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WATER TREATMENT USING NATURAL COAGULANTS: A REVIEW ON THE POTENTIAL UTILISATION OF BANANA WASTE

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Abstract: The massive industrial and agricultural development in the past few years has increased the pollution level of water bodies. Several studies have concluded that the global depletion of freshwater resources will result in difficulties accessing clean water. Plant-based water treatment techniques have attracted great interest in the past few years due to their safety and cost-effectiveness compared with chemical-based techniques. Natural coagulants have been extensively studied in terms of the type of plant and the mechanism of coagulation. Banana is one of the most famous tropical fruits from the Musa genus in the Musaceae family. It is widely consumed in Malaysia, especially Musa acuminata, Musa balbisiana, and Musa paradisiaca, resulting in tremendous amounts of biomass residue, including peels, stems, and leaves, with high potential use for wastewater treatment applications. This review aims to highlight the advantages of natural coagulants and to discuss the potential use of different banana wastes in water treatment applications.

Keywords: *Plant-based coagulant, banana wastes, water treatment, natural coagulant, waste biomass*

1. Introduction

Since the last century, with a dramatic increase during the Industrial Revolution, many industrial wastes have increased proportionally, increasing a worldwide primary source of severe pollution (Mohan et al., 2019). Air, water, and even soil have been polluted by anthropogenic activities. For instance, water pollution may occur due to the use of multiple chemical reagents, ranging from inorganic compounds to polymers and even organic products (Salmasi et al., 2020). Many developing societies lack appropriate wastewater treatment techniques, leaving waste without treatment. Many of these communities consume non-treated or badly treated water daily, which eventually affects their health and leads to severe waterborne disease (Ravindra et al., 2019).

Coagulation is a water treatment technique used to assist in colloidal particle removal (Lv et al., 2018), lime softening (Ghernaout et al., 2018), water clarification (López et al., 2021), sludge thickening (Atamaleki et al., 2020), and solid dewatering (Feng et al., 2022). Due to the potential cause of health problems, the use of chemical coagulants, such as alum, is not a preferable option (Bahrodin et al., 2021). Its use is restricted to turbidity removal, and it is not recommended for use in developing countries (Sulaiman et al., 2017). The use of plant-based materials as natural coagulants for water purification is simple, safe for human health, eco-friendly, and effective.

In a previous study by Nath et al. (2021), the authors showed that several chemical coagulants have the ability to change the physicochemical properties of treated water. The same authors encouraged the use of natural coagulants to replace chemical ones. Several studies have concluded that natural coagulants show a significant improvement in the environment and ecosystem as a sustainable solution to wastewater treatment issues (Mumbi et al., 2018). The use of natural coagulants has been practiced since ancient times and has been proven in water treatment, while retaining natural benefits (Nandini and Sheba (2016).

Banana is one of the most consumed tropical fruits in Malaysia and many tropical countries (Soluri, 2021). The banana tree has been reported to produce around 3 to 20 fruits in a single cluster once in its lifetime. However, after consumption, this tree leaves a large amount of biomass that can be used for several applications. In our previous research, we investigated the potential use of banana peels in river water treatment applications and found great potential for such waste as a natural coagulant (Azamzam et al., 2022). Worldwide, the volume of bananas produced in 2020 has reached approximately 119.83 million tonnes, which increased from 117.53 million tonnes in 2019 (Duraiprasanth et al., 2022). Therefore, one million tonnes of peels are mostly discarded and are rarely utilised. It has been reported that Malaysians consume bananas, either ripe (fried banana) or unripe (fresh fruits), for making chips and juice (Aida et al., 2016). However, it has been reported that chip and juice factories generate tonnes of banana peel waste every year, which, in most cases, is not utilised and is dumped in landfills (Ahmad and Danish (2018). These wastes have a high quantity of beneficial organic compounds, including cellulose, lignin, pectin

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substances, pigments, and chlorophyll (Kandeeban and Malarkodi (2019). Banana stem juice has promising potential for use as a natural coagulant in water treatment (Hilal et al., 2004). İn the past few years, many review articles have been published discussing natural coagulants (Nath et al., 2020), polymeric coagulants (Nath et al., 2020), and plant-based coagulants (Choy et al., 2015). Several studies have been conducted on the potential use of banana wastes in water treatment applications, but they have never been reviewed. The present review attempts to deliver collective information about the potential use of banana waste materials as natural coagulants for water treatment. It also briefly discusses the advantages, disadvantages, and mechanisms of natural-based coagulants in water treatment applications.

2. Coagulation Treatment Method

Coagulation is an extensively used method for wastewater treatment that reduces turbidity and removes suspended colloidal particles. This method is usually performed using a sizeable chemical reactor that enters the basin and influences wastewater. The wastewater is then homogenised with a suitable coagulant agent and mechanically mixed until the sedimentation process takes place. Eventually, gravity settling is performed to remove the particulate matter (Amran et al., 2018). Concerted research and development efforts have been conducted in the past two decades to discover new plant species and constituents that can be used as natural coagulants, further boosting the effectiveness of existing plant-based natural coagulants (Liao et al., 2017). The coagulation process is a physicochemical process that reduces the repulsive potential of an electrical double layer of colloids using various coagulants. This will lead to the agglomeration and development of colloidal microparticles into larger particles or flocs (Mazloomi et al., 2019). This agglomeration can be formed by several mechanisms, including polymer bridging, charge neutralisation, and sweep coagulation, as discussed in the following section.

Coagulation Mechanisms

The coagulation process of both natural and synthetic coagulants can be performed using several mechanisms, including polymer bridging, sweep coagulation, double layer compression, and charge neutralisation (Bolto and Gregory (2007) as presented in Figure 1. Naceradska et al. (2019) investigated the removal mechanism of algal organic matter using Jar tests with either aluminium sulphate or polyaluminium chloride. The authors reported that high-molecular weight organic matter, such as saccharides, was more amenable to coagulation than lower weight compounds. The low surface charge of the removed fraction indicated that the prevailing coagulation mechanism was the adsorption of non-proteinaceous matter onto aluminium hydroxide precipitates. In comparison, Adeleke et al. (2021) showed different mechanisms for the natural coagulant *Moringa oleifera* and reported that the amino acid residues in *Moringa* had certain interactions with pollutant ligands, indicating that coagulation may occur.

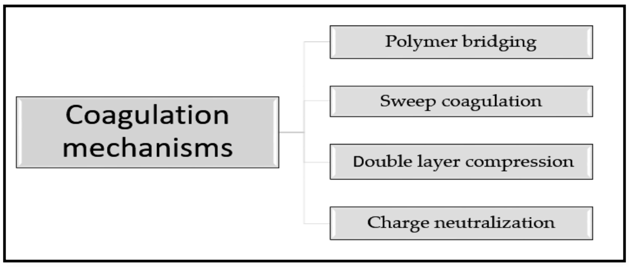

Figure 1. Diverse types of coagulation mechanisms. Reprinted from Bolto and Gregory (Bolto et al., 2007), with permission from Elsevier.

Many polysaccharides, such as cellulose, starch, gelatine, alginate, and chitosan, have been investigated as natural coagulants due to their biosafety to humans (Nath et al., 2020). The coagulation/ flocculation mechanism of these polysaccharides involves charge neutralisation and polymer bridging (Nath et al., 2020). In charge neutralisation, the positively charged coagulant attracts the negatively charged pollutant particles, which are adsorbed on its surface, neutralising the colloid particle charges. As shown in Figure 2, the surface charge difference between the pollutant and the coagulant leads to a decrease in electrostatic repulsion in the colloid particles, which then results in the attraction of the particles and coagulation (Henderson et al., 2008). In polymer bridging mechanisms, the adsorption of the particles takes place in a long chain of linear and high molecular weight polymers, leaving dangling heavy coagulant polymer segments to bridge all the particles of the pollutants together, as presented in Figure 2b (Diddens and Heuer (2019). Sweep coagulation is another mechanism that occurs in the presence of chemical coagulants (i.e., metal salts), which are usually added to the water at higher dosages than the solubility of the amorphous hydroxides, and the colloid particles eventually become entrapped within the precipitate and are removed from the suspension (Nan et al., 2016). High electrolyte concentrations in colloidal solutions cause double-layer compression. The colloids become unstable and increase the possibility of coagulation by lowering the colloid particles' repulsive force (Kristianto, 2017).

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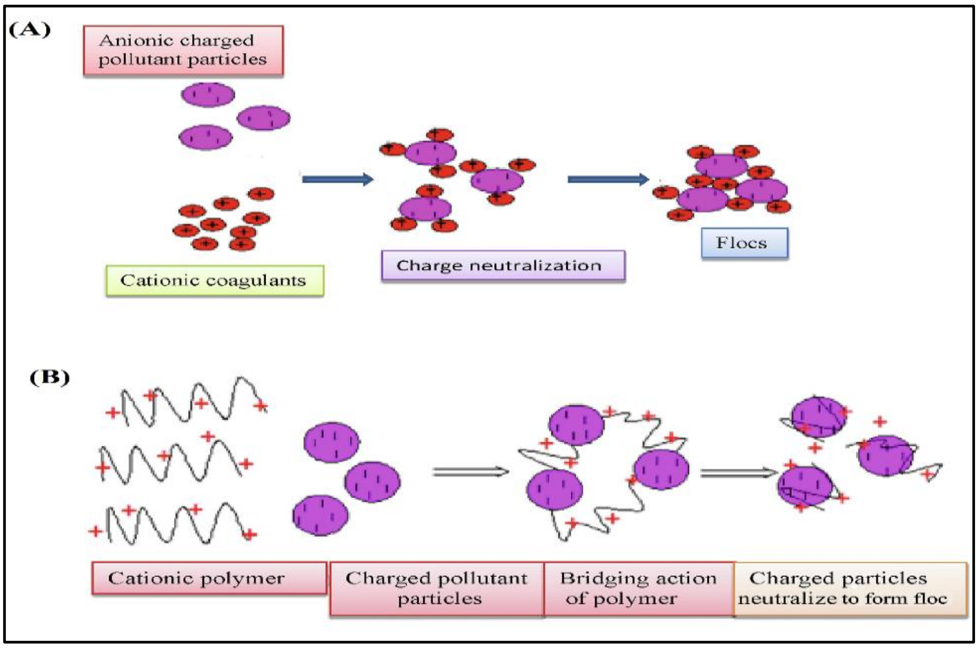

Figure 2. Coagulation mechanisms: (A) charge neutralisation and (B) polymer bridging. Reprinted from Nath et al. (2020) with permission from Elsevier.

The efficiency of coagulants in water treatment depends on the type of coagulants, their quantity, mixing conditions, and pH, as well as the properties of the solution to be treated, such as particle size, particle charges, the presence of divalent cations, the hydrophobicity level of the particles, and destabilising anions, such as sulphate ions, bicarbonate, and chloride (Sillanpää et al., 2018).

Synthetic and Inorganic Coagulants

Many synthetic and inorganic coagulants have been used in water treatment applications, such as metal salts and polymeric polysaccharides from a non-plant source, such as chitosan (Nath et al., 2020). Most polymeric coagulants 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 coagulants, and negatively charged polymers are called anionic polymer coagulants. In contrast, a mixture of two or more types of polymers is called a polyelectrolyte coagulant. Chitosan, for example, is one of the most frequently used polymers in different applications, including absorption. The presence of the cationic charge in their structure (i.e., amino groups) enables the polymer to efficiently absorb various metal ions (Alsharari et al., 2018). Metal salts and pre-hydrolysed coagulants have also been used in primary wastewater treatment (Theodoro et al., 2013).

Aluminium-based salts (such as aluminium sulphate 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). Lately, the application of ferric-based coagulants is preferable compared to aluminium-based coagulants, which have been linked to 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 are the most commonly used pre-hydrolysed coagulants. However, the use of these materials has the limitation of reducing the water pH to become close to acidic. Additionally, they have been reported to cause some health issues in humans after the consumption of water, such as presenile dementia and Alzheimer's disease (Gurumath and Suresh (2019). Another limitation of using metal salts as coagulant agents is the resulting large volume of sludge and the relatively high coagulant cost (Kristianto, 2017). Shi et al. (2004) reported that using ferric salt and poly-ferrous sulphate as coagulants could accelerate pipe corrosion. Table 1 presents an illustration of various non-plant-based coagulants applied for water treatment.

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Table 1. Applications of synthetic and inorganic coagulants in water treatment applications

Coagulant	Type of pollutant	Type of water	Reference
Titanium salts	Sludge dewatering and algae-laden	Seawater and wastewater	(Shrestha et al., 2017; Zhang et al., 2017)
Titanium (III) chloride	Dissolved organic matter	Surface water	(Hussain et al., 2019)
polymeric zinc–ferric–silicate–sulphate	Humic acid, algae and oils	Wastewater	(Liao et al., 2017; Sun et al., 2017)
Ferric salts	Organic matter, sludge, and turbidity	Wastewater and drinking water	(Chua et al., 2020; Mazaheri et al., 2018)
Poly-ferric-titanium-silicate-sulphate	Organic dyes	Disperse and reactive dye wastewaters treatment	(Huang et al., 2020)
Aluminium salts	Organic matter and turbidity	Wastewater	(Mazloomi et al., 2019; Wan et al., 2019)
Titanium-Based Xerogel	Turbidity and cyanobacteria	Wastewater treatment of	(Wang et al., 2016; Wang, Wang et al., 2018)

Natural-Based Coagulants

Natural coagulants have been investigated with immense potential in water treatment applications (Ang and Mohammad (2020). The water treatment process removes suspended and colloidal materials and particles in water, such as organic matter, microbes, and inorganic matter (Jayalakshmi et al., 2017). Various plant-based materials have been used in many parts of the world, such as China, India, and even Africa (Asrafuzzaman et al., 2011; Kristianto, 2017). Due to the health, costs, and environmental aspects of many inorganic and synthetic coagulants, numerous studies have recently been conducted to search for a sustainable, eco-friendly, and non-toxic alternative to inorganic coagulants for water treatment purposes. The use of plant-based coagulants for water treatment has gained more interest as a natural, cost-effective, and renewable method that has been widely studied in the past few decades.

Plant-based coagulants have become more popular in recent research as an alternative and safer material to chemical and synthetic-based coagulants, especially in drinking water treatment. Table 2 presents a few examples of commonly used plants for water treatment. Plant-based coagulants are non-toxic, lower cost, safe, biodegradable, available, and sustainable; plant-based coagulants from different parts of plants have been utilised in water treatment, including roots, stems, fruits, fruit shells, leaves, and even seeds. In some plants, such as banana and *Moringa*, many parts of the plant tree have been utilised as natural coagulants.

Table 2. Plant-based coagulants and their applications.

Plants and their used parts	Treatment applications	Optimum result	Reference
Oryza sativa Rice starch	Treatment of palm oil mill	Removing up to 88.4% TSS at the small dosage of 0.55 g/L	(Teh et al., 2014)
Hibiscus sabdariffa Roselle seeds	Treatment of wastewater containing Congo red dye	Removing up to 91.2% of colour at 190 mg/L coagulant dosage at a 400 ppm dye concentration	(Yong and Ismail (2016)
Moringa oleifera Moringa 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. Nalta jute	Treatment of humic acid wastewater	Removing up to 95% turbidity and 100% of total organic carbon	(Altaher et al., 2016)
Ocimum basilicum Basil	Treatment of Landfill leachate	When combined with alum, it was able to reduce 64.4% of COD and 77.4% of water colour	(Rasool et al., 2016)
Zea mays Cornstarch	Treatment of kaolin and microorganisms	At a 0.5 mg/L dose, it was able to remove up to 98% of kaolin, *E. coli*, and *S. aureus*	(Liu et al., 2017)
Citrus sinensis Orange peel	Dairy wastewater treatment	At only 0.2 g/L, it was able to remove up to 97% turbidity	(Anju and Mophin-Kani (2016)
Artocarpus heterophyllus Jackfruit	Treatment of kaolin	60 mg/L dose was able to reduce turbidity by 43%	(Choy et al., 2017)

Abbreviations: TSS, Total suspended Solids; COD, Chemical Oxygen Demand

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A plant-based coagulant is sustainable and cheaper than a chemical coagulant, as most chemical coagulants require other material to effectively treat high turbidity, which raises the cost of the treatment process and makes it difficult to use 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 the accumulation of microbial activity. This issue has been solved by the addition of chlorine at safe doses to sanitise treated water (Amran et al., 2018). The sedimentation time is another limitation that can be mentioned regarding plant-based coagulants, which require more time than chemical coagulants (Kumar et al., 2017).

3. Banana Wastes as a Bio-Coagulant

In recent years, the utilisation of banana waste as a bio-coagulant in water treatment has earned growing interest due to its eco-friendliness, sustainability, and biodegradability. A variety of banana waste, including banana peels, leaves and stems, fruit peels, and stem juice, has been investigated for removing water turbidity. Different banana parts, such as banana peels, piths, trunks, and leaves, have been studied in terms of utilising them in many applications, including water treatment (Mokhtar et al., 2019). Table 3 presents selected studies on banana waste as a bio-coagulant for water treatment applications.

Table 3. Recent studies on the use of banana waste as a natural coagulant in water treatment applications

Banana part	Type of water	Pollutants	Finding	Ref
Banana pith	River water	Physicochemical and heavy metal	Significant reduction in turbidity, COD, TSS, nitrates, sulphates, and heavy metals	(Kakoi et al., 2016)
Banana peels	Wastewater	Turbidity	Removal of up to 96% of water turbidity	(Azamzam et al., 2022)
Banana Pith Juice	Textile Wastewater	Turbidity and TSS	Slight reduction of TSS and significant removal of turbidity	(Gopika and Kani, 2016)
Banana peel	Storage water tanks	Turbidity and colour	A decrease in both turbidity and colour	(Fu et al., 2019)
Banana peel with fenugreek seeds	palm oil mill effluent	Turbidity	Better removal efficiencies than commercial flocculant	(Ling et al., 2018)
Banana Peel	Domestic wastewater	Turbidity, COD, and NH_4-N	89.9%, 80.0%, and 62.5% reduction	(Ting et al., 2022)
Banana Peel	Synthetic Wastewater	Turbidity	88% turbidity reduction under optimum conditions	(Mokhtar et al., 2019)
Lemon and Banana Peel	Synthetic raw water	Turbidity and BOD	Turbidity and BOD were significantly removed	(Subashree et al., 2018)
Banana peel gel	Acid mine drainage water	Heavy metal	Removed Cd, Cu, Pb, and Zn	(Yabuki et al., 2020)
Banana Pith Starch	River water treatment	Turbidity, colour, and TDS	Turbidity reduced by 94.4% and colour by 87.46%	(Yushananta and Ahyanti, 2022))

Banana Peels

Banana fruit peels have been analysed as coagulation agents for the removal of different physicochemical parameters (Mokhtar et al., 2019; Olaoye et al., 2018). Banana peel establishes approximately 40% of the overall weight of fresh banana fruit, generating a large amount of unusable waste (Pelissari et al., 2017). Banana peels contain many active organic compounds, such as polysaccharides, cellulose, pectin, and hemicellulose, in addition to pigments and other low molecular weight compounds (Khawas and Deka, 2016). They are an excellent source of starch, cellulose, galacturonic acid, and pectin, with different ratios based on banana type, analysis method, and maturation level (Chaturvedi et al., 2018). Chaturvedi et al. (2018) used the aqueous extract from banana peel and were able to remove up to 88% of water turbidity of household wastewater under the optimised conditions of the tested parameters. İn a separate study, the powdered extract of banana peels (*Musa paradisica*) removed up to 83% of water turbidity at all tested pH values, and the maximum removal of turbidity was recorded at a pH between 5 and 9, which was 98.8% (Daverey et al., 2019), suggesting the promising potential of utilising banana peels as a safe and cost-effective water treatment technique for turbidity removal. In our previous research, we found that the high positive charge in banana peel attracts the negatively charged particles that cause turbidity in the water (Azamzam et al., 2022). We used microwave treatment to enhance the attraction, which showed greater coagulation performance compared with the non-modified particles and the solution (Figure 3). The modification led to the formation of larger flocs around the banana peel particles and, thus, better precipitation. In a different study, Pathak and Mandavgane (2015) followed the same modification process and reported that the surface of their banana peel became rough and porous after microwave treatment, confirming its efficiency in releasing active compounds (Pathak and Mandavgane (2015). The results of these studies confirm that the mechanism of the banana peel coagulation process is performed by the charge neutralisation of different charges.

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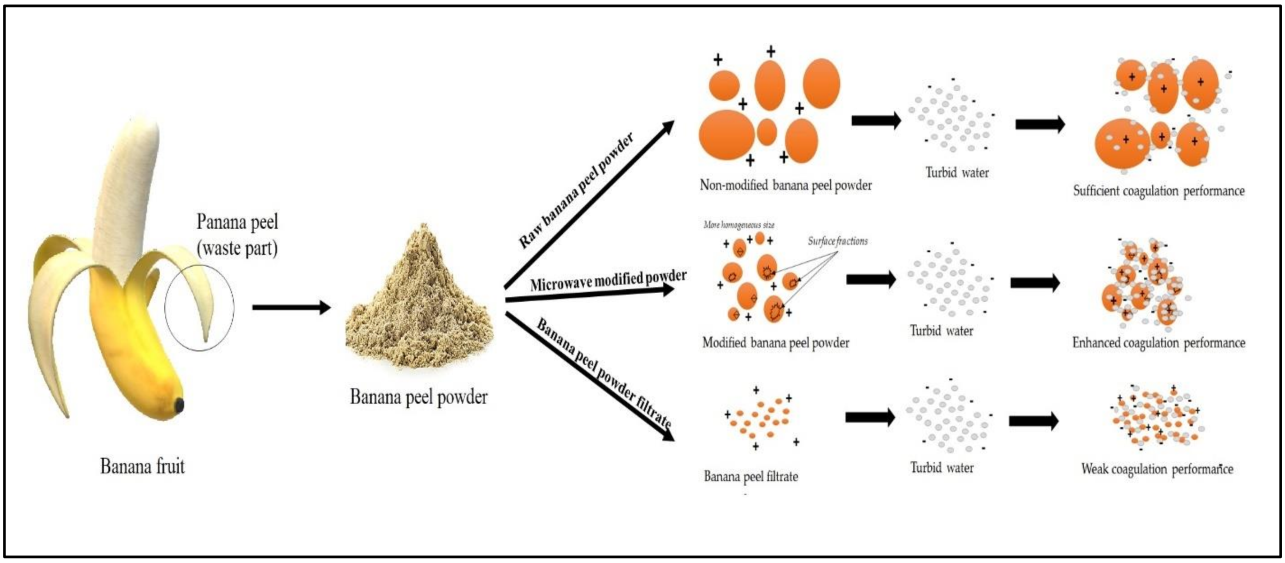

Figure 3. Coagulation mechanism of microwave-modified and non-modified banana peel particles. Adapted from Azamzam et al. (2022)

Banana Stem

The banana stem has been reported to be rich in polyelectrolytes that have not yet been significantly utilised for water treatment or any other economic purposes (Alwi et al., 2013). Two distinct methods have been reported for the mechanism of polyelectrolytes present in the banana pith, namely (i) bridging between particles and (ii) formation of bridges between particles (Kakoi et al., 2016). However, after harvesting banana fruit, the stem in most cases is left in the plantation as a mulching agent and fertiliser. The inner part of the stem is referred to as the pith, which is rarely used as food for livestock and, most of the time, is considered waste. After drying, powdered banana pith (Kakoi et al., 2016) is used for the direct removal of turbidity, suspended solids, and some heavy metals from river water. According to Feng et al. (2015), there are two steps in polyelectrolyte action: neutralisation of charges and the creation of bridges between particles. Chemically, banana pith is composed of a large number of significant functional groups, such as carboxylic, ether, and hydroxyl groups (Nayak et al., 2018).

The juice of banana pith has been used by Alwi et al. (2013) to reduce water physicochemical parameters, including turbidity, suspended solids, and chemical oxygen demand; they successfully reduced turbidity by 98.5%. They proposed that the juice contains inulin, which is the active coagulant agent responsible for creating bridges and thus decreasing the mentioned parameters. Namasivayam et al. (1993) reported that banana stems removed up to 87% of Rhodamine B from textile wastewater, even at low pH levels (pH 4). The use of banana stems has also been studied for the direct removal of red colour and acid brilliant blue, using the adsorption technique. It was able to absorb up to 5.92 mg of natural red and 4.42 mg of bright blue per gram of pith (Namasivayam et al., 1998), indicating the promising potential of using banana pith in adsorption and water treatment applications. Yushananta and Ahyanti (2022) synthesised banana pith starch using agricultural waste and used it for river water treatment applications. The authors also modified their isolated starch using cations from GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) into the main chemical structure of starch using microwave radiation. Modified banana pith starch reduces up to 94.4% of water turbidity, in addition to 87.46% and 57.33% of colour and Total Dissolved Solids (TDS), respectively. These findings put additional value on banana waste, which can be further modified to enhance its water treatment performance.

4. Challenges and Propositions of Using Bio Coagulants in Water Treatment

Different types, parts, and composites of natural coagulants have been investigated for treating synthetic or wastewater, aiming to provide safer options than conventional chemical methods. The use of banana waste does not give the best results compared to other natural coagulants. Most natural coagulants generate flocs, but issues regarding the removal of those flocs have still not been settled. Composite coagulants can overcome many problems linked with a sole coagulant, including the generation of small flocs and the non-optimal removal of some parameters (Mohd-Salleh et al., 2019). Similarly, composite coagulants generate more recoverable aggregated flocs and more resistance to shear than aggregated flocs caused by non-composite coagulants (Wang, Yue et al., 2018). However, the materials induced the growth rate and antibiotic sensitivity mutations in the tested microorganisms (Yahya, Abdulsamad, et al., 2020). Most studies, such as Kalemelawa et al. (2012) and Mosa et al. (2015) focus on the good side of naturally occurring products and ignore the potential lousy side related to a human directly or indirectly. Therefore, more microbiological studies regarding the use of natural coagulants need to be done (Yahya, Alfallous, et al., 2020).

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5. Conclusion

The use of renewable and sustainable sources of low-cost agricultural biomass waste, such as banana peels and stems, to produce natural coagulants is considered a better choice for water treatment. They can be used as a coagulant to treat various water parameters, such as turbidity, colour, COD, BOD, and even heavy metals. Future studies can be carried out to further investigate the efficiency of this plant-based treatment technique, which can be used as an option to overcome the issue of clean water scarcity, especially in rural areas.

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