Recent Advancements in Plant-Based Natural Coagulant Application in the Water and Wastewater Coagulation-Flocculation Process: Challenges and Future Perspectives

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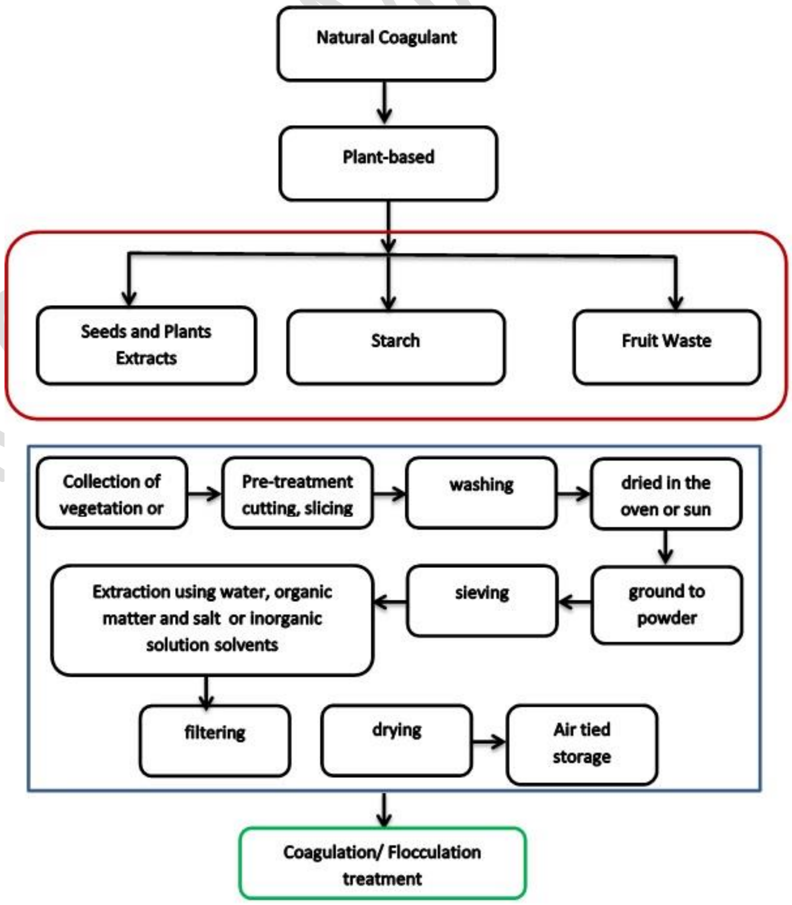
GRAPHICAL ABSTRACT



Abstract

Growing environmental awareness coupled with stricter regulations has pushed various industries to look for appropriate technologies to treat wastewater. Natural coagulants have recently been used as part of water and wastewater technologies due to their performance, cost-effectiveness when compared to chemical coagulants, they produce less sludge and are safer for humans. A huge effort has also been taken to find new plant species and ingredients that may be used as natural coagulants, and hence improve the efficacy of existing plant-based natural coagulants. The purpose of this research is to assess the progress of various natural coagulants used for water and wastewater treatment. Besides, several plant-based natural coagulants are also summarized and reviewed. The results reveal that natural coagulants are more successful than chemical coagulants at removing heavy metals and suspended particles from wastewater, with better removal efficiency exceeding chemical coagulants and being more environmentally friendly.

Keywords:

Plant-based Coagulant; Natural Coagulant; Wastewater Treatment; Removal Efficiency.

1. Introduction

Water is a crucial, essential element for humans and all existing organisms. Without water, there would be no life (Abujazar et al., 2018; Asrafuzzaman et al., 2011; Desta and Bote, 2021; Nath et al., 2019). In recent times, water consumption has increased due to industrialization and urbanization, as well as population growth (M. Y. D. Alazaiza et al., 2022; Ali and Tien Seng, 2018; Tang et al., 2016; Wang et al., 2021), and accordingly, wastewater from the various industrialization and urbanization activities is also increasing, making access to clean and safe water a critical issue.

Inappropriately treated or untreated wastewater contains contaminants, for example, suspended solids, pathogens, nutrients, and heavy metals that are toxic and hazardous to the environment and lead to numerous pollution problems (Mukherjee et al., 2016). These contaminants also give rise to waterborne diseases, which affect human health and are responsible for about 80% of the world's diseases (Lundqvist et al., 2019; Ugwu et al., 2017). Therefore, treating and reusing wastewater is the best way to combat freshwater scarcity and waterborne diseases (Abujazar et al., 2018).

The production of effluents from different industrial processes due to rapid industrialization has posed several environmental risks. Water and soil contamination can result from the indiscriminate dumping of these effluents. This case has a considerable impact on the health of living organisms, in addition to adverse effects on abiotic components such as soil and water (Chhonkar et al., 2000; Pal et al., 2010; Zhang et al., 2021). Every day, massive amounts of industrial effluent are generated worldwide. Industrial effluent is rife with chemical colors, the sodium salts of organic acids, lignin, and a considerable concentration of COD, oils, detergents, and suspended solids (Hassan et al., 2009; Muralimohan et al., 2014; Tobajas et al., 2014; Wang et al., 2011).

Sometimes, wastewater also contains heavy metals. This wastewater has anthropogenic origins, such as farms (i.e., it comes from pesticides spraying through raining runoff to surrounding environments). Agriculture is one of the most significant anthropogenic activities contributing to releasing bioavailable heavy metals into the environment (Malan et al., 2015).

On the other hand, mining activities which refer to the extraction of metals and minerals from the ground, mining of ore deposits that contain large amounts of sulfide minerals and heavy metals, is an essential source of heavy metals in the environment (Banks et al., 1997). The main pathways include airborne distribution of polluted dust from mine tailings or waste rock piles and discharge of acid mine drainage waters. The solubility of heavy metals such as Pb, Cd, Zn, Cu, Cr, Ni, and Fe occurs in the water body due to pyrite (FeS_2) or pyrrhotite (FeS) oxidation with lower pH (pH 3-4) (Wingenfelder et al., 2005).

Natural disasters such as earthquakes, storms, and volcanoes introduce large amounts of contaminants into our surface water and groundwater sources, many of which are collectively classified as metal pollutants' by environmental and health protection agencies such as Cu, Zn, As, Pb, Se) (Ahmed et al., 2020; Ali and Tien Seng, 2018; Helen Kalavathy and Miranda, 2010; Ilyinskaya et al., 2021; Ugya AY et al., 2018).

Heavy metals such as arsenic (As), lead (Pb), copper (Cu), cadmium (Cd), nickel, zinc (Zn), and chromium (Cr) are highly toxic. Even at low concentrations, these metals are considered the most harmful type of waste in freshwater reservoirs because they are non-biodegradable, and inherently long-lived (Azimi et al., 2017; Nath et al., 2019; Vikashni et al., 2012).

To overcome these problems, water/wastewater containing heavy metal ions are purified via conventional methods such as ultrafiltration, reverse osmosis, ion exchange, solvent extraction, sedimentation and chemical precipitation, coagulation/flocculation, etc. (Dąbrowski et al., 2004; Gao et al., 2014; Kabir et al., 2014; Pang et al., 2011; Shaidan et al., 2012; Tang et al., 2016; Vega Andrade et al., 2021; Zhang et al., 2021). However, these treatment methods have limitations and challenges, as well as different scopes and limitations of application, so the type of water/wastewater treatment should first be considered before any of these methods can be used.

Nevertheless, most of the above processes have disadvantages, such as insufficient removal efficiency. Besides, they also generate significant amounts of sludge, which contain hazardous chemical precipitates. These precipitates cause problems to human health, and the environment by causing secondary pollution from added chemical substances (Adhoum et al., 2004; Bazrafshan et al., 2015). Moreover, these methods also require high capital and high operating costs and advanced technology, as they are required in higher doses to be effective, and are not cost-effective (Asrafuzzaman et al., 2011; Bratby, 2006; Ghorpade and Ahammed, 2018; Parson, S.; Jefferson, 2006; Saleem and Bachmann, 2019)

Coagulation/flocculation in water/wastewater treatment generally includes the use of chemical reagents (coagulants or flocculants) to change the physical state of dissolved and suspended particles, allowing them to be removed more easily via the sedimentation process (Hameed et al., 2020; Johnson et al., 2008; Liu et al., 2021; Pang et al., 2011).

There are many types of coagulants; chemical-based coagulants and natural coagulants, which are based on natural materials. Chemical coagulants can be divided into three main types according to their chemical composition: inorganic-based coagulants, organic-based flocculants, and hybrid materials (Abujazar et al., 2022;

Lee et al., 2012). However, the usage of these chemical coagulants has had a number of negative consequences, such as aluminum residuals invading the human body and collecting in the brain, as well as harmful bulky sludge creation (Amagloh and Benang, 2009). Furthermore, because good coagulation requires a certain pH and set of conditions, the pH of the effluent must be adjusted beforehand. Furthermore, at low temperatures, these coagulants perform inefficiently (Bratby, 2006; Parson, S.; Jefferson, 2006; Ribau Teixeira et al., 2017; Rong et al., 2013).

The pursuit of eco-friendly and sustainable treatment procedures is an ongoing process and has received much support, as they have proven benefits. Reducing or replacing problematic coagulating ingredients will provide value to the sources of origin while minimizing the chemical and physical side effects of these methods on the treated media (Debora Peruço Theodoro et al., 2013). According to Ndabigengndabigengesere et al. (1995), natural coagulants can be obtained from plants, animals, and microorganisms. Natural coagulants have been used in water/wastewater treatment even before the advent of synthetic chemicals such as aluminum and ferric salts, with many recent studies highlighting the importance of natural coagulants (Hamidi et al., 2021). Plant-based materials can act as coagulants because they have inherent coagulation mechanisms, namely the neutralization of charge in colloidal particles and polymer bridging (Al-Sahari et al., 2020; Hariz Amran et al., 2018; Lee et al., 2012).

To solve this problem and to overcome the disadvantages of efficient and ineffective treatment, numerous natural coagulants have been implemented to treat water/wastewater effluents. These coagulants are more cost-effective and environmentally friendly than chemical coagulants, with many researchers reporting them as a viable alternative. Natural coagulants may be grown on-site and are biodegradable. Compared to chemical coagulants, they also generate less sludge and are safer for humans (Abidin et al., 2013; Dotto et al., 2019; Muralimohan et al., 2014; Prabhakaran et al., 2020; Shankar et al., 2014; Tie et al., 2015).

Because of numerous limitations in using chemical and synthetic coagulants, the trend has recently shifted toward studying and developing natural coagulants for effective wastewater treatment. In this paper, we offer an overview of the most recent information on wastewater treatment via the coagulation/flocculation process using natural coagulants, with an emphasis on the applications and assessment of natural coagulants for successful coagulation. In particular, the performance of natural coagulants was compared against artificial coagulants in the treatment of industrial wastewater. Furthermore, optimizing parameters impacting natural coagulant efficacy in industrial wastewater treatment was also investigated. Moreover, this study also examined the various coagulants used in industrial wastewater treatment, highlighting their optimum coagulation features and limitations. Future views on natural coagulant usage are given at the conclusion of this paper.

2. Characterization and Mechanisms of Natural Coagulants

Coagulation/flocculation in water/wastewater treatment is the process of adding chemicals to modify the physical state of suspended and dissolved particles in order to enhance removal efficiency (Teh et al., 2016). Coagulation can be defined as the process by which colloids, particles, and dissolved solids are combined to form larger flocs. Meanwhile, flocculation is the process by which destabilized particles are made to coalesce into larger flocs and settle at the bottom as sediment (Alexander et al., 2012; Bratby, 2006).

Polymer bridging, charge neutralization, double layer compression, and sweep coagulation are the four types of known coagulation mechanisms (Al-Sahari et al., 2020; Hariz Amran et al., 2018). For natural plant-based coagulants, however, only charge neutralization and polymer bridging are potential coagulation processes (Bolto and Gregory, 2007; Kristianto, 2017; Yin, 2010).

In the charge neutralization mechanism, the charges of colloidal particles are neutralized by ion-containing coagulants, which reduce the repulsive force between adjacent particles, as seen in Figure 1, by stabilizing highly negatively-charged suspended particles in water bodies, causing them to resist each other via the ensuing Van der Waals contact (Bolto and Gregory, 2007; Kristianto, 2017; Oladoja, 2015; Tripathy and De, 2006). A zero zeta potential is predicted at optimal coagulant dose; but, in fact, a slightly positive (Ndabigengndabigengesere et al., 1995) or negative (Henderson et al., 2008) zeta potential is obtained. The electrostatic repulsion between colloidal particles is decreased when the surface charges of the particles are reduced, enabling coagulation and resulting in bigger flocs (Kristianto, 2017).

According to the findings of Malik et al. (2015), treating tannery wastewater with aloe vera gel as a coagulant led to the creation of H^+ ions, which assisted in the charge neutralization of anionic colloidal particles. In another work on grafting polymerization of starch, corn starch (St) was grafted along with 3-Chloro-2-hydroxypropyl trimethylammonium chloride (CTA) in four different ratios (St:CTA) to remove kaolin, Escherichia coli, and Staphylococcus aureus from water samples (Liu et al., 2017). A recent study used *Moringa Oleifera* seed as a natural coagulant and found that it exhibited a positive charge and charge neutralization mechanisms (Mateus et al., 2020).

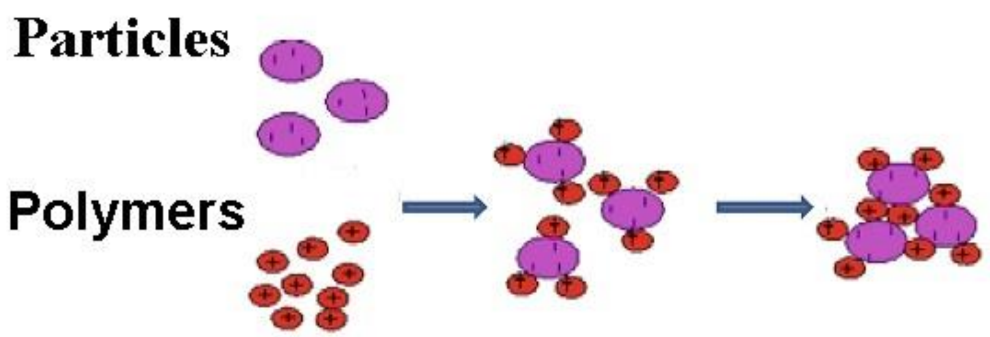


Figure 1: Mechanisms of flocculation charge neutralization (Nath et al., 2019)

In the polymer bridging mechanism, long-chain polymers are generally adsorbed by colloidal particles, leaving behind dangling, coagulating polymer segments to bind the particles together and form large flocs. This means that only a fraction of the polymers will adhere to the particles, while other polymers will form loops and tails (Hariz Amran et al., 2018). As illustrated in Figure 2, the basic structure of polymer bridging is made up of loops and tails, which allow attachment to other colloidal particles and hence the creation of bigger flocs (Bolto and Gregory, 2007). To achieve effective polymer bridging, a sufficient unoccupied particle surface area must be present, and the bridging must be long enough to overcome inter-particle repulsion flocs (Li et al., 2008). These conditions are met with the addition of a sufficient amount of polymers by using a natural coagulant that provides sufficient bridging bonds and a bare particle surface for strong polymer bridging (Gumfekar et al., 2017; Lu et al., 2016; Tripathy and De, 2006; Yin, 2010).

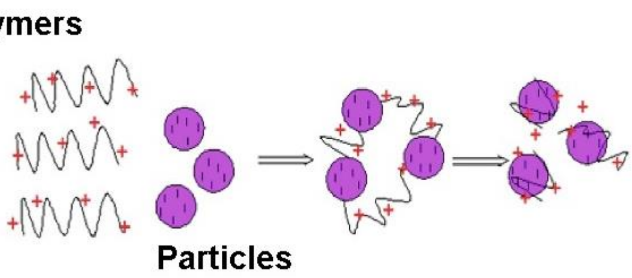


Figure 2: Mechanisms of flocculation bridging neutralization (Nath et al., 2019)

Factors that affect coagulation/flocculation process are, amongst others, temperature, pH, effluent quality, coagulant dose and type, mixing time, and optimal pH. One past study noted the optimal parameters for flocculation at pH 6 to pH 8 (Chaibakhsh et al., 2014; del Real-Olvera et al., 2016) with a coagulant dosage of (100, 200, 300, and 400) mg/L; and specific mixing speed procedures—rapid mixing at 100 rpm for 10 min and slow mixing at 30 rpm for 20 min followed by rapid mixing at 80 rpm for 2 min and slow mixing at 20 rpm for 20 min (Chaibakhsh et al., 2014). Low temperature is not favorable for flocculation because the reduced temperature will increase the solution’s viscosity. A more viscous solution at a lower temperature can have a detrimental impact on floc settling. Besides, low temperature also reduces the aggregation rate of particles in a solution (Tie et al., 2015; Xiao et al., 2009).

3. Plant-based Natural Coagulants

Natural coagulants had been used for treating water/wastewater long before chemical coagulants came into the picture (Choy et al., 2015; Yin, 2010). Microorganisms, plant-based coagulants, and animal-based coagulants are the three primary types of natural coagulants. Isinglass from shredded fish bladders (Biggs S, 2007) and chitosan from *crustacean shells* are the most widely used animal-based coagulants (Bratby, 2006; Zhao et al., 2021). Plant-based coagulants, on the other hand, have far more readily available sources than animal-based coagulants, implying that the former might be viable alternatives to chemical coagulants and that their importance has grown over time. However, in this study, we will only concentrate on natural plant-based coagulants, which may be divided into three categories: seed and plant extracts, starch, and fruit waste (Al-Sahari et al., 2020; Hariz Amran et al., 2018; Zhao et al., 2021), as shown in Figure 3.

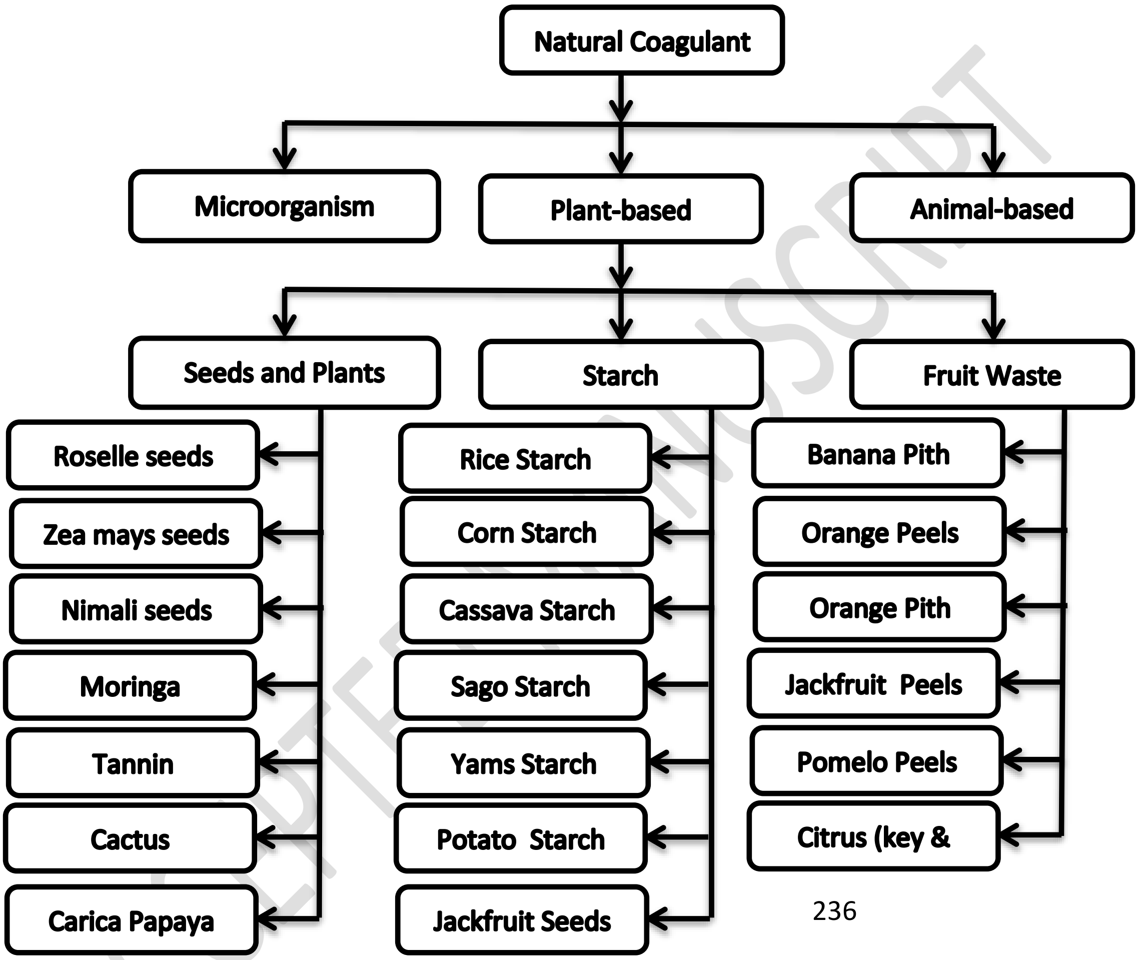


Figure 3 Schematic diagram of plant-based natural coagulant categories

According to one study Yin, (2010), there are three stages to preparing natural (plant-based) coagulants: the primary phase (pulverization), the secondary phase (extraction), and the tertiary phase (purification), as shown in Figure 4. In the main phase, the undesirable parts of the plant are detached by cutting, slicing or peeling, etc., while the required parts are usually dried in an oven or under the sun and then pulverized or ground into powder along with plant tissues and other organic matter that may pollute the water (Ghebremichael et al., 2005). In the second stage of extraction, the coagulants are removed from the powder using a variety of solvents, including water, organic matter, salt solutions (Okuda et al., 2001) or inorganic solvents (acetone or alcohol) (Pichler et al., 2012).

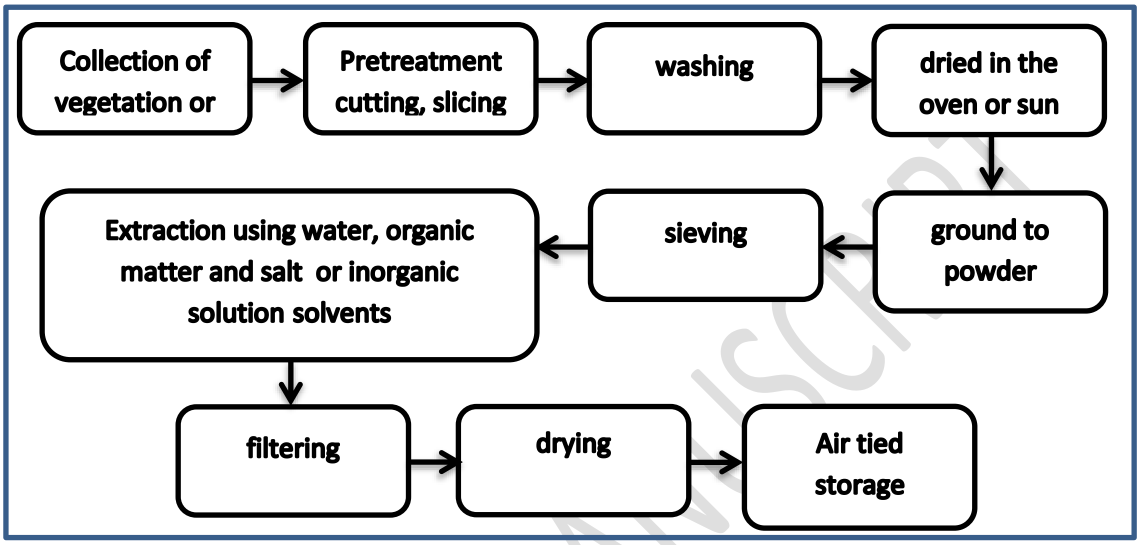


Figure 4: Schematic diagram showing the preparation of plant-based natural coagulants

According to Othmani et al., (2020), extraction using just water or saline solution (NaCl) should be preferred over the use of organic solvents because of the availability and inexpensive cost of water, as well as its reduced environmental effect. Several investigations have found salt having a greater impact on extraction efficiency than water, with results demonstrating that coagulants extracted with NaCl were more efficient at eliminating kaolinite turbidity at a dose 7.4 times lower than coagulants extracted with purified water.

Due to its high expense, the third step (purification) mentioned above generally involves laboratory-size work and is done in the academic research phase. Various purification techniques, such as freeze-drying, have been proposed in earlier research (Grossmann et al., 2018). Two techniques, ion exchange (Megersa and Triest, 2018) and dialysis (Idris et al., 2016), can be used for purifying extracts (particularly Moringa Oleifera) that may be integrated into a scaled-up machine to handle a higher turbid water throughput.

According to past studies (Al-Sahari et al., 2020; Saranya et al., 2014), plant-based natural coagulants can be categorized into polyelectrolytes such as cationic, anionic, or non-ionic and two cationic polymers with positive charges, namely chitosan and starch, which are typically investigated at pH 7 or less (Saleem and Bachmann, 2019). Starch is a polysaccharide that is made up of amylose and amylopectin (Kristianto, 2017). When used alone as a coagulant, starch has poor efficiency. As a result, cationic starch derivatives are frequently used to improve their efficiency. Cationic starch uses charge neutralization to stabilize strongly negatively-charged colloidal particles in aqueous fluids (Oladoja, 2015). Examples of cationic plant-based coagulants are seed proteins from *Moringa Oleifera* (Camacho et al., 2017) and *Cocos nucifera* (Camacho et al., 2017). Anionic coagulants are polymeric molecules that have a net negative charge, such as natural tannins commonly obtained from the bark or wood of *Schinopsis Acacia*, *Castanea* and other plants, and the corncob *of Quercus macrolepi* and sulfated polysaccharides and modified lingo sulfates (Freitas et al., 2018; Saleem and Bachmann, 2019).

4. Application of plant-based natural coagulants in water treatment

The natural plant-based coagulants used to treat wastewater include starchy plants and legumes. Coagulants with natural properties are said to be environmentally friendly and do not harm human health. They are also cost-effective, biodegradable, and non-toxic, whereas chemical coagulants can cause neurological and pathological diseases (Rajendran et al., 2015). On the other hand, the excessive use of inorganic coagulants such as aluminum sulfate in domestic water treatment processes causes high negative environmental impacts and risks to living organisms (Patchaiyappan and Devipriya, 2022).

Chestnut and *acorn* seed extracts (Šćiban et al., 2009), *Moringa peregrine* seeds (Bazrafshan et al., 2013), *Cactu* (Kazi and Virupakshi, 2013), *Moringa Oleifera* seeds (Kazi and Virupakshi, 2013), *tannin* (Banch et al., 2020), Acacia mearnsii (Beltrán-Heredia et al., 2011), and *Cassia angustifolia seeds* (Shak and Wu, 2014) are some of the natural coagulants that have been used to reduce turbidity and remove suspended solids, color, and other pollutants from aqueous solutions.

Previous research has used starch as a natural coagulant in coagulation and flocculation processes. For example, rice starch was used to treat palm oil mill wastewater, corn starch in dye wastewater, tapioca starch in semiconductor wastewater, and sago starch in synthetic wastewater (Aziz et al., 2000; Fatehah et al., 2013a; Louis and Sudha, 2013; Teh et al., 2014). The majority of natural coagulants that are in use are polysaccharides or proteins (Yin, 2010).

One study of Ali and Tien Seng, (2018) used *Moringa Oleifera* to remove heavy metals from wastewater. The study investigated Fe, Cu, and Cr at concentrations of 10000 ppm, 5000 ppm, and 15000 ppm, respectively, and found that the plant-based coagulant showed a removal efficiency of 69.99% Fe, 88.86% Cu, and 93.73% Cr. In another study of Al-Gheethi et al., (2017), *Moringa oleifera* was observed to have high efficiency at eliminating turbidity (83.6%) and reducing $FeSO_4$ (59%). However, *Moringa Oleifera* had comparable performance in removing the amount of COD (47.2%) and $FeSO_4$ (54%) in laundry effluents. In another research work (Patil and Mugeraya, 2013), it was found that the leaves and latex of *Calotropisprocera* could be used in heavy metal bioremediation. Table 1 summarizes the recent research on plant-based natural coagulants in treating various wastewaters.

Table 1: Recent studies on plant-based natural coagulants used in various water treatments

Coagulant type	Wastewater type	Experiments conditions		Removal efficiency (%)	Reference
		pH	Coagulant dosage		
Arachis hypogea	Turbid water	-	1000mg/L	Turbidity (96.70%)	(Mbogo, 2008)
Banana pith	River water	4	1kg/m^3	Turbidity (98.50%) COD (54.30%) SS (96.03%) Sulphates (98.90%) Nitrates removal (88.70%) Copper (100%) Chromium (100%) Iron (92%) Zinc (81%) Lead (100%)	(Kakoi et al., 2016)
cactus extracts	Synthetic wastewater	11.5	1.2mL	Suspended matter (99%) Zn (96%)	(Mounir et al., 2014)
Carica papaya seed	River water	-	100 seeds/100L	Turbidity (90%) E. coli bacteria (88%)	(Yongabi et al., 2011)

Glycine max	Turbid water	-	1000mg/L	Turbidity (98.90%)	(Mbogo, 2008)
Jackfruit	kaolin water	-	60mg/L	Turbidity (43%)	(Choy et al., 2017)
Jatropha curcas	synthetic wastewater	3	120mg/L	Turbidity (99%)	(Abidin et al., 2013)
mango seed kernels	Synthetic turbid water	7	25mg/L	Turbidity (92%)	(Seghosime et al., 2017)
Mango seed kernels	Sewage wastewater	5.2	168mg/L	Suspended matter (31.60%) COD (33.40%)	(Dange and Lad, 2015)
Mango seed kernels	Synthetic turbid water	13	0.5mg/L	Turbidity (98.60%)	(Qureshi et al., 2011)
Margaritarea discoidea	synthetic turbid water	0	10mL/L	Turbidity (98%)	(Oladoja et al., 2017)
Moringa Oleifera	Model turbid water	-	50mg/L	Turbidity (83.70%) E. coli (88.00%)	(Pritchard et al., 2009)
Moringa Oleifera	Reservoir water	-	750mg/L	Colour (52.40%) TC (73.60%)	(Pritchard et al., 2009)
Moringa Oleifera	Low turbidity river water	-	500mg/L	Colour (41.40%) TC (76.80%) E. coli (81.80%)	(Pritchard et al., 2009)
Moringa Oleifera	Turbid water	-	250mg/L	Colour (75.80%) Turbidity (74.00%) TC (93.90%) E. coli (92.60%)	(Pritchard et al., 2009)

Moringa Oleifera	Hybrid water	-	1000mg/L	Colour (82.50%) Turbidity (97.20%) E. coli (65.80%)	(Pritchard et al., 2009)
Moringa Oleifera	Turbid water	-	50mg/L	E. coli (88.00%) Turbidity (83.70%)	(Pritchard et al., 2010)
Moringa Oleifera	Reservoir water	-	750mg/L	Colour (52.40%) TC (73.60%)	(Pritchard et al., 2010)
Moringa Oleifera	Turbid river water	-	250mg/L	Colour (75.80%) Turbidity (74.00%) TC (93.90%) E. coli (92.60%)	(Pritchard et al., 2010)
Moringa Oleifera	Chicago Sky Blue 6B synthetic wastewater	8	250mg/L	Azo dye Chicago sky blue (99%)	(Beltran-Heredia and Sánchez-Martín, 2008)
Moringa Oleifera	River water	7.5	1000ppm	Fe (69.99%) Cu (88.86%) Cr (93.73%) COD (34.94%) Turbidity (81.60%)	(Ali and Tien Seng, 2018)
Moringa Oleifera	Water containing heavy	7	200mg/L	Pb (81%)	(Barnes et al., 2016)

	metal		400mg/L (ratio 1:1)	Ni (74%)	
Moringa Oleifera Seed	River water	6.9	30,000mg/L	Fe (100%) Cu (98%) Cd (98%) Pb (78.10%) Turbidity (94%)	(Shan et al., 2017)
Moringa Oleifera Seed	Domestic wastewater	7.2	200mg/L	Turbidity (90.43%) BOD_5 (55.77%) Phosphorus (88.84%) Nitrogen (54.54%) Calcium (62.38%) $CaCO_3$(50.48%) Coliform (97.50%)	(Ugwu et al., 2017)
Moringa Oleifera Tapioca starch	Wastewater	-	150mg/L	BOD (99.60%) COD: (99.70%) TSS (91.50%)	(Suhartini et al., 2013)
Moringa oleifera with Musa Cavendish (banana peel)	Water containing heavy metal	7	400mg/L (ratio 1:1)	Cd (97%)	(Barnes et al., 2016)
Okra extracts	Synthetic turbid water	7	25mg/L	Turbidity (92)	(Fahmi et al., 2014)

Okra extracts	Synthetic wastewater	6	5mg/L	Turbidity (97.30%)	(Anastasakis et al., 2009)
Orange peel	Synthetic turbid waste water	4	1mg/L	Turbidity (99.01%)	(Ho et al., 2010)
Orange peel	Laundry wastewater	-	6mL/L	Turbidity (99.01%) TSS (81.50%) TDS (56.40%) Anionic surfactant (77.40%)	(Mohan, 2014)
Parboiled rice husk ash	Turbid water	7.55	12000mg/L	Turbidity (95%)	(Adams and Mulaba-Bafubiandi, 2014)
Peanut seeds	Synthetic water	-	20mg/L	Turbidity (92%)	(Birima et al., 2013)
Pisum sativam	Turbid water	-	1000mg/L	Turbidity (99%)	(Mbogo, 2008)
Plantago ovata		7	0.25mg/L	Turbidity (99%)	(Ramavandi et al., 2015)
Rice starch	Microalgae sample	4	120mg/L	Microalgae removal (80%)	(Choy et al., 2016)
Rice starch	Kaolin wastewater	6.5	120mg/L	Kaolin removal (50%)	(Chua et al., 2021)
Rice starch	Synthetic turbid water	4	120mg/L	Turbidity (50%)	(Choy et al., 2016)
Roselle seeds	synthetic wastewater	4	40mg/L	Turbidity (93%)	(Mark J. Hammer and Mark J. Hammer, 2004)

Sago (dry tapioca root)	Water from Mudasarlova, India	6–8	100–200mg/L	Colour (99.93%) Chloride (78.57%) Hardness (87.50%) Turbidity (70–100%)	(Srinivas and Vuppala, 2015)
Tamarind seed powder	Detergent wastewater	-	400mg/L	COD (43.50%) Turbidity (97.78%)	(Ayangunna et al., 2016)
Tamarindus indica	River water	1	2mg/L	Turbidity (76%)	(Pengchai, P.; Keawkhun, K.; Suwapaet, 2012)
Unparboiled rice husk ash	Turbid water	8.15	8000mg/L	Turbidity (94.99%)	(Adams and Mulaba-Bafubiandi, 2014)

5. Application of plant-based natural coagulants in landfill leachate treatment

Globally, landfills are still the most used method to dispose of large quantities of solid waste, as it has a relatively low cost (Cheng et al., 2020; Daniel and Bhada-Tata, 2012; Wiszniowski et al., 2006). However, despite their economic advantages over alternative techniques (Renou et al., 2008), landfills generate leachate that contaminates surface and groundwater resources if not properly treated. This leachate can permeate through soils and sub-soils and cause the widespread pollution of ground and surface waters (Aziz and Sobri, 2015; Tatsi et al., 2003). Landfill leachate is hard to handle due to its complex composition, which is made up of many substances such as polycyclic aromatic hydrocarbons (Ates and Argun, 2018), antibiotics (Chung et al., 2018), heavy metals (Deng et al., 2018), and so on. Leachate also includes a significant quantity of organic pollutants, which are often specified by (COD) and (BOD), making it challenging to treat.

There are many techniques for treating leachate, including physical, chemical, and biological processes. Coagulation-flocculation is a comparatively simple technique that has been successfully used to treat landfill leachate (Ghafari et al., 2010; Maleki et al., 2009), with aluminum sulfate (alum), ferrous sulfate, ferric chloride, and polyaluminum chloride (PAC) being the most commonly-used coagulants for leachate treatment (Samadi et al., 2010; Wang et al., 2009; Zainol et al., 2011). However, several drawbacks have been linked to the use of these chemicals, such as their relatively high cost and harmful effects to human health (Alzheimer's disease), toxicity to live fish (Muisa, 2010), and the production of large amounts of sludge (Patale and Pandya, 2012). As a result, natural coagulants are seen as a crucial ecological crucial solution to the problem of landfill leachates.

Coagulation and flocculation processes using natural coagulants such as *Hibiscus Rosa-Sinensis*, *chitosan*, *cassava peels*, and palm oil starch have been the subject of many studies, as they are renewable resources and can be used in remote areas. Besides, they have also been proven safe for terrestrial and marine ecosystems (Camacho et al., 2017). However, the use of natural coagulants to remove refractory chemicals and humic acids from stabilized leachate has not been documented as much (Rusdizal et al., 2015). One investigation (Awang and Aziz, 2012) was carried out to assess the efficacy of *Hibiscus Rosa-sinensis* leaf extract in removing color, iron (Fe^{3+}), suspended particles, turbidity, and ammonia nitrogen (NH_3-N) from landfill leachate. Significant removal efficiency was reported, with the Fe^{3+} removal rate increasing to 100% when 4,000 mg/L alum was combined with 500 mg/L *Hibiscus rosa-sinensis* leaf extract (HBaqs). Another study of Aziz and Sobri, (2015) investigated native *sago* starch (NSTS) and commercial sago starch (CSS) as a sole coagulant in the presence of polyaluminium chloride (PAC) as a coagulation aid and investigated their efficiency at removing color, suspended solids (SS), NH_3-N, turbidity, chemical oxygen demand (COD), organic UV_{254}, Cd, and Ni from semi-aerobic landfill leachate. Table 2 shows the recent studies on plant-based natural coagulants used in landfill leachate treatment. Tannin has recently been used for leachate treatment. Tannin is extracted from the bark of the Black Acacia (Acacia mearnsii). One study of Banch et al., (2019) employed modified tannin for organic and heavy metal removal from stabilized leachate. The study reported a COD, TSS, NH_3–N, and color removal performance of 53.50%, 60.26%, and 91.39%, respectively, while the removal of selected heavy metals Fe^{2+}, Zn^{2+}, Cu^{2+}, Cr^{2+}, Cd^{2+}, Pb^{2+}, As^{3+} and cobalt Co^{2+} were 89.76%, 94.61%, 94.15%, 89.94%, 17.26%, 93.78%, 86.43%, and 84.19%, respectively. The study recommended tannin for removing heavy metal and ammonia from stabilized landfill leachate, but highlighted its limited performance at removing organic matter.

Table 2: Recent studies on plant-based natural coagulants in landfill leachate treatment

Coagulant type	water type	Experiments conditions		Removal efficiency (%)	Reference
		pH	Coagulant dosage		
Commercial sago starch (CSS)	landfill leachate	4	6000mg/L	Colour (15.10%) SS (29.50%) NH_3-N (10.70%) COD (28.00%) Organic UV_{254} (51.60%) Cd (33.20%) Ni (16.30%)	(Aziz and Sobri, 2015)
Cross linked Oil Palm Trunk Starch (COPTS)	landfill leachate	8.3	1000mg/L	Turbidity (43.20%) Colour (65.10%) SS (25%) NH3-N (5%) COD (39%) Mn (100%) Cu (49%) Zn (79.5%)	(Yusoff et al., 2019)

				PO4 (100%)	
guar gum	landfill leachate	8.5	44.4mg/L	COD (22.5%)	(Cheng et al., 2020)
Hibiscus rosa-sinensis	landfill leachate	6	400 mg/L alum +50 mg/L	Turbidity (60%) Color (61%) Fe3+ (100%) SS (72%) NH_3-N (28%)	(Awang and Aziz, 2012)
Jackfruit Seeds Starch	landfill leachate	5	PAC 600mg/L; JSS 500mg/L	Turbidity (92.30%) Colour (15.10%) SS (94.50%) NH3-N (14.40%) COD (33.50%)	(Yusoff and Mohamad Zuki, 2015)
Native sago trunk starch (NSTS)	landfill leachate	4	7000mg/L	Colour (13.10%) SS (27.90%) NH_3-N (8.20%) COD 1.70% Organic UV_{254} (43.80%) Cd (25.50%)	(Aziz and Sobri, 2015)

				Ni (44.10%)	
Oil Palm Trunk Starch (OPTS)	landfill leachate	7	500mg/L	Turbidity (38%) Colour (36%) SS (94.50%) NH3-N (14.4%) COD (33.50%)	(Yusoff et al., 2019)
Psyllium Husk	landfill leachate	7.5	7.2mg/L PAC + 0.4mg/L	COD (64%) Colour (90%) TSS (96%)	(Al-Hamadani et al., 2011)
Sago Trunk Starch	landfill leachate	6	2000mg/L PAC+ 6000mg/L NSTS	Turbidity (98.90%) Color (94.70%) SS (99.20%) NH_3 (2.40%) COD (35.50%) Organic UV_{254} (69.50%) Cd (53.80%)	(Aziz and Sobri, 2015)
Salvia Hispanica	landfill leachate	7	40mg/L	COD (39.80%)	(Tawakkoly et al., 2019)
Tapioca starch flour	landfill leachate	5	0.2mg/L	COD (60%)	(Zin et al., 2014)
Tobacco Leaf (TL)	landfill leachate	6	1000mg/L PAC	Turbidity (21%)	(Rusdizal et al., 2015)

	+1000mg/L TL	Colour (86%) SS (48%) NH3-N (54%) COD (91%)

6. Applications of plant-based natural coagulants in industrial wastewater treatment

Due to the obvious potential environmental and human health issues linked with chemical coagulants, it is now essential to explore the use of natural coagulants in industrial wastewater treatment. Natural coagulants are fast gaining popularity these days as a viable alternative to chemical coagulants (Yin, 2010).

According to one study (Geng et al., 2018), plant-based coagulants can be used to treat industrial wastewater originating from numerous manufacturing plants because of their simple operation, high efficiency, and low cost, such as for textile (Abidin et al., 2013; Dotto et al., 2019; Muralimohan et al., 2014; Prabhakaran et al., 2020; Shankar et al., 2014), dairy (Anju S and Mophin-Kani, 2016; Sivakumar, 2015; Sivakumar et al., 2016, 2014; Triques et al., 2020), tannery (Ahmed et al., 2020; Kazi and Virupakshi, 2013), laundry wastewater (Freitas et al., 2015), etc. In another study (Kazi and Virupakshi, 2013), *Cicer arietinum*, *Moringa Seed,* and *Cactus Opuntiaficus* were used as natural coagulants, and these plants removed turbidity with 81.20%, 82.02%, and 78.54% efficiency, respectively, and reduced the maximum COD in tannery effluents by 90%, 83.33%, and 75%, respectively.

Another study showed that *Moringa oleifera, Azadirachta indica, Trigonella foenum graecum,* and *Cicer arietinum* showed high efficiency at reducing the turbidity in wastewater from the dairy industry by 61.60%, 71.74%, 58.20%, and 78.33%, respectively (Gayathri, 2017). Table 3 summarizes recent research that has used plant-based natural coagulants to treat industrial wastewater.

398 **Table 3: Recent studies on plant-based natural coagulants to treat industrial wastewater**

Coagulant Type	Water Type	Experiments Conditions		Removal Efficiency	Reference
		pH	Coagulant Dosage	(%)	
Azadirachta indica	Dairy industry	7.6	0.05g/500ml	Turbidity (71.74%)	(Gayathri, 2017)
Cactus Opuntiaficus	Tannery Wastewater	5.5	0.2mg/500ml	Turbidity (78.54%) COD (75%)	(Kazi and Virupakshi, 2013)
Cicer arietinum	Tannery Wastewater	5.5	0.1g/500ml	Turbidity (81.20%) COD (90%)	(Kazi and Virupakshi, 2013)
Cicer arietinum	Dairy wastewater	9	0.1g/L	Turbidity (78.33%) COD (83%)	(Patil and Hugar, 2015)
Cicer arietinum	Dairy industry	7.6	0.1g/500ml	Turbidity (78%)	(Gayathri, 2017)
Chitosan	Olive mill wastewater	4.3	400mg/L	Turbidity (84%) TSS (81%)	(Rizzo et al., 2008)
Chitosan	Textile wastewater	4	30mg/L	Turbidity (95%) COD (65%)	(Hassan et al., 2009)
Dolicus lablab	Dairy wastewater	9	0.2g/L	Turbidity (71.74%) COD (75%)	(Patil and Hugar, 2015)
Guar	Cosmetic	7	300mg/L	Turbidity (67.82%)	(Carpinteyro-Urban et al., 2012)

	wastewater				
Moringa oleifera	Dairy wastewater	7.2	1g/L	Turbidity (98%) Colour (95%) COD (55%)	(Vieira et al., 2010)
Moringa oleifera	Oil refinery wastewater	6	70mg/L	Turbidity (63.70%) TSS (81.52%) COD (50.41%)	(Dehghani and Alizadeh, 2016)
Moringa oleifera	Dairy industry	7.6	0.4g/500ml	Turbidity (61.60%)	(Gayathri, 2017)
Okra	Coal washery wastewater treatment	7-8	400ppm	Turbidity (61%)	(Babarao, T.D.; Verma, 2015)
Roselle seeds	Glove Manufacturing	4	60mg/L	Turbidity (78.20%)	(Saharudin and Nithyanandam, 2014)
Surjana seed	Textile wastewater	--	25g/L	Colour (58.51%) COD (74.11%)	(Patel and Vashi, 2013)
Trigonella foenum-graecus	Dairy wastewater	9	0.1g/L	Turbidity (85%) COD (63%)	(Patil and Hugar, 2015)
Trigonella foenum graecum	Dairy industry	7.6	0.4g/500ml	Turbidity (58.20%)	(Gayathri, 2017)

7. Replacing chemical coagulation with plant-based natural coagulation

Natural coagulants might be a viable alternative to chemical coagulants and a step forward in green water treatment technology, reducing environmental pollution and health hazards while also boosting green technology in water and wastewater treatment (Mathuram et al., 2018).

A wealth of research has been done to assess the effectiveness of natural coagulants in removing contaminants from water and wastewater, and it has been established that natural coagulants can have comparable removal efficiency to that of chemical coagulants (Kumar and Quaff, 2019). Combining chemical coagulants with natural coagulants may improve the performance of coagulation. In one study, *chitosan* and ferric chloride ($FeCl_3$) were used to treat Palm Oil Mill Effluent (POME) wastewater. Chitosan at 400 mg/L achieved a removal efficiency for turbidity, TSS, oil & grease, COD, NH_3-N, and color of (98.35%), (98.95%), (94.86%), (68.31%), (95.88%), and (96.44%), respectively, while ferric chloride ($FeCl_3$), at 500 mg/L, showed a removal efficiency for turbidity, TSS, oil & grease, COD, NH_3-N, and color of (95.99 %), (99.09%), (65%), (72.04%), (93.74%), and (66.44%), respectively (Jagaba et al., 2020).

Considerable contrast was also detected in the reduction of turbidity in surface water when 0.75 g/L *Moringa Oleifera* and 1 g/L Alum were used and removed (75%) and (70%) of turbidity, respectively (M and Rohini, 2017). This finding reveals that natural coagulants have promise in treating wastewater.

According to prior research, natural coagulants may remove contaminants just as well as chemical coagulants. Table 4 compares the efficacy of plant-based natural and chemical coagulants; in most experiments, the natural coagulants achieved comparable removal efficiency to that of chemical coagulants. However, the majority of the studies reveal that the chemical coagulants had a somewhat greater clearance efficiency than the natural coagulants. It is worth noting, however, that the efficacy of natural coagulants can be improved further by improving the extraction and purification processes to get the active coagulant ingredient.

Table 4: Comparison between the performance of plant-based natural coagulants and chemical coagulants in wastewater treatment

Type of wastewater	Natural coagulant	Removal efficiency (%)	Chemical coagulant	Removal efficiency (%)	Reference
Arsenic (As(III)) contaminated surface water	Cellulose	As(III) (84.62%) @ 1mg/L	Ferric chloride ($FeCl_3$)	As(III) (69.3%) @ 40mg/L	(Kumar and Quaff, 2019)
	Chitosan	As(III) (75.87%) @ 25mg/L			
Semiconductor CMP wastewater	Potato flour	Total suspended (TS) (90%) @ 0.02 to 3.38g/L		Total suspended (TS) (78%) @ 0.02 to 0.06g/L	
	Tapioca flour	Total suspended (TS) (89%) @ 0.02 to 3.01g/L	Alum		(Fatehah et al., 2013b)
	Corn flour	Total suspended (TS) (77%) @ 0.02 to 0.07g/L		Total suspended (TS) (53%) @ 0.02 to 0.03g/L	
	Rice flour	Total suspended (TS) (73%) @ 0.02 to 1.14 g/L	PAC		
Surface water	Moringa oleifera	Turbidity (75%) @ 0.75g/L	Alum	Turbidity (70%) @ 1g/L	(M and Rohini, 2017)
Synthetic greywater	Tamarind seeds	Turbidity (61.33%) @ 400mg/L	Alum	Turbidity (96.49%) @ 1000mg/L	(Chitra and Muruganandam, 2020)

		COD (41.37%) @ 400 mg/L		COD (94.70%) @ 1000mg/L	
	Moringa oleifera	Turbidity (85.75%) @ 600mg/L COD (65.71%) @ 600 mg/L			
	Banana peels	Turbidity (90.42%) @ 1000mg/L COD (76.10%) @ 1000mg/L			
Raw waters	Moringa oleifera	Turbidity (99%) @ 0.9g/L	Aluminum sulphate	Turbidity (99%) @ 0.05g/L	(Anderson et al., 2021)
Paper mill industry	Moringa oleifera seed	Turbidity (96.20%) @ 150mg/L COD (97.30%) @ 150mg/L	Alum	Turbidity (97.10%) @ 4000mg/L COD (92.70%) @ 4000mg/L	(Boulaadjoul et al., 2018)

Wastewater Palm Oil Mill Effluent POME ,	Moringa oleifera	Turbidity (88.30%) @ 2000mg/L TSS (95.42%,%) @ 2000mg/L Oil & grease(87.05%) @ 2000 mg/L COD (51.99%) @ 3000mg/L NH_3-N (89.81%) @ 2000mg/L Colour (90.15%) @ 2000mg/L	Aluminum sulphate	Turbidity (98.68%) @ 4000mg/L TSS (98.72%) @ 4000mg/L Oil & grease(97.97%) @ 3000mg/L COD (75.01%) @ 4000mg/L NH_3-N (98.46%) @ 4000mg/L Colour (94.01%) @ 3000mg/L	(Jagaba et al., 2020)
	chitosan	Turbidity (98.35%) @ 400mg/L TSS (98.95%) @ 400mg/L Oil & grease(94.86%) @ 400mg/L COD (68.31%) @ 400mg/L	Ferric chloride ($FeCl_3$)	Turbidity (95.99 %) @ 1500mg/L TSS (99.09 %) @ 1000 mg/L Oil & grease(65%) @ 1000mg/L COD (72.04 %) @ 1000mg/L	

Paint industry	Cactus	NH_3-N (95.88%) @ 400mg/L Colour (96.44%) @ 400mg/L Turbidity (88.50%) @ 3g/L COD (83.40%) @ 3g/L Colour (89.40%) 3g/L	Ferric chloride	NH_3-N (93.74 %) @ 1000mg/L Colour (66.44%) @ 500mg/L Turbidity (82.60%) @ 0.7 g/L COD (78.20%) @ 0.7 g/L Colour (88.40%) @ 0.7 g/L	Vishali and Karthikeyan,) (2015
Synthetic turbid water	Banana peel	Turbidity (65.60%) @ 150ml/400ml	Alum	Turbidity (73.10%) @ 150ml/400ml	Kian-Hen and Peck-Loo,) (2017

8. Limitations and future perspectives regarding the use of plant-based natural coagulants

Natural coagulants have an advantage over chemical coagulants in treating wastewater for a variety of reasons; they are safer, more environmentally friendly, less expensive, and more effective at treating high-turbidity water than chemical coagulants (Abidin et al., 2013; M. Alazaiza et al., 2022; Dotto et al., 2019; Garde et al., 2017; Muralimohan et al., 2014; Prabhakaran et al., 2020; Shankar et al., 2014; Tie et al., 2015).

According to Awad et al. (2013), aside from the above benefits, natural coagulants also have several drawbacks. For example, natural coagulants may increase the organic matter in water, and thus increase microbial activity. The residual organic matter may alter the odor, taste, and color of the treated water, necessitating the use of a disinfectant such as chlorine (Choy et al., 2014). As a result, more research into the relationship between natural coagulants and their influence on chlorine should be considered. Some coagulants require a longer sedimentation time than chemical coagulants, and while other coagulants have antibacterial properties that can treat E. coli-infested water, they do not completely remove them, and thus pose a risk of secondary bacterial growth (Ghebremichael, 2007).

The disadvantages of using natural coagulants, on the other hand, are mostly due to the coagulant itself, such as the influence of seasons and storage length, which can impair the production and ongoing supply of the natural coagulant. The greatest impediment to the commercialization of natural coagulants is the difficulty in producing raw materials in large quantities; plant species.

Raw materials necessary to make natural coagulant should be accessible in big quantities for a successful and practical use. Technical assistance, professional advice, and new equipment are required for the long-term deployment of natural coagulants, resulting in higher manufacturing costs. In the near run, this is not particularly cost effective, thus market acceptability will be low. As a result, the lack of mass cultivation of resources hinders a consistent supply of raw materials and long-term uses. However, several studies have been undertaken to date in order to mitigate the effects of the natural coagulant's downsides. Current study on a mixture of plant waste, such as banana and pomegranate peels, demonstrated the appropriateness of combining multiple raw material sources, offering insight into examining additional alternative sources in small quantities to be utilized as natural coagulants (Mohana, 2019).

Another major limitation is that natural coagulants are less effective in the coagulation process due to the current lack of knowledge about the characteristics and properties of the resultant flocs that form. Knowledge of these parameters, such as floc strength, structure, and compactness, could be critical in determining the most likely coagulation theory (Choy et al., 2014; Idris et al., 2013).

9. Conclusion

Untreated or inadequately treated wastewater is harmful to the environment. Chemicals have long been employed in wastewater treatment as coagulants. Chemical coagulants have intrinsic downsides, including the formation of non-degradable sludge and high expense, despite their high treatment effectiveness. As an alternative to chemical agents, natural plant-based coagulants have been introduced. Many of these plant-based coagulants are being studied for their usefulness in wastewater pollution treatment and have showed promise in treating wastewater in an efficient and ecologically beneficial manner.

It can be concluded that natural-plant based coagulants are more efficient and safer than chemical coagulants, with the former demonstrating high efficiency at turbidity removal and maximum COD reduction in tannery effluents by using Cicer arietinum, Moringa Seed, and Cactus Opuntiaficus as natural coagulants in the treatment of wastewater pollution, respectively, compared to chemical coagulants, which have many drawbacks such as carcinogenic properties. Therefore, resorting to nature will undoubtedly aid in replacing chemical reagents in water bodies, which, in turn, would pave the way for a more sustainable environment and cleaner technology.

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