Global NEST Journal, Vol 24, No 4, pp 687-705

Recent advancements in plant-based natural coagulant application in the water and wastewater coagulation-flocculation process: challenges and future perspectives

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Received: 24/06/2022, Accepted: 03/10/2022, Available online: 16/10/2022

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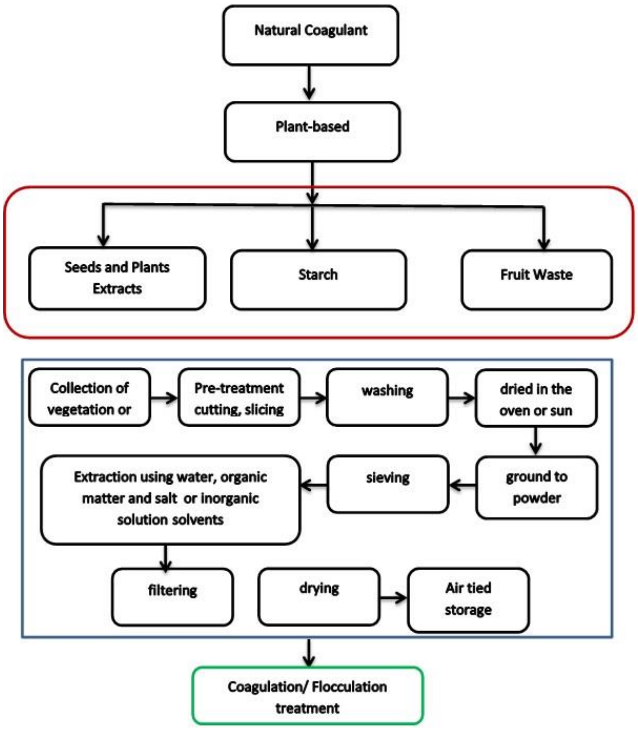
https://doi.org/10.30955/gnj.004380

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 and 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).

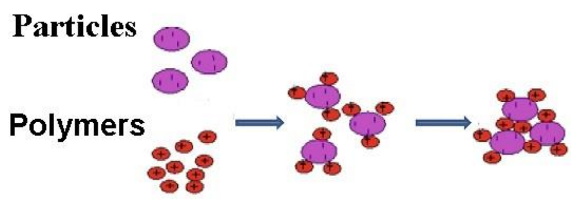


Figure 1. Mechanisms of flocculation charge neutralization (Nath *et al.*, 2019)

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).

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).

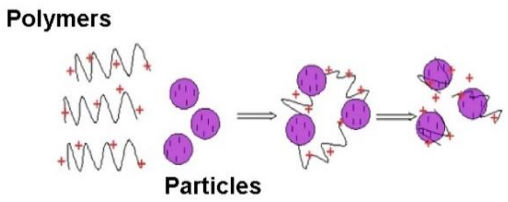


Figure 2. Mechanisms of flocculation bridging neutralization (Nath *et al.*, 2019)

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.

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).

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.

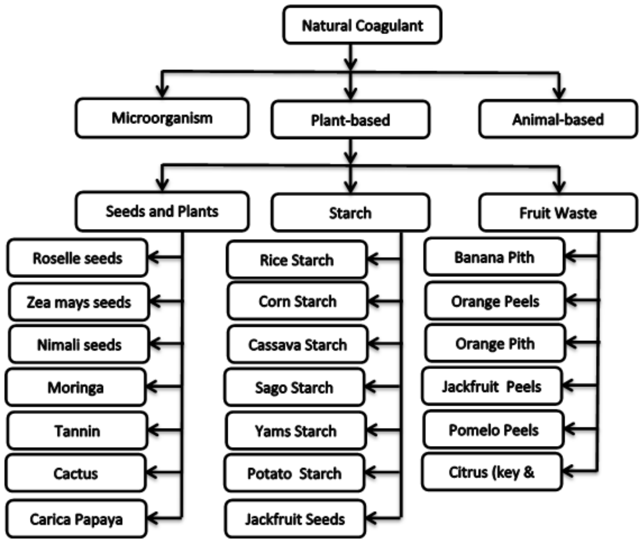


Figure 3. Schematic diagram of plant-based natural coagulant categories

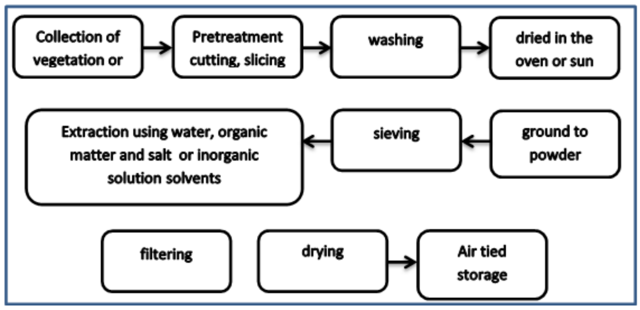


Figure 4. Schematic diagram showing the preparation of plant-based natural coagulants

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 (%)	
		pH	Coagulant dosage	Reference	
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%) Manganese (60%)	(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%)	(Dange and Lad, 2015)

				COD (33.40%)	
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 metal	7	200mg/L 400mg/L (ratio 1:1)	Pb (81%) Ni (74%)	(Barnes *et al.*, 2016)
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	7	25mg/L	Turbidity (92)	(Fahmi *et al.*, 2014)

	water				
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 (Fe3+), suspended particles, turbidity, and ammonia nitrogen (NH3-N) from landfill leachate. Significant removal efficiency was reported, with the Fe3+ 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), NH3-N, turbidity, chemical oxygen demand (COD), organic UV254, 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, NH3–N, and color removal performance of 53.50%, 60.26%, and 91.39%, respectively, while the removal of selected heavy metals Fe2+, Zn2+, Cu2+, Cr2+, Cd2+, Pb2+, As3+ and cobalt Co2+ 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%)	(Aziz and Sobri, 2015)
				SS (29.50%)	
				NH_3-N (10.70%)	
				COD (28.00%)	
				Organic UV_{254} (51.60%)	
				Cd (33.20%)	
				Ni (16.30%)	
Cross linked Oil Palm Trunk Starch (COPTS)	landfill leachate	8.3	1000mg/L	Turbidity (43.20%)	(Yusoff *et al.*, 2019)
				Colour (65.10%)	
				SS (25%)	
				NH3-N (5%)	
				COD (39%)	
				Mn (100%)	
				Cu (49%)	
				Zn (79.5%)	
				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	Turbidity (60%)	(Awang and Aziz, 2012)
			50+mg/L	Color (61%)	
				Fe3+ (100%)	
				SS (72%)	
				NH_3-N (28%)	
Jackfruit Seeds Starch	landfill leachate	5	PAC 600mg/L;	Turbidity (92.30%)	(Yusoff and Mohamad Zuki, 2015)
			JSS 500mg/L	Colour (15.10%)	
				SS (94.50%)	
				NH3-N (14.40%)	
				COD (33.50%)	
Native sago trunk starch (NSTS)	landfill leachate	4	7000mg/L	Colour (13.10%)	(Aziz and Sobri, 2015)
				SS (27.90%)	
				NH_3-N (8.20%)	
				COD 1.70%	
				Organic UV_{254} (43.80%)	
				Cd (25.50%)	
				Ni (44.10%)	
Oil Palm Trunk Starch (OPTS)	landfill leachate	7	500mg/L	Turbidity (38%)	(Yusoff *et al.*, 2019)
				Colour (36%)	
				SS (94.50%)	
				NH3-N (14.4%)	

				COD (33.50%)	
Psyllium Husk	landfill leachate	7.5	7.2mg/L PAC + 0.4mg/L	COD (64%)	(Al-Hamadani *et al.*, 2011)
				Colour (90%)	
				TSS (96%)	
Sago Trunk Starch	landfill leachate	6	2000mg/L PAC+	Turbidity (98.90%)	(Aziz and Sobri, 2015)
			6000mg/L NSTS	Color (94.70%)	
				SS (99.20%)	
				NH_3 (2.40%)	
				COD (35.50%)	
				Organic UV_{254} (69.50%)	
				Cd (53.80%)	
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)
			1000+mg/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.

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%)	Kazi and Virupakshi, 2013)
				COD (75%)	
Cicer arietinum	Tannery Wastewater	5.5	0.1g/500ml	Turbidity (81.20%)	Kazi and Virupakshi, , 2013)
				COD (90%)	
Cicer arietinum	Dairy wastewater	9	0.1g/L	Turbidity (78.33%)	Patil and Hugar, 2015)
				COD (83%)	
Cicer arietinum	Dairy industry	7.6	0.1g/500ml	Turbidity (78%)	(Gayathri, 2017)
Chitosan	Olive mill wastewater	4.3	400mg/L	Turbidity (84%)	(Rizzo *et al.*, 2008)
				TSS (81%)	
Chitosan	Textile wastewater	4	30mg/L	Turbidity (95%)	(Hassan *et al.*, 2009)
				COD (65%)	
Dolicus lablab	Dairy wastewater	9	0.2g/L	Turbidity (71.74%)	(Patil and Hugar, 2015)
				COD (75%)	
Guar	Cosmetic wastewater	7	300mg/L	Turbidity (67.82%)	(Carpinteyro-Urban *et al.*, 2012)
Moringa oleifera	Dairy wastewater	7.2	1g/L	Turbidity (98%)	(Vieira *et al.*, 2010)
				Colour (95%)	
				COD (55%)	
Moringa oleifera	Oil refinery	6	70mg/L	Turbidity (63.70%)	(Dehghani and

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

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	Alum	Total suspended (TS) (78%) @ 0.02 to 0.06g/L	(Fatehah *et al.*, 2013b)
	Tapioca flour	Total suspended (TS) (89%) @ 0.02 to 3.01g/L			
	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			
	Moringa oleifera	Turbidity (85.75%) @ 600mg/L			
		COD (65.71%) @ 600 mg/L		COD (94.70%) @ 1000mg/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	Alum	Turbidity (97.10%) @ 4000mg/L	(Boulaadjoul *et al.*, 2018)
		COD (97.30%) @ 150mg/L		COD (92.70%) @ 4000mg/L	
Wastewater Palm Oil Mill Effluent POME,	Moringa oleifera	Turbidity (88.30%) @ 2000mg/L	Aluminum sulphate	Turbidity (98.68%) @ 4000mg/L	(Jagaba *et al.*, 2000)
		TSS (95.42%,%) @ 2000mg/L		TSS (98.72%) @ 4000mg/L	
		Oil &		Oil &	

		grease(87.05%) @ 2000 mg/L		grease(97.97%) @ 3000mg/L	
		COD (51.99%) @ 3000mg/L		COD (75.01%) @ 4000mg/L	
		NH3-N (89.81%) @ 2000mg/L		NH3-N (98.46%) @ 4000mg/L	
		Colour (90.15%) @ 2000mg/L		Colour (94.01%) @ 3000mg/L	
	chitosan	Turbidity (98.35%) @ 400mg/L	Ferric chloride ($FeCl_3$)	Turbidity (95.99 %) @ 1500mg/L	
	chitosan	TSS (98.95%) @ 400mg/L	Ferric chloride ($FeCl_3$)	TSS (99.09 %) @ 1000 mg/L	
	chitosan	Oil & grease(94.86%) @ 400mg/L	Ferric chloride ($FeCl_3$)	Oil & grease(65%) @ 1000mg/L	
	chitosan	COD (68.31%) @ 400mg/L	Ferric chloride ($FeCl_3$)	COD (72.04 %) @ 1000mg/L	
	chitosan	NH3-N (95.88%) @ 400mg/L	Ferric chloride ($FeCl_3$)	NH3-N (93.74 %) @ 1000mg/L	
	chitosan	Colour (96.44%) @ 400mg/L	Ferric chloride ($FeCl_3$)	Colour (66.44%) @ 500mg/L	
Paint industry	Cactus	Turbidity (88.50%) @ 3g/L	Ferric chloride	Turbidity (82.60%) @ 0.7 g/L	(Vishali and Karthikeyan, 2015)
Paint industry	Cactus	COD (83.40%) @ 3g/L	Ferric chloride	COD (78.20%) @ 0.7 g/L	(Vishali and Karthikeyan, 2015)
Paint industry	Cactus	Colour (89.40%) 3g/L	Ferric chloride	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)

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.

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.

Acknowledgments

The authors wish to extend sincere thanks to the many individuals and organizations for their kind support in the study, in particular, the Institute of International Education-Scholar Rescue Fund (IIE–SRF). New York, NY 10007 USA, for supporting this work. Also, the research leading to these results has received funding from Ministry of Higher Education, Research, and Innovation (MoHERI) of the Sultanate of Oman under the Block Funding Program, MoHERI Block Funding Agreement No. MoHERI/BFP/ASU/01/2021

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