***NEPHELIUM LAPPACEUM* SEED AS NATURAL COAGULANT/FLOCCULANT FOR LANDFILL LEACHATE TREATMENT**

by

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List of Abbreviations

APLS	Alor Pongsu Landfill Site
BOD	Biochemical Oxygen Demand
COD	Chemical Oxygen Demand
SS	Suspended Solids
DO	Dissolved Oxygen
FESEM	Field Emission Scanning Electron Microscopy
FTIR	Fourier Transform Infrared Spectroscopy
HDPE	High Density Polyethylene
$SnCl_4$	Tin tetrachloride
SVI	Sludge Volume Index
Vs	Sludge settling velocity
PZC	Point of Zero Charges
MLSS	Mixed Liquor Suspended Solids
NH_3-N	Ammonia-Nitrogen
TDS	Total Dissolved Oxygen
HCl	Hydrochloric Acid

BIJI *NEPHELIUM LAPPACEUM* SEBAGAI BAHAN PENGGUMPAL/PENGELOMPOKAN SEMULA JADI DALAM OLAHAN LARUT LESAPAN

ABSTRAK

Penjanaan larut lesapan di tempat pelupusan sampah telah memberi banyak risiko pencemaran alam sekitar yang ketara kerana kandungannya yang berbahaya dan bertoksik boleh mendatangkan mudarat kepada ekosistem jika terus dilepaskan ke dalam alam sekitar. Bahan penggumpalan dan pengelompokan berasaskan kimia biasa digunakan dalam proses penggumpalan – pengelompokan. Akan tetapi, keburukan penggunaan bahan-bahan kimia telah menyebabkan para penyelidik mencari bahan-bahan semula jadi untuk menggantikan atau mengurangkan jumlah bahan penggumpal dan pengelompokan berasaskan kimia. Biji *Nephelium lappaceum* telah dipilih dalam kajian ini sebagai bahan penggumpal/pengelompok semula jadi untuk mengolah larut lesapan dari tapak pelupusan sampah Alor Pongsu (APLS). Keberkesanan biji *Nephelium lappaceum* ini ditentukan melalui satu siri ujian balang dengan mencari keadaan operasi yang terbaik (pH dan dos) untuk biji *Nephelium lappaceum* dan $SnCl_4$ sebagai bahan penggumpal. Prestasi penggumpalan dan pengelompokan diukur melalui kecekapan penyingkiran warna, pepejal terampai (SS), dan keperluan oksigen kimia (COD). Keputusan menunjukkan biji *Nephelium lappaceum* sebagai bahan penggumpal tunggal boleh menghapuskan 19.48% warna, 35.62% COD, dan 0% SS dalam kondisi yang terbaik iaitu pH 6 dengan dos 2 g/L. Sementara itu, $SnCl_4$ sebagai bahan penggumpal tunggal boleh memberikan peratusan penyingkiran yang tinggi iaitu 98.22% SS, 88.86% warna, dan 84.22% COD pada pH 7 dengan 10.50 g/L $SnCl_4$.

Dengan membandingkan kecekapan penyingkiran oleh kedua-dua bahan penggumpal, biji *Nephelium lappaceum* tidak memberi kesan yang memuaskan, tetapi biji ini menunjukkan prestasi yang sangat baik dalam menghapuskan bahan pencemar sebagai bahan bantuan penggumpalan atau pengelompokan dengan $SnCl_4$ sebagai pengumpalan utama dan justeru dapat mengurangkan kepekatan $SnCl_4$ dari 10.50 g/L ke 8.40 g/L dengan dapat merawat 88.86% SS, 87.57% warna, dan 75.94% COD. Selain itu, keberkesanan biji *Nephelium lappaceum* dapat meningkatkan prestasi enap cemar dan kebolehan enapan. Biji *Nephelium lappaceum* meningkatkan saiz flok dari ketumpatan flok, pengenapan halaju enap cemar dan juga indeks isipadu enap cemar. Kesimpulannya, biji *Nephelium lappaceum* mempunyai potensi untuk digunakan sebagai pengelompokan semula jadi dengan bahan pengumpalan berasaskan kimia untuk mengolah larut lesapan dari tapak pelupusan sampah dan ini boleh mengurangkan penggunaan bahan pengumpalan berasaskan kimia dan meningkatkan prestasi pengenapan enap cemar.

***NEPHELIUM LAPPACEUM* SEED AS NATURAL COAGULANT/FLOCCULANT IN LANDFILL LEACHATE TREATMENT**

ABSTRACT

Landfill leachate produced from landfilling has caused pollution risk to environment due to the harmful and toxic content if directly discharged into environment. Chemical based coagulants and flocculants are commonly used in coagulation – flocculation process. However, the drawbacks of using these chemical materials have triggered researchers to find natural materials to substitute or reduce the amount of chemical based coagulants and flocculants. *Nephelium lappaceum* seed has been chosen in this study as natural coagulant/flocculant to treat landfill leachate from Alor Pongsu Landfill Site (APLS). The effectiveness of the seed was determined through a series of jar tests by finding the best operational conditions (pH and dosages) for *Nephelium lappaceum* seed and $SnCl_4$ as coagulant. The performance of coagulants was measured through the removal efficiencies of colour, suspended solids (SS), and chemical oxygen demand (COD). Results indicated that *Nephelium lappaceum* seed as sole coagulant could remove 19.48% of colour, 35.62% of COD, and 0% of SS at the best conditions which is pH 6 with dosage of 2 g/L. Meanwhile, $SnCl_4$ as sole coagulant could remove high percentage of pollutants which is 98.22% of SS, 88.86% of colour, and 84.22% of COD at pH 7 with 10.50 g/L of $SnCl_4$. *Nephelium lappaceum* seed is not effective as $SnCl_4$ when used as sole coagulant, but it shows excellent performance in removing pollutants as flocculant with $SnCl_4$ as primary coagulant and thus, has accomplished the aim of reducing the amount of $SnCl_4$ from 10.50 g/L to 8.40 g/L with removal efficiencies of 88.86% colour, 87.57% SS, and 75.94% COD. Besides

that, the effectiveness of *Nephelium lappaceum* seed can be further proven with sludge performance and settleability. *Nephelium lappaceum* seed helps in improving the sludge performance and settleability by increasing flocs size, density of flocs, sludge settling velocity as well as sludge volume index. In conclusion, *Nephelium lappaceum* seed has a potential to be used as natural flocculant with chemical based coagulant to treat landfill leachate treatment and this could reduce the usage of chemical based coagulant and improve the sludge settling performance.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Solid waste management has become a major concern in the world, especially in developing countries. This is due to solid waste generation that keeps increasing year by year corresponding to the growth of population, economic prosperity, and rapid urbanization (Ho et al., 2017). It is about 95% of solid waste generated has been dumped in landfill (Bashir et al., 2010). Malaysia highly depends on landfilling as a main waste disposal method due to the simple disposal procedure and low cost as compared to other disposal methods such as incineration and anaerobic waste treatment (Moh and Latifah, 2017) However, this kind of disposal method has led to overflowing of landfill that may pollute the surrounding environment with its toxicity.

Landfill leachate is a complex wastewater generated when water percolates through solid waste at landfill that may contain high concentrations of biodegradable and non – biodegradable organic matters, ammonia nitrogen, phosphate, colour, and suspended solid (Ching et al., 2011). Disposal of raw leachate directly without undergoing any treatment could cause soil, surface and groundwater contamination, which indirectly would affect living organisms, including degeneration of human health. For that reason, Malaysian government has enforced environmental rules and regulations that require landfill leachate to be treated and monitored prior discharge. This was done to prevent contamination of water resources and reduce the harmful impacts of landfill leachate to the environment.

Several treatments that are used to treat wastewater and water have been employed to treat landfill leachate. For instance, biological treatment, chemical treatment, coagulation – flocculation, and reverse osmosis that are commonly used in wastewater treatment have been applied in landfill leachate treatments to minimize the contaminants and reduce the negative impacts on the environment (Aziz et al. 2011). However, some of these treatments are not suitable to be used in certain landfill leachate due to the variability of leachate characteristics. Moreover, landfill age also plays an important role in deciding suitable treatment methods due to the changes of leachate characteristics (Zin et al., 2013). Young landfill leachate releases large amount of volatile fatty acid content and can be characterized by high BOD which is more than 10 g/L and high ratios of BOD/COD (>0.7) (Kurniawan et al., 2006). Therefore, the most suitable treatment to be used for young landfill leachate is biological method due to its high concentration of biodegradable substances in the landfill leachate (Comstock et al., 2010). On the other hand, matured and stabilized landfill leachate has high strength of COD (500 - 4500 mg/L), pH higher than 7.5, and low biodegradability (BOD/COD<0.1) (Bashir et al., 2010). During this phase, chemical and physical treatments are the best methods to be used, as biological treatment is ineffective in removing pollutants.

Coagulation – flocculation process is a typical physico – chemical treatment process that is broadly used in most raw water and industrial wastewater treatment. This process involves the mechanisms of destabilization of colloidal particles by adding coagulants and promoting agglomeration of flocs (Teh et al., 2014). Generally, inorganic coagulants are used in the coagulation – flocculation process, such as alum, polyaluminium chloride (PAC), ferrous sulfate, and ferric chloride (Mojiri et al., 2014). Inorganic metal salts are effective pollutant removal, but due to the increasing awareness on toxicity from excessive use of inorganic coagulants, there have been many studies conducted to alternatively replace or reduce inorganic coagulants with natural coagulant or flocculant in wastewater treatment (Antov et al., 2012). Nevertheless, these natural coagulants are still not commonly used in landfill leachate treatment even though they are biodegradable, eco-friendly, low in price, as well as abundant in source.

1.2 Problem Statement

Landfill leachate is rich in chemical oxygen demand (COD), colour, suspended solids (SS), and heavy metals (Mojiri et al., 2014). Therefore, landfill leachate needs to be treated before being discharged into the environment. Untreated landfill leachate may deteriorate the quality of receiving water bodies, such as lake, river, and stream, near the landfill sites.

The Malaysian legislation has consented a list of parameters of landfill leachate discharge limit in Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009. The concentrations of SS, colour, and COD in landfill leachate are often found to be multiple times higher than the permissible discharge limit (Zakaria et al., 2015). Hence, these parameters need to be lowered within the range of permissible discharge limit before being discharged into the environment.

There are various treatments of landfill leachate, including biological and physico-chemical treatments. Alor Pongsu Landfill Site (APLS) is a mature and stabilize landfill which has no treatment applied on its landfill leachate. Physico-chemical treatment is more suitable to be used in this kind of landfill leachate as it has low BOD content (Ghafari et al., 2010). Coagulation – flocculation is one of the physico-chemical treatment methods that has been used widely in water, wastewater and landfill leachate treatment. Coagulation – flocculation process is commonly practiced using inorganic metal salts as coagulant/flocculant. However, excessive usage of these inorganic metal salts may cause adverse effects to the environment and pose a risk to human health (Renou et al., 2008). Therefore, it is vital to develop and utilize natural coagulants/flocculants for landfill leachate treatment in order to reduce and eliminate the adverse effects to the living organisms and environment.

Malaysia is one of the tropical countries that is rich with various kinds of fruits. Thereby, many food industries take advantage of this richness by producing canned fruits. *Nephelium lappaceum* canning industry is well-established in Malaysia and Thailand, and this industry involves the production of *Nephelium lappaceum* fruits in syrup (Abidin et al., 2014). During the canning process, *Nephelium lappaceum* fruits are deseeded and the seeds are usually disposed, thus becoming wasted by-products. Therefore, utilization of *Nephelium lappaceum* seed is needed in order to improve sustainability of the fruit canning industry waste management, as well as reducing solid waste generation. There was a preliminary study done on *Nephelium lappaceum* seed polysaccharide as natural coagulant in treating turbidity of water (Abidin et al., 2014), but there is no study done to date to test the usage of *Nephelium lappaceum* seed as natural coagulant or flocculant in treating landfill leachate.

Therefore, this leaves a research gap that must be filled, which triggered the interest to study on the effectiveness of *Nephelium lappaceum* seed as coagulant or flocculant in landfill leachate treatment. In this research, the investigation on *Nephelium lappaceum* seed covers the usage of the seed as sole coagulant, and as flocculant with $SnCl_4$ as coagulant to remove SS, colour, and COD from landfill leachate.

1.3 Objectives

This research aims to determine the effectiveness of using *Nephelium lappaceum* seed as coagulant/flocculant in removing colour, suspended solid (SS), and chemical oxygen demand (COD) in landfill leachate treatment. In order to accomplish this, the following objectives are outlined:

1) To investigate the characteristics of *Nephelium lappaceum* seed as coagulant/flocculant in term of pH, particle size, molecular weight, zeta potential, functional groups, surface morphology

2) To determine the best operational conditions (pH and dosage) of *Nephelium lappaceum* seed and $SnCl_4$ as coagulants for treating landfill leachate by evaluating and comparing the performance of the treatment in terms of colour, COD, and SS

3) To investigate the possibility of reducing the quantity of Tin (iv) chloride ($SnCl_4$) as primary coagulant in the presence of *Nephelium lappaceum* seed as flocculant

4) To examine the performance of sludge formed in coagulation – flocculation process by using $SnCl_4$ as coagulant with and without the aid of *Nephelium lappaceum* seed as flocculant at the best operational conditions.

1.4 Scope of Work

The research focuses on determining the efficiency of natural material (*Nephelium lappaceum* seed) as an alternative coagulant or flocculant besides metal salts materials that are currently widely used, such as alum and PAC in landfill leachate treatment. The research was done by conducting laboratory tests to achieve the main research objective. Leachate samples were taken from Alor Pongsu Landfill Site (APLS), which is located in Perak. Characteristics of the leachate samples on their chemical and physical properties were observed for four months, which is from January to April 2017. The effectiveness of coagulant was evaluated through removal of chemical oxygen demand (COD), suspended solids (SS), and colour from landfill leachate. The best operational conditions of coagulation were identified through determination of optimum pH and optimum dosage of both *Nephelium lappaceum* seed and $SnCl_4$. The evaluation was made through jar test operation that mimicked the coagulation process in the treatment plant. The jar test process involved rapid mixing, slow mixing, and settlement with different time limit. These three operations were derived from similar past studies. Sludge formed resulting from the coagulation process at best operational conditions of two coagulants was evaluated based on sludge settling rate, sludge volume index, functional groups, floc size, density, and surface morphology.

1.5 Dissertation Outline

The dissertation consists of five chapters. Chapter 1 comprise the introduction to this project, which includes background of the present study, problem statement, objectives, and scope of the study. Chapter 2, focuses on literature review, and similar past studies done on topics related to the present study, which include studies and articles on solid waste management in Malaysia, coagulation – flocculation process in treating landfill leachate, and natural coagulants. Chapter 3 stipulates the methodologies used in this research. This chapter describes the details on the research design and procedures of the experiment. Chapter 4 contains comprehensive discoveries, analysis, and results gained from the present study. Elaboration and details on the findings are covered in this chapter as well. The last chapter, which is Chapter 5, presents the overall conclusion to the research findings. It also includes useful and beneficial recommendations for future research work improvement and enhancement.

CHAPTER 2

LITERATURE REVIEW

2.1 Solid waste management

Solid waste management refers to all activities and actions required to manage solid waste from its inception to its final disposal stage including collection, transportation, waste treatment and disposal of solid waste, together with monitoring and regulation of disposal (UNSD, 1997). It also comprises the legal and regulatory framework that relates to solid waste management. Hence, solid waste management is an obligation placed upon everyone in reducing the adverse effects of waste on health, environment, and aesthetic values.

However, solid waste management has become a crucial issue to be solved as the volume of solid waste generated keeps on increasing every year due to many factors in particularly the growth of population and economic activities. The increase in world population causes global urbanization and economic expansion of developing countries, which in turn contributes to the accelerated increase of rate of solid waste production. According to a recent study by World Bank (2012), the global solid waste generation is estimated to be 1.3 billion tonnes per year or an average of 1.2 kg/capital/day.

However, it can be seen that the waste generation rates per capita differ across countries and cities depending on the level of urbanization and economic wealth. For example, developing countries such as North America and European Union produced high generation of solid waste, and it is expected that Asia, Latin America, and South

Africa would also experience the same in decades to come. It is predicted that middle and low-income countries would follow the same trends with the increase of urbanization levels and economic development (UNEP, 2010).

In Malaysia, solid waste volume grows from 9.0 million tonnes in 2000 to about 10.9 million tonnes in 2010, to about 12.8 million tonnes in 2015 and is expected to reach 15.6 million tonnes in 2020 (Agamuthu & Dennis, 2011). This recorded amount has become a significant concern to Malaysia as this shows that the country needs to provide more options of waste disposal methods such as incineration, recycle, and composting. However, these options are costly and time consuming, hence, landfilling becomes the main disposal methods to dispose solid wastes. On top of that, habits and attitudes of Malaysian in managing their solid wastes have resulted in difficulty to use other disposal methods than landfilling (Moh & Latifah, 2017). For that reason, Malaysia depends heavily on landfilling to dispose the solid wastes collected.

2.2 Landfill

Many disposal methods such as incineration, composting, anaerobic treatment, and recycling have been used in many countries especially in Singapore, Japan, and America. Regardless of many alternatives of solid waste disposal methods, landfilling is still being practiced in most of the countries in the world (Mohajeri et al., 2010). This is because the final disposal of unrecovered wastes, which are the by-products from the alternative methods, is usually dumped at landfills (Moh & Latifah, 2017). Nevertheless, the situation is not the same in some countries as they choose landfilling as their primary disposal method without applying the alternative methods beforehand.

Aforementioned, Malaysia depends on landfill to dispose solid wastes collected. About 95% of waste collected is dumped at the landfill and another insignificant percentage of solid wastes are treated, processed, or being dumped illegally. For this reason, more than 290 landfills in Malaysia have existed and almost half of the landfills are old dumpsites that are not operating anymore. Table 2.1 shows the number of operating and closed landfill sites in Malaysia.

Table 2.1: Number of operating and closed landfills in Malaysia (JPSPN, 2017)

States	Number of operating landfills	Number of closed landfill	Total
Johor	14	23	37
Kedah	8	7	15
Kelantan	13	6	19
Melaka	2	5	7
Negeri Sembilan	7	11	18
Pahang	16	16	32
Pulau Pinang	2	1	3
Perak	17	12	29
Perlis	1	1	2
Sabah	19	2	21
Sarawak	49	14	63
Selangor	8	14	22
Terengganu	8	12	20
WP Kuala Lumpur	0	7	7
WP Labuan	1	0	1
Total	165	131	296

Construction of new landfill becomes a challenge as the land has become scarce parallel to the growing population. The problem has become the alarming issue to the respective authority to provide the land for landfill especially in large city.

Besides that, the dependency on landfill as main disposal method has caused a major concern to the environment including human health. Landfill produces methane gases through anaerobic decomposition of waste, leachate through percolation of water through the wastes in landfill, odour as well as vector borne diseases that are spread by birds and birds (Bashir et al., 2011). This may cause contamination to atmosphere, surface and groundwater, as well as harm the public health.

2.3 Landfill Leachate Generation and Characteristics

Disposal of solid waste using landfill will result in generations of landfill leachate as it is an unavoidable circumstance. Landfill leachate is the dark aqueous effluent which is a complex and high-strength wastewater that exhibits acute and chronic toxicity. It is produced through the percolation of liquid, either from rainwater or water from the waste itself, combined with the decomposition formed by physico-chemical and biological reactions (Zhang et al., 2016).

Landfill leachate may contaminate surface and groundwater if it is directly discharged into the environment due to its complex characteristics. Characteristics of landfill leachate vary from different landfills as they are affected by several factors including composition of solid wastes at landfill, climate conditions, site hydrogeology, moisture routing through the landfill, design and operation of the landfill and landfill age (Ghafari et al., 2010). Despite that, the main factor that indicates the overall characteristics and composition of landfill leachate is landfill age.

Landfill leachate can be categorized into young leachate which is produced from young landfill (<5 years), intermediate leachate from landfill age between 5 to 10 years, and mature and stabilized leachate that is produced from mature landfill (>10 years). There are significant differences of leachate characteristics between these landfill leachates. Table 2.2 demonstrates the characteristics of leachate from young landfill to mature landfill respectively.

Table 2.2: Characteristics of landfill leachate respective to its age.

Parameters	Value, mg/L			References
	Young leachate	Intermediate leachate	Mature leachate	
pH	4.5 - 6.5	6.5 - 7.5	More than 7.5	Ghafari et al., 2010
COD	15,000 – 60,000	3,000 – 15,000	Less than 3,000	Ghafari et al., 2010
BOD_5/COD ratio	0.5 – 1.0	0.1 – 0.5	Less than 0.1	Ghafari et al., 2010
Ammonia Nitrogen	Less than 400	-	More than 400	Ghafari et al., 2010
Heavy metal	Low - medium	Low	Low	Davis and Cornwell, 2013

On account of the high presence of pollutants in the landfill leachate, Malaysia government has gazetted and enforced legislation related to the discharge of landfill leachate into the environment in order to minimize the impact of landfill leachate towards the environment. Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 states that no person should be allowed to discharge leachate that contains substances in concentration greater than those acceptable conditions into any soil, or into any inland waters (Yong & Aziz, 2016). Table 2.3 tabulates the permissible conditions for discharging landfill leachate into the environment in Malaysia. By referring to this legislation, it can be known which pollutant's concentration that needs to be lowered down prior to discharging.

Table 2.3: Landfill leachate discharge standards in Malaysia (EQA, 2009)

Parameter	**Unit**	**Standard**
Temperature	^oC	40
pH Value	-	6.0-9.0
BOD_5 at 20^oC	mg/L	20
COD	mg/L	400
Suspended Solids	mg/L	50
Ammoniacal Nitrogen	mg/L	5
Mercury	mg/L	0.005
Cadmium	mg/L	0.01
Chromium, Trivalent	mg/L	0.20
Arsenic	mg/L	0.05
Lead	mg/L	0.10
Copper	mg/L	0.20
Nickel	mg/L	0.20
Tin	mg/L	0.20
Zinc	mg/L	2.00
Boron	mg/L	1.00
Iron (Fe)	mg/L	5.00
Silver	mg/L	0.10
Barium	mg/L	1.00
Fluoride	mg/L	2.00
Formaldehyde	mg/L	1.00
Phenol	mg/L	0.001
Sulphide	mg/L	0.50
Oil and grease	mg/L	5.00
Colour	mg/L	100

2.4 Landfill Leachate Treatment

The presence of high concentration of pollutants in landfill leachate has become a major environmental issue in landfill site. Through direct discharge landfill leachate into the environment, this will cause a decline in surface and groundwater quality, and hence, may affect living organisms, including human health (Kamaruddin et al., 2015). Therefore, it is necessary to treat landfill leachate in order to remove all pollutants prior to discharge into the environment.

Landfill leachate treatments have been widely studied to find various possible treatments for landfill leachate, which include biodegradation, chemical method, physical methods, and membrane processes. Moreover, most of the treatments, either single or any combination of treatment methods, that are applied on landfill leachate originated from water wastewater treatment methods (Zin et al., 2013). Table 2.4 shows various treatment of landfill leachate.

Table 2.4: Various landfill leachate treatments (Metcalf & Eddy, Inc., 2014)

Treatment category	Treatment Approach	Brief description
Biological	Aerobic – suspended growth	Based on the growth and maintenance of a suspension of microorganisms. Examples: aerated lagoons, sequence batch reactors, activated sludge
	Aerobic – attached growth	The bacteria attach themselves on the supporting mediums such as rotor blades, plastics, and gravel. Examples: Rotating biological contactors (RBC), and trickling filters

Table 2.4: Various landfill leachate treatments (Continued from page 14)

	Anaerobic	The treatment takes place with the absence of oxygen. It involves two stages. First stage, the facultative microorganisms undergo conversion to change complex organic matter into organic matter. Meanwhile, second stage involves conversion of volatile organic acids to carbon dioxide and methane gas by anaerobic microorganisms.
Physico-chemical	Chemical precipitation	This technique is effective in removing of non-biodegradable organic compounds, ammonium, and heavy metal in landfill leachate. Conversion of dissolved ions in the solution to insoluble solid phase through chemical reactions. Different components have different optimum pH of precipitation
	Flotation	Floating matters including colloids, oil and grease are being removed from landfill leachate by using configuration such as dissolved air flotation units.

However, not all of these treatments are suitable to be used on landfill leachate due to its complex characteristics. Generally, some of the treatments are suitable on young landfill leachate, whereas some may be suitable on matured and stabilized landfill leachate. According to Bashir et al. (2011), young landfill leachate with high biodegradability is more suitable to be employed with biological treatment. Matured landfill leachate contains low biodegradability that makes it difficult to treat using biological treatment. Hence, physico-chemical is the most preferred technique to treat matured and stabilized landfill leachate. Therefore, prior to applying any of the treatments on the landfill leachate, it is vital to examine the characteristics of the landfill leachate. Table 2.5 shows the past research done on characteristics of APLS raw leachate.

Table 2.5: Previous study on characteristics of APLS leachate

Parameters	**Zakaria et al. (2015)**	**Rahim & Aziz (2016)**
COD (mg/L)	2180	3925
BOD_5 (mg/L)	227	131
NH_3-N	1897	1296
Colour	4650	13786
pH	7.93	7.99
Temperature (^{0}C)	30.2	31.34
Conductivity (µs/cm)	8208	-
Dissolved Oxygen (mg/L)	0.09	0.17
Total Dissolved Solid (mg/L)	4855	-

Based on the previous studies, it can be concluded that APLS is a matured and stabilized landfill leachate as the landfill leachate from APLS exhibits the characteristics of matured and stabilized landfill leachate when compared to Table 2.2. Consequently, the most suitable landfill leachate treatment that is suitable to be applied in APLS is physico-chemical treatment including coagulation – flocculation process.

2.5 Coagulation and Flocculation Process

Coagulation – flocculation process is one of the physico-chemical methods that has been widely used in pre-treatment of water and wastewater treatment due to its simple and low cost operation. Coagulation – flocculation process involves two distinct mechanisms, which are charge neutralization of negatively charged colloids by cationic hydrolysis products, and incorporation of impurities in an amorphous hydroxide precipitate through sweep flocculation (Ghafari et al., 2010). The coagulation – flocculation process is mainly used to agglomerate colloidal particles that are contributed through deterioration of water quality into bigger flocs, either as precipitates or suspended particles. The flocs can then be removed in subsequent processes (Davis & Cornwell, 2013).

Coagulation – flocculation process has been employed successfully in pre-treatment of landfill leachate (Ghafari et al., 2009). Besides, this treatment is also capable of removing pollutants such as COD, NH_3-N, colour, turbidity, and SS in the post-treatment of landfill leachate (Rusdizal et al., 2015). Several studies using coagulation – flocculation process on the removal of pollutants such as organic matters, ammoniacal nitrogen, and heavy metals had been reported. Table 2.6 shows some studies that have been done by using coagulation – flocculation process in landfill leachate treatment.

Table 2.6: Studies on coagulation - flocculation process in landfill leachate treatment using various coagulants

Coagulant	Research findings	Source
$FeCl_3$ and chitosan	$FeCl_3$ effectively remove 97.78% of turbidity and 95.54% of colour at optimum pH 6 and optimum dosage of 3.6 g/L whereas chitosan was found to be less effective to remove pollutants from landfill leachate	Ramli & Abdul Aziz, 2015
Alum and PAC	Best operating condition for alum and PAC were obtained. PAC was able to remove 99.18% turbidity, 97.26% colour, 56.76% COD and 99.22% TSS. Meanwhile for alum, it was found to remove 84.50% COD, 94.82% turbidity, 92.23% colour, and 95.92% TSS.	Ghafari et al., 2010
Iron chloride	Using iron chloride as coagulant to remove pollutants from Matang Landfill Site. It was reported that iron chloride able to remove 97% SS, 95% colour, and 66% COD effectively but less effective in removing NH_3-N	Zin et al., 2013
PAC and Longan seed	PAC was found to effectively remove 95% SS, 70% colour, and 94% COD at pH 6, but Longan seed was less effective where it removed 15.1% colour, 28.0% COD, and 29.5% SS	Rahim & Aziz, 2016

In general, coagulation – flocculation treatment involves two stages. In the first stage, coagulation process destabilizes colloidal particles charges by neutralizing charges among colloidal particles. It is followed by flocculation process where aggregation of colloidal particles takes place during coagulation process. Sometimes, a coagulant aid or also known as flocculant that has high molecules polymers is added to strengthen, bind, and add weight to the flocs and thus, increasing the flocs settling rate. After that, the bigger flocs formed can be removed by sedimentation, filtration, and flotation (Brostow et al., 2009)

2.5.1 Coagulation

Coagulation – flocculation is typically used prior to sedimentation and filtration in water and wastewater treatment. Coagulation is the first step in the process which destabilizes charges of particles and then forms bridges between the particles in order to form larger flocs. Coagulation involves the addition of coagulants with opposite charges of those colloidal particles in order to neutralize the charges on dispersed on-settleable solids (Abidin et al., 2014). In theory, most particles in water are negatively charged. Therefore, any cation (positive ion) can be employed as a coagulant.

Neutralization of colloidal particles' charges occurs very rapidly. High energy of rapid mixing can be an aid in neutralization process in promoting particle collision and hence, enhancing the aggregation of colloidal particles (Engelhardt, 2010). Therefore, it is vital to design rapid mixing system in coagulation process.

Even though the precise method of coagulation cannot be determined, scientists have come up with four theories on coagulation mechanisms that are thought to occur during the process. These include double layer compression (Van der Waals bonding factor), adsorption and charge neutralization, adsorption and interparticle bridging, and sweep flocculation (Abidin et al., 2014). The four mechanisms of coagulation are further explained in Table 2.7.

Table 2.7: Mechanisms in coagulation - flocculation process (Sahu & Chaudhari, 2013)

Mechanisms	Brief description
Double layer compression	Double layer compression consists inner and outer region, which are stern and diffuse layer respectively. In diffuse layer, it is required to maintain the electrical neutrality of colloidal suspension. When chemical agents with counter ions are added, the volume of diffuse layer will be reduced, and thus, resulting destabilization of colloidal dispersion by electrical double layer compression. During particle collision, Van der Waals attraction forces decreases while the aggregation among the colliding particles increases after compression of double layer.
Interparticle bridging	Coagulation process uses polymer with high molecular weight as coagulant to promote the interparticle bridging between the particles. Long-chained in polymer aids in attaching to particles and form bridges between particles and thus, causes particles to destabilize. In general, the polymer with higher molecular weight will result in a better and more effective flocculation. Moreover, these polymers have tendency to produce relatively large and packed, instantly settled, and dense flocs.
Charge neutralization	Typically, polluted water particles have negative charges. Addition of coagulants into the polluted water will be resulted in the formation of several hydrolysis species that have positively charged surface. Therefore, the negatively charges are neutralized with positively charged surface. The neutralization causes the reduction of electrical repulsion between particles and thus may cause aggregation to occur. However, overdosing may cause the particles to re-stabilize and the particles become positive charges and thus caused repulsion between the particles.
Sweep flocculation	Sweep coagulation occurs when a coagulant encapsulates suspended particulates in a soft colloidal floc. Unlike charge neutralization, overdosing of coagulant does not hinder the elimination of particles as the excessive coagulant will precipitate.

2.5.2 Flocculation

Coagulation process is incomplete without flocculation process. Flocculation process is defined as a slow mixing phase to encourage the collision of particles in the water in order to form larger mass of flocs (Engelhardt, 2014). The slow mixing may be carried out for more than 20 minutes and it is then followed by settlement process.

There are two types of flocculation which are microflocculation and macroflocculation. Microflocculation, also known as perikinetic flocculation occurs when the particles are aggregated due to the simple random motion of particles that is known as Brownian motion. As the name implies, the effect of microflocculation is only significant on submicron particles (Engelhardt, 2014).

On the other hand, macroflocculation which is also known as orthokinetic flocculation occurs when the aggregation of particles is caused by inducing velocity gradients and mixing in the solution that contains particles. For flocculation, these two processes are important to agglomerate the colloidal particles into flocs which later will be removed by particle-separation methods such as sedimentation and filtration (Metcalf & Eddy, Inc., 2014).

The efficiency of flocculation is affected by the aptitude of floc formation and the ability of flocs to settle. The flocs formed from the addition of coagulation may not be ideal for settling and filtration. Hence, coagulant aids or flocculants may be introduced into the process in order to enhance the formation and settleability of flocs. Similar to coagulant, polymers with high molecular weight are preferred to be used as flocculant.

2.5.3 Colloidal System

A colloidal system is generally defined as a mixture of one substance (dispersed phase) that is finely divided and distributed thoroughly into the second substance (dispersion medium). Colloids have extremely large surface area and the smallest size of dispersed phase in the range of 1 to 1000 nm. These features of colloidal particles have caused it to absorb numerous ions from the surrounding medium and give an electrostatic charge to the colloids. Subsequently, the obtained electrostatic forces prevent the colloids from bonding and aggregating with each other (Ghernaout & Ghernaout, 2012).

Based on the properties and interaction of colloids with water, colloids can be categorized into two groups, which are hydrophilic colloids and hydrophobic colloids. Hydrophilic colloids are water-loving colloids, whereby it has great affinity for water. These colloids are considered stable as there are strong attractive forces between dispersed phase and dispersion medium, of which in this case is water. For that reason, the possibility of particles aggregation is reduced and thus affecting the floc settling rates. In contrast, hydrophobia colloids are water-hating and have no affinity towards water. These colloids are less stable due to the weak attractive forces between the two said phases. Nevertheless, the hydrophobic colloids may settle by itself even if it takes as long as several years to do so.

Colloidal stability in water is mostly affected by the electrical repulsion of the colloids. All the colloids in water theoretically have negative charges, and this has caused the repulsion between the colloids in the water whenever collision occurs (Yong & Aziz, 2016). For that reason, the aggregation of colloids into bigger flocs will not happen. Therefore, it is necessary to neutralize the negative charged colloids by adding electrolyte. As the concentration of electrolyte increases, the repulsive electrostatic interaction will be reduced and thus, indirectly reducing the energy barrier and facilitating effective particle collision by destabilizing the colloidal system.

DVLO theory is adapted in order to investigate the dependency of various parameters for colloidal stability (Duan & Gregory, 2003; Rahim & Aziz, 2016). DVLO theory states that the net energy of particles interaction is caused by repulsion from double electrical layers, and the attraction is a result from Van der Waals force (Kulshreshtha et al., 2010). This interaction can be pictured as two blocks which are connected by a spring as shown in Figure 2.1. The Van der Waals attraction is shown when the spring is stretched at long distance and a net force is applied to pull the block together. Meanwhile, the electrostatic repulsion is visualized when the spring is compressed at close approach, producing a net repulsive force pushing the blocks apart. However, forces will be in equilibrium when the blocks are placed at some intermediate distance (Lee, 2017).

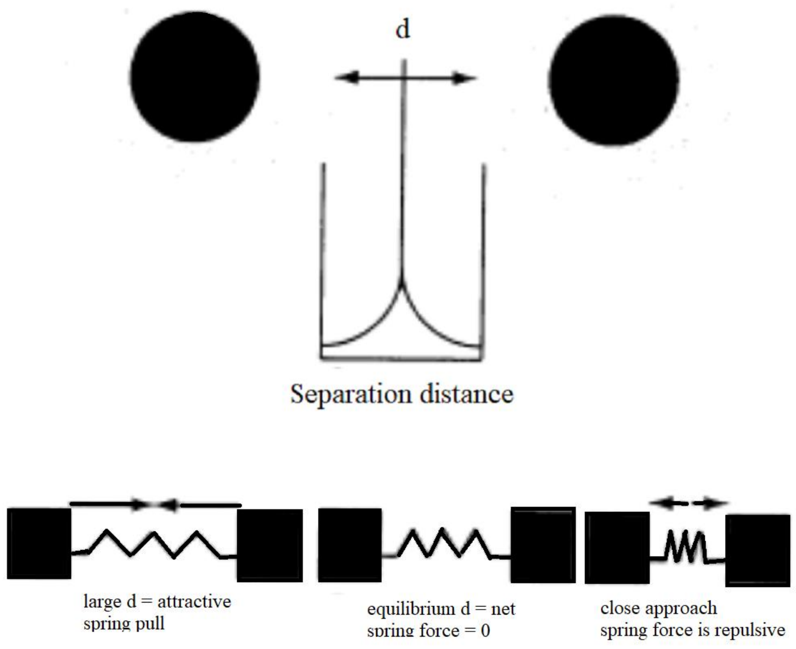

Figure 2.1: Interaction between two particles using DVLO theory

2.5.6 Factors Affecting Coagulation and Flocculation

Coagulation – flocculation process is effective in removing pollutants during pre-treatment and post treatment of polluted water. However, there are several factors affecting the effectiveness of coagulation – flocculation process. The factors are type and dosage of coagulant and coagulant aids, pH, contact time of coagulant with pollutants, rate of speeding mixture, alkalinity, and temperature (Jamil, 2005). Among all the factors, pH and coagulant dose are two factors that significantly affect the effectiveness of coagulation – flocculation process.

Value of pH, which indicates the acidity and alkalinity of fluid, is the main factor that affects the efficiency of coagulation – flocculation process. During the treatment, pH of water influences the chemical properties of coagulant, namely charge on particles, natural organic matter, solubility and speciation. This has made the flocculation process more challenging (Kemunto, 2016).

Besides that, the coagulant dosage is important in determining the effectiveness of coagulation – flocculation process. The dosage of coagulant added should correspond to the concentration of pollutants in the solution. This is to ensure that the process of destabilization is effective and thus, making the coagulation – flocculation process more efficient. Low dosage will cause incomplete destabilization of particles, whereas overdosing will cause re-stabilization. Therefore, optimum dosage, which refers to the coagulant dosage that has the highest removal efficiency of pollutants, needs to be determined under certain conditions of experiment (Sahu & Chaudari, 2013).