work and the combination of electrocoagulation with the process of ultrasonic is also studied to differentiate the effective treatment between single and combined methods.

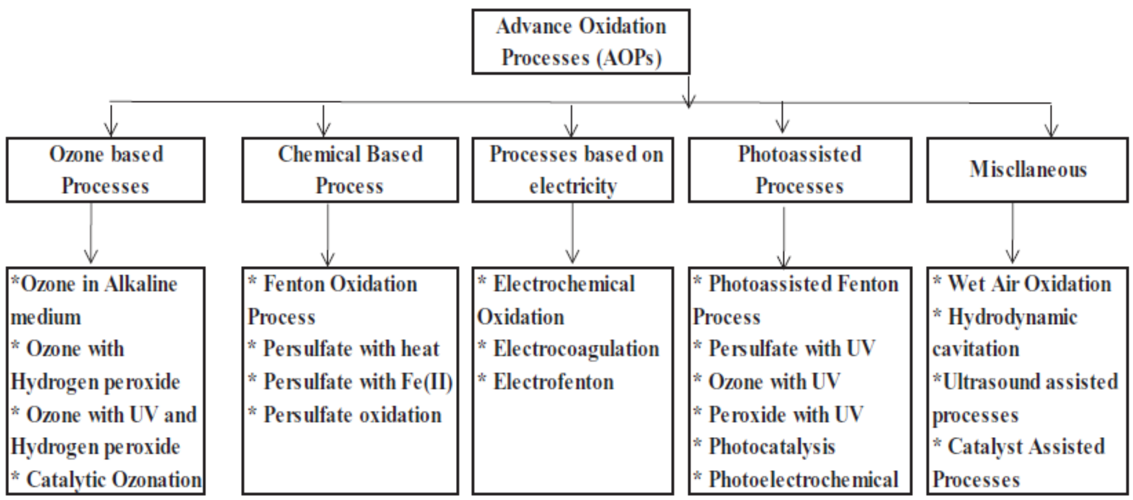

Figure 2.5: Advanced Oxidation Process
(Source: Gautam et al., 2019)

2.6 Electrocoagulation

In recent years, electrochemical methods have been established as one of the potential techniques in wastewater treatment and they also have been used in landfill leachate treatment as a pre-treatment stage (Ghanbari et al., 2020; Huda et al., 2017). This treatment method is categorized as one of the AOP technologies that involve electrolysis, ionization, hydrolysis, and generation of free radicals that can enhance the treatment of leachate (Gautam et al., 2019). Electrocoagulation (EC) is one of electrochemical which involves the use of electrical current for treatment without adding any chemical coagulants and the process of flotation and precipitation occurs simultaneously (Apaydin and Özkan, 2020; Aziz and Ramli, 2018). This treatment method is also known as an environmentally friendly and simple method in which it is not required any addition of chemical substances and less treatment time (Zailani and Zin, 2018). Furthermore, one of the most significant advantages of the EC method is that it can be scaled up to pilot and industrial levels (Shahedi et al., 2020). Other than leachate treatment, the applications of the EC method have been used widely in various types of wastewater such as textile wastewater, tannery, pulp and paper industry wastewater, laundry wastewater, and other types of wastewater that contain hazardous contaminants (Mohammadizaroun and Yusoff, 2014). This shows that the EC method has been used successfully in purifying and treating different types of water and wastewater.

The EC method usually consists of pairs of electrodes arranged in pairs of two which are anodes and cathodes. The treatment of wastewater by using the EC method applies the principles of electrochemistry where the cathode is oxidized and water is reduced. The reaction starts when the metal electrode is emitted into the apparatus and exposed to the electric supply which generates metal cations. The hydroxyl ions also formed when the cathode electrode makes contact with the wastewater sample (hydrolysis process). When the anode and cathode make contact with the wastewater, the particulates will be neutralized due to the reaction of generated metal hydroxides with the pollutants (Nidheesh et al., 2021). Then, it will cause the formation of hydroxide complexes at the cathode electrode that stabilizes and form agglomerates. The chemical equation from the reaction that occurred at the anode and cathode is shown below (Nidheesh et al., 2021).

At the anode, $$M_{(S)} \rightarrow M_{(aq)}^{n+} + ne^- \quad (2.1)$$

$$H_2O \rightarrow 4H^+_{(aq)} + O_{2\,(g)} + 4e^- \qquad (2.2)$$

At the cathode, $$nH_2O + ne^- \rightarrow \left(\frac{n}{2}\right) H_{2\,(g)} + nOH^-_{(aq)} \qquad (2.3)$$

By referring to the study from Nidheesh et al., (2021), it stated that during the EC process, the major process involve in pollutant removal is due to coagulation, flocculation, and adsorption. It also stated that metal hydroxide acts as an excellent adsorbent and it is responsible for the generation of the floc. The generation of hydrogen gas will assist the floc which acts as floating sludge to the surface of the wastewater. The agglomerates also can form at the bottom of wastewater. However, according to Butler et al., (2011), the particulates that form at the top of the tank from the electrocoagulation-flotation apparatus are caused by the hydrogen bubbles that are created from the anode. Figure 2.6 below shows the example of the EC apparatus setup and the reaction that occurred during the treatment process.

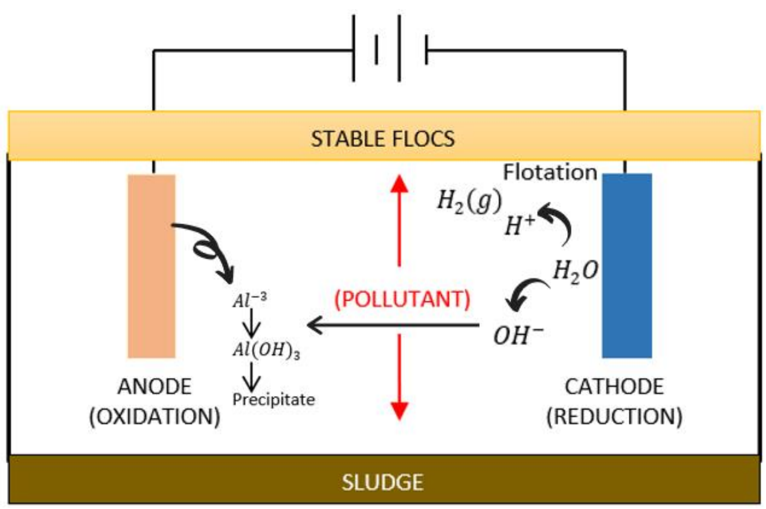

Figure 2.6: Schematic diagram of the experimental setup

Various studies of electrochemical processes such as EC have resulted in good efficiency in removing sources of color, ammonia, suspended solids, and COD from