APPLICATION OF AZADIRACHTA INDICA LEAVES AS NATURAL COAGULANT FOR KITCHEN WASTEWATER TREATMENT

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

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ABSTRAK

Salah satu kaedah alternatif untuk merawat air buangan adalah dengan kaedah penggumpalan-pengelompokan. Bahan penggumpal yang digunakan dalam proses ini, mungkin memberi kesan negatif kepada alam sekitar kerana kebanyakannya berasaskan bahan kimia dan tidak mesra alam. Hasilnya, penghasilan bahan penggumpal yang mesra alam diperlukan untuk mencapai pembangunan yang mampan. Penyelidikan sebelumnya yang dilakukan tidak membincangkan lebih lanjut mengenai penggunaan ekstrak daun Azadirachta indica (neem) sebagai bahan penggumpal semula jadi untuk mengurangkan parameter lain kecuali kekeruhan air. Keupayaan serbuk daun neem, daun neem dirawat dengan air suling dan asid hidroklorik (AI-powder, AIp-DW, AIp-0.1M HCl dan AIp-0.025M HCl), untuk bertindak sebagai bahan penggumpal dalam kajian ini dinilai menggunakan air sisa domestik melalui ujian balang. Aktiviti penggumpalan dan kecekapan penyingkiran (RE) kekeruhan, warna, minyak dan gris dan keperluan oksigen kimia (COD) dikira dari hasil ujian balang. Tahap pH air sisa dikawal pada pH 7 untuk meningkatkan kecekapannya semasa ujian balang. AIp-0.1M HCl mencatatkan RE tertinggi untuk semua parameter yang masing-masing adalah 58%, 44% dan 6.44% kekeruhan, warna dan minyak dan gris. Walau bagaimanapun, nilai COD yang dicatat meningkat untuk semua jenis bahan penggumpal yang digunakan dalam projek ini kerana bahan organik yang tinggal dalam serbuk daun neem tidak dikeluarkan. Justeru, penggunaan ekstrak dari daun A. indica sebagai bahan pembekuan dapat diusulkan sebagai alternatif dalam pengolahan air buangan bukan hanya dari sumber domestik tetapi juga dari sumber industri melalui penggumpalan-pengelompokan.

ABSTRACT

One of the alternative methods for treating wastewater effluent is by coagulation-flocculation treatment. The conventional coagulants used in this process, on the other hand, may have a negative impact on the environment because they are primarily chemical-based and not environmentally friendly. As a result, producing an environmentally friendly coagulant is required to achieve sustainable development. Previous research conducted did not further discussed about the use of extracts from Azadirachta indica (neem) leaves as a coagulant to reduce other parameters except for turbidity. The ability of neem leaves powder, neem leaves treated with distilled water and hydrochloric acid (AI-powder, AIp-DW, AIp-0.1M HCl and AIp-0.025M HCl), to act as a coagulant in this study was evaluated using domestic wastewater using jar test. The coagulation activity and removal efficiency (RE) of turbidity, colour, oil and grease and chemical oxygen demand (COD) were calculated from jar test results. The pH level of wastewater was controlled at pH 7 to increase its efficiency during jar test. AIp-0.1M HCl recorded the highest RE for all parameters, which are 58%, 44% and 6.44% of turbidity, colour and oil and grease, respectively. However, the COD value recorded was increasing for all types of coagulant used in this project due to the remaining organic matter in neem leaf powder has not been removed. From the result, the use of extracts from A. indica leaves as coagulants has a potential to be proposed as an alternative coagulant in the treatment of wastewater not only from domestic sources but also from industrial sources via coagulation-flocculation.

TABLE OF CONTENTS

LIST OF TABLES

Page

LIST OF FIGURES

Page

LIST OF ABBREVIATIONS

UNESCO	United Nations Educational, Scientific and Cultural Organization
BOD	Biochemical oxygen demand
MOH	Ministry of Health
COD	Chemical oxygen demand
DAF	Dissolved air flotation
$AlCl_3$	Aluminium chloride
PAC	Polyaluminium chloride
$FeCl_3$	Ferric chloride
A. indica	Azadirachta indica
SS	Suspended solid
AC-H_2O	Acorn leaf treated with distilled water
NaCl	Sodium chloride
NaOH	Sodium hydroxide
HCl	Hydrochloric acid
MO	Moringa oleifera
TSS	Total suspended solids
RE	Removal efficiency
Zp	Zeta potential
FOG	Fat oil and grease
WW	Wastewater
NTU	Nephelometric Turbidity unit
pH	Potential Hydrogen
DO	Dissloved oxygen
TDS	Total dissolved solids
ORP	Oxidation reduction potential
AIp-DW	A. indica leaves treated in distilled water
AIp-0.1M HCl	A. indica leaves treated in HCl with concentration of 0.1M
AIp-0.025M HCl	A. indica leaves treated in HCl with concentration of 0.025M
rpm	Revolution per minute

CHAPTER 1

INTRODUCTION

1.1 Background of the Study

Water is necessary for all living things on the planet. Water occupied a large part of the planet, but clean water contributes to only just a small amount of it. Furthermore, a portion of the remaining water could be polluted to the point that they are unsafe for direct use. Water is a crucial factor in economic growth since it is widely used in various productive sectors, including manufacturing, livestock and agricultural production, and urban supply, resulting in water overuse. According to UNESCO, a reduction in water quality will lead to water scarcity (Villaseñor-Basulto *et al.*, 2018).

Wastewater treatment is the decomposition process of highly complex, putrescible, organic solids into mineral or reasonably stable organic solids. During primary and secondary treatment, the bulk of biochemical oxygen demand (BOD) and suspended solids in wastewater are eliminated. However, it is becoming increasingly clear that this level of care is insufficient to protect receiving waters or provide reusable water for industrial and domestic recycling (Sonune and Ghate, 2004).

Contaminated drinking water is a significant carrier of disease-causing agents that can endanger human health (Ashbolt, 2015). Therefore, proper water treatment is needed to eliminate turbidity and other pathogenic bacteria, which can be provided by coagulants (Choy *et al.*, 2014). Mainly in water treatment, the first step is coagulation and flocculation, which can be accomplished with either natural or chemical-based coagulants.

Several previous studies focused on the coagulation-flocculation process using chemical-based (inorganic) coagulants such as alum and ferric salts. However, there are concerns about introducing chemical coagulants into the environment. For example, there are several severe disadvantages to using aluminium-based coagulants (the main component of PACI and alum), such as Alzheimer's disease. Nowadays, there is a greater emphasis on using natural coagulants to alleviate water and wastewater treatment challenges in developing countries. Fortunately, there is little published information on the use of natural coagulants for wastewater purification.

Before introducing chemical-based coagulants such as aluminium and ferric salts, natural coagulants of vegetable and mineral origin were used in water and wastewater treatment. Natural coagulants are biodegradable and cost-effective for developing countries. Moreover, they can be grown locally and have a more comprehensive effective dosage range for flocculation of different colloidal suspensions in water purification. This study is focusing on the treatment of domestic wastewater by using natural coagulants.

1.2 Problem Statement

Domestic wastewaters from the kitchen are among the environmentally damaging input to the ecosystem because they contain turbid wastewater that is heavily polluted to the environment. Kitchen wastewater is a form of grey wastewater that primarily contains organic, oil, and grease contaminants. Human wellbeing is directly affected by the purity of water consumed (Nair *et al.*, 2019). It also has detrimental environmental consequences, such as eutrophication of water sources and clogging of sewage pipes. As a result, before being discharged, this form of wastewater must be treated.

Coagulation and flocculation are two processes that occur in sequence to counteract the forces that keep the suspended particles in the aqueous solution stable. Coagulants are used in coagulation are the most crucial physicochemical process in the treatment of potable water to reduce the electrical charge between suspended particles (Katrivesis *et al.*, 2019). As the main coagulant, aluminium sulphate and iron salts are commonly used in the coagulation process (Rubini et al., 2019). However, chemical coagulants are undesirable to both humans and the environment. Many studies published on the association of some neurodegenerative and neurotoxic diseases (Adesina *et al.*, 2019) between Alzheimer's disease and redundant aluminium compounds. As suggested by the World Health Organisation, the level of aluminium in potable water should not reach 0.2 mg/L as well as in Malaysia's Drinking Water Standard (Ministry of Health (MOH), 2019).

Coagulant plays a vital role in the primary step of coagulation and flocculation process wastewater treatment. However, spending a hefty amount of chemical coagulants will increase the total treatment cost of wastewater treatment. Therefore, several studies have been carried out to identify healthy and environmentally sustainable natural coagulants to replace inorganic and chemical coagulants in potable water (Camacho *et al.*, 2017). Hence, a natural coagulant is chosen in this project to replace the dependency on chemical coagulant. Natural coagulant of plant-based chosen is from the leaves of Azadirachta indica (neem). It is a convenient, environmentally sustainable, and cost-effective form of water treatment. Although there are some data and information related to the topic, fewer studies in the literature review regarding the analysis performed. As a result, the current research aims to analyze the use of neem leaf as coagulants and turbidity removers and how it can purify wastewater from domestic sources.

1.3 Objectives

The aims of this study are:

i. To test the efficiency of neem leaf as a natural coagulant for wastewater treatment in improving selected water quality parameters.
ii. To compare the efficiency obtained from neem leaf with chemical coagulants applied in past research.

1.4 Scope of Work

The work concentrates on identifying and determining any possible plant that can be used as a plant-based coagulant in water and wastewater treatment. Any part of the plant can be used as a coagulant, including the seed, husk, flower, leaf, fruit, stem, and so on. One plant-based coagulant is chosen from the leaf part to test its efficiency in improving the selected water quality parameters. The result obtained then will be compared with the efficiency of chemical coagulants used in past research. Comparison of the efficiency of water treatment is based on pH, turbidity, colour, COD and oil and grease.

1.5 Justification of Research

The primary goal of this research is to test the efficiency of neem leaves powder as a plant-based coagulant for wastewater treatment in improving selected water quality parameters. Furthermore, the efficiency obtained from neem leaves powder will be compared with the efficiency of chemical coagulants applied in past research. In the future, neem leaves may be used as a natural coagulant in improving the wastewater treatment process. The output from this objective may reduce the chemical coagulant dependency, thus produce treated water with better quality. Furthermore, when opposed to chemical coagulants, which are non-biodegradable and usually create hazardous sludge, natural coagulants can be more environmentally friendly and sustainable since they are biodegradable.

The sample used in this research is collected from the downstream of outlet waste from the kitchen stall. By having a natural coagulant to treat the domestic wastewater, the sludge produce is low in volume but higher in nutritional sludge value. It has also been demonstrated in previous research that the use of natural coagulants is non-corrosive, which eliminates the risk of pipe erosion. Thus the treated water produced is safe for consumption and could be discharged to the environment without having too many pollutants.

1.6 Dissertation Outline

This dissertation is categorized into five chapters. Chapter 1 is an introduction that contains the background of this study, problems statement, objectives and scope of work. The outline of subsequent chapters is stated below.

Chapter 2 discuss literature review, which describes various subjects relevant to the study from past research. This chapter focuses on different plant-based coagulants and chemical coagulants used in past research and characteristics of wastewater used. This chapter shows strong evidence that plant-based coagulants can help in improving water quality.

Chapter 3 explains the methodology to carry out the project. First, this chapter discusses how to prepare the selected plant-based coagulant and coagulation process by jar test apparatus. Next, it also includes the technique used in analytical procedures to determine the efficiency of selected plant-based coagulants after being applied.

Chapter 4 explains the results and discussions of this project. The result obtains such removal efficiency according to selected parameters such as pH, turbidity, colour, COD, and oil and grease. The result is tabulated in Microsoft excel and shown in graph format to get a clear view of the efficiency after treatments. Result obtained from this project was then compared with the efficiency of chemical coagulant used in past research.

Lastly, Chapter 5 discussed the conclusion and future recommendations of the overall project. It is concluded that all objectives are achieved in this project.

CHAPTER 2

LITERATURE REVIEW

2.1 Coagulation and flocculation process

The method of combining small particles into larger aggregates (flocs) and adsorbing dissolved organic matter onto particulate aggregates to remove impurities in subsequent solid/liquid separation processes is known as coagulation and flocculation. The Schultz–Hardy rule and Smoluchowski's particle collision function were first used to explain the coagulation process, which forms the theoretical basis of coagulant demand and particle number changes in the flocculation system (Jiang, 2015).

Coagulation is the mechanism of neutralising the negative charge of suspended particles in water using cationic coagulants, which have positive electric bills to balance the colloids' negative charges. The particles collide as a result of the addition of coagulants, forming larger particles known as flocs. Rapid mixing is allowed to disperse the coagulant and facilitate particle collisions, maximising the effectiveness during the process. Extra caution should be taken to ensure that the solution does not contain excessive coagulants, resulting in a complete charge reversal and the colloidal complex being re-stabilised. The primary reactions and processes that occur during coagulation are depicted in Figure 2.1.

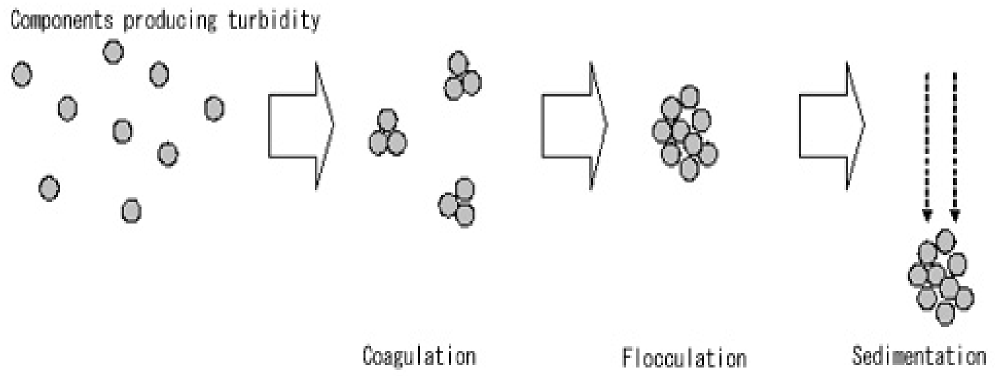

Figure 2.1: Process of Coagulation, Flocculation and Sedimentation (Conventional Water Treatment: Coagulation and Filtration (2019)

The flocculation process converts unstable particles into bulky flocs, which followed directly after the coagulation process to settle easier. The action of coagulants in forming bridges between flocs to connect the particles into large agglomerates or clumps is known as flocculation.

Meanwhile, bridging occur when segments of the coagulants chain adsorbed on various particles and helped them to aggregate (Lee et al., 2014). Slow stirring is necessary during flocculation to enable contact between small flocs and agglomerate them into larger flocs, improving subsequent removal. To remove the large flocs, solid removal processes such as sedimentation, rapid or membrane filtration, or DAF can be used.

2.2 Coagulants

Coagulant The coagulant is an essential part of the treatment process for both surface water and industrial wastewater. Coagulants are chemicals that are added to water or wastewater to cause coagulation. The coagulation method involves the addition of conventional chemical-based coagulants, such as alum ($AlCl3$), ferric chloride ($FeCl3$), and polyaluminium chloride (PAC), to remove dissolved chemical species and turbidity from water (Yin, 2010). These coagulants improve solid/liquid separation by aggregating fine particles into large agglomerates, resulting in entities that settle faster and are easier to capture in filters. More than a century ago, coagulants such as ferric chloride and aluminium sulphate were used in full-scale water treatment plants for the first time (Jiang, 2015). However, chemical coagulants have weaknesses in addition to their effectiveness.

When coagulant is used to treat water, there is a chance that coagulant residue will remain in the water after the operation (Amran *et al.*, 2018). Alum residue is undesirable and potentially unhealthy because it can cause illness. In humans, aluminium-based coagulants have been related to the development of Alzheimer's disease (Rondeau *et al.*, 2000; Flaten, 2001). Hence, the coagulant for the specific wastewater must be carefully chosen based on its feasibility. This is required for an efficient coagulation process and leave the smallest possible residual of the chemical in the treated water. Furthermore, a natural coagulant is gaining popularity as a result of the findings due to its advantages over chemical coagulants.

2.3 Plant-Based Coagulant

Plant and animal-based coagulants are the two types of naturally derived coagulants. Isinglass from shredded fish bladders and chitosan from crustacean shells are two popular animal-based coagulants (Bina *et al.*, 2009). On the other hand, plant-based coagulants have far more readily accessible sources than animal-based coagulants, implying that they may be viable alternatives to chemical coagulants and have grown in popularity. Since 2000 years ago, ancient civilisations in India, China, and Africa have used plant derivatives as natural coagulants in their water sources (Asrafuzzaman et al., 2011). In addition to being water clarifying agents, natural coagulants have antimicrobial and heavy metal removal properties in some instances (Choy *et al.*, 2015).

Plant-based coagulants gain popularity as a natural, non-hazardous, degradable, and potentially carbon-neutral alternative to the current coagulants. Plant-based coagulants contain rich proteins and polysaccharides termed polyelectrolytes for being polymers (Bolto and Gregory, 2007). Since effluent production is inevitable, the water obtained after treatment could be recycled in the industry's outside areas. In addition, the produced sludge will be easier to dispose of due to its biodegradability (Kumar et al., 2020). In colloid-free water and solutions, plant-based coagulants form random patterns, restricting the irreversibility of loops, tails, and trains to adsorbed colloid particles (Bolto and Gregory, 2007). In addition, plant-based coagulants act by neutralising charges (Miller *et al.*, 2008) and inter-particle bridging (Antov et al., 2010; Hameed *et al.*, 2016), which helps to destabilise colloidal suspensions and form micro-and macro-flocks.

The use of these natural coagulants is an environmentally friendly aspect, and their advantages outweigh their drawbacks. The material that has gotten the most attention is the seed of M. oleifera. The M. oleifera tree, also known as drumstick and horseradish, is native to Northern India and is widely planted throughout the tropics. According to research, the cactus has similar properties to M. oleifera seeds and thus can act as a coagulant (Yin, 2010; Vijayaraghavan, G.; Sivakumar, 2011). Cactus is native to the torrid and subtropical regions of the world. It has recently gained popularity due to its nutritional and medicinal components, including proteins, amylose, malic acid, resin, vitamin, and cellulose.

Based on the fact stated above, the current study is geared toward the potential use of A. indica (neem) leaves as a coagulant to treat domestic wastewater via a jar test experiment of coagulation-flocculation process. The use of A. indica leaves as a potential coagulant for domestic wastewater treatment, on the other hand, has never been investigated further in the literature. Based on past research, most of it only covers the removal efficiency of turbidity using neem leaves. To evaluate the removal performance of A. indica leaves, its potential transformation from plant to natural coagulant must be further investigated in this project.

2.3.1 Azadirachta Indica (Neem) leaf

The Azadirachta indica, commonly known as neem, is native to India and can be found in most tropical and subtropical countries. Many biologically active compounds can be derived from neem's chemical constituents, with azadirachtin being the most biologically active (Hashmat et al., 2012). The neem tree is a common medicinal plant in Asia and Africa used for various medicinal purposes since ancient times. Because of its high concentration of biologically active constituents, it is used in multiple traditional remedies. Thus, it has a lot of medicinal value and has been used all over the world. The various active compounds extracted from each plant's parts account for their use and attribution as a medicinal plant (Eid et al., 2017). Figure 2.2 below show the A. indica leaves.

Figure 2.2: A. indica leaves

The chosen part from neem to be used as a natural coagulant in this study is extracted from its leaf. During long-term agitation treatment, the surface structures of neem leaf will remain stable and can also be obtained without unnecessary expense (Mohan *et al.*, 2019). Hence, it clearly shows that it can serve as a potential coagulant to remove contaminants from the water. Moreover, neem leaves are also used in the preparation of litter compost and as green leaf manure. Furthermore, it was found that neem leaf extracts had insecticidal properties. It is used as a foliar spray and for seed treatment in rice cultivation (Lokanadhan et al., 2012).

Sowmeyan et al. (2018) found that neem leaf reduced 94% of fluoride with an optimum dose of 2 mg/L in a study on the efficacy of herbs in community water treatment. In comparison to other studies, this research shows that fluoride elimination from raw water is exceptionally high. As a result, we can conclude that neem contains certain elements that can help to reduce the harmful effects of excessive fluoride. Another experiment was conducted by taking a wastewater sample from a paper mill. Among the chosen natural coagulants, A. indica showed a better coagulation and turbidity removal due to a high percentage of 63.01% turbidity removal with an optimum dosage of 6.5g (Saravanan *et al.*, 2018). The efficiency from neem leaf powder is then compared with the chemical coagulant used, aluminium sulphate. The turbidity removal of alum is only 75.01%.

Findings on another literature review by Anju (2016) resulted in turbidity removal of 98% when neem leaf was used as a coagulant. The pH was alkaline, in the range of 8 and above but less than 9. Therefore, neem leaf powder can be used as a water purifier, and an environmentally friendly water purification method could be created (Mohan *et al.*, 2019). The optimum dose was found to be 30 mg/L in an experiment performed by Nair *et al*. (2019), with the percentage turbidity removal efficiency of neem leaves being 88.75% and the optimum pH range being 6.80-7.02.

According to a study conducted by Mohan *et al.* (2019), neem leaves powder has a significant impact on the removal of physicochemical parameters such as pH, total solids, turbidity, COD, BOD, and other parameters, as shown in Table 2.1 below. The pH of the water was reduced by using neem leaf, with the pH level falling between 7 and 8 after treatment, which was within the acceptable range. Neem leaves have been recommended for the decolourisation of aqueous solutions of coloured effluents from factories because they are less expensive but much more effective than commercially available activated charcoal.

Table 2.1: Results of using neem leaves to treat wastewater (Mohan *et al.*, 2019)

No.	Parameters	Before treatment values	After treatment values
1.	pH	9.2	7.7
2.	Total solids mg/l	5819	4628
3.	TDS mg/l	5770	4516
4.	TSS mg/l	49	42
5.	EC mhos/cm	8261.3	7010
6.	Turbidity NTU	24	20
7.	T. alkalinity mg/l	90	80
8.	Bi-carbonate alkalinity mg/l	30	20
9.	T. hardness mg/l	80	70
10.	C. hardness mg/l	45	40
11.	MG hardness mg/l	35	30
12.	Calcium mg/l	18	10
13.	Magnesium mg/l	8.4	6
14.	Carbonate hardness mg/l	80	50
15.	Non carbonate hardness mg/l	Nil	Nil
16.	Chloride mg/l	4301.23	3176.32
17.	Sulphate mg/l	1620	1609
18.	MLVSS mg/l	31	18
19.	Iron mg/l	0.8	0.5
20.	Silica mg/l	2.92	2.81
21.	COD mg/l	1173	601
22.	BOD mg/l	282.6	206
23.	Copper mg/l	<0.01	<0.01
24.	Chromium mg/l	1.2	<0.01

2.3.2 Ocimum sanctum (Tulsi) leaf

Tulsi, or Ocimum sanctum, is an aromatic plant that belongs to the Lamiaceae family. This plant comes in two colours; black, Krishna Tulsi, and green, known as Rama Tulsi (N *et al.*, 2017). Both varieties, however, are considered equivalent in terms of their chemical constituents. It is known as "holy basil" or "sacred basil" and is present in almost every Indian household due to its medicinal, nutritional, and spiritual properties. It has been discovered that regularly consuming the leaves and their products helps prevent diseases, encourage health, longevity, and well-being, and relieve physical and mental stress (Sah et al., 2018). In addition, Tulsi leaves contain naturally occurring chemicals such as eugenol, beta-cymene, and carvacol, which have a wide range of antibacterial applications (Pandiri and Moni, 2018). The O. sanctum plant is shown in Figure 2.3 below.

Figure 2.3: O. sanctum leaf and flower

In a study conducted by Dhruva and Suresh (2016), the effect of Tulsi leaf on reducing turbidity, COD, and pH from sewage water was demonstrated. The results show that the optimal dose for turbidity removal is 150mg/L, with a removal rate of 69.82% for turbidity and 43.11% for COD. Another research by Nair *et al.* (2019) found that the optimal dose with a turbidity value removal of 14 NTU was 30 mg/L. The optimal pH range and percentage turbidity elimination are 8.48-9.06 and 94.17%, respectively. Holy basil leaves are non-toxic and can be used as a coagulant in water treatment. It is a simple, environmentally friendly, and cost-effective water treatment system. In rural areas, this form of care is more efficient and cost-effective. Moreover, in rural areas, the sludge that settles at the bottom of the tank after treatment can be used as bio-fertilisers (Dhruva and Suresh, 2016).

2.3.3 Hibiscus rosa sinensis (leaf)

The herb Hibiscus rosa sinensis L. (Malvaceae) is native to China, as Rosa sinensis means "rose of China" (Sivaraman and Saju, 2021). It's a tropical shrub with a wide range of flower colours widely used as an ornamental plant in the tropics. Hibiscus is a common herbal tea ingredient that also has medicinal properties. In medicine, the red-flowered variety is favoured. According to various reports, specific Hibiscus plants have different medicinal properties (Khristi and Patel, 2017). For example, the leaves and flowers of Hibiscus rosa-sinensis are used as an antiseptic for boils and ulcers (Divya *et al.*, 2013). In addition, the leaves of this plant can help with body burning, urinary discharges, seminal weakness, piles, uterine and vaginal discharges, and foetal formation (Kumar, 2012). Figure 2.4 below show the Hibiscus rosa-sinensis plant.

Figure 2.4: Hibiscus rosa sinensis flower and leaf

Aziz et al. (2012) evaluate the effectiveness of Hibiscus rosa sinensis leaf extract in the treatment of textile wastewater. This natural coagulant was discovered in textile wastewater treatment to serve as a coagulant and coagulant aid with poly aluminium chloride (PAC). COD and colour removal was found to be ineffective when hibiscus rosa sinensis leaf extract was used as a coagulant, with removal rates of 10.9% and 17.14%, respectively. Suspended solids (SS) removal, on the other hand, is high, at 43%. The sticky nature of the polysaccharides present in Hibiscus rosa-sinensis, which resulted in the formation of larger flocs, may have contributed to the successful removal of SS. Since the larger flocs settle more rapidly, a modest decrease in SS concentration was observed. Furthermore, at an optimal pH of 6, where the leaf extract performed best, the parameter such as COD, SS, and colour removal efficiency was 8.33, 42.2, and 14.3%, respectively.

Furthermore, the antimicrobial properties of this plant demonstrated the ability to inhibit some of the bacteria tested, including Escherichia coli and Staphylococcus aureus, which are commonly found in surface water.

2.3.4 Acorn (leaf)

Quercus L. (Oak) is a member of the Fagaceae family, which includes a woody plant with simple alternate leaves distinguished by their wood, wind-pollinated flowers, acorns as fruits, and the ability to live for many years (Tantray et al., 2017). The leaf of oak was studied as a plant-based coagulant to improve water quality and reduce raw water turbidity (Benalia *et al.*, 2018). Figure 2.5 below show the acorn leaf and its fruit.

Figure 2.5: Acorn leaf

The fine powder made from acorn leaves was pre-treated with various solvents in order to boost the coagulant's behaviour, for AC-H2O and AC-powder recorded the highest turbidity efficiency at 84.77% and 71.6%, respectively. Liquid agents are more mobile and open to colloids than solid agents, which explained the difference in its removal. Following that, the turbidity removal efficiency of the treated powder with other solutions was 91.07%, 85.92%, and 92.92% for NaCl (0.5 M), NaOH (0.05 M), and HCl (0.1M), respectively. The turbidity removal efficiency increased as the NaCl concentration increased from 0.25 M to 0.5 M. This finding is consistent with previous research and is linked to two phenomena. The first is the "salting-in effect," in which more coagulant agent is extracted from the acorn leaves and dissolved in the extracting solvent solution at higher NaCl concentrations. Because the coagulant agent is a protein, as the salt concentration increased, so did the coagulant agent's solubility and thus its concentration in the solution. The second phenomenon is the effect of salt on particle aggregation. For this case, increasing the salt concentration resulted in intense particle aggregation owing to double-layer compression.

The flection point in turbidity removal efficiency after 0.05 M, with a minimum for the coagulant obtained from the 0.1 M NaOH extracting solution can be explained that some proteins may be denatured at the NaOH concentration, reducing protein solubility in the extracting solution.

2.3.5 Moringa Oleifera (MO)

Moringa oleifera, also known as the "drumstick tree" or "horseradish tree," is a tropical and subtropical tree native to India that grows worldwide. In areas with less than 400 mm of annual rainfall, it is a rapidly growing tree where it can reach 6–7 metres in height in a year (Foidl et al., 2001). M. oleifera is now predominantly found in the Middle East, Africa, and Asia, but due to its adaptability, it is spreading to new areas, especially drought-stricken tropical and subtropical lands (Leone *et al.*, 2016). Moringa's leaves, pods, and seeds contain several essential phytochemicals, making it a nutrient-dense plant (Gopalakrishnan et al., 2016). Any part of the tree is ideal for nutritional or commercial purposes due to its high nutritive value. M. oleifera seed, as shown in Figure 2.6, is extensively used as a natural coagulant in water treatment.

Figure 2.6: MO seeds

Moringa seed powder was often used to treat industrial wastewater and proven to be more effective than the tamarind seed powder to treat tofu industrial wastewater based on a study conducted by (Setyawati, 2017). The efficiency obtained for the removal of BOD, COD and TSS is 85.09%, 88% and 85.72%, respectively. However, the usage of MO as a coagulant did not significantly affect the COD value in the treatment of batik industrial wastewater due to its value remain the same after the treatment process (Effendi et al., 2015). The percentage removal obtained for turbidity is 95.5%, and TSS is 87.5%. Another study conducted by Fagundes-klen and Dotto (2019) on textile wastewater using MO seed as coagulant proved that apparent colour and COD removal are high at 82.2% and 83.05%, respectively.

Proteins contain amino acids that are soluble in water and positively charged to bind with particles to form flocs are found in MO seed and can serve as an effective natural coagulant in the pre-treatment of water purification systems (Chu *et al.*, 2017). The amino acid mentioned contains glutamine, arginine, proline, and 60 residuals, whereas the protein-peptide contains 8 positively charged amino acids, 7 arginines and histidine, and 15 glutamine residuals (Thanh, 2018). Because of their antimicrobial and coagulant properties, extracted low molecular weight proteins, chitin-binding proteins, cationic polypeptides, and lectins are widely used in water treatment to remove hardness and microorganisms.

2.3.6 Tamarindus indica

Tamarind (Tamarindus indica) is a tropical African leguminous tree that belongs to the Fabaceae family. Tamarind is widely spread throughout Asia and Southeast Asia, especially in India and Thailand, where the species is planted in large plantation scales and is economically significant (Aziz *et al.*, 2018). Tamarindus indica is known as a medicinal plant that has been used for centuries to treat wounds, snake bites, stomach pain, colds, inflammations, diarrhea, helminth infections, and fever (Menezes *et al.*, 2016). Each part of the plant plays various advantages according to its chemical constituents. Figure 2.7 below show a tamarind fruit, including the seed.

Figure 2.7: Tamarindus indica fruit and seed

Tamarind seeds can be used as plant-based coagulants because of the seeds' protein content, which functions as a polyelectrolyte. Polyelectrolytes are polymers that have ionised groups of positive or negative charges (Dobrynin and Rubinstein, 2005). The group can dissociate in a polar solvent like water, leaving the charge on the polymer chain and releasing the opposing ion into the solution. When a polyelectrolyte concentration is applied, colloidal stability is decreased, and the rejecting force between particles is reduced, allowing the precipitation process to continue (Setyawati, 2017).

Tamarind seeds have also been said to have the potential to lower fluoride levels. The high polyphenolic content, especially tannins, may be responsible for these behaviours. Tannins are a safer alternative to chemical coagulants due to their structure and degree of tannin alteration (Mathuram et al., 2018). It is well recognised that phenolic groups deprotonate easily to form phenoxide, which is stabilised by resonance. This is said to boost the coagulation effect, and since tannins contain a lot of phenolic groups, they naturally have higher coagulation ability.

In a study conducted by Setyawati (2017), they used tamarind seed as a coagulant to treat tofu industrial wastewater. The parameters examined were BOD, COD and TSS. The results obtained after the treatment process are 79.02%, 84%, and 71.43% for BOD, COD, and TSS. Table 2.2 below show the research summary of natural coagulants used in wastewater treatment.

Table 2.2: Research summary of natural coagulants used in wastewater treatment.

No.	Coagulant	Reference	Type of wastewater	Removal parameter (%)	Remarks
1.	Azadirachta indica (leaf)	(Sowmeyan et al., 2018)	Raw water sample	Fluoride (94%); optimum dosage of 2 mg/L	The potential of Neem leaf to treat turbid wastewater is very efficient due to the percentage of turbidity removal is high compared to other natural coagulants. The range of pH is also within the limit, which is 6.5-8.
		(Saravanan *et al.*, 2018)	Paper mill wastewater	Turbidity (63.01%); optimum dosage 6.5 g	
		(Anju, 2016)	Dairy industry	Turbidity (98%), optimum pH (8-9)	
		(Nair *et al.*, 2019)	Kaolin synthetic wastewater	Turbidity (88.75%), optimum pH (6.80-7.02); optimum dosage of 30 mg/L	
		(Mohan *et al.*, 2019)	Textile wastewater	pH (19.48%), TDS (27.76%), TSS (16.67%), Turbidity (20%), Hardness; T (14.29%), Ca (12.5%), Mg (16.67%), COD (95.17%), BOD (37.18%)	
		(Rubini et al., 2019)	Kitchen wastewater	Turbidity (65%); optimum dosage 400 mg/L	
2.	Ocimum sanctum (leaf)	(Dhruva and Suresh, 2016)	Sewage	Turbidity (69.82%), COD (43.11%); optimum dosage of 150 mg/100mL	Tulsi leaf can be used as a coagulant in the treatment of wastewater due to the percentage removal is high. Holy basil leaf can be used in rural areas where no facilities are available for drinking water treatment.
		(Nair *et al.*, 2019)	Canteen wastewater	Turbidity (94.17%), optimum pH (8.48-9.06); optimum dosage of 30 mg/L	

Table 2.2: Continued

No.	Coagulant	Reference	Type of wastewater	Removal parameter (%)	Remarks
3.	Hibiscus rosa sinensis (leaf)	(Aziz et al., 2012)	Textile wastewater	COD (10.9%), Colour (17.14%), SS (43%)	The percentage removal of COD, colour and SS is high when hibiscus leaf was used as coagulant aid with PACl
4.	Acorn (leaf)	(Benalia *et al.*, 2018)	Raw water	Turbidity removal; AC-powder (71.6%); AC-H_2O (84.77%); 0.5M NaCl (91.07%); 0.05M NaOH (85.92%); 0.1M HCl (92.92%)	The efficiency of acorn leaf was improve when it is treated with different solution.
5.	Moringa oleifera (seed)	(Setyawati, 2017)	Tofu industrial wastewater	BOD (85.09%), COD (88%), TSS (85.72%)	Moringa seed powder is oftenly used to treat industrial wastewater. But, it did not significantly affect COD value in treatment of batik industrial wastewater.
		(Effendi et al., 2015)	Batik effluent	Turbidity (95.5%), TSS (87.5%)	
		(Fagundes-klen and Dotto, 2019)	Textile wastewater	Apparent colour (82.2%), COD (83.05%)	
6.	Tamarindus indica (seed)	(Setyawati, 2017)	Tofu industrial wastewater	BOD (79.02%), COD (84%), TSS (71.43%)	Compared to MO seed, tamarind seed is less effective to treat industrial wastewater (low percentage removal)