SYNTHESIS OF BANANA PEEL FLOCCULANT FOR THE REMOVAL OF TURBIDITY AND BACTERIA INACTIVATION

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by

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LIST OF ABBREVIATIONS

µL	Microliter
µm	Micrometer.
CFU	Colony Forming Unit.
DSC	Differential Scanning Calorimetry
DTG	Derivative Thermo-Gravimetry.
E. coli	Escherichia coli.
FTIR	Fourier Transform-Infra-Red.
GC-MS	Gas Chromatography–Mass Spectrometry.
GPa	Giga Pascal.
mL	Milliliter
mm	Millimeter.
Nm	Nano Meter.
SEM	Scanning Electron Microscope.
SODIS	Solar disinfection
SSA	Specific Surface Area
TGA	Thermo Gravimetric Analysis
USM	Universiti Sains Malaysia
UV	Ultra Violet.
XRD	X-ray Powder Diffraction

LIST OF SYMBOLS

%	Percent
±	Plus-minus
°	Degree
°C	Degree Celsius
≥	Greater than or equal to
≤	Less than or equal to
mV	Millivolts
mS/cm	MilliSiemens/cm
NTU	Nephelometric turbidity units

SINTESIS EJEN PEMBEKUAN DARIPADA KULIT PISANG UNTUK PENYINGKIRAN KEKERUHAN DAN PENYAHAKTIFAN BAKTERIA

ABSTRAK

Penggunaan dan kegunaan air secara global telah meningkat secara signifikan dalam beberapa tahun terakhir akibat pertumbuhan penduduk. Badan air mengandung jumlah partikel mikroskopis dan zat terlarut yang besar, menjadikannya tidak sesuai untuk digunakan oleh manusia, terutama di negara-negara tropis. Tujuan kajian ini adalah untuk meningkatkan prestasi bahan berbuku bio kulit pisang sebagai salah satu sisa terbanyak di Malaysia dalam aplikasi rawatan air dan menggabungkannya dengan proses pemusnahan solar (SODIS) untuk membasmi air yang dirawat. Hasil kajian menunjukkan bahawa rawatan gelombang mikro pada bahan berbuku bio kulit pisang secara signifikan meningkatkan prestasi pemflokulan. Bahan berbuku bio kulit pisang dalam kajian ini terdiri daripada 22.2 ± 3.1 % hemiselulosa, 15.9 ± 1.7 selulosa, 9.65 ± 2.1 pektin dan hanya 8.1 ± 0.8 % lignin. Analisis protein kulit pisang menunjukkan jumlah protein yang agak rendah iaitu hanya 6.2 ± 0.9 %. Biopolimer ini memiliki pelbagai jenis kumpulan fungsional permukaan dan dengan itu, pelbagai jenis cas permukaan. Oleh itu, apabila bahan berbuku bio kulit pisang digunakan pada air yang tercemar, saiz pin-flok terus meningkat dengan cepat dalam mekanisme penghubung polimer, dan ikatan dan perlanggaran tambahan diperlukan untuk meningkatkan saiz zarah. Penilaian pengubahsuaian proses penggilingan menunjukkan saiz zarah yang lebih kecil, yang juga meningkatkan prestasi pemflokulan dengan meningkatkan kawasan permukaan zarah. Hasil kajian juga menunjukkan pengurangan yang signifikan dalam kekeruhan air sungai, warna, pepejal terlarut, pepejal tersuspensi, permintaan oksigen kimia, permintaan oksigen biokimia dan logam berat. Prestasi

SODIS berkadar songsang dengan tahap kekeruhan, semakin tinggi kekeruhan, semakin rendah inaktivasi bakteria. Pada kekeruhan air terendah, lebih daripada 98% pemusnahan berlaku, berbanding dengan yang tertinggi yang hanya menunjukkan 60% pengurangan. Botol kecil membenarkan penembusan solar yang lebih baik, menghasilkan pemusnahan yang lebih baik. Kejutan, bekas terbesar, walaupun kedalaman airnya besar, menunjukkan pemusnahan yang paling kurang berkesan, walaupun tahap kekeruhan pada tahap terendahnya. Bahan berbuku bio kulit pisang yang diubahsuai berjaya digunakan sebagai bahan pra-rawatan untuk sampel air untuk mengurangkan kekeruhan air sungai sebelum proses SODIS. Bahan berbuku bio berjaya mengurangkan kekeruhan air secara signifikan, yang meningkatkan prestasi SODIS. Semua koliform faecal dan koliform total telah dibersihkan sepenuhnya dari air sungai, manakala 96.3% bakteria hidup berjaya dieliminasi. Menggabungkan bahan berbuku bio kulit pisang dengan SODIS boleh menjadi pendekatan yang baru dalam rawatan air dan digunakan dalam aplikasi rawatan berskala besar pada masa akan datang.

SYNTHESIS OF BANANA PEEL FLOCCULANT FOR THE REMOVAL OF TURBIDITY AND BACTERIA INACTIVATION

ABSTRACT

The consumption and usability of water worldwide have significantly increased in the past few years due to the surge in population. Water bodies contain huge amounts of microscopic particles and dissolved impurities, making them unfavourable for human use, especially in tropical countries. The aim of this study was to enhance the performance of banana peels bio-flocculant as one of the most common wastes in Malaysia in water treatment applications and couple it with solar disinfection process (SODIS) to disinfect the treated water. The results showed that microwave treatment of banana peel bio-flocculant significantly enhanced the flocculation performance. Banana peel bio-flocculant in this study consisted of 22.2 ± 3.1 % hemicellulose, 15.9 ± 1.7 cellulose, 9.65 ±2.1 pectin and only 8.1± 0.8 % lignin. Protein analysis of banana peel showed relatively low amount of protein of only 6.2 ± 0.9 %. These biopolymers are possessing variety of surface functional groups and thus variety of surface charges. Therefore, once the banana peel bio-flocculant applied to polluted water, the size of pin-flocs continues to increase rapidly in a polymer bridging mechanism, and additional bonds and collisions are somehow needed to furtherly increase the particle size. The modification assesses the grinding process, resulting in smaller particle size, which also enhanced the flocculation performance by increasing the surface area of the particles. The results also showed significant reduction in river water turbidity, colour, total dissolved solids, total suspended solids, chemical oxygen demand, biochemical oxygen demand and heavy metals. SODIS performance was inversely proportion with the turbidity level, the higher turbidity the lower bacterial

inactivation. At the lowest water turbidity, more than 98% disinfection was achieved, compared with the highest one that only show 60 % removal. Small bottles allowed better solar penetration, resulting in better disinfection. Surprisingly, the largest container, despite its substantial water depth, exhibited the least effective disinfection, even when the turbidity level was at its lowest. Modified banana peel bio-flocculant was successfully used as pre-treatment material for water sample to reduce river water turbidity prior SODIS process. The bio-flocculants were able to significantly reduce water turbidity, which enhanced the SODIS performance. All the faecal coliform and total coliforms were completely disinfected from raver water, while 96.3 % of total viable bacteria were successfully eliminated. Coupling banana peel bio-flocculant with SODIS could someday be a novel approach in water treatment and applied in the large-scale treatment applications.

CHAPTER 1

INTRODUCTION

1.1 Overview

Water is essential for all the living organisms including humans, animals, plants and even microorganisms, it is the second most important natural resource necessary for the survival of life on earth, comes after the air (Amos et al., 2020). Since the last century, with a dramatical increase in the industrial revolution, many industrial wastes have been increasing proportionally, raising one of the worldwide primary sources of serious pollution problems (Mohan et al., 2019). Air, water and even soil have been polluted because of anthropogenic activities. Water pollution for instance is due to the use of multiple chemical reagents ranged from inorganic compounds to polymers and even organic products (Salmasi et al., 2020).

1.1.1 Water treatment plants

Water treatment is performed in order to improve water quality. The processes employed for water treatment depend on the quality of the water supply. Several approaches have been developed for water treatment depending on the initial level of water quality, the type of pollution and the availability (Saleh et al., 2022). The applicability of these approaches has been always a great challenge in many countries, taking into consideration the cost of installation, safety and the effectiveness. However, most of these methods are designed to have two major phases: solid-liquid separation phase, which is running as pre-treatment step. This phase must be followed by disinfection phase (Li et al., 2009). The pre-treatment phase could be physical, chemical or biological approaches, which applied to reduce water turbidity.

Disinfection phase is used to eliminate the microorganisms using UV or solar treatments to meet the national or international requirements (Chibowski and Szcześ, 2018). Despite the cost and applicability, pre-treatment approaches can be classified into three methods including physical, chemical and biological treatment. Physical treatment are mostly used pre-treatment steps, which include filtration, adsorption and distillation (Ezugbe et al., 2022).

In some cases, physical treatment may be sufficient for water treatment, but in term of water microorganisms' further step may be required. Chemical treatment such as coagulation, precipitation and hydrolysis, are not favourable in term of drinking water, due to the potential health effect for the chemical agents. Biological treatments using rotating microbial contactors have been also utilized in water treatment (Tang and Hadibarata, 2022). Figure 1.1 presents different physical, chemical and biological processes for water treatment.

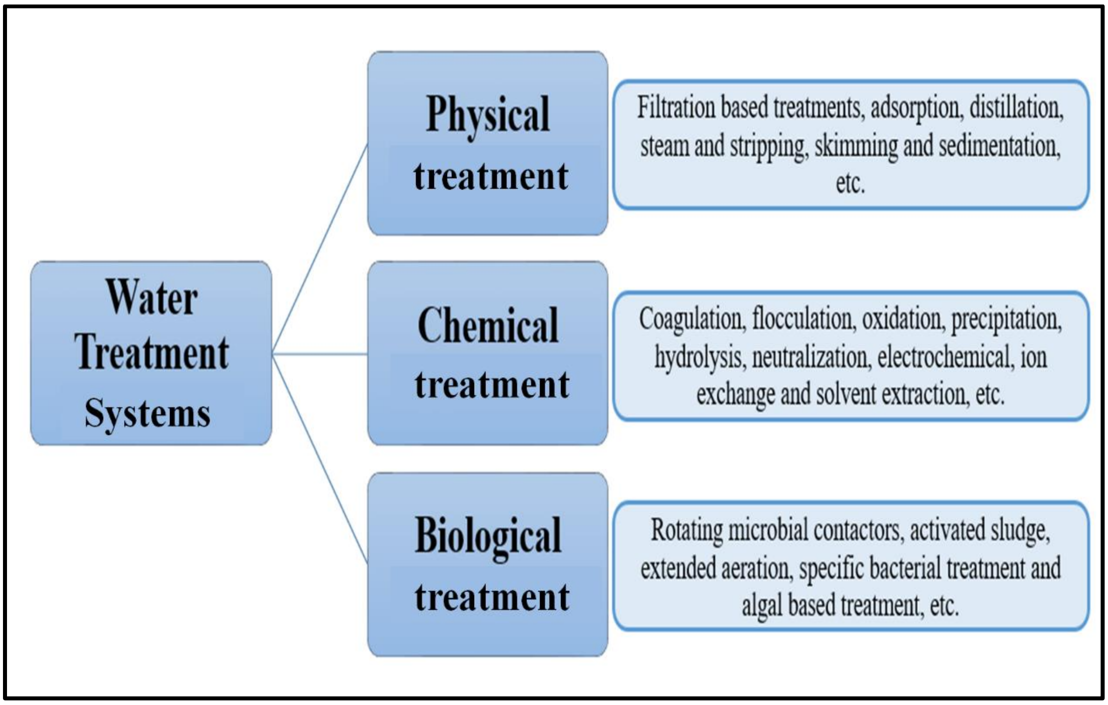

Figure 1.1 Different used water treatment systems

1.1.1(a) Physical water treatment

The physical water treatment approaches typically consists of filtration techniques that involve using any particle size based separation such as screens, sand or cross flow filtration membranes, in addition to adsorption, sedimentation and distillation techniques (Cho et al., 2005). Physical treatment is typically used in numerous undeveloped and developing countries as a pre-treatment method for removing of larger suspended material from water. Thines and co-workers stated two main types of adsorption for different pollutants including physisorption and chemisorption (Thines et al., 2017).

The physisorption occurs due to the attraction of the organic-pollutant and adsorbent by the force of van der Waals, which is a weak force (0.4-4.0 kJ/mol) resulted from electrical interactions between two or more atoms or molecules that are very close to each other, compared with the strong chemical bond such as hydrogen bonds, ionic interactions, and hydrophobic interactions (15, 20 and <40 kJ/mol respectively) that may result between the surface of the adsorbent and the organic pollutant in chemisorption. Table 1.1 presents the summary of conventional physical approaches for water treatment.

Table 1.1 Illustration of different physical treatments for water treatment

Physical treatment	The principal	Reference
Adsorption	The adsorbent attracts organic and inorganic pollutants, leading to their accumulation onto its surface and eventually precipitate.	(Panahi et al., 2018)
Filtration	Passing of water through permeable membrane contains small pores able to trap the desired pollutant.	(Anjum et al., 2019)
Electrodialysis	Separation and movement of ions through ion-exchange membranes under the influence of an electric field. It does not involve chemical reactions to remove ions; instead, it relies on the selective permeability of ion-exchange	(Bernardes et al., 2016)

	membranes to separate specific ions based on their charge and size. The movement of ions is driven purely by electrostatic forces created by the applied electric field.	
Distillation	Separating the pure water from the dissolved pollutant by using selective boiling and condensation approaches.	(Ghernaout, 2019)
Air stripping	Moving air through contaminated water to remove certain pollutant known as volatile organic compounds, which easily evaporate.	(Quan et al., 2009)
Sedimentation	Removing the suspended solids from water by means of gravity or separation.	(Qian et al., 2021)

Various plant-based coagulants and flocculants have been used for water treatment applications, including *Moringa oleifera* (Taiwo et al., 2020), *Cicer arietinum* (Lek et al., 2018), and *Dolichos lablab* (Daverey et al., 2019). Coagulants and flocculants from natural sources are often seen to be safe for human health (Kristianto, 2017). Some natural coagulants and flocculants have been studied and are known to have several advantages. However, many of these natural coagulants and flocculants are valuable, expensive, and not highly available, which limit their usage. The banana tree has been reported to produce from 3 to 20 fruits in a cluster only once a lifetime, and after the fruit has been consumed, different parts of the banana tree are not utilized, such as banana peels and stems, which are thus considered as waste (Baharin et al., 2016). This waste has a high quantity of many useful organic compounds, including cellulose, lignin, pectin substances, pigments, and chlorophyll, etc., in addition to low molecular weight organic compounds (Kandeeban and Malarkodi, 2019). Worldwide, following the consumption of bananas, million tons of peels are mostly discarded and are rarely utilized. In Malaysia, banana is ranked as the second most widely cultivated fruit, and it can be utilised either ripe or unripe.

1.1.1(b) Chemical water treatment

The chemical water treatment approach involves using of chemicals in an array of procedures to expedite purification or separation of the water pollutant. Chemical approaches consists of various reactions that eventually lead to neutralization and hydrolysing the water pollutants into harmless chemicals (Meghwal et al., 2020). Salimi and co-workers stated that coagulation and advanced oxidation process are the main chemical methods in water treatment processes, which also include ozonation and Fenton treatment (Salimi et al., 2017). These approaches are usually followed by photocatalysis and/or membrane techniques. Erickson and co-workers have used iron-enhanced sand filters for dissolved phosphorus removing from storm-water (Erickson et al., 2012). The authors reported that 88 % of phosphate was captured for at least 200 meters of targeted treated depth, compared with the conventional sand filtration technique.

Chlorination technique has been also used prior to powdered activated carbon filtration as two steps water treatment approach (Li et al., 2016). Advanced oxidation is a technique consist of using oxidizing agents for the oxidation of the pollutant (Wang and Zhuan, 2020). Various advantages have been reported to this technique such as rapid reaction rates compared to similar techniques, un-required of large area for processing the water in addition to less retention time (Marican and Durán-Lara, 2018). Vela et al. (Vela et al., 2019) reported the possibility of using $Na_2S_2O_8$ as oxidizing species in the removal of some pesticides with a removal efficiency reached 87 %. However, the complex structure of many types of pesticides, herbicide and toxins make their removal a complex process and may form other toxic compounds depending on their type, despite the time required for the reaction, as they are present in very tiny quantities.

1.1.1(c) Biological water treatment

The utilization of microorganism in water treatment has attracted tremendous attention in the past few years, and become a part water treatment approach. Two basic approaches have been reported to handle the used microorganism in water treatment process (Kamarudin et al., 2021); The first approach is preserving the microorganisms in liquid suspension, in which decomposing the pollutants is taking place by appropriate mixing methods. The second approach is attaching or immobilizing certain microorganisms on specific solid-material such as rock, slag or any similar material (Pal and Kumar, 2020).

Pandey and co-workers developed novel approach consist of two-stage anaerobic packed bed reactor (Pandey and Sarkar, 2017). The authors immobilized certain microorganisms, on porous polyvinyl alcohol-gel beads and reported high efficiency in COD removal of up to 89%. Activated sludge augmented by 2,4-dichlorophenol degrading bacteria has been used in different study, which was isolated from soil contaminated with 2,4-dichlorophenol and reported significant improvement in the removal efficiency (Quan et al., 2004).

1.1.2 Solar disinfection (SODIS) in water treatment applications

Disinfection of contaminated water using solar radiation (SODIS) is known to inactivate bacteria (Dessie et al., 2014). Conventional SODIS has been used in many isolated regions in Africa and south-east Asia as a household water treatment method (McGuigan et al., 2012). The utilization of solar power in water treatment has recently gained tremendous attention due to its accessibility, cost-effectiveness, and availability in most undeveloped countries. Despite the effectiveness and low costs of the most currently applied water treatment approaches such as ozonation, chlorination, and advanced oxidation processes they have major drawbacks including the generation of potential harmful by-products, decomposition of organic pollutants, and/or inactivation instead of killing of water pathogenic microorganisms (Aguilar et al., 2018; Nasuhoglu et al., 2018).

Different approaches have been used for solar disinfection, such as the filling of transparent polyethylene bottles with untreated water and then exposing them to sunlight for variable time, depending on the climate (Nalwanga et al., 2014). This simple procedure was found to be sufficient for killing the bacteria and other water microorganisms and considered a successful approach in different regions with illuminated periods such as Cameroon, India, Senegal, and South Africa (Helali et al., 2014). However, conventional SODIS technology has been associated with several drawbacks, including being a slow process, low disinfection performance and limited volume of water being processed. The efficiency of the process mainly depends on several factors including the initial level of water contamination, solar irradiance, water turbidity, time of day, and the atmospheric conditions (Azamzam et al., 2021). However, great advances have been made in this field to enhance efficiency and overcome such issues in the past few years including several modification techniques either for the water to be treated or for the solar rays.

1.2 Problem statement

Every living organism on earth, especially human being directly or indirectly requires clean water as an essential resource for its living. The use of plant-based treatment techniques has attracted great interest in the past few years due to their safety, sustainability and cost effective compared to chemical-based techniques (Owodunni and Ismail, 2021).

Many chemical coagulants and flocculants have been used in water treatment applications, such as iron and aluminium salts, in addition to some polymeric polysaccharides (Nath et al., 2021). Aluminium-based salts (such as aluminum sulfate and chloride) and ferric-based salts (such as ferrous sulphate and ferric chloride) are the most widely used metal saltwater treatment coagulants (Bahadori et al., 2013). Ferric chloride and aluminium sulphate are the most widely used metal-based coagulants that are characterized by their excellent performance in wastewater treatment; however, their use has the limitation of reducing the water pH to become close to acidic. In addition, they have been reported to cause some health issues to humans after the consumption of water, such as presenile dementia and Alzheimer’s disease (Gurumath and Suresh, 2019b). Another limitation of using metal salts as coagulant agents is the resulting large volume of sludge and the relative high coagulant cost (Kristianto, 2017). Thus, plant-based coagulants and flocculants are the suitable and safer alternatives for these chemicals.

Several attempts have been made to enhance the coagulation performance of banana peels by either mixing them with other coagulants or conducting chemical modifications to the peels. Fu and co-workers used chemical modification to produce oxidized banana peel in order to enhance the coagulation activity (Fu et al., 2019a). Chemical modification may lead to chemical leaching in treated water and cause further issues. In this study, A facile and affordable preparation approach was used to enhance the coagulation performance of banana peel as a natural coagulant. This approach consists of multiple microwave radiation treatments followed by multiple grinding to reduce the particle size and modify the surfaces of the particles without the need for any chemical reactions.

1.3 Hypothesis

Several banana peel-based materials have been recently used in water treatment applications including activated carbon, pseudo-graphite, biochars and carbon foam. In spite of these advances, eco-friendly and cost-effective preparation and modification of banana peel flocculant can be fabricated using microwave treatment to modify the surfaces of the bio-flocculant. Solar rays have the potential to disinfect water microorganisms, making it smart choice to overcome the limitation of plants-based flocculation that associated with increasing the number of microorganisms. This hypothesis is using microwave treatment to modify banana peel flocculant and enhance its performance in water treatment application and couple solar disinfection to overcome increasing the number of microorganisms.

1.4 Objectives of the study

The aim of this study is to enhance the performance of banana peels flocculant as one of the most common wastes in Malaysia in water treatment applications and couple it with solar for disinfection purpose. However, in order to achieve this, the research has several specific objectives including:

1- To prepare and characterize banana peels bio-flocculation for water treatment applications.

2- To modify banana peels bio-flocculation using microwave treatment and study the flocculation mechanism of banana peels bio-flocculation.

3- To determine the effect of water pre-treatment using banana peels bio-flocculation on enhancing solar disinfection (SODIS) performance in the removal of *facal coliform* microorganisms.

1.5 Scope of the study

The main focus of the present research is to utilize banana peel waste in the preparation of natural flocculant and enhance its performance in water treatment by using microwave treatment. The flocculation process in this study focussed solely on the water treatment process; modified bio-flocculant was compared with non-modified one to prove the effectiveness of the modification. The study also includes fully characterization of the prepared flocculants including particle size analysis, zeta potential study, pectin content and SEM analysis for its surface morphology to investigate of the potential flocculation mechanism of banana peel in water treatment.

Jar test was used for studying the flocculation process using different parameters including contact time, pH, dosage, initial turbidity and setting time rates for both the modified and non-modified bio-flocculants. Besides that, this study also includes coupling solar disinfection with bio-flocculation study to solve the issue of increasing the number of microorganisms. Different parameters including initial bacterial number, different water depth, turbidity levels, total suspended solids and duration time were investigated for studying the potential of SODIS in water disinfection. Finally, coupling banana peel bio-flocculants as water pre-treatment with SODIS was investigated to provide fully water treatment approach.

1.6 Significant and limitations of the study

The use of natural products in water treatment applications are widely used since ancient times, which have proved their ability in water treatment, while retaining their natural benefits (Nandini and Sheba, 2016). Recently, numerous studies on the use of plant-based materials as coagulants have been reported (Danish et al., 2018; Mouiya et al., 2019). Banana wastes such as peels, leaves and even stems are not giving poisonous effect to the water. Most of them are wastes which have not been utilized. But they are readily available, eco-friendly, and they can be a reliable technique for wastewater treatment.

This study is important as it develops a more systematic system to overcome the shortcomings of the conventional bio-coagulants and flocculants. The performance of raw (non-modified) banana peel bio-flocculant wasn't sufficient in both synthetic and river water treatment. In this study, microwave treatment significantly enhanced the flocculation performance of banana peel in the removal of water turbidity. The whole treatment approach is eco-friendly and doesn't include any chemical usage. This study also eases the determination of the optimal time, pH level and dosage of flocculation process for water treatment.

This study also utilizes SODIS as a sustainable source to overcome the issue associated with the conventional coagulation and flocculation processes in terms of increasing the number of microorganisms. It eases the determination of optimum duration time, pottle size and initial bacterial number in solar water disinfection. Finally, this study establishes sustainable water treatment approach by coupling banana peel bio-flocculant and solar disinfection; by pre-treating the water by banana peel bio-flocculant and then disinfect the treated water with solar, providing an eco-friendly and sustainable water treatment.

1.7 Organization of thesis

This thesis basically consists of five chapters. The first chapter include an overall background and the scope of the research, a simplified picture of water treatment techniques and the advantages of using the natural coagulant in this field. It also includes the problem statement and the justification of using each approach and banana peel materials including natural coagulant, activated carbon in both the conventional and modified ones, in addition to the use of solar for water disinfection.

Chapter one also includes the objectives of this study addressing the generalized aim and the three specific objectives. Chapter two addresses a review of literatures and previous research that have been conducted in each field of this research starting from water treatment techniques, and ending with the specific used approaches and materials. The materials and methods that have been used in this research are addressed in details in chapter 3, explanation of the preparation, modification and characterization approaches of each material.

The fourth chapter talk and presents the results of each experiment, with a critical discussion of presented results and comparison the obtained results with previous works. Finally, chapter five presents the conclusions reached by the present study from each objective and the recommendations for future research in the area of study.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Access to safe, microbial-free drinking water is essential to health, which has been always a major concern as being one of the basic human health fundamental rights worldwide. More than 2 billion people around the world are lacking to proper sanitation and hygiene in term of drinking water (Cowie et al., 2020). Treatment techniques for drinking water in many undeveloped and developing countries are either insufficient or inaccessible, and thus, millions are lacking the access to safe water service. However, it has been reported that roughly 144 million mainly rely on surface water as the main source for drinking, cooking and daily usage, resulting in half million death annually, caused by diarrheal alone apart from other waterborne diseases (Pichel et al., 2019).

In many urban areas, the governments supply households with clean water, after intensive treatment with a combination of traditional treatment techniques including chlorination, filtration, ultraviolet irradiation (UV irradiation), ozonation, flocculation or Fenton and photo-Fenton approach, etc. However, with the crazy increase in world population, these methods may not adequately address the requirement of all the households from drinking water, especially in the poor villages. The past few years witnessed the development of many cost-effective and proper water treatment technologies, using sustainable and eco-friendly principals instead of chemical treatment processes (McGuigan et al., 2012).

The utilization of solar in water treatment has recently gained tremendous attention due to its accessibility and the availability in most of undeveloped countries. Despite the effectiveness and low costs of the most currently applied water treatment approaches such as ozonation, chlorination and advanced oxidation processes they have major drawbacks including the generation of potential harmful by-products, decomposition of organic pollutants and/or inactivation instead of killing of water pathogenic microorganisms (Aguilar et al., 2018; Levchuk et al., 2015; Nasuhoglu et al., 2018).

2.2 Coagulation and flocculation in water treatment

Coagulation and flocculation are two widely used processes for wastewater treatment due to their efficiency and simplicity (Bratby, 2016; Kim, 2016). Coagulants and flocculants are chemicals used in the water treatment process to remove impurities and clarify water. Coagulants initiate the process by neutralizing charges on particles, allowing them to come closer together, while flocculants further aid in the clumping of these particles into larger flocs, making it easier to remove them from the water.

The colloidal material in the wastewater in coagulation and flocculation processes is being destabilize by adding the coagulants and/or flocculants, causing the agglomeration of small particles into larger settleable aggregates and can be removed effortlessly (Lapointe et al., 2020). These processes have been extensively used in water pre-treatment prior to biological treatment, to reduce water turbidity and to enhance the biodegradability of the wastewater during the biological treatment (Singh and Kumar, 2020). Coagulation and flocculation also are proven techniques when they used with sedimentation process for the treatment of high suspended solids wastewater especially those formed by colloidal matters (Bratby, 2016; Kim, 2016). These processes require the addition of certain compounds known as coagulants and flocculants for coagulation and flocculation respectively, which can be either natural or chemicals (Kurniawan et al., 2020).

2.2.1 Inorganic coagulants and flocculants

Negatively charged polymers are widely used in wastewater treatment, notably as high molecular weight flocculants (Lapointe and Barbeau, 2020). The chemical compounds commonly used as coagulants and flocculants include iron salts ($FeCl_3$ or $Fe_2(SO_4)_3$) (Moran, 2018b), aluminum salts ($Al_2(SO_4)_3$) (Aragonés-Beltrán et al., 2009), hydrated lime, magnesium carbonate (Moran, 2018b), and synthetic polymers (aluminum chlorohydrate, polyaluminum chloride (PAC), polyaluminum sulfate chloride, and polyferric sulfate) (Weydts et al., 2015). Some of the mentioned compounds have been shown to be effective in reducing suspended and even dissolved solid concentrations in water (Nath et al., 2020).

Most of the polymeric coagulants and flocculants are positively charged due to the presence of charged function groups on their surfaces (Rizal et al., 2020). Positively charged groups are called cationic polymer coagulant, negatively charged polymers are called anionic polymer coagulant. In contrast, the two or more kind of polymers is called polyelectrolytes coagulant and/or flocculants. Polyelectrolytes are a class of polymers that contain ionizable groups along their molecular chains. These ionizable groups can be either positively charged (cationic) or negatively charged (anionic). Polyelectrolytes, including both coagulants and flocculants, play crucial roles in water and wastewater treatment processes (Yousefi et al., 2020). These specialized polymers help in the removal of suspended particles and impurities from water, making it safe for consumption or suitable for discharge into the environment. The presence of the cationic charge in in some chemical compounds (such as amino groups) indorse the polymer in the efficient absorption of various metal ions (Alsharari et al., 2018). Chemical coagulation and flocculation are complex phenomena involving various interrelated parameters. They can be categorised in the three parts as shown in Figure 2.1.

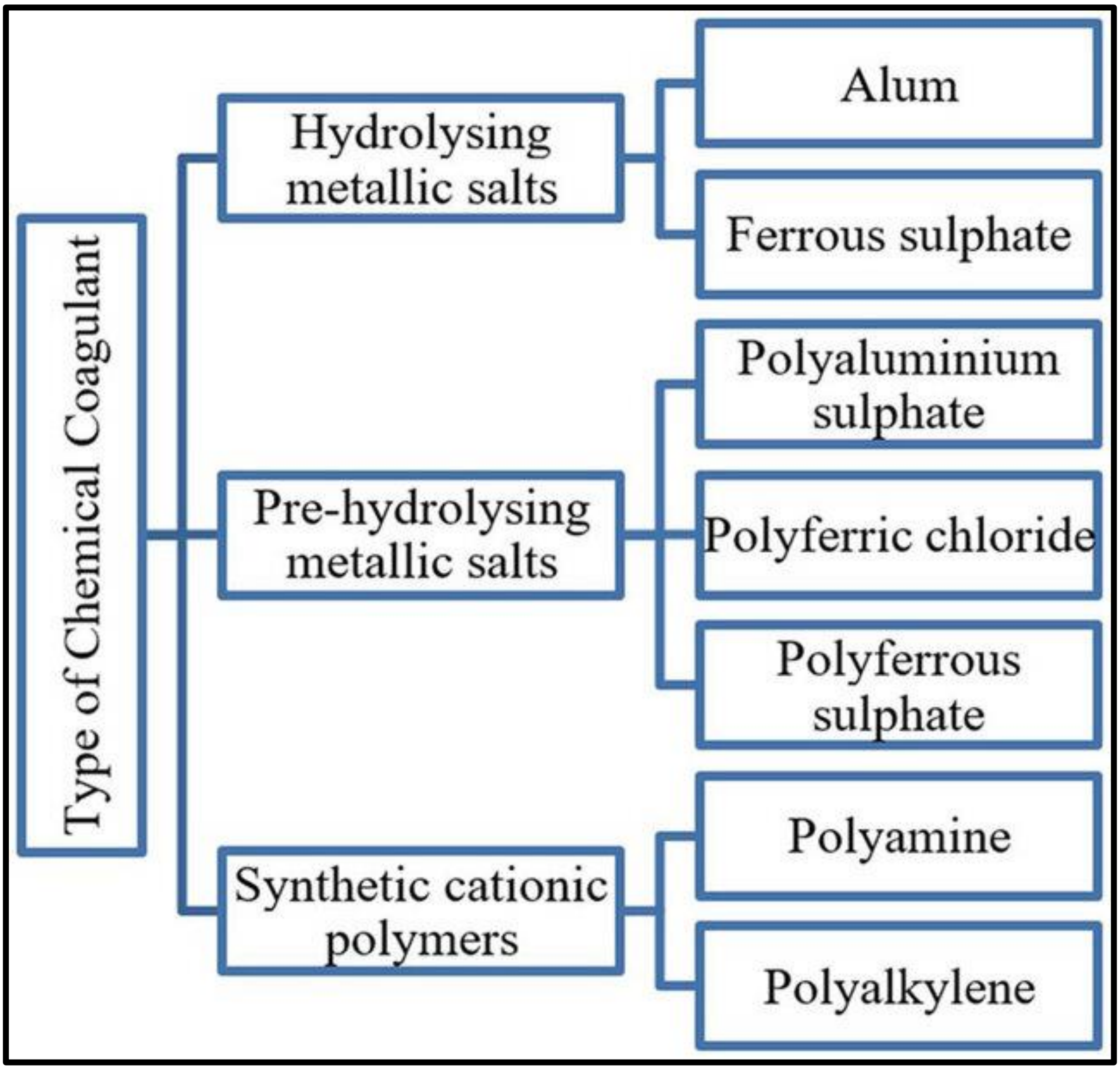

Figure 2.1 Categorisation of chemical coagulants according to their effectiveness. Adapted with permission from (Mohamad et al., 2023)

Metal salts and pre-hydrolyzed coagulant have also been used as primary wastewater treatment (Theodoro et al., 2013). Aluminum based salts and ferric based salts are the most widely used metal saltwater treatment coagulants (Bahadori et al., 2013). Lately, the application of ferric-based coagulants is more preferably compared to aluminium-based coagulant that has been linked with many health risks. Ferric chloride and aluminium sulphate have excellent performance as wastewater treatment agents.

Poly-ferric chloride, poly-ferrous sulphate and poly-aluminium chloride the most used pre-hydrolyzed coagulant. However, the use of these material has the limitation of reducing water pH to become close to acidic. Also, they have been reported to cause some health issues to humans after the consumption of water, such as presenile dementia and Alzheimer’s disease (Gurumath and Suresh, 2019a). Another limitation of using metal salts as coagulant agents is the resulted large volume of sludge, and the relative high coagulant cost (Kristianto, 2017). In a different study, (Shi et al., 2004) have reported that using each of ferric salt and poly-ferrous sulphate as coagulants could accelerate pipe corrosion. Table 2.1 presents an illustration of various non-plant-based coagulants applied for water treatment.

Table 2.1. Inorganic-based coagulants and their applications

Coagulant	Applications	Reference
Titanium salts	Tanium Removal of dissolved organic matter, seawater reverse osmosis pre-treatment, wastewater sludge dewatering, and algae-laden surface water treatment	(Chi et al., 2019; Hussain et al., 2019)
polymeric zinc–ferric–silicate–sulfate	Treatment of humic acid/ algae polluted water, and high-concentration oily wastewater treatment	(Liao et al., 2017; Sun et al., 2017)
Ferric salts	Removal of organic matter, turbidity reduction, and water treatment sludge dewatering	(Chua et al., 2020; Wan et al., 2019)
poly-ferric-titanium-silicate-sulfate	Disperse and reactive dye wastewaters treatment	(Huang et al., 2020)
Aluminum salts	Wastewater treatment, removal of organic matter, and turbidity removal	(Mazloomi et al., 2019; Wan et al., 2019)

Titanium-Based Xerogel	Wastewater treatment of low turbidity, and removal of cyanobacteria	(Wang et al., 2018c)

2.2.2 Natural coagulants and flocculants

Natural or plants-based coagulants and flocculants are group of plants (or their compounds) have been investigated with immense potential in water treatment applications (Wang et al., 2020). Water treatment have been defined as the removal of all the suspended and colloidal materials and particles in water such as organic matter, microbes, and inorganic matters (Jayalakshmi et al., 2017). Various plant-based materials have been used in many parts in the world such as China, India and Africa (Asrafuzzaman et al., 2011; Kristianto, 2017). Due to the health, costs and environmental aspects of many inorganics and/or synthetic compounds, numerous studies have been developed in recent years aiming to search for a sustainable, eco-friendly, and non-toxic alternative to inorganic coagulants and flocculants for water treatment purposes (Barros et al., 2022).

The use of plant-based coagulants and flocculants for water treatment gained more interest, as they are natural, cost-effective, and renewable (Gholami et al., 2022). Table 2.2 presents few examples of commonly used plants for water treatment. Plant-based coagulants and flocculants from different parts of plants have been utilized in water treatment; including roots, stems, fruits, fruit shells, leaves and even seeds. However, in some plants, such as banana and Moringa, many parts of the plant tree have been utilized as natural coagulants.

Table 2.2. Organic-based coagulants and their applications

Part of plant	Treatment applications	Optimum result	Reference

Rice starch	Treatment of palm oil mill	Removing of up to 88.4% TSS at the small dosage of 0.55 g/L	(Teh et al., 2014)
Roselle seeds	Treatment of wastewater containing Congo red dye	Removing of up to 91.2 % of colour at 190 mg/L coagulant dosage at 400 ppm of dye concentration	(Yong and Ismail, 2016)
Moringa oleifera seeds	Cyanobacteria and natural organic matter (OM) treatment	Removing 80% of chlorophyll a, 80–90% of dissolved OM, and 80% of cyanobacteria cells	(Teixeira et al., 2017)
Corchorus olitorius L.	Treatment of humic acids wastewater	Removing of up to 95 % of turbidity and 100% of total organic carbon.	(Altaher et al., 2016)
Basil (Ocimum basilicum)	Treatment of Landfill leachate	When combined with alum was able to reduce 64.4% of COD and 77.4% of watercolour.	(Rasool et al., 2016)
Corn starch	Treatment of kaolin and microorganisms	At 0.5 mg/L dose was able to remove up to 98% of kaolin, E. coli and S. aureus.	(Liu et al., 2017)
Orange peel	Dairy wastewater treatment	At only 0.2 g/L was able to remove up to 97% of turbidity.	(Anju and Mophin-Kani, 2016)
Jackfruit	Treatment of kaolin	60 mg/L dose was able to reduce turbidity by 43%.	(Choy et al., 2017)

The use of plant-based coagulant such as moringa and flocculants such as banana peels in water treatment has many advantages over chemical coagulant; the possibilities of residue coagulant make it safer and preferable than chemical coagulant, which have been linked to many diseases such as Alzheimer if consumed (Garde et al., 2017). Another advantage is the cost, a plant-based coagulant is sustainable, and cheaper compared to chemical coagulant, as most of the chemical coagulant require other material aid to effectively treat high turbidity, which raises the cost of the treatment process and make it difficult to be used in developing countries (Antov et al., 2010). However, a significant increase in water organic material is one of the top disadvantages of using a plant-based coagulant, which results in accumulation of microbial activity. This issue has been solved by the addition of chlorine in safe doses to sanitize the treated water (Amran et al., 2018). The sedimentation time is another limitation can be mentioned regarding plants-based coagulants which requires more time compared to chemical coagulants (Kumar et al., 2017).

2.2.3 Limitations and challenges of natural coagulants and flocculants

Although, the use of natural coagulants and flocculants for water purification is simple, safe for human health, eco-friendly and effective, but the efficiency and performance of many of them may not be acceptable in term of water quality compared with chemical coagulants and flocculants. For this reason, the processes of flocculation using natural resources mostly require the addition of enhanced compounds to enhance the overall charges of the materials. These enhancing compounds in most of causes are chemical such as iron salts ($FeCl_3$ or $Fe_2(SO_4)^3$) (Moran, 2018a), aluminum salts ($Al_2(SO_4)^3$) (Aragonés-Beltrán et al., 2009), hydrated lime (Kurniawan et al., 2020), magnesium carbonate (Moran, 2018a), and polymers (aluminum chlorohydrate, polyaluminum chloride (Armstrong and LeHew, 2011), and polyferric sulfate) (Armstrong and LeHew, 2011).

Despite the significant improvement that these compounds can do to the natural coagulants and flocculants, they are associated with several environmental problems (Barakwan et al., 2019) due to their chronic toxicity, specifically for environmental observers worldwide (Imron et al., 2020). Several studies have been done to enhance the coagulation and flocculation performance of cheap and available plant waste to enhance their performance in water treatment. Natural coagulants and flocculants are also associated with sludge formation even in tiny amounts, which suspend together, fall, and gather at the bottom of the tank (Kurniawan et al., 2020). The impact of sludge formation on microorganisms depends on various factors such as the composition of the sludge, treatment processes, environmental conditions, and the presence of toxic substances. Sludge provides a habitat for various microorganisms, including bacteria, fungi, and protozoa. Microorganisms adhere to the sludge particles and form biofilms, which can contain diverse microbial communities (Zhao et al., 2010).This phenomenon may lead to significant increase in the number of microorganisms in the treated water. Using polysaccharides-based coagulants and/or flocculants such as banana peels may lead to partially dissolving of these molecules in water, especially if they were in fine powder form, which make disinfection stage required in the water treatment process.

2.2.4 Mechanisms of coagulation and flocculation processes

Coagulation is a chemical process widely used in water treatment applications that involves particle charge neutralization (Yin, 2010). A coagulant (organic or inorganic) is a compound with mostly positive charges (divalent) that able to interact with suspended particles in the wastewater and create a neutral form of combined compounds (Okaiyeto et al., 2016). This process usually followed by another process known as flocculation and then setting stages as presented in Figure 2.2.

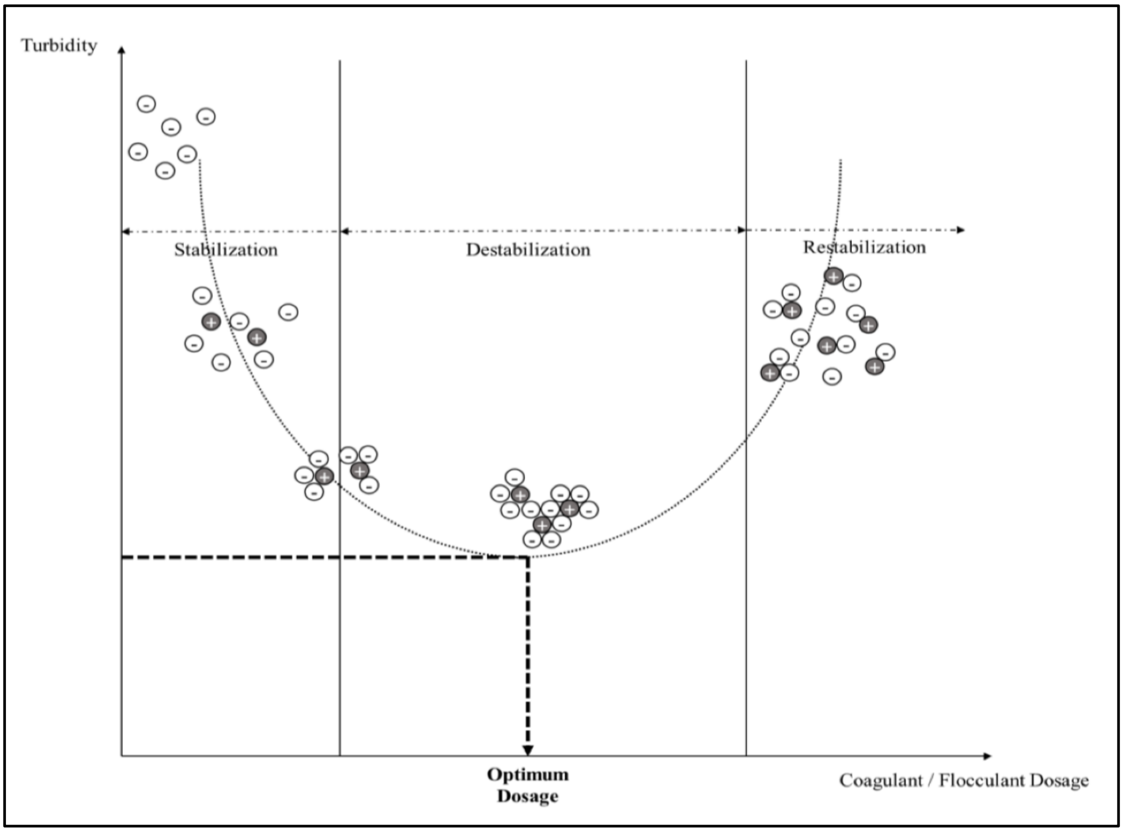

Figure 2.2 Phases through the coagulation–flocculation process. Adapted with permission from (Kurniawan et al., 2020)

In order for these processes to happened, they require a certain mixing speed, dosage, time and intensity, which can accelerate the rate of particle collision (Ahmed et al., 2015). However, the collision and accumulation of flocs within the wastewater will form an agglomeration of particles as a result of neutralization process that have a high settling velocity (Yunos et al., 2017). Four mechanisms are known for coagulation (Figure 2.3), including polymer bridging, sweep coagulation, double layer compression, and charge neutralization (Bolto and Gregory, 2007).

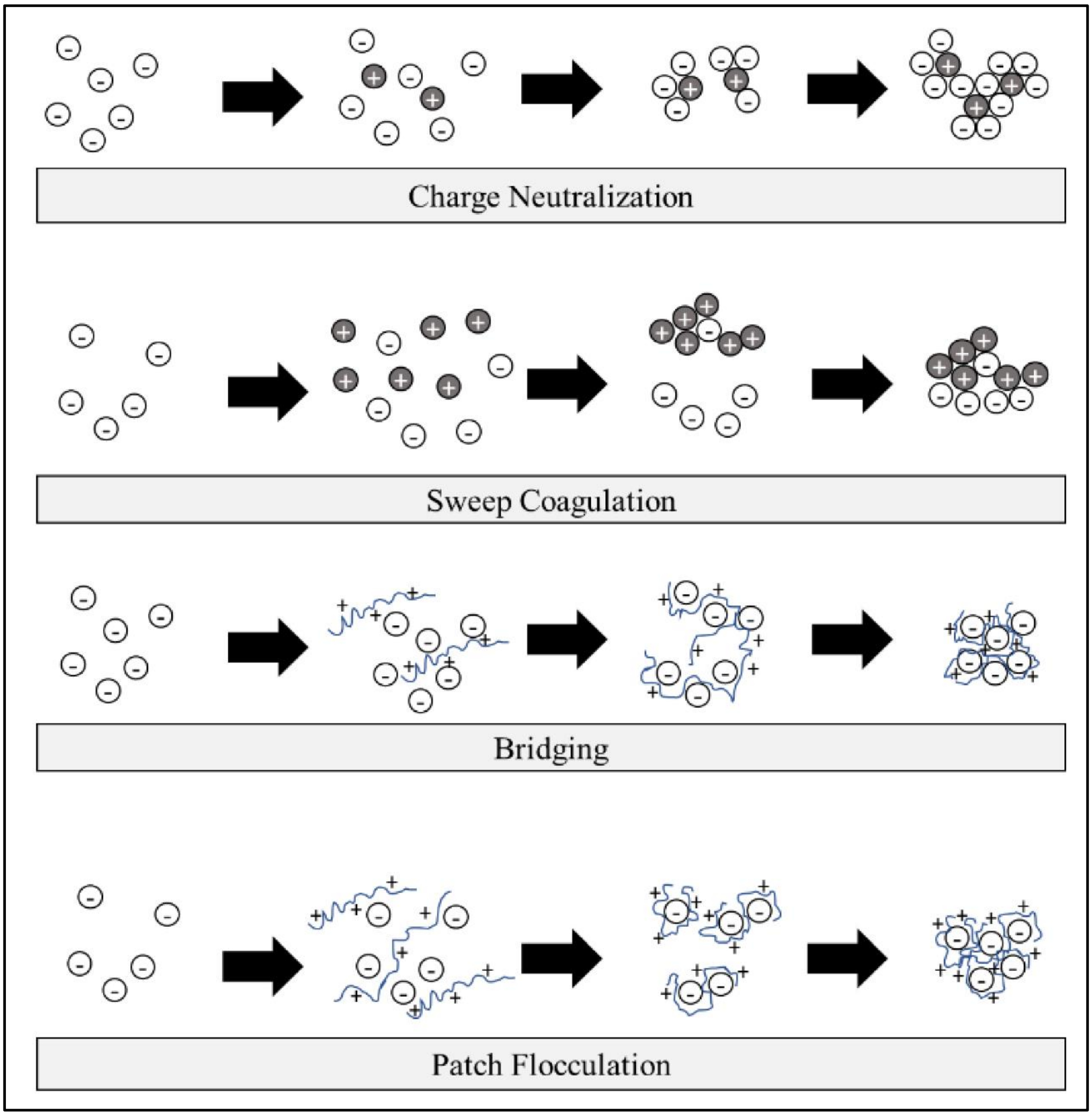

Figure 2.3 The four suggested mechanisms of coagulation and flocculation. Adapted with permission from (Kurniawan et al., 2020)

Many polysaccharides such as cellulose, pectin, starch, gelatine, alginate and chitosan have been investigated as a natural coagulant due to their biosafety to human, unlike the chemical-based coagulants (Nath et al., 2020). The coagulation/ flocculation mechanism of these polysaccharides involves charge neutralization and polymers bridging (Nath et al., 2020). In charge neutralization the positively charged coagulant attract the negative charged pollutant particles, which adsorbed on its surface leading to neutralizing of the colloid particle charges.

The significant decrease of electrostatic repulsion that presents in the colloid particles lead to the reduction of particles surface charges, which make the coagulation possible, as figure 2.4 presents (Henderson et al., 2008). In polymer bridging mechanisms, the adsorption of the particles is taking place in long-chain of linear and high molecular weight polymer and leaving dangling heavy coagulant polymer segments to bridge all the particles of the pollutants together (Diddens and Heuer, 2019).

Sweep coagulation is another mechanism occurs with the help of chemical coagulants (i.e; metal salts), which are usually added to the water in higher dosages than the solubility of the amorphous hydroxides, and the colloid particles eventually entrapped within the precipitate and then removed from the suspension (Nan et al., 2016). The solution with high electrolytes occurs in double-layer compression. The colloids become unstable and increase the possibilities of coagulation, lowering the colloid particles repulsive force in the solution occur due to the compression in the volume that happens due to the high ionic concentration (Hsiung et al., 2016).