DEVELOPMENT OF ENVIRONMENTAL FRIENDLY COAGULANT FOR THE INNOVATIVE PURIFICATION OF URBAN AND AGRICULTURAL RUNOFF

by

NOR SYUHADA BINTI TAJUDIN

A dissertation submitted in partial fulfillment of the requirement for the Degree of Bachelor Health Sciences (Honours) (Environmental and Occupational Health)

JUNE 2015

ACKNOWLEDGEMENT

Alhamdulillah, I am grateful to the Allah for the strength, good health and wellbeing that were necessary to complete this thesis. I wish to express my sincere thanks to my supervisor, Dr Foo Keng Yuen for his valuable guidance, expertise, patience and most importantly continuous motivation along my degree studies. His challenges brought this work towards a completion. I am also deeply thankful to all members in River Engineering and Urban Research Centre (REDAC) USM Engineering Campus, especially to Tina, Yuh Nien and Siti Fairos for their supported and helped in laboratory works. I pray for them to succeed and achieve their dreams.

My special and heartily thanks to my group members, Nazwatul Iman, Rosnizan Roslan and Aniff Asri for their help and support. Completing this work would have been all the more difficult were it not for the support and friendship given by them and our memories will not be forgotten. Not forgotten to my roommate, Fisya Hamzah and my entire course mate.

Most importantly, I also thank to my family who encouraged me and prayed for me throughout the time of my degree study. This thesis is heartily dedicated to my parents, Tajudin and Che maznah for their love, full support, and continuous pray. Not forgotten for my sisters and brothers, thanks. May the Allah, Almighty God richly bless all of you.

Nor Syuhada Tajudin

1 Ramadan 1436H

TABLE OF CONTENT

LIST OF TABLES

LIST OF FIGURES

LIST OF PLATES

LIST OF ABBREVIATION

AC	Activated carbon
ATR	Attenuated Total Reflection
BC	Before century
BOD	Biological Oxygen Demand
COD	Chemical Oxygen Demand
DID	Department of Irrigation and Drainage
DO	Dissolved oxygen
DOE	Department of Environment
FTIR	Fourier Transform Infrared Spectrometer
MSMA	Manual Saliran Mesra Alam
MOH	Ministry of Health
NOM	Natural organic matter
NPS	Nonpoint source
NTU	Nephelometric Turbidity Unit
PAC	Polyaluminium chloride
POME	Palm oil mill effluent
POU	Point of use
PPME	Paper and pulp mill effluent
RPM	Rotation per minute
SEM-EDX	Scanning Electron Microscope-Energy Dispersive X-ray
TDS	Total dissolve solids
TOC	Total organic carbon
TN	Total nitrogen
TSS	Total suspended solid
US EPA	United State Environmental Protection Agency
WHO	World Health Organization
WQI	Water Quality Index

LIST OF SYMBOLS

cm^3	Centimetre cubic
mg/L	Milligram per litter
μm	Micrometre
mm	Millimetre
m^3	Meter cubic
ml	Millilitre
meq/gm	Milliequivalent per milligram
mg	Milligram
g	Gram
%	Percent
km^2	Kilometre square
km^3	Kilometre cubic
^oC	Degree Celsius
rpm	Rotation per minute
w/v	Weight per volume
Wt%	Weight percentage
>	More than
<	Less than

PENYEDIAAN KOAGULAN MESRA ALAM UNTUK PENULENAN INOVATIF AIR LARIAN BANDAR DAN PERTANIAN

ABSTRAK

Pada masa ini, koagulan semula jadi merupakan ciptaan organik yang berasas daripada polisakarida atau polimer semula jadi menjadi perhatian global kerana keringkasan pengoperasian, mudah didapati, mesra alam, dan kos rendah. Kajian ini bertujuan untuk menyediakan koagulan mesra alam daripada sekam padi untuk penulenan air larian bandar dan pertanian dengan mengkaji keberkesanan untuk kekeruhan, jumlah pepejal terampai (TSS) dan permintaan oksigen kimia (COD). Parameter operasi termasuk dos koagulan, pH, suhu, keamatan pencampuran perlahan dan masa sentuhan telah dinilai. Hasil kajian menunjukkan bahawa dos optimum untuk air larian bandar dan pertanian masing-masing adalah 75 mg/L dan 175 mg/L. Air larian bandar mencatatkan 90% pengurangan kekeruhan, 73% daripada COD dan TSS penyingkiran sehingga 89% dan pengurangan tertinggi kekeruhan, COD dan TSS untuk air larian pertanian masing-masing sebanyak 95%, 74% dan 88%. Koagulan yang baru dibangunkan ini adalah pH bebas dan dapat mengekalkan prestasi tinggi pada julat pH yang luas dengan keperluan kurang daripada 50 minit untuk penyelesaian mendapan. Kajian morfologi menunjukkan koagulan berasaskan sekam padi mengandungi tinggi unsur silikon. Penemuan ini mengambarkan koagulan berasaskan sekam padi berpotensi untuk penulenan air sisa tercemar.

DEVELOPMENT OF ENVIRONMENTAL FRIENDLY COAGULANT FOR THE INNOVATIVE PURIFICATION OF URBAN AND AGRICULTURAL RUNOFF

ABSTRACT

Presently, natural coagulant is an organic creation founded from polysaccharides or natural polymers has emerged to be a global interest due to the simplicity of operation, wide availability, biodegradable, and relatively low cost. This study aims to develop an environmental friendly coagulant from rice husk for the purification of urban and agricultural runoff by examined the effectiveness for turbidity, Total Suspended Solid (TSS) and Chemical Oxygen Demand (COD). The operating parameters including the coagulant dosage, pH, temperature, slow mixing intensity and contact time were evaluated. Result showed that the optimum dosage for urban and agricultural runoff was 75 mg/L and 175 mg/L, respectively. Urban runoff recorded 90% turbidity reduction, 73% of COD and TSS removal of up to 89% and the highest of turbidity, COD and TSS reduction for agricultural runoff of 95%, 74% and 88%, respectively. The newly developed coagulant was pH independent to maintain at relatively high performance at a wide pH range with the requirement of less than 50 minutes for the settlement of flocs. Morphological study showed that the derived rice husk coagulant contains high silicon element. The findings illustrated the potential of rice husk derived coagulant for the purification of contaminated wastewater.

CHAPTER ONE

INTRODUCTION

1.1 Urban and agricultural runoff breakdown in Malaysia

Water covered 70% of the earth surface, and hence, it becomes one of the most precious natural resource of the world. Growing numbers of population indirectly increased economic activities and industrialization has not only created an increased demand for fresh water but then also give rise in severe misuse of it (Jodi *et al.*, 2012). Malaysia is a tropical country that located near to equator with climate hot and humid all along the year. According to the Malaysia Metrological Department, the characteristic features of the climate of Malaysia are uniform temperature, high humidity and copious rainfall (Alam *et al.* 2010; Mohd *et al.* 2010) (Table 1.1). Malaysia receives an annual average rainfall approximately more than 2500 mm or 990 billion m^3 (River Basin Initiative, 2015).

High intensity and long duration of rainfall common are common events in the humid tropic region in Malaysian urban cities (Abustan *et al.*, 2008). Therefore, the country is rich in water resources as compared to the other regions in the world (Toriman and Mokhtar, 2012). As much as 97 % of water supply in the country is from surface water while the remaining is derived from groundwater. Groundwater is used as an alternative source of water, especially in areas where surface water supply is inadequate or unavailable (Compendium of Environment Statistics Malaysia, 2013). Even though the country is rich in water resource, the management of water resources should be addressed to meet with the increasing demand.

Table 1.1: Total number of rainfall at selected meteorological stations, Peninsular Malaysia from January to June 2013 (Compendium of Environmental Statistics Malaysia, 2013)

City	Total rainy days	Amount of rainfall (mm)
Cameron Highland	114	1,204.6
Petaling Jaya	105	1,820.8
Kuantan	82	1,135.0
Kuala Terengganu	71	949.4
Ipoh	91	1,604.8
Mersing	73	1,629.4
Batu Embun, Jerantut	84	907.8
Muadzam Shah	77	1,183.3
Subang	97	1,351.2
Bayan Lepas	79	757.6
Batu Pahat	89	692.3
Alor setar	63	737.6
KLIA Sepang	62	623.1
Kluang	76	1,033.2
Sitiawan	71	815.8
Butterworth	72	1,030.8
Chuping	65	806.0
Melaka	67	473.4
Kota Bharu	64	900.8

From 990 billion m^3 of rainfall receive annually, approximately 360 billion m^3 (36%) of rainfall water would return to the atmosphere as evapotranspiration, 566 billion m^3 (57%) appears as surface runoff and the remaining 64 billion m^3 (7%) of it will recharge as groundwater (Figure 1.1). Table 1.2 shows the total 566 billion m^3 of surface runoff, 147 billion m^3 of it is found in Peninsular Malaysia, 113 billion m^3 in Sabah and 306 billion m^3 in Sarawak (Alam *et al.* 2010; Mohd *et al.* 2010). The total internal water resource of Malaysia was estimated at 580 km^3 per year. The World Resources Institute had estimated that in 2007, the annual renewable water supply of Malaysia to be 22,100 m^3 per person per year, a fall of about 2% from 22,484 m^3 per person per year in 2006. But by 2025 it is projected that this would fall to approximately 10,000 m^3 per person per year. Malaysia is still very much "rich" in water resources but all these could come to naught if this precious resource is not conserved and preserved (Randall, 2008).

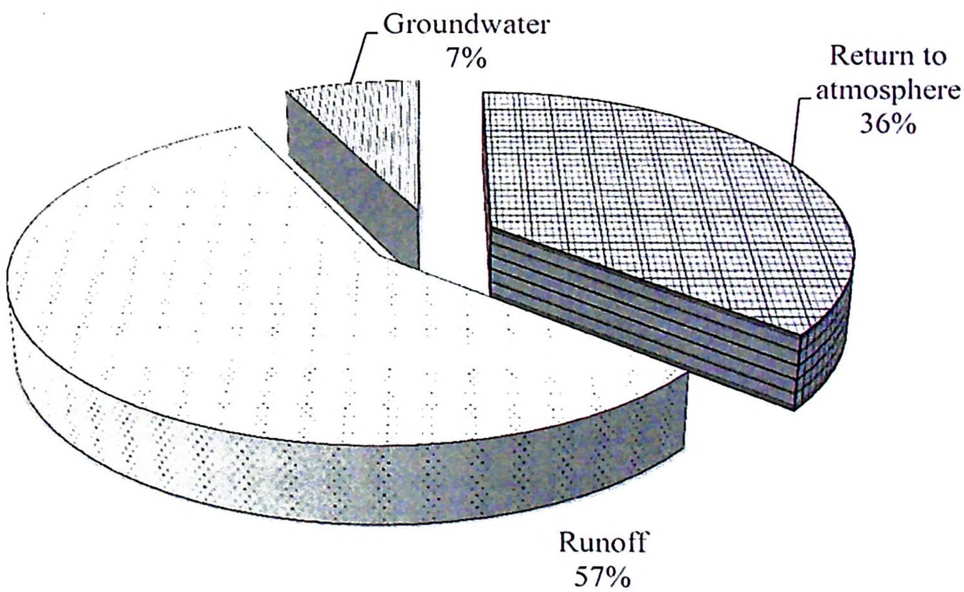

Figure 1.1: Breakdown of rainfall received annually in Malaysia (Alam *et. al.*, 2010)

Table 1.2: Breakdown of surface runoff in Malaysia (Alam, *et. al.*, 2010)

	Peninsular	Sabah	Sarawak	Total
Surface runoff	147 billion m^3	113 billion m^3	306 billion m^3	**566 billion m^3**

Continuous growth of population, increasing standard of living, climate change, industrialization include agriculture as well as urbanization are initiating the decrease in water resource worldwide (Wu *et al.*, 2013). In the year 2000 to 2030, urban populations are expected to be double in Asia region and water scarcity is now a global pandemonium not limited to arid zones (Wu *et al.*, 2013). Urban runoff is responsible for water quality deterioration of receiving water downstream of urban areas (Ismail, 2008). Urban storm water runoff are known as a major source of pollutants to receiving waters and a number of recent investigations have evaluated storm water runoff quality characteristics and best management technologies to minimize pollutant input to receiving waters (Davis *et al.*, 2001). Poor management of urban storm water indirectly one of the causes of flash flood in urban area. In Malaysia, monsoon and flash flood have been dominating the entire urban areas in Peninsular Malaysia, for example in Penang, upper Kinta Valley, Malacca Basin, East Coast of Kelantan, Terengganu, Pahang and Johor (Izham *et al.*, 2008). The frequent occurrence of flash flood in the last several years especially in urban cities and town areas (DID, 2000) has initiated awareness to better understanding within urban storm water management (Abustan *et al.*, 2008).

The management of urban storm water in Malaysia is managed by Department of Irrigation and Drainage (DID). DID also issues a guideline for the management of urban storm water; Urban Storm Water Management Manual for Malaysia or in Bahasa, Manual Saliran Mesra Alam (MSMA). The manual contains the latest standard and practice for storm water management (SWM), technologies used, best engineering practice that were generally based from foreign countries. The two main issues concerning storm water is concern for two main issues, one; related to the volume and timing of runoff water (flood control and water supplies) and second related to the potential contaminants that the water is carrying such as water pollution (Abustan *et al.*, 2008). According to Erickson (2013), the impact of increasing volume of urban storm water runoff and pollutant loads is substantial. Urban runoff is a global issue especially in developing countries.

Malaysia has more than 7 million hectares agricultural land that produces an abundant supply of food and other products. Agriculture is one of the major economic sectors in Malaysia. In the Tenth Malaysia Plan (10^{th} MP), agriculture sector is acknowledged to be one of 12 National Key Economic Area (NKEA) that would generate more income to the country where oil palm becomes the major production of crops followed by rubber and paddy (Compendium of Environment Statistics Malaysia, 2013). The land use for agriculture sector is also increased from year 2008 to 2012 (Table 1.3). The rise of agriculture sector not only gives benefits to economy, but it will turn bad if the practice is not managed well. The increase of land use for agriculture sector indirectly will increase the volume of agriculture runoff. According to Dayton Daily News, global warming aside, agricultural runoff becomes one of today's biggest environmental threats. A 2000 study by the U.S.

EPA found that agriculture was responsible for nearly half of the impairment to river far more than other sources, including urban runoff and storm sewers (Tom, 2015).

Table 1.3: Planted area of selected crops for period 2008 to 2012, Malaysia (Selected Agricultural Indicator, 2013)

	Area ('000 hectares)				
Crops / Year	**2008**	**2009**	**2010**	**2011**	**2012**
Oil palm	4,487.9	4,691.1	4,853.8	5,000.1	5,076.9
Rubber	986.2	1,015.1	1,015.2	1,012.8	1,059.7
Paddy	656.6	674.9	677.9	687.5	692.3
Cocoa	20.9	19.3	20.8	21.2	21.7
Tobacco	6.7	7.6	3.7	4.2	2.5
Vegetables	37.7	41.1	52.8	52.6	53.3
Fruits	264.8	250.0	239.4	226.8	241.8
Others	147.8	137.5	143.1	144.6	150.7
Total	**6608.6**	**6836.6**	**7006.7**	**7149.0**	**7298.9**

1.2 The application, advantages and disadvantages of coagulants

There are many technologies include traditional and modern have been utilised aims to eliminate the colloidal particles from wastewater such as technique of ion exchange, membrane filtration, precipitation, floatation, solvent extraction, adsorption, coagulations, flocculation, biological and electrolytic techniques (Radoiu *et al.*, 2004). According to Renault *et al.* (2009a), coagulation-flocculation is one of popular and widely used methods works by solid-liquid separation process for removal of suspended and dissolve solids, colloids as well as organic matter. Tzoupanos and Zouboulis (2008) also agreed that a very important step in water and wastewater treatment is the coagulation-flocculation process, which is extensively used, due to its simplicity and cost-effectiveness.

Over the years, substantial research has been carried out to develop the performance of coagulation or flocculation in different types of water and wastewater. Pre-treatment of young leachate or the post treatment of partially stabilized leachate is one of the studies by coagulation process (Yilmaz *et al.*, 2010). According to Vijayaraghavan *et al.* (2011), coagulation is an essential process in the treatment of surface water and industrial wastewater. Currently, there are numerous studies have been conduct to recognize the applications of coagulations. Different types of wastewater such as palm oil mill effluent (POME), textile wastewater, pulp mill wastewater, oily wastewater, sanitary landfill leachate and other are study (Ahmad *et al.*, 2005; Tatsi *et al.*, 2003; Wong *et al.*, 2006; Yue *et al.*, 2008; Zhong *et al.*, 2003). Further, coagulation also serves as a primary treatment to remove toxicity, colour, Chemical Oxygen Demand (COD) and Biological Oxygen Demand (BOD_5) (Pradeep *et al.*, 2011).

The advantages of coagulation process make it a good choice for water treatment. Coagulation process are strongly affects the overall treatment performance; hence, it is effectively the increase the process stage (Tzoupanos and Zouboulis, 2008). Polymeric flocculants, synthetic as well as natural coagulants have become very popular in industrial effluent treatment as it is natural inertness to pH changes, high efficiency with low dosage, and easy for handling (Singh *et al.*, 2000). Further, coagulation process is also effective in removing high concentration of organic pollutants (Wang *et al.*, 2002), heavy metals and some anions (Kreith, 1994). Aluminum and iron coagulants are inorganic coagulants that are widely used for the removal of humic substances from water (Lu *et al.*, 1999; Wang *et al.*, 2002; Amokrane *et al.*, 1997; Ching *et al.*, 1994). Coagulation-flocculation are more effective in removing higher molecular weight hydrophobic natural organic compounds compare with hydrophilic low molecular weight organic matter (Matilainen *et al.*, 2010).

Besides water and wastewater treatment, coagulation is also suitable to use in treating leachate water. Chemical-based coagulants have been adapted in leachate treatment even though they are frequently used in water and wastewater treatment (Zin *et al.*, 2013). Previous studies by using different chemical coagulant on landfill leachate have been conducted (Tatsi *et al.*, 2003; Sletten *et al.*, 1995; Amokrene *et al.*, 1997). A study on new landfill leachate performed by Yilmaz *et al.*, (2010), demonstrated that alum are able to removed 44% of chemical oxygen demand (COD) and 99% of colour, while ferric could chloride removed 45% of COD and 98% of colour.

Aluminium salts are able to decrease organic matter, colour, turbidity, and microorganism levels in water (WHO, 1998). The usage of aluminium as coagulant gives greatest result in treating water and become choices widely. According to Flaten (2001), there are two major sources of aluminium in drinking water, either it occur by naturally as a result of leaching from mineral in soil and bedrock or aluminium in water treatment as coagulants. The large particles has difficulties to filtered from the water and most of added aluminium is eliminated by filtration and sedimentation together with particles and humic compounds cause increasing of aluminium concentration in water.

Many studies have emphasized to the consequence of uses of such salts especially to health effect. Chemicals coagulants include polyaluminium chloride and alum adds impurities such as epichlodine that are carcinogenic (Gherbremicheal *et al.*, 2005; Muyibi and Alfugara, 2003). According to Pise and Halkude (2012), poisoning factor that cause dialysis encephalopathy is said to be aluminium in water. According to Flaten (2001), the first reporting the effect of aluminium to the brain of patient with Alzheimer disease was published in the year 1973 by Crapper *et al.* (1973). In 2001, 9 out of 13 published epidemiology studies of aluminium in drinking water and Alzheimer disease have shown statistically significant positive relations. The study was conducted at a few countries including Switzerland, United Kingdom, Norway, France and Canada. Miller *et al.* (1984) pointed out the effect of aluminium salts may cause Alzheimer's disease and other similar health related problems.

The usage of inorganic coagulants has been reduced currently, as it is inefficiency in wastewater treatment due to the narrow application (Lee *et al.*, 2014). Chemical coagulant has constrains of pH and alkalinity of wastewater (Pise and Halkude, 2012). Besides, as reported in many studies, its usage would cause two important environmental consequences, the production of large volumes of metalhydroxide (toxic) sludge which will create disposal problem, and an increase in metal (example: aluminium) concentration in the treated water which may have human health implications (Flaten, 2001; Ward *et al.*, 2006).

Up to now, a wide range of flocculants (also known as coagulant aids) have been developed to help improving the treatment process by introducing synthetic or natural organic flocculants and grafted flocculants (Lee *et al.*, 2014). Inorganic coagulant is highly sensitive to pH of water and it has likelihood to produce secondary contaminant of drinking water with traces synthetic polymeric flocculants or residual iron and aluminium (Bratskaya *et al.*, 2004). The reactions of aluminium with water caused the pH of water decrease and make the process less ineffective in cold water. According to Vijayaraghavan *et al.* (2011), these coagulants are ineffective in low temperature water, relatively consume high costs, produced side effects on human health, and create large number of sludge volumes as well as the less significant in pH of treated water. The costs of these chemicals have also been increasing rate thus causing local manufacturing companies cannot cope with the demand for these chemicals in other industrial applications. Therefore, the shortfall has to be imported with scarce convertible foreign currency (Muyibi *et al.*, 2004).

Many studies showed that sludge that formed during coagulation with synthetic polymer is non-biodegradability, thus, it has limited potential to be recycled (Bratskaya *et al.*; 2004, Pan *et al.*; 1999, Divakaran and Pillai; 2001). Coagulation is usually combined with other treatment techniques to ensure the effective removal of pollutants (Zin *et al.*, 2013). According to Anastasakis *et al.* (2009) aluminium salts are regularly expensive on the market and in some of countries, they have to import from outside. Since the usages of chemicals are associated with human health and environmental problems (Kaggwa *et al.*, 2001), consequently, natural coagulants become the demand. The limitation of availability and relative expense using of alum and ferric for water purification (WHO, 2008) caused expansion of the research to find natural plant basis coagulants.

Natural coagulant is organic creation which can be founded by polysaccharides or natural polymers become the interest since they are produce by natural and environmentally friendly behaviour (Lee *et al.*, 2014). The practice of natural-plant based coagulant in treating water give various advantages compared to chemicals as shown in Table 1.5. Plant based natural coagulant can originated from seeds, leaves, fruits, roots and barks (Zin *et al.*, 2013). Natural coagulants have the significant benefits over the commercial ones as they are available in abundance, cheaper, environmental friendly, multifunction, and biodegrades naturally in water purification (Sciban *et al*, 2005).

Polysaccharides shows shear stability and exhibit capability in coagulation process (Browstow *et al.*, 2013). In addition, these organic polymers are interesting because, comparative to the use of synthetic organic polymers containing acrylamide monomers, there is no human health danger and the cost of these natural coagulant would be less expensive than the conventional chemicals alike since it is locally available in most rural communities (Asfaruzzaman *et al.*, 2011). Yin (2010) considered coagulants that occur naturally are safe for human health. According to Narasiah *et al.* (2002), natural coagulants are biodegradable as well as produce less voluminous sludge that amounts only 20 to 30% that alum treated counterpart. Natural macromolecular coagulants are promising and have attracted the attention of many researchers because of their abundant source, low price, multi-purposeness and biodegradable product (Katayon *et al.*, 2006; Muyibi *et al.*, 2002a; Zhang *et al.*, 2006). However, the development of most natural coagulants application has so far been confined to laboratories.

Table 1.4: Application of coagulation-flocculation process with chemical coagulants and flocculants (Lee *et al.*, 2014)

Coagulants	**Flocculants**	**Types of wastewater**	**Reference**
Ferric chloride, aluminium sulphate and lime	Neutral (N200), two cationic (K1370 and K506) and an anionic (A321) polyelectrolytes	Sanitary landfill leachate	Tatsi *et al.* (2003)
Alum and polyaluminium chloride (PAC)	Cationic polymer (KP 1200B) and anionic polymer (AP 120C)	Ceramic industry wastewater	Chong *et al.* (2009)
Ferric chloride	Non-ionic polyacrylamide	Beverage industrial wastewater	Amuda and Amoo (2007)
Alum/ferric salt	Synthetic cyanoguanidine-formaldehyde based polymer	Synthetic reactive dyes wastewater	Joo *et al.* (2007)
Modified alum (Envifloc-40L)	Industrial grade flocculant (Profloc 4190)	Palm oil mill effluent	Ahmad *et al.* (2005)
Alum, ferric chloride and ferric sulphate	Anionic polyacrylamide	Abattoir wastewater	Amuda and Alade (2006)

Table 1.5: The application of coagulation process from natural coagulants

Natural coagulants	Types of water treatment	Reference
Moringa Oleifera	Palm Oil Mill Effluent (POME)	Bhatia *et al.* (2006)
	Heavy metal water	Vikashni *et al.* (2012)
Rice husk	Fluoride water	Vardhan and Karthikeyen (2011)
Okra gum	Clay water	Renuka and Jadhav (2013)
	Biologically treated effluent	Anastasakis *et al.* (2009)
Grape seed	Cationic dyes	Jong *et al.* (2009)
Common bean	Synthetic turbid water	Antov *et al.* (2009)
Mallow mucilage	Biologically treated effluent	Anastasakis *et al.* (2009)
Cicer arietinum	Synthetic turbid water	Asrafuzzaman *et al.* (2011)
Nirmali seed	Synthetic turbid water	Rani and Talikoti (2013)
Cactus *opuntia ficus indica*	Paint effluent	Vishali and Karthikeyen (2014)
Cassia Obtusifolia	Agro industrial wastewater	Shak and Wu (2014)
Tapioca starch	Semiconductor wastewater	Fatehah *et al.* (2013)
Peanut	Synthetic turbid water	Birima *et al.* (2013)
Rambutan seed	Synthetic turbid water	Zurina *et al.* (2014)
Chitosan	Surface water	Rizzo *et al.* (2008)
Bentonitic clay	Oily and mining wastewater	Abdelaal (2004)

1.3 Problem Statement

Pure water is a vital to the human life, drinking, cooking, and washing as well as for the socio economic activities, industries, agriculture, aquaculture, and transportation. Today, the preservation of good water quality has become the challenge as from activities of irresponsible human that the discharging contaminants into the water resource. According to Toshio *et al.* (2013), 32% of wastewater that are generated in the Asian countries is being treated, and (Randall, 2008) about six million tons of sewage every year is generated in Malaysia, most of which is treated and released into the rivers.

Malaysia is a developing country that is characterized by rapid urbanization, which was indirectly, alters natural vegetation, and the physical characteristics of the landscape. Hence, it is resulting lower infiltration capacity of the soil, and an increase in the runoff volume (Elliot, Trowsdale, 2007; Gupta, Saul, 1996). Water qualities, aquatic habitats, riparian vegetation and social problems including aesthetics, recreation and economics are among the environmental issues raised from poor management of storm water (MSMA, 2012). These runoffs would move and enter into water bodies: river, wetland, lakes, ponds, reservoirs, and estuary and coastal and finally causing water pollution. Today, many river basins in the country are slowly experiencing serious environmental problems. This is due to the humid tropical condition in Malaysia, rate of rainfall that is high, as well as the volumes of surface runoff and sedimentation of receiving water bodies have been greatly increased (Abas and Hashim, 2014).

With high the sediment levels, it will cause garbage float and rot in the river, and hence, leading to the pollution of heavy metals, toxic chemicals as well as domestic and farm waste (Lee, 2000). Runoff not only brings threat to economic, but more worst to environmental one (National Geographic Education, 2015). According to Izham *et al.* (2008), many river basins in Malaysia are now under intense pressure from urban, industrial, and infrastructural development where downstream receiving water bodies such as rivers, lakes, ponds, reservoirs, and estuary and coastal waters have become sensitive to increased rates and volumes of runoff and pollutant discharge.

Furthermore, the costing of chemical coagulants are also high. According to Renuka and Jadhav (2013), the cost of water treatment is increasing and the quality of the river is not stable due to the suspended and colloidal particles load source by land development as well as high storm runoff during the rainy season, as experienced by Malaysia and other countries that receive copious rainfall. According to Azmi Mat Nor, Chief Executive Officer of the Company Manufacturers Air Sungai Selangor Sdn. Ltd. (Splash), the cost of water production is higher compared with the charge that being paid by the consumers (Utusan Online, 2015).

The main issues that are correlated to the process of water treatment by chemicals coagulant are the side effect to human health. The most commonly used chemicals in coagulation process are inorganic metal salts such as aluminium sulphate, $Al_2(SO_4)^3$ or also known as alum and Ferum sulphate, $Fe_2(SO_4)^3$ or ferric. Aluminium is classified as the third most abundant metallic element, with about 8%

of the earth's crust. It present in all food stuffs includes drinking water and other beverages as well as a dust in the air (Ganrot; 1986 and WHO; 1998).

The Environmental Health Criteria document for aluminium by WHO concluded that there were positive linking of aluminium in drinking water to cause Alzheimer disease (WHO, 1998). Surprisingly, the consumption of alum is very high in water the treatment in Malaysia (Ali *et al.*, 2009). The Ministry of Health (MOH) had fixed the parameter for aluminium in drinking water to not exceed 0.2 mg/L (Table 1.6). Purification of water by using aluminium salts might lead to the increase of concentrations of aluminium and deposit in the human distribution system (WHO, 1998).

Water treatment is an important process to generate safe and clean drinking water, and to avoid the transmission of waterborne disease such as cholera, and typhoid. According to the WHO, it has been documented that 3.4 million lives were sacrifice from the waterborne disease every year, and it is the leading cause of disease and death among children with 5 years old or below (Jessica, 2015). According to the Compendium of Environment Statistics Malaysia (2013), the number of cases of waterborne disease in Malaysia in 2012 is 14,232 cases. These cases are including cholera, typhoid, dysentry, viral hepatitis A and food poisoning. Globally, 2 million of tons of sewage, industrial and agriculture waste are discharged every day (UN WWAP, 2003) and in Malaysia, the estimated volume of wastewater generated by the municipal and industrial sectors is 2.97 billion cubic meters per year (DOE, 2011). As a result, a cost effective purification approach is required to reach the decontamination objectives required by law (Renault *et al.*, 2009a).

Table 1.6: National Drinking Water Quality Standard (Ministry of Health, 2010)

Parameters	**Maximum Acceptable value (treated water)**
Turbidity	< 5 NTU
pH	6.5 – 9.0
Colour	< 15 TCU
Free residual Chlorine	0.2 – 5.0 mg/L
Aluminium	< 0.2 mg/L
Ferum	< 0.3 mg/L
Biological Oxygen Demand (BOD)	6 mg/L (raw water)
Chemical Oxygen Demand (COD)	10 mg/L (raw water)

In Malaysia, 98% to the total national water use is harvested from the 150 river basins systems around the country (Randall, 2008). Rivers plays important role as they contribute more than 90% of the raw water supply, and major river management issues are all linked to the water quality. Since 97% of Malaysia's water supply comes from rivers, poor river water quality will severely affect water supply (Chan, 2012). As the country moving towards realizing the Vision 2020 through the implementation of its policy agenda for heavy industrialization, infrastructures, and urban-expansions, water demand has increased steeply, and greater pressure is on preserving the current water resources as well as alternative courses of action for water quality improvement (Muyibi *et al.*, 2008).

The quality of surface water and groundwater is identified in terms of its physical, chemical and biological parameters (Othman *et al.*, 2012). The water quality is monitored by the Department of Environment (DOE) based on Water Quality Index (WQI) (Table 1.8). WQI is a mathematical instrument used to transform large quantities of water quality data into single number (Nives, 1999). The parameters comprised of Biochemical Oxygen Demand (BOD_5), Chemical Oxygen Demand (COD), Ammonical Nitrogen (NH_3N), Suspended Solids (SS), pH and Dissolved Oxygen (DO) (Compendium of Environment Statistics Malaysia, 2013).

Table 1.7: National Water Quality Standard for Malaysia (DOE, 2000)

Parameters	Unit	Class				
		I	II	III	IV	V
Chemical Oxygen Demand (COD)	mg/L	<10	10-25	25-50	50-100	›100
pH		>7	6-9	5-6	<5	>5
Total Dissolved Solid (TDS)	mg/L	500	1000	-	4000	-
Total Suspended Solid (TSS)	mg/L	<25	25-50	50-150	150-300	>300
Turbidity	NTU	5	50	-	-	-

Table 1.8: The Interim National River Water Quality Standards for river (DOE, 2000)

Class	**Uses**
Class I	Conservation of natural environment. Water Supply I - Practically no treatment necessary. Fishery I - Very sensitive aquatic species.
Class IIA	Water Supply II - Conventional treatment. Fishery II - Sensitive aquatic species
Class III	Water Supply III - Extensive treatment required. Fishery III - Common, of economic value and tolerant species; livestock drinking.
Class IV	Irrigation
Class V	None of the above.

1.4 Research Objectives

The objectives of this study are listed as below:

1.4.1 General objective

To develop an environmental friendly coagulant for the innovative purification of urban and agriculture runoffs.

1.4.2 Specific objectives

1. To identify a potential renewable resource for preparation of natural coagulant.
2. To examine the coagulating activity for the purification of urban and agricultural runoffs.
3. To optimize the best operating conditions for the treatment process.
4. To determine the physical and chemical behavior of the natural coagulant.

1.5 Organization of Thesis

This thesis is structured into five chapters, including an introduction (Chapter 1), literature review (Chapter 2), methods and materials (Chapter 3); results and discussions (Chapter 4); and conclusions and recommendations (Chapter 5). Chapter one contains an overview of urban and agricultural breakdown in Malaysia. This chapter also presented about the application, problem statement and the objectives of the study. Literature review contains the detail about coagulation, the classification, physicochemical properties and the preparation steps of natural coagulant. Also, chapter two are about urban and agricultural runoff, hazard and its environmental impact as well as the treatment technologies. Methodology of the study is presents in chapter three. This are include the preparation of coagulant, performance analysis and parameter used. The result and discussion from the study are elaborated in chapter 4. The result are discussing about the objectives of the study including all the parameter, morphological studies. Chapter five are the conclusion and recommendation for the improvement respectively.

CHAPTER TWO

LITERATURE REVIEW

2.1 Coagulation

2.1.1 Definition and historical studies

Coagulation is a term that are used to describes the effect produced when certain chemicals are added to raw water containing slowly settling or non settleable particles. According to US EPA (2002), the chemicals hydrolyse and neutralise the electrical charges on the colloidal particles, which begin to form agglomerations termed floc which will be removed by clarification and filtration. Coagulation is known as a simple pollutant removal process that are involving the addition of coagulant either chemical or organic to untreated water to destabilize suspended matter, colloids and other substance by causing them to be transformed into a larger size (Zin *et al.*, 2013).

Meanwhile, coagulant is defined by US EPA (2002) as the chemicals that have been used to destabilize colloids and produce the fine colloidal particles to form a larger particle that can be separated from the water by settlement or flotation. Often the terms of coagulation and flocculation are used synonymously even though it is quite different (Hughes, 1990; Havelson and Panzer, 1980). The process of coagulation induced destabilization through charge neutralisation by the addition of inorganic chemicals. Coagulation describes the initial colloid destabilisation, principally by charge neutralisation after adding the coagulant (Jiang, 2001). According to Tripathy and Ranjan (2006), flocculation process are differ from coagulation by forming larger agglomerates of particles in suspension or of small agglomerates already formed as a result of coagulation through high molecular weight polymeric materials. Generally, flocculation describes the subsequent aggregation of µm and sub µm size particles into mm size flocs. Because coagulation and the early phase of flocculation occur very rapidly, in practice there is little distinction. Hence, the term either 'coagulation' or 'flocculation' could be used to describe the overall treatment process Nevertheless, in wastewater treatment, referring to both processes as coagulation is acceptable (Jiang, 2001).

According to the water treatment guideline suggested by the US EPA (2002), chemicals that are used in coagulation or flocculation process are usually categorized either as primary coagulants or as coagulants aids. Coagulant aids aims to destabilize the water by enhance the density for slow-settling flocs or toughness. Therefore, coagulation can be defined as the process of adding a coagulant to a liquid to destabilize suspended matter, colloids, and other substances, and allow them to be transformed into a larger form in which finally it can be easily removed from the process (Zin *et al.*, 2013).

Commonly used coagulants are inorganic metal salts such as aluminium sulphate and ferric chloride (Lee *et al.*, 2014) that are supplied in powder or in liquid form (Tzoupanos and Zouboulis, 2008). Zin *et al.*, (2013) also agreed that the most commonly used chemical coagulants are alum, ferrous sulfate, ferric chlorosulfate, and ferric chloride. Other chemical based coagulants used that are including lime and polyaluminium chloride (PAC). Principal aluminium compounds are commercially available for suspended solids removal are dry and liquid alum. Commercial dry alum that is most often used in wastewater treatment is known as filler alum with the approximate formula $Al_2(SO_4)_3 14H_2O$ and a molecular weight of about 600 (Tripathy & Ranjan, 2006).

The current operational procedures at many treatment plants in developing countries are works based on arbitrary guidelines, particularly in relation with the dosage of chemicals used (Ali *et al.*, 2009). Although the drawbacks of aluminium in drinking water are known, surprisingly, it is still widely used around the world due to the effectiveness in treating water. The main purpose to use natural coagulants for water treatment is to making it as point of use (POU). There are many studies conducted on natural coagulants include in Malaysia. The research on it is still on going to find other types of plants and non-plant source that can has high potential to be use as coagulant. The process of coagulation-flocculation starts by sedimentation, filtration, and followed by disinfection, often by chlorine, that is commonly used worldwide in the water treatment industry before distribution of treated water to consumers (Ndabigengesere and Narasiah, 1998).

Coagulation are derives from the *Latin* word 'coagulare' which means to agglomerate (SNF Floerger, 2003). In general, water treatment originally focused to improve the aesthetic qualities of the drinking water. Ancient Sanskrit and Greek writings recommended water treatment methods such as filtering through charcoal, expose to sunlight, boiling, and straining. Coagulation and flocculation could be achieved using either natural coagulants or chemical-based coagulants. Among the two, natural coagulants have long been acknowledged for their application in traditional water purification which was derived from various ancient records cited (Dorea, 2006; Bratby, 2006). The use of plant-based coagulants to purify turbidity of