Wastewater treatment using plant-derived bioflocculants: green chemistry approach for safe environment

Nilanjana Das, Nupur Ojha and Sanjeeb Kumar Mandal

ABSTRACT

The rapid expansion of global trade and human activities has resulted in a massive increase in wastewater pollution into the atmosphere. Suspended solids, organic and inorganic particles, dissolved solids, heavy metals, dyes, and other impurities contained in wastewater from various sources are toxic to the atmosphere and pose serious health risks to humans and animals. Coagulation–flocculation technology is commonly used in wastewater treatment to remove cell debris, colloids, and contaminants in a comfortable and effective manner. Flocculants, both organic and inorganic, have long been used in wastewater treatment. However, because of their low performance, non-biodegradability, and associated health risks, their use has been limited. The use of eco-friendly bioflocculants in wastewater treatment has become essential due to the health implications of chemical flocculants. Because of their availability, biodegradability, and protection, plant-derived coagulants/flocculants and plant-based grafted bioflocculants have recently made significant progress in wastewater treatment. This study will undoubtedly provide a clearer understanding of the current state, challenges, and solutions for bioflocculation in wastewater remediation using green materials for the sake of a cleaner climate.

Key words | biodegradable green bioflocculants, eco-friendly, environment, flocculation activity, plant-derived bioflocculants, wastewater treatment

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HIGHLIGHTS

- Plant mucilage as a cost-effective and eco-friendly natural bioflocculant.
- It can serve as a potential bioflocculant for treatment of wastewaters released from different sources.
- Significant removal of pollutants from wastewaters revealed the novelty of plant mucilage as a bioflocculant.
- It throws light on their applications aiming to exploit the green materials for remediation of wastewater.

INTRODUCTION

Water contamination has become more serious as a result of human economic growth all over the world. The persistent release of water polluted with various toxins into the atmosphere poses a risk to marine life as well as human health. As a result, there is a need to develop low-cost, high-efficiency methods for handling polluted water (Ahmed *et al.* 2020). Environmental pollution is largely caused by industrialization, urbanization, and intensive agricultural development, which produces vast volumes of wastewater that must be treated.

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Most dyes are poisonous, mutagenic, teratogenic, and carcinogenic to microorganisms, humans, and aquatic organisms, so wastewater containing dyes is considered a major environmental threat (Aljeboree *et al.* 2017). Textile, rubber, coating, and paper industries all use 'dyes' in their various manufacturing processes. Even at extremely low concentrations, these compounds can cause substantial pollution and, as a result, kill ecosystem elements. Photocatalysis, biodegradation, chemical coagulation, ion exchange, and adsorption are some of the currently available remediation techniques. Because of its high performance, ease of operation, low cost of operation, and ease of desorption, adsorption is one of the most useful methods in the treatment of aqueous solutions. However, the key operative point of adsorption is the selection or fabrication of the adsorbents, which must be highly stable, cost effective, environmentally friendly, and certainly successful. Activated carbon, zeolite, clay, chitosan, montmorillonite, and vermiculite are among the reactive materials that have been tested. However, these materials have many drawbacks, including a high cost, poor adsorption ability, and a low reuse rate, all of which restrict their use (Ahmed *et al.* 2020). Congo-red (CR) dye, a highly toxic and carcinogenic pollutant, has recently been degraded using gamma radiation (Shah *et al.* 2020). Furthermore, the removal of an organic dye by sonocatalysis using a TiO_2/Montmorillonite nanocomposite was previously reported (Khataee *et al.* 2015).

Heavy metal contamination, in addition to dyes, is a significant environmental issue, and heavy metal remediation remains a major problem. With rapid industrialization, the environmental hazards posed by heavy metals are becoming more serious (Vimala *et al.* 2020). Heavy metals from wastewater must be remedied for environmental protection, which necessitates the use of efficient, environmentally sustainable, and cost-effective materials.

The wastewater treatment technologies used should be cost effective, have a short processing period, and have a low environmental impact (Abdollahi *et al.* 2018). For the removal of various contaminants from wastewater, various treatment approaches such as ion exchange, membrane filtration, activated carbon, and coagulation/flocculation processes have been used (Al-Wasify *et al.* 2017; Rachdi *et al.* 2017). Coagulation/flocculation systems are the most commonly used wastewater treatment methods for eliminating dissolved pollutants, suspended solid (SS) particles, and emulsified oil (Ortiz-Oliveros & Flores-Espinosa 2019). These techniques enhance the aesthetic appearance of turbid water and are used to extract suspended particles by destabilizing them and creating larger, heavier flocs that help them settle.

Inorganic and organic polymer flocculants are the most common flocculants used in wastewater treatment. Inorganic flocculants are mostly made up of aluminium and iron, and they have a strong treatment effect on suspended solids and colloidal particles, but they have some disadvantages. Sludge produced during wastewater treatment with an aluminium salt flocculant is often used as a fertilizer in agriculture. However, high levels of aluminium in the soil pollute the atmosphere and damage plants. Furthermore, free aluminium ions pollute water by infiltrating lakes, rivers, and groundwater by infiltration, diffusion, deposition, and migration, resulting in water contamination and health risks. According to Tietz *et al.* (2019), traces of aluminium in foods cause acute carcinogenic and genotoxic diseases. Alzheimer's disease may be related to residues of aluminium left in treated wastewater or marine environments in this context (Zhang *et al.* 2015). The use of iron flocculant is also discouraged due to its corrosive nature. It has the ability to speed up the aging of equipment and boost the cost of water treatment (Porwal *et al.* 2015). Polyacrylamide and its derivatives are major ingredients in organic polymer flocculants, which have advantages such as quick flocculation, low dosage, and easy separation. However, the most widely used water treatment products, polyacrylamide or acrylamide sodium acrylate, have harmful health effects due to the presence of trace acrylamide monomers, which are potentially toxic and carcinogenic (Im *et al.* 2019; Liu *et al.* 2019). Because of the disadvantages of traditional flocculants, bioflocculants or natural flocculants should be used instead.

Bioflocculants have gained much attention because of their unique flocculating properties, as well as their non-toxic, biodegradable nature, which helps to create an environmentally friendly climate (Kothari *et al.* 2017; Shahadat *et al.* 2017). Bioflocculants have been shown to contain large quantities of nucleic acids, proteins, glycoproteins, and polysaccharides (Dao *et al.* 2016). Drinking water and wastewater treatment, removal of synthetic dyes and heavy metals, removal and recovery of cell biomass, emulsification, nanoparticle synthesis, mining, cryoprotection, and other fields all use extracellular biopolymer flocculants (Mu *et al.* 2018). Bioflocculants can be used as excellent alternatives to clarifying agents and have a wide variety of uses in the dairy, poultry, and other industries (Lee & Chang 2018). Several researchers have documented the use of bioflocculants for wastewater treatment (David *et al.* 2019; Dlamini *et al.* 2019; Maliehe *et al.* 2019; Pham & Bui

2020), heavy metal removal (Tawila *et al.* 2018; Agunbiade *et al.* 2019; Fan *et al.* 2019; Biswas *et al.* 2020), and dye decolorization (Chouchane *et al.* 2018).

Chemical flocculants are now limited in many industrial applications by many authorities because they are expensive, pose health risks, and may not be available locally. As a result, other flocculants that provide a new sustainability approach must be considered. This environmentally friendly process relies on the use of bioflocculants extracted from plant sources in the coagulation–flocculation treatment of wastewater to remove contaminants. Plant-based flocculants have recently gained more publicity because they are healthy, cost effective, biodegradable, non-toxic, do not cause secondary contamination, and can coagulate fine particles. This article reviews the production and current status of plant-derived flocculants and plant-based grafted bioflocculants, with the intention of shedding light on their potential applications in wastewater remediation. We hope that the research reports presented here will enable potential researchers to use plant-based bioflocculants as environmental remediation agents.

PLANT-DERIVED COAGULANTS/FLOCCULANTS

Plant-based coagulants/flocculants made from plant gum, vegetable extracts, tannins, seed kernels, and other sources are being studied in the field of bioflocculation. Amran *et al.* (2018) published a mini analysis on the coagulation/flocculation potential of plant-based materials based on ten years of research reports. Saleem & Bachmann (2019) published a recent study on the efficacy of various plants as sources of cationic, anionic, and non-ionic coagulants, suggesting their potential for widespread application and commercialization. Mohd-Salleh *et al.* (2019) evaluated over 16 plants and reported on the potential of natural materials as aids and their value as sustainable composite coagulants/flocculants.

Coagulants/flocculants have been isolated from a number of plant sources, including *Aloe vera* (Irma *et al.* 2015; Jaouadi *et al.* 2020), *Lepidium sativum* (Allafchian *et al.* 2019), *Moringa oleifera* (Agarwal *et al.* 2019; Zaid *et al.* 2019), *Salvia hispanica* (Tawakkoly *et al.* 2019), guar gum (Dwari & Mishra 2019), *Abelmoschus esculentus* (Lee *et al.* 2018), *Opuntia ficus-indica* (Miller *et al.* 2008; Sellami *et al.* 2014; Nharingo *et al.* 2015; Bouaouine *et al.* 2019), *Hibiscus esculentus* and *Trigonella foenum-graecum* (Jones & Bridgeman 2019), *Albizia* gum (Afolabi & Adekanmi 2017), malva nut gum (Ho *et al.* 2014), guar, mesquite seed gum and *Opuntia* mucilage (Carpinteyro-Urban & Torres 2013), *Plantago ovata* (Al-Hamadani *et al.* 2011), date palm (Khiari *et al.* 2010) and *Tamarindus indica* (Mishra *et al.* 2006). Because of their natural abundance, safe and cost-effective existence, natural products from acorn leaves (Benalia *et al.* 2019), banana fruit peel (Zaidi *et al.* 2019), lime seeds (Seghosime *et al.* 2017), and tamarind (Buenaño *et al.* 2019) have been identified as promising coagulants/flocculants (Muruganandam *et al.* 2017). The different sources of plant-based flocculants are depicted in Figure 1.

EXTRACTION OF PLANT-BASED COAGULANTS/FLOCCULANTS

Extraction of plant-based bioflocculants for wastewater treatment is a significant step toward creating a green and sustainable technology. Plant-derived flocculants have a mucilaginous texture, a neutral pH, and are polysaccharides in their natural state. The flocculating behaviour of mucilage is due to certain active ingredients in the mucilage. Mucilage is a heterogeneous mixture of monosaccharides present in plants, including arabinose, glucose, galactose, mannose, rhamnose, xylose, and uronic acid. In water, it exhibits viscous colloidal dispersion characteristics (Mirhosseini & Amid 2012). For the extraction of plant-derived coagulants/flocculants, two methods have been used so far: (i) solvent extraction and precipitation, and (ii) drying and grinding (Lee *et al.* 2014).

The mature seeds of *Moringa oleifera* were separated from their pods, shelled, ground, and sieved to extract the coagulant. The powder was then dried and used as a coagulant immediately (Sethupathy 2015). Active protein was extracted in another process by dissolving the powder in water, organic solvents such as alcohol or acetone, or salt solutions such as $MgCl_2$, KCl, or NaCl. Since the proteins in *M. oleifera* seeds are active agents, salt extraction was favored. Salt increases protein solubility, which aids in increasing *M. oleifera*'s coagulating ability against contaminants.

The preparation of flocculant from cactus (*Opuntia ficus-indica*) was reported by Sellami *et al.* (2014). Fresh cladodes were thoroughly rinsed, washed, cut into small parts, and crushed in a mixer to create a viscous mucilage that could be used as a coagulant or flocculant. To maintain the coagulating ability, the mucilage was held at a temperature below 4 °C for up to two weeks. The powdered coagulant and/or flocculant from cactus was made by oven-drying the cladodes and then grinding them. The

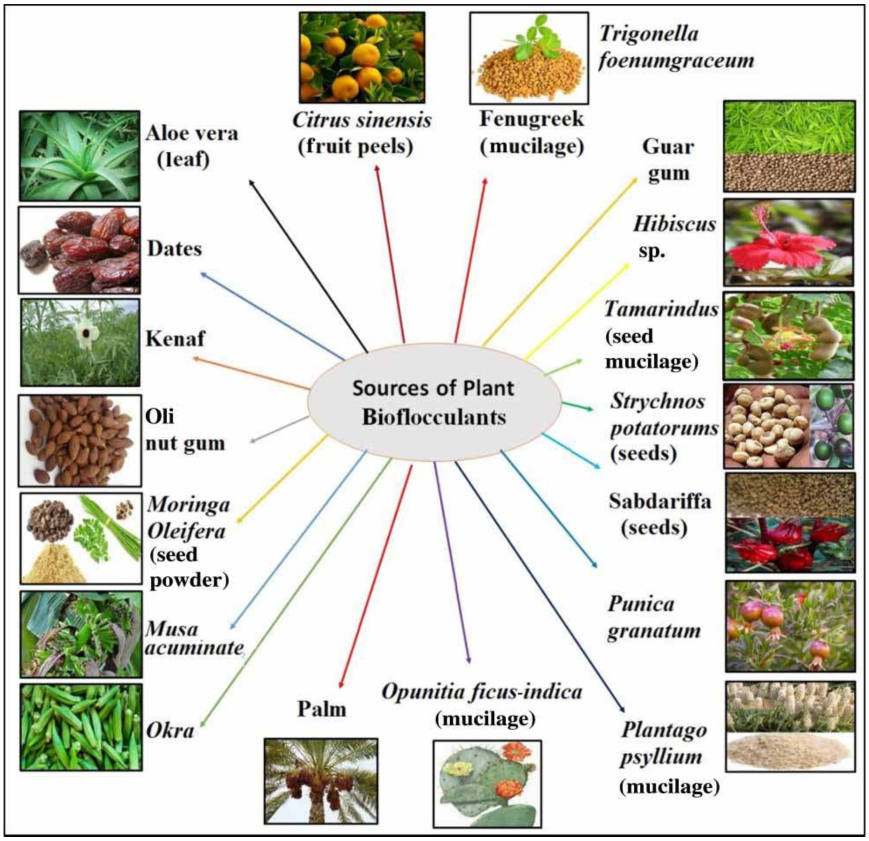

Figure 1 | Sources of plant-based bioflocculants.

coagulation function of cactus mucilage was lowest when oven dried at 120 °C or when only the skin of cladode was used (Miller *et al.* 2008). Furthermore, cactus coagulant/flocculant may be made from the mucilage or powder by combining it with water, salty water, or organic solvents. The use of water as a solvent should be promoted because it is environmentally friendly. In certain cases, however, the extract can be purified and then precipitated by alcohol, which can then be oven dried (Lassoued *et al.* 2018) or lyophilized (Bouaouine *et al.* 2019).

Ameena *et al.* (2010) documented the isolation of mucilage from *Hibiscus rosasinensis*. They gathered fresh leaves, washed them, and dried them. The dried powdered leaves were soaked in water for 5–6 minutes, then boiled for half an hour before being set aside for 1 hour to allow for full mucilage release into the water. The substance was squeezed out of a muslin cloth bag, and acetone was applied to the filtrate to precipitate the mucilage, which was then removed, dried, and stored at 50 °C. The dried powder was gathered, sieved, and stored in desiccators until required.

Mango seeds, like other plant sources, have a major coagulating effect due to their kernel content. The seed kernels were generally removed in the first step, then cleaned with distilled water and sliced. Then they were dried at various temperatures, such as 105 °C for 24 hours (Dange & Lad 2015a) or 120 °C for 1 hour (Qureshi *et al.* 2011), or sun-dried for 7 days and ground to make powder (Seghosime *et al.* 2017).

Although much research has been performed, there is no published report on the relationship between extraction process and flocculation quality. It is vital to understand whether the extraction process affects flocculation parameters such as pH, flocculant dose, and contact time. It may be important to optimize the extraction conditions for the development of a potential flocculant that can be compared to commercial flocculants in terms of efficiency and cost.

WASTEWATER TREATMENT USING PLANT-BASED FLOCCULANTS

Because of their non-toxic and biodegradable nature, plant-based bioflocculants have become popular as an alternative to synthetic flocculants in wastewater treatment. They are abundantly available from renewable resources and have no negative environmental effects (Lee *et al.* 2014). Plant-based flocculation was used to treat wastewater obtained from a number of sources, including textile, compost leachate, landfill leachate, tannery, palm oil mill effluent, sewage, food industry, glue industry, laundry, and dairy wastewater. To maximize flocculant yields, flocculation operation, and pollutant removal, culture conditions such as pH, temperature, and flocculant dosage were optimized. The results obtained differed depending on the plant sources used for bioflocculant processing and the characteristics of the wastewater.

The use of plant-derived biopolymers in wastewater treatment has been noted. Several plant parts, such as stems, leaves, seeds, husks, piths, and kernels, have been evaluated for their ability to coagulate/flocculate turbidity (TN), chemical oxygen demand (COD), and total suspended solids (TSS) from various types of wastewaters (Adwuyi & Adwumi 2018). To remediate a range of wastewaters (Torres *et al.* 2014), researchers have examined the coagulating–flocculating behaviour of various plant parts and extracts extracted from stems (Alwi *et al.* 2013), leaves (Sellami *et al.* 2014), seeds (Ngbolua *et al.* 2016), fruits shells and kernels waste (Jayalakshmi *et al.* 2016).

Textile wastewater

Moringa oleifera seeds have recently been identified as a source of coagulant for textile wastewater treatment (Agarwal *et al.* 2019). The seeds contain cationic and dimeric proteins that could work as a coagulant to eliminate turbidity and color. The findings showed that 95% of textile wastewater was degraded at pH 10 with a dose of *M. olifera* seeds of 16 mg/L. The application of the coagulant was suggested as a cost-effective approach for large-scale water treatment because of the substantial reduction in COD. In another research study, mucilage from the cactus cladodes (*Opuntia ficus-indica*) was used as a bioflocculant in conjunction with alum to treat textile effluent (Bouatay & Mhenni 2014). COD (88.76%), TN (91.66%), and dye (99.84%) were found to have the highest removal rates. The use of cress seed mucilage magnetic nanocomposite to extract methylene blue dye from water has been confirmed (Allafchian *et al.* 2019). Tannins are polyphenol compounds found in the leaves, roots, bark, seeds, wood, and fruits of plants. Lopes *et al.* (2019) examined the decolorization of synthetic dye effluents using a tannin-derived coagulant in a coagulation–flocculation process and compared it to a conventional inorganic coagulant, iron (III) sulfate. Two effluents were examined containing: (1) direct azo dye and salts and (2) dyeing auxiliary chemicals. The tannin coagulant was found to be efficient for color removal in both synthetic and natural textile effluents, eliminating all dyes completely over a pH range of 4–9. pH had a greater effect on the efficiency of iron (III) sulfate. At higher pH, the tannin coagulant performed better than the iron coagulant. Since it did not demonstrate substantial dye removal after a long period of contact time, the bioflocculant derived from *Tamarindus indica* (tamarind) could not be used as an effective flocculant for the removal of dyes such as direct (direct quick scarlet) and vat (golden yellow) from textile wastewater. For the highest flocculating efficiency of plant-based bioflocculants, the acceptable pH range was neutral in most of the recorded studies. Depending on the form and characteristics of treated wastewater, some were found to be stable in acidic or alkali environments.

Compost leachate wastewater

Tawakkoly *et al.* (2019) used a natural coagulant, the mucilaginous extract of *Salvia hispanica* seeds, to treat compost leachate wastewater. COD and TN removal from compost leachate wastewater was tested. Response surface methodology was used to optimize the coagulation–flocculation phase. Under optimal conditions, using 40 g/L of coagulant at pH 7, the TN removal and COD reduction percentages were found to be 62.4% and 39.7%, respectively.

Landfill leachate

The ability of psyllium husks as a coagulant aid and coagulant for the treatment of landfill leachate was investigated. When used as a primary coagulant, psyllium husk was found to be ineffective. When combined with polyaluminium chloride (PACl), it was more successful as a coagulant aid, removing 64% of COD, 90% of color, and 96% of TSS (Al-Hamadani *et al.* 2011).

Dairy wastewater

Moringa oleifera powder with particle sizes ranging from 150 to 425 μm was used to treat dairy wastewater with a turbidity of 230 NTU and a COD of 2,240 mg/L (Pallavi & Mahesh 2013). In terms of COD (64.2%) and TN (86%) exclusion, the finest particle gave the best performance.

Glue industry wastewater

Sellami *et al.* (2014) used cactus juice as a bio-flocculant to treat a glue industry effluent (pH 6.7, suspended matter (SM): 270 mg/L, and COD: 99,200 mg/L). The removal rates for SM and COD were found to be 83.3% and 67.5%, respectively. The removal percentages for SM (79.6%) and COD (59.1%) were noted during polyacrylamide (PAM) testing.

Tannery wastewater

A cactus juice-based coagulant was used to treat tannery effluent with a turbidity of 66–960 NTU and COD of 28,000 mg/L. The removal of TN (78.5%) and COD (80.6%) was found to be successful. They also mentioned *Moringa oleifera* seeds being used to treat tannery wastewater. The removal of TN and COD was found to be 82.0 and 83.3%, respectively (Kazi & Virupakshi 2013).

Laundry wastewater

The efficacy of an okra-based flocculant in treating laundry wastewater (pH: 5–6) was demonstrated (Freitas *et al.* 2015). The use of this natural resource as a flocculant improved the coagulation–flocculation process. The authors indicated that color (93.6%), TN (97.2%), and COD (85.7%) were all significantly removed. De-Souza *et al.* (2014) used *Opuntia ficus-indica* mucilage to treat jeans laundry effluent and found that COD removal was approximately 64.8% and TN elimination was 91.3%. Al-Gheethi *et al.* (2017) found that *Moringa oleifera* seeds had a higher TN removal efficiency (84%) than $FeSO_4$ in a study on laundry wastewater treatment (59%). However, COD removal was 47.2%, which appears to be equivalent to $FeSO_4$ removal (54%). *Moringa oleifera* was also found to have better settling characteristics than $FeSO_4$.

Palm oil mill wastewater

The seeds of *M. olifera* were used to treat a palm oil mill effluent with COD (40.2 mg/L) and an initial SM (17.9 mg/L) as a possible source of effective coagulant. It allowed for the removal of COD (92%) and SM (52%) respectively. Furthermore, they found that combining *M. olifera* with a commercial flocculant (NALCO 7751) greatly improved COD and SM elimination. Ullah & Rathnasiri (2015) also stated that using mango seed powder to treat a palm oil mill effluent allowed the effective removal of SM (96%), COD (89%), and color (97%) respectively.

Sewage wastewater

The capacity of mango kernels to coagulate was investigated for the purpose of cleaning wastewater (Dange & Lad 2015a, 2015b). Mango kernels were used as the sole coagulant, resulting in removal efficiencies of 31.6% for SM and 33.4% for COD. When alum and mango were combined, however, the removal rate increased to 66.8% for SM and 69.2% for COD. Mango extract has gained importance as an important coagulant, according to research studies. Aloe gel (Alg), a natural extract from the *Aloe* sp. plant, was recently tested to see whether it could be used as a bio-flocculant in the treatment of urban wastewater sewage sludge (Jaouadi *et al.* 2020). Under monitored laboratory conditions, the gel was used alone and in combination with water glass (WG). Alg was found to be successful in settling the flocculated sludge quickly and removing the sludge's unpleasant odours. It was discovered to be pH resistant, with no effect on the pH of the wastewater. To improve Alg's treatment efficiency, WG was tested as an alkali agent, which decreased ammonia (NH_4-N) and COD levels in the wastewater.

Wastewater containing heavy metals

Plant-based bioflocculants were found to be capable of removing heavy metals from both synthetic and real wastewater, in addition to organic pollutant removal. Adsorption and coagulation methods were used to remove cadmium (II) from wastewater using *M. oleifera* seeds (Swelam *et al.* 2019). pH, contact time, initial Cd(II) concentration, and temperature were used to compare the increase in Cd(II) removal percentage with the coagulation and adsorption technique. This research was critical in deciding on an environmentally safe water treatment strategy for removing Cd(II) from contaminated effluent. Nharingo *et al.* (2015) investigated the use of the *Opuntia ficus-indica* cactus as a low-cost, readily accessible, and environmentally friendly bioflocculant for the removal of heavy metals from wastewater. The pH, contact time, temperature, initial concentration of lead (II) ions, particle size, dosage, and ionic strength of the coagulation–flocculation process were all optimized. After using *Opuntia ficus-indica* powder, heavy metals such as Cd (84.16%), Zn (85.74%), Cu (93.02%), and Pb (100%) were removed under optimal conditions. The bioflocculant removed lead from wastewater with impressive effectiveness, which could be tested on a commercial scale. Table 1 displays a variety of wastewaters that have been treated with plant-derived flocculants.

TURBIDITY REMOVAL

Turbidity is a critical parameter that must be tested for both wastewater treatment and healthy drinking water use. The presence of fine clay, silt, inorganic and organic elements, plankton, and microorganisms, among other things, causes it. According to a World Health Organization (WHO) survey, the maximum turbidity that can be used for disinfection is 0.1 Nephelometer Turbidity Units (NTU) (WHO 2008). The efficacy of disinfection methods can be greatly improved by reducing turbidity. *M. oleifera* seed powder was used as a biocoagulant in the water treatment process to remove turbidity (Zaid *et al.* 2019). Response surface methodology (RSM) was used to assess the effects of different parameters on turbidity removal. Lower turbidity (<5 NTU) obtained from the analysis demonstrated the possible use of *M. oleifera* seeds as an important coagulant for water treatment. The turbidity of the water was found to be less than the recommended value under optimal conditions, confirming the capacity of *M. oleifera* seed powder for the treatment of industrial and domestic river water. In the presence of KNO_3, Dwari & Mishra (2019) recorded the use of guar gum as an effective flocculant that showed excellent turbidity removal performance. The bioresource *Opuntia ficus-indica* was discovered to be a strong source of bioflocculant for turbidity removal (Bouaouine *et al.* 2019). There were two molecules found in the bioflocculant that had synergistic flocculating power and operated in a bridging adsorption mode. The removal of turbidity was estimated to be 70%. Jones & Bridgeman (2019) used crude extracts of okra, kenaf, and sabdariffa as coagulants to investigate the characteristics of treated water using fluorescence excitation–emission matrices (EEMs). Purified proteins from okra, sabdariffa, and kenaf were also tested for their effect. Fluorescence analysis verified that the purified

Table 1 | Plants as sources of bioflocculants used for wastewater treatments

Common name	Scientific name	Plant parts used for bioflocculant preparation	Types of wastewater	References
Aloe	*Aloe vera*	Gel	Urban wastewater sewage sludge	Jaouadi *et al.* (2020)
Salvia	*Salvia hispanica*	Mucilage from seed	Compost leachate wastewater	Tawakkoly *et al.* (2019)
Cress	*Lepidium sativum*	Mucilage	Dye bearing wastewater	Allafchian *et al.* (2019)
Okra	*Abelmoschus esculentus*	Seed pods	Laundry wastewater	Freitas *et al.* (2015)
		Seed pods	Synthetic turbid water	Fahmi *et al.* (2014)
		Seed pods	Synthetic wastewater:	Anastasakis *et al.* (2009)
Cactus	*Opuntia ficus-indica*	juice	Industrial wastewater containing heavy metals viz. cadmium, chromium	Srarfi *et al.* (2018)
		Powder	Wastewater containing heavy metal ((lead)	Nharingo *et al.* (2015)
		gum	laundry effluent	De Souza *et al.* (2014)
		Juice	Textile effluent:	Bouatay & Mhenni (2014)
		Juice	Glue industry wastewater	Sellami *et al.* (2014)
		Juice	Food industry effluent	Sellami *et al.* (2014)
		Cladode	Synthetic wastewater	Belbahloul *et al.* (2014)
		Powder	Tannery effluent	Kazi & Virupakshi (2013)
		Cladode	Industrial effluent	Abid *et al.* (2008)
Drumstick	*Moringa oleifera*	Seed	Textile wastewater	Agarwal *et al.* (2019)
		Seed	Wastewater containing heavy metal (cadmium)	Swelam *et al.* (2019)
		Seed	Laundry wastewater	Al-Gheethi *et al.* (2017)
		Juice	Urban wastewater	Rachdi *et al.* (2017)
		Seed	Tannery effluent	Kazi & Virupakshi (2013)
		Seed	Dairy wastewater	Pallavi & Mahesh (2013)
		Seed	Dye bearing wastewater	Beltrán-Heredia *et al.* (2009)
Mango	*Mangifera indica*	Seed kernels	Synthetic turbid water	Seghosime *et al.* (2017)
		Seed kernels	Palm oil mill effluent	Ullah & Rathnasiri (2015)
		Seed kernels	Sewage wastewater	Dange & Lad (2015a, 2015b)

proteins could extract dissolved organic carbon (DOC) from raw water. Srarfi *et al.* (2018) described the use of cactus *Opuntia* bioflocculant to treat industrial wastewater. TN was removed from industrial wastewater by the bioflocculant (19.59%). Rachdi *et al.* (2017) used a bioflocculant from the same plant to treat urban wastewater and found that TN (93.65%), SS (82.75%), and COD (64.30%) were all removed. Malva nut gum (MNG) was used as a safe and sustainable source for water treatment because it had excellent flocculation efficiency at low concentrations (Ho *et al.* 2014). It worked well as a powerful anionic flocculant, conformed well to the surface of particles with a branch-like surface structure, and performed well in water treatment. MNG (0.06 mg/L) and Fe (0.08 mg/L) were combined to make the composite Fe-MNG, which was used to treat water at pH 3.01. The *Aloe vera* leaf gel was evaluated as a flocculant and turbidity removal was assessed (Irma *et al.* 2015). The plant's clarification tests revealed a high level of SS, TN (72%), and color removal (15%). *Strychnos potatorum* seeds are said to act as a natural coagulant, allowing turbid waters to be clarified (Deshmukh *et al.* 2013). For the coagulation of SS, wastewater was treated with natural and synthetically prepared *S. potatorum*. The natural coagulant properties of *S. potatorum* were compared to those of alum, a conventional chemical coagulant. Lower dosages of *S. potatorum*, ranging from 0.25 to 3.5 mg/L, were found to be effective as coagulants, with higher turbidity of 1000–3000 NTU. Table 2 compares the effectiveness of different plant-derived flocculants in removing TN.

MECHANISM OF FLOCCULATION

There has been a surge in interest in the coagulation–flocculation method for wastewater treatment. Polymer bridging, electrostatic patching, and charge neutralization are all common flocculation mechanisms. As shown in Figure 2, polymer bridging is the most widely accepted mechanism for plant-based bio-flocculation. Depending on the particle present in wastewater, biopolymers function as a bridge, creating a particle–polymer–particle complex (Bolto & Gregory 2007). Polymer bridging has been proposed as a potential mechanism for flocculation during the treatment of textile effluent with *Tamarindus indica* mucilage (Mishra & Bajpai 2006) and *Plantago psyllium* mucilage (Mishra & Bajpai 2005). According to studies, the molecular

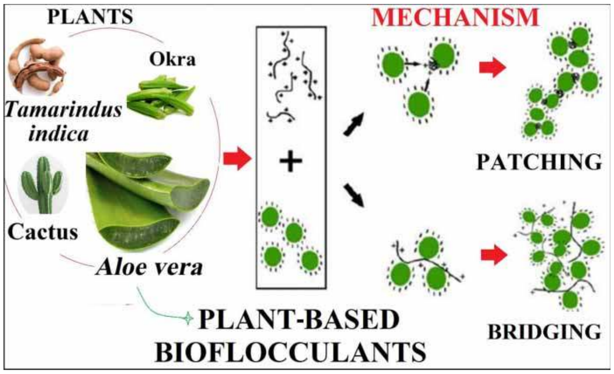

Figure 2 | Mechanism of flocculation using plant-based bioflocculants.

Table 2 | Comparison of turbidity (TN) removal efficiency using various plant-derived flocculants

Types of wastewater	Turbidity removal efficiency (%)	Plants as sources of flocculants	References
Compost leachate wastewater	62.4	*Salvia hispanica*	Tawakkoly *et al.* (2019)
Synthetic turbid water	<5.0 NTU	*Moringa oleifera*	Zaid *et al.* (2019)
Industrial wastewater	19.5	*Opuntia ficus-indica*	Srarfi *et al.* (2018)
Urban wastewater	93.6	*Opuntia ficus-indica*	Rachdi *et al.* (2017)
Laundry wastewater	97.2	*Abelmoschus esculentus*	Freitas *et al.* (2015)
	91.2	*Opuntia ficus-indica*	De Souza *et al.* (2014)
	83.6	*Moringa oleifera*	Al-Gheethi *et al.* (2017)
Textile effluent	91.6	*Opuntia ficus-indica*	Bouatay & Mhenni (2014)
Tannery effluent	82.0	*Moringa oleifera*	Kazi & Virupakshi (2013)
	78.5	*Opuntia ficus-indica*	Kazi & Virupakshi (2013)
Synthetic turbid water	72,0	*Aloe vera*	Irma *et al.* (2015)
Synthetic turbid water	86.0	*Moringa oleifera*	Qureshi *et al.* (2011)
	98.0	*Mangifera indica*	Qureshi *et al.* (2011)

weight and charge density of the flocculant played a critical role in determining its effectiveness (Morrissey *et al.* 2020). Experiments on municipal sludge dewatering revealed that hydrophobic association caused by copolymerization increased bridging and sweeping effects, promoting floc growth (Zhou *et al.* 2020). Furthermore, the flocs produced had a higher mechanical strength and were less compressible, resulting in improved permeability to mechanical dewatering.

Charge neutralization is a process that only works when the flocculants and colloid suspended particles have opposite charges. Adsorption and charge neutralization were the key mechanisms of coagulation/flocculation in *Moringa oleifera*. Proteins (34%) are contained in the seeds, as well as carbohydrates (15%) and lipids (15.5%). This high protein content may be the most effective coagulation agent. The protein's short chain and cationic surface charge allow it to adsorb and neutralize negatively charged colloids in water (Bolto & Gregory 2007).

Impurity particles may also be found to be negatively charged. Bioflocculants isolated from psyllium, okra, and Isabgol have been stated to have anionic charges, while fenugreek flocculants have neutral charges. The majority of confirmed bioflocculants have either anionic or neutral charges, according to reports (Lee *et al.* 2014). The interaction of solid waste with mucilage can result in flocculation. There was contact between the free hydroxyl groups of the polysaccharide and the contents of the wastewater when solid waste was treated with polysaccharides. During the flocculation phase, improvements in the composition of crystalline waste materials in wastewater were observed (Mishra *et al.* 2002, 2003; Mishra *et al.* 2004a, 2004b).

The flocculation mechanism of *Opuntia ficus-indica* mucilage was discovered. Galacturonic acid (8–12.7%), L-arabinose (24.6–42%), D-xylose (22–22.2%), D-galactose (21–40.1%), and L-rhamnose (7–13.1%) are all found in the mucilage of *Opuntia ficus-indica* (Nobel *et al.* 1992). Galacturonic acid, also known as polygalacturonic acid, is the most active compound that has a high coagulating activity (over 50%) when used alone to remove TN. Mucilage containing arabinose, galactose, rhamnose, and xylose, conversely, only reduces TN by 30% (Miller *et al.* 2008). Polygalacturonic acid interacts with anionic or cationic pollutants through its long anionic chain, which includes carboxyl (–COOH), carbonyl (–C = O), and hydroxyl (–OH) classes. As a result, the mechanism of flocculation is deprotonation of the OH group, which increases anionic pollutant adsorption and causes agglomeration. Furthermore, according to Theodoro *et al.* (2013), the oxidation of the terminal carbon OH edge to a COOH group induces cationic pollutant adsorption and coagulation–flocculation, and pollutant elimination from wastewater occurs by adsorption and bridging. Nharingo *et al.* (2015) also agreed with this mechanism.

According to Lee *et al.* (2014), bioflocculation is a direct flocculation process that is more effective than conventional coagulation–flocculation due to the high charge density of amphoteric plant-derived coagulants/flocculants. Because of the dual presence of anionic and cationic active compounds, biopolymers are capable of bridging at any pH condition, so pH adjustment may not be necessary quite often (Mukhtar *et al.* 2015). In the case of a direct flocculation operation, bridging mechanisms and charge neutralization can occur simultaneously.

The fractionated components of *Opuntia ficus-indica* were used to create a bi-molecular principle in flocculation (Bouaouine *et al.* 2019). The flocculant, which was created using a three-step protocol of extraction, precipitation, and fractionation, showed 84% flocculation efficiency. Bridging and adsorption were found to be the underlying mechanisms on synthetic kaolin. Hassimi *et al.* (2020) demonstrated the importance of biomolecules in flocculation by studying novel anionic bioflocculants derived from fermented fractions of sago mill effluent and palm oil mill effluent. Due to charge neutralization and intra-particle bridging, fractions containing xylose and glucose were found to have a high flocculation quality.

It is safe to assume that research into the flocculation process for the possible use of plant-based flocculants in wastewater treatment is still minimal. As a result, researchers should keep looking for the mechanism behind the successful use of new coagulants/flocculants extracted from different plant sources that has yet to be discovered.

WASTEWATER TREATMENT USING PLANT-BASED GRAFTED BIOFLOCCULANTS

Plant-based grafted bioflocculants have received much attention in recent years as a way to boost flocculation properties and resolve disadvantages like unregulated biodegradability and varying efficiency due to different processing conditions (Ferasat *et al.* 2020). To extend shelf life and boost flocculating efficiency, natural polysaccharide biodegradability must be carefully monitored. Synthetic flocculants, conversely, are highly efficient and have a long shelf life, but they are non-biodegradable and environmentally harmful (Makhtar *et al.* 2020). The development of grafted copolymers can be used to enhance the properties of natural and synthetic polymers.

The aim of plant-based grafted copolymerization is to create a unique modified polymer that combines the best features of both groups. It has been demonstrated that grafting synthetic polymer branches onto the rigid backbone of natural polymers can result in efficient, relatively shear stable, and environmentally friendly flocculants. According to some research studies, acrylamide-grafted natural polymers like amylopectin, guar gum, and xanthan gum, as well as starch and sodium alginate, are widely used as flocculants. Polyacrylamide (PAM) was grafted onto the backbone of plant-based biopolymers (*Plantago ovata*, *Plantago psyllium*, and *Tamarindus indica*) and demonstrated significantly improved flocculating properties as compared to non-grafted natural polysaccharides (Ferasat *et al.* 2020). Table 3 summarizes the flocculating efficiency of all plant-based grafted bioflocculants published in the literature.

Textile water treatment

Wastewater treatment is critical for ensuring a sustainable supply of clean water, especially for human consumption. In wastewater treatment, green or natural bioflocculants could resolve the drawbacks of chemical flocculants. For flocculant grafting, galactomannans, a branched natural biopolymer with good biodegradability, biocompatibility, and sustainability, was recently used. Sharma *et al.* (2020) used Taguchi's method of rigorous design of experiments with 3-chloro-2-hydroxypropyltrimethylammonium chloride (CHPTAC) under alkaline heterogeneous conditions to demonstrate the quaternization of galactomannan derived from *Cassia tora* gum (QCTG) under alkaline heterogeneous conditions, which could be used in a variety of industries for end-use applications (Sharma *et al.* 2020).

Municipal sewage water treatment

Lee *et al.* (2018) published a study on a techno-economic analysis of industrial scale bioflocculant processing using okra as a biomass feedstock. Using continuous mode microwave extraction at 90 °C, a residence period of 10 minutes, a water loading of 3.5 w/w, and a production rate of 220 tonnes per year, the biosludge flocculant's dewatering capacity was optimized and showed significant results (Lee *et al.* 2018).

Furthermore, Guo *et al.* (2019) described a new modified rice straw biochar (RSB) that improved sludge dewaterability and served as a skeleton construct. Charge neutralization was the process used to improve the dewatering of the sludge. The sludge system was dosed with a changed RSB filled with positively charged aluminium, which destroyed the sludge colloidal system's stability and resulted in easier sludge particle congregation, which increased sludge dewatering. Another important mechanism was the development of skeleton structures in the sludge cake to facilitate water passage by lowering the sludge's extracellular polymeric substances (EPS) (Guo *et al.* 2019).

Tannery water treatment

Furthermore, Chen *et al.* (2020) demonstrated an octopus-like lignin-grafted cationic polyacrylamide (L-CPA) with excellent flocculation performance, was inexpensive, environmentally friendly, and technically feasible, indicating a promising future in wastewater treatment. Using acrylamide (AM), methylacryloyloxyethyltrimethyl ammonium chloride (DMC), and enzymatic hydrolysis lignin (EHL) as raw materials, a high-efficiency L-CPA bioflocculant was synthesized. In this case, a linear pre-polymer of cationic polyacrylamide (CPA) terminated with chlorine was first prepared, and then grafted onto EHL using chlorine reactions with phenolic hydroxyl groups in lignin molecules.

The main mechanism behind this formulation was that the prepared L-CPA could self-assemble into octopus-like nanospheres with CPA segments suspended in water and hydrophobic lignin skeletons condensed in the center, giving the L-CPA excellent flocculation efficiency for kaolin suspension under acidic, neutral, or alkaline conditions ($pH = 5$–9). For flocculation of the kaolin suspension, only a small amount of L-CPA (4.0–4.5 mg/L) was needed (Figure 3).

Coal mine and iron ore water treatment

Furthermore, a recent study demonstrated the formulation of polyacrylamide-grafted fenugreek gum (FG-g-PAM) as a bioflocculant. Fenugreek gum was modified in this study using two different graft copolymerization methods: microwave-assisted and thermal synthesis. The effective-grafted product was created using an optimized concentration of acrylamide monomer and ceric ammonium nitrate (CAN) initiator. In the refloccculation method, the flocculation efficacy was significantly increased from 92.05 to 95.98% in this analysis. In kaolin, coal fine, and iron ore suspensions, the flocculation efficacy was found to be 95.98%,

Table 3 | Wastewater treatment using plant-based grafted bioflocculants

Plant-based grafted bioflocculant	Grafting method	Treated wastewater	Parameters	Grafted-bioflocculant dosage (ppm)	Flocculation efficiency	References
Quaternization of *Cassia tora* gum galactomannan (QCTG) i.e. 3-chloro-2-hydroxypropyltrimethylammonium chloride (CHPTAC), quaternary ammonium group grafted on the backbone of galactomannan	Taguchi L'16 (4^5) orthogonal array design	Textile, cosmetics, pharmaceutical and as wet-end additives in paper industry	2 h at 30 °C, sodium hydroxide, (0.0125 mol), CHPTAC, (0.00319 mol) and gum–liquor ratio (1: 15)	QCTG (DS = 0.112)	Turbidity	Sharma *et al.* (2020)
Rice straw biochar (RSB) modified by aluminium chloride	Charge neutralization, incompressible and permeable sludge cakes (skeleton structures builder)	Dewatering of the sludge from municipal sewage treatment plant	30 mins, SV_{30}%, specific resistance to filtration (SRF) (79.8%, 1.2×10^{12} m/kg), moisture content (MC) (81.4%) and capillary suction time (CST) (38 s)	0.3 g RSB/g (dry sludge)	19.4 kg/(m^2·h) of dewatering	Guo *et al.* (2019)
Octopus-like lignin-grafted cationic polyacrylamide (L-CPA) flocculant	Self-assembly, grafting to strategy using acrylamide (AM), methylacryloyloxyethyltrimethyl ammonium chloride (DMC) and enzymatic hydrolysis lignin (EHL)	Water flocculation	30 mins, pH = 5	4.0–4.5 mg/L of L-CPA	Kaolin suspension, L2-CPA-3 (92%), L2-CPA-4 (90%) and L2-CPA-5 (82%)	Chen *et al.* (2020)
Polyacrylamide-grafted fenugreek gum (FG-g-PAM)	Microwave-assisted and thermal synthesis technique of graft copolymerization	Wastewater treatment by deflocculation and refloccculation	15 min, pH 4 at 25 °C	1 ppm	96% turbidity removal by microwave-assisted process, 89% efficiency of turbidity by thermally synthesized grade	Mishra & Kundu (2019)
Plant-based *Tacca leontopetaloides* biopolymer flocculant (TBPF)	Gelatinization	Leachate treatment	pH 3	240 mg/L	Turbidity reduced from 218 to 45.8–54.5 NTU, total suspended solids (TSS) from 214 to 19.3–19.9 mg/L and color from 14201 to 852–994 PtCo	Makhtar *et al.* (2020)

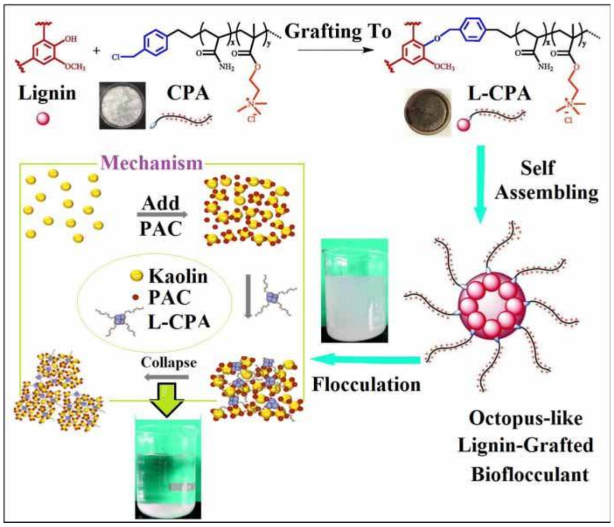

Figure 3 | Formulation of octopus-like lignin-grafted bioflocculant and its mechanism.

40.52%, and 68.94%, respectively. In comparison to chemical flocculants, the novel grafted flocculant was predicted to be a highly effective flocculant in the future (Mishra & Kundu 2019).

Leachate water treatment

Furthermore, Makhtar *et al.* (2020) investigated a plant-based *Tacca leontopetaloides* biopolymer flocculant (TBPF) for leachate treatment, which showed excellent removal of TN, TSS, and color. The tuber of the *Tacca leontopetaloides* plant was used to extract a new natural-based bioflocculant, which was then processed via gelatinization to create TBPF for leachate treatment. The presence of –COOH and –OH groups in TBPF matched bioflocculant properties for leachate therapy. In this analysis, 240 mg/L of TBPF at pH 3 was found to be very effective in reducing turbidity, TSS, and color from 218 NTU, 214 mg/L, and 14201 PtCo to 45.8–54.5 NTU, 19.3–19.9 mg/L, and 852–994 PtCo, respectively, indicating that this new plant-based TBPF has much potential for leachate and other wastewater treatment applications.

The development of superior flocculation technology has so far focused on combining the properties of chemical and biological flocculants, with the primary goal of lowering flocculant dosage and improving flocculation efficiency. PAM has been considered as a suitable co-polymer graft for this reason. PAM's neurotoxicity, conversely, may be the big discrepancy. As a result, further research into the development of natural and healthy organic co-grafts that could replace acrylamide is needed. In this regard, Ferasat *et al.* (2020) have recently identified yeast cell wall (YCW) as a so-called safe polymer. This polymer has recently been shown to be an efficient kaolin coagulation matrix in an aqueous environment.

CONCLUSION

The possible use of plant-derived flocculants and plant-based grafted bioflocculants as a replacement for traditional chemical coagulants/flocculants in wastewater treatment was examined in this study. Bioflocculants derived from various plant sources have been studied extensively, and the potential use of these green flocculants for wastewater treatment has been identified. They also shown important results in the removal of SS, TN, and COD, total nitrogen, dye, and heavy metals from wastewater. These products' eco-friendliness has been well known, and they have paved the way for the implementation of green chemistry, clean, healthy, and cost-effective technology. Plant-based flocculants can, in reality, be used to replace chemical flocculants, which have been shown to have negative effects on the environment and human health. The data on green flocculant production conditions are needed to develop a comprehensive strategy for scientific research and commercial implementation in wastewater treatment. Many plant-based flocculants showed substantial flocculating behaviour in the laboratory, with pollutant removal efficiency exceeding 90% depending on the type of plant material used to make the flocculant and the characteristics of the wastewater. Plant-based grafted copolymerization has been shown to be an efficient green flocculant for wastewater treatment, containing the best properties of both the plant and polymer groups. Green flocculants for wastewater treatment have been found to be efficient, shear stable, and environmentally friendly by researchers. However, extensive research is needed to optimize bioflocculation procedures for each form of wastewater treatment based on the plants and polymers that should be statistically optimized to improve flocculation operation. Statistical optimization, which should be the primary focus and potential research prerequisite for realistic applications, has received little attention from researchers so far. Furthermore, several studies are required to understand the process of bioflocculation, taking into account the changes in various treatment systems. Finally, the effectiveness of green flocculants should be tested on a broad scale, in real-world environments, and for a wide range of wastewater systems, followed by a techno-economic evaluation.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

Abdollahi, K., Yazdani, F., Panahi, R. & Mokhtarani, B. 2018 Biotransformation of phenol in synthetic wastewater using the functionalized magnetic nano-biocatalyst particles carrying tyrosinase. *3 Biotech* **8**, 419.

Abid, A., Zouhri, A. & Ider, A. 2008 Utilisation d'un nouveau bio-floculant extrait du cactus marocain dans le traitement des rejets chargés de chrome (VI) par le procedé de coagulation floculation. *Revue Des Energies Renouvelables* **11** (2), 251–257.

Adwuyi, A. S. & Adwumi, J. R. 2018 Optimisation of coagulation-flocculation process for the treatment of wastewater using inorganic and three natural coagulants. *Journal of Engineering and Engineering Technology* **12** (2), 260–265.

Afolabi, T. A. & Adekanmi, D. G. 2017 Characterization of native and graft copolymerized albizia gums and their application as a flocculant. *Journal of Polymers* **8**, 1–8.

Agarwal, V., Dixit, D. & Bhatt, M. J. 2019 Use of *Moringa oleifera* seeds as a primary coagulant in textile wastewater treatment. In: *Waste Management and Resource Efficiency* (S. Ghosh, ed.). Springer, Singapore, pp 1231–1236.

Agunbiade, M. O., Pohl, C., Heerden, E. V., Oyekola, O. & Ashafa, A. 2019 Evaluation of fresh water actinomycete bioflocculant and its biotechnological applications in wastewaters treatment and removal of heavy metals. *International Journal of Environmental Research and Public Health* **16** (18), 3337.

Ahmed, D. N., Naji, L. A., Faisal, A. A., Al-Ansari, N. & Naushad, M. 2020 Waste foundry sand/MgFe-layered double hydroxides composite material for efficient removal of Congo red dye from aqueous solution. *Scientific Reports* **10** (1), 1–12.

Al-Gheethi, A. A., Mohamed, R. M. S. R., Wurochekke, A. A., Nurulainee, R. N., Rahayu, J. M. & Amir, H. M. K. 2017 Efficiency of *Moringa oleifera* seeds for treatment of laundry wastewater. In: MATEC Web of Conferences, *EDP Sciences*, **103**, 06001.

Al-Hamadani, Y. A., Yusoff, M. S., Umar, M., Bashir, M. J. & Adlan, M. N. 2011 Application of psyllium husk as coagulant and coagulant aid in semi-aerobic landfill leachate treatment. *Journal of Hazardous Materials* **190** (1–3), 582–587.

Aljeboree, A. M., Alshirifi, A. N. & Alkaim, A. F. 2017 Kinetics and equilibrium study for the adsorption of textile dyes on coconut shell activated carbon. *Arabian Journal of Chemistry* **10**, S3381–S3393.

Allafchian, A., Mousavi, Z. S. & Hosseini, S. S. 2019 Application of cress seed mucilage magnetic nanocomposites for removal of methylene blue dye from water. *International Journal of Biological Macromolecules* **136**, 199–208.

Al-Wasify, R. S., Ali, M. N. & Hamed, S. R. 2017 Biodegradation of dairy wastewater using bacterial and fungal local isolates. *Water Science and Technology* **76** (11), 3094–3100.

Alwi, H., Idriss, J., Musa, M. & Hamid, K. H. K. 2013 A preliminary study of banana stem juice as a plant-based coagulant for treatment of spent coolant wastewater. *Journal of Chemistry* **2013**, 1–7.

Ameena, K., Dilip, C., Saraswathi, R., Krishnan, P. N., Sankar, C. & Simi, S. P. 2010 Isolation of the mucilages from *Hibiscus rosasinensis* Linn. and Okra *(Abelmoschus esculentus* Linn.) and studies of the binding effects of the mucilages. *Asian Pacific Journal of Tropical Medicine* **3** (7), 539–543.

Amran, A. H., Zaidi, N. S., Muda, K. & Loan, L. W. 2018 Effectiveness of natural coagulant in coagulation process: a review. *International Journal of Engineering and Technology* **7** (3.9), 34–37.

Anastasakis, K., Kalderis, D. & Diamadopoulos, E. 2009 Flocculation behaviour of mallow and okra mucilage in treating wastewater. *Desalination* **249** (2), 786–791.

Belbahloul, M., Anouar, A. & Zouhri, A. 2014 Low technology water treatment: investigation of the performance of cactus extracts as a natural flocculant for flocculation of local clay suspensions. *International Journal of Engineering Research and Technology (IJERT)* **3** (3), 2240–2245.

Beltrán-Heredia, J., Martin, J. S., Delgado-Regalado, A. & Jurado-Bustos, C. 2009 Removal of dyes by *Moringa oleifera* seed extract. Study through response surface methodology. *Journal of Chemical Technology and Biotechnology* **84**, 1653–1659.

Benalia, A., Derbal, K., Panico, A. & Pirozzi, F. 2019 Use of acorn leaves as a natural coagulant in a drinking water treatment plant. *Water* **11** (1), 57.

Biswas, J. K., Banerjee, A., Sarkar, B., Sarkar, D., Sarkar, S. K., Rai, M. & Vithanage, M. 2020 Exploration of an extracellular polymeric substance from earthworm gut bacterium (*Bacillus licheniformis*) for bioflocculation and heavy metal removal potential. *Applied Sciences* **10** (1), 349.

Bolto, B. & Gregory, J. 2007 Organic polyelectrolytes in water treatment. *Water Research* **41** (11), 2301–2324.

Bouaouine, O., Bourven, I., Khalil, F., Bressollier, P. & Baudu, M. 2019 Identification and role of *Opuntia ficus indica* constituents in the flocculation mechanism of colloidal solutions. *Separation and Purification Technology* **209**, 892–899.

Bouatay, F. & Mhenni, F. 2014 Use of the cactus cladodes mucilage (*Opuntia ficus-indica*) as an eco-friendly flocculants: process development and optimization using statistical analysis. *International Journal of Environmental Research* **8** (4), 1295–1308.

Buenaño, B., Vera, E. & Aldás, M. B. 2019 Study of coagulating/flocculating characteristics of organic polymers extracted from biowaste for water treatment. *Ingeniería e Investigación* **39** (1), 24–35.

Carpinteyro-Urban, S. & Torres, L. G. 2013 Use of response surface methodology in the optimization of coagulation-flocculation of wastewaters employing biopolymers. *International Journal of Environmental Research* **7** (3), 717–726.

Chen, N., Liu, W., Huang, J. & Qiu, X. 2020 Preparation of octopus-like lignin-grafted cationic polyacrylamide flocculant and its application for water flocculation. *International Journal of Biological Macromolecules* **146**, 9–17.

Chouchane, H., Mahjoubi, M., Ettoumi, B., Neifar, M. & Cherif, A. 2018 A novel thermally stable heteropolysaccharide-based bioflocculant from hydrocarbonoclastic strain *Kocuria rosea* BU22S and its application in dye removal. *Environmental Technology* **39** (7), 859–872.

Dange, P. S. & Lad, R. K. 2015a Upgrading conventional sewage treatment process by using *Mangifera indica*. *International Journal for Scientific Research and Development* **3** (2), 1584–1588.

Dange, P. S. & Lad, R. K. 2015b A treatment of sewage using natural coagulant. *International Journal of Emerging Technology and Advanced Engineering* **Special issue**, 58–64.

Dao, V. H., Cameron, N. R. & Saito, K. 2016 Synthesis, properties and performance of organic polymers employed in flocculation applications. *Polymer Chemistry* **7** (1), 11–25.

David, O. M., Oluwole, O. A., Ayodele, O. E. & Lasisi, T. 2019 Characterisation of fungal bioflocculants and its application in water treatment. *Current Journal of Applied Science and Technology* **34** (6), 1–9.

Deshmukh, B. S., Pimpalkar, S. N., Rakhunde, R. M. & Joshi, V. A. 2013 Evaluation performance of natural *Strychnos potatorum* over the synthetic coagulant alum for the treatment of turbid water. *International Journal of Innovative Research in Science, Engineering and Technology* **2** (11), 6183–6189.

de Souza, M. T. F., Ambrosio, E., de Almeida, C. A., de Souza Freitas, T. K. F., Santos, L. B., de Cinque Almeida, V. & Garcia, J. C. 2014 The use of a natural coagulant (*Opuntia ficus-indica*) in the removal for organic materials of textile effluents. *Environmental Monitoring and Assessment* **186** (8), 5261–5271.

Dlamini, N. G., Basson, A. K. & Pullabhotla, V. S. R. 2019 Optimization and application of bioflocculant passivated copper nanoparticles in the wastewater treatment. *International Journal of Environmental Research and Public Health* **16** (12), 2185.

Dwari, R. K. & Mishra, B. K. 2019 Evaluation of flocculation characteristics of kaolinite dispersion system using guar gum: a green flocculant. *International Journal of Mining Science and Technology* **29** (5), 745–755.

Fahmi, M. R., Hamidin, N., Abidin, C. Z. A., Fazara, U., Ali, M. D. & Hatim, M. D. 2014 Performance evaluation of okra (*Abelmoschus esculentus*) as coagulant for turbidity removal in water treatment. *Key Engineering Materials* **594–595** (2014), 226–230.

Fan, H. C., Yu, J., Chen, R. P. & Yu, L. 2019 Preparation of a bioflocculant by using acetonitrile as sole nitrogen source and its application in heavy metals removal. *Journal of Hazardous Materials* **363**, 242–247.

Ferasat, Z., Panahi, R. & Mokhtarani, B. 2020 Natural polymer matrix as safe flocculant to remove turbidity from kaolin suspension: performance and governing mechanism. *Journal of Environmental Management* **255**, 109939.

Freitas, T. K. F. S., Oliveira, V. M., De Souza, M. T. F., Geraldino, H. C. L., Almeida, V. C., Fávaro, S. L. & Garcia, J. C. 2015 Optimization of coagulation-flocculation process for treatment of industrial textile wastewater using okra (*A. esculentus*) mucilage as natural coagulant. *Indus Crops and Products* **76**, 538–544.

Guo, J., Jiang, S. & Pang, Y. 2019 Rice straw biochar modified by aluminum chloride enhances the dewatering of the sludge from municipal sewage treatment plant. *Science of The Total Environment* **654**, 338–344.

Hassimi, A. H., Hafiz, R. E., Muhamad, M. H. & Abdullah, S. R. S. 2020 Bioflocculant production using palm oil mill and sago mill effluent as a fermentation feedstock: characterization and mechanism of flocculation. *Journal of Environmental Management* **260**, 110046.

Ho, Y. C., Norli, I., Alkarkhi, A. F. & Morad, N. 2014 Extraction, characterization and application of malva nut gum in water treatment. *Journal of Water & Health* **13** (2), 489–499.

Im, D., Nakada, N., Kato, Y., Aoki, M. & Tanaka, H. 2019 Pretreatment of ceramic membrane microfiltration in wastewater reuse: a comparison between ozonation and coagulation. *Journal of Environmental Management* **251**, 109555.

Irma, N. Y., Philippe, S., Abdoukarim, A., Alassane, Y. A., Pascal, A. C., Daouda, M. & Dominique, S. K. 2015 Evaluation of *Aloe vera* leaf gel as a natural flocculant: phytochemical screening and turbidity removal trials of water by coagulation flocculation. *Research Journal of Recent Sciences* **4** (12), 1–9.

Jaouadi, T., Hajji, M., Kasmi, M., Kallel, A., Chatti, A., Hamzaoui, H., Mnif, A., Tizaoui, C. & Trabelsi, I. 2020 *Aloe* sp. leaf gel and water glass for municipal wastewater sludge treatment and odour removal. *Water Science and Technology* **81** (3), 479–490.

Jayalakshmi, G., Dwarapureddi, B. K. & Saritha, V. 2016 Legitimate use of plant waste products for drinking water treatment. *Journal of Environmental Research and Development* **11** (02), 351–359.

Jones, A. N. & Bridgeman, J. 2019 A fluorescence-based assessment of the fate of organic matter in water treated using crude/purified *Hibiscus* seeds as coagulant in drinking water treatment. *Science of the Total Environment* **646**, 1–10.

Kazi, T. & Virupakshi, A. 2013 Treatment of tannery wastewater using natural coagulants. *International Journal of Innovative Research in Science Engineering and Technology* **2** (8), 4061–4068.

Khataee, A., Sheydaei, M., Hassani, A., Taseidifar, M. & Karaca, S. 2015 Sonocatalytic removal of an organic dye using TiO_2/Montmorillonite nanocomposite. *Ultrasonics Sonochemistry* **22**, 404–411.

Khiari, R., Dridi-Dhaouadi, S., Aguir, C. & Mhenni, M. F. 2010 Experimental evaluation of eco-friendly flocculants prepared

from date palm rachis. *Journal of Environmental Sciences* **22** (10), 1539–1543.
Kothari, R., Pathak, V. V., Pandey, A., Ahmad, S., Srivastava, C. & Tyagi, V. V. 2017 A novel method to harvest *Chlorella* sp. via low cost bioflocculant: influence of temperature with kinetic and thermodynamic functions. *Bioresource Technology* **225**, 84–89.
Lassoued, B. M. G., Djobbi, B. & Hassen, R. B. 2018 Influence of operating factors on turbidity removal of water surface by natural coagulant indigenous to Tunisia using experimental design. *Journal of Water Chemistry and Technology* **40** (5), 285–290.
Lee, D. J. & Chang, Y. R. 2018 Bioflocculants from isolated strains: a research update. *Journal of the Taiwan Institute of Chemical Engineers* **87**, 211–215.
Lee, C. S., Chong, M. F., Robinson, J. & Binner, E. 2014 A review on development and application of plant-based bioflocculants and grafted bioflocculants. *Industrial & Engineering Chemistry Research* **53** (48), 18357–18369.
Lee, C. S., Chong, M. F., Binner, E., Gomes, R. & Robinson, J. 2018 Techno-economic assessment of scale-up of bio-flocculant extraction and production by using okra as biomass feedstock. *Chemical Engineering Research and Design* **132**, 358–369.
Liu, Z., Wei, H., Li, A. & Yang, H. 2019 Enhanced coagulation of low-turbidity micro-polluted surface water: properties and optimization. *Journal of Environmental Management* **233**, 739–747.
Lopes, E. C., Santos, S. C. R., Pintor, A. M. A., Boaventura, R. A. R. & Botelho, C. M. S. 2019 Evaluation of a tannin-based coagulant on the decolorization of synthetic effluents. *Journal of Environmental Chemical Engineering* **7** (3), 103125.
Makhtar, N. S. M., Idris, J., Musa, M., Andou, Y., Hamid, K. H. K. & Puasa, S. W. 2020 Plant-based *Tacca leontopetaloides* biopolymer flocculant (TBPF) produced high removal of turbidity, TSS, and color for leachate treatment. *Processes* **8** (5), 527.
Maliehe, T. S., Basson, A. K. & Dlamini, N. G. 2019 Removal of pollutants in mine wastewater by a non-cytotoxic polymeric bioflocculant from *Alcaligenes faecalis* HCB2. *International Journal of Environmental Research and Public Health* **16** (20), 4001.
Miller, S., Fugate, E., Craver, V. O., Smith, J. A. & Zimmerman, J. B. 2008 Toward understanding the efficacy and mechanism of *Opuntia* spp. as a natural coagulant for potential application in water treatment. *Environmental Science and Technology* **42** (12), 4274–4279.
Mirhosseini, H. & Amid, B. T. 2012 A review study on chemical composition and molecular structure of newly plant gum exudates and seed gums. *Food Research International* **46** (1), 387–398.
Mishra, A. & Bajpai, M. 2005 Flocculation behaviour of model textile wastewater treated with a food grade polysaccharide. *Journal of Hazardous Materials* **118** (1–3), 213–217.
Mishra, A. & Bajpai, M. 2006 The flocculation performance of *Tamarindus* mucilage in relation to removal of vat and direct dyes. *Bioresource Technology* **97** (8), 1055–1059.
Mishra, S. & Kundu, K. 2019 Synthesis, characterization and applications of polyacrylamide grafted fenugreek gum (FG-g-PAM) as flocculant: microwave vs thermal synthesis approach. *International Journal of Biological Macromolecules* **141**, 792–808.
Mishra, A., Agarwal, M., Bajpai, M., Rajani, S. & Mishra, R. P. 2002 *Plantago psyllium* mucilage for sewage and tannery effluent treatment. *Iranian Polymer Journal* **11** (6), 381–386.
Mishra, A., Agarwal, M. & Yadav, A. 2003 Fenugreek mucilage as a flocculating agent for sewage treatment. *Colloid and Polymer Science* **281** (2), 164–167.
Mishra, A., Srinivasan, R., Bajpai, M. & Dubey, R. 2004a Use of polyacrylamide-grafted *Plantago psyllium* mucilage as a flocculant for treatment of textile wastewater. *Colloid and Polymer Science* **282** (7), 722–727.
Mishra, A., Yadav, A., Agarwal, M. & Bajpai, M. 2004b Fenugreek mucilage for solid removal from tannery effluent. *Reactive and Functional Polymers* **59** (1), 99–104.
Mishra, A., Bajpai, M., Pal, S., Agrawal, M. & Pandey, S. 2006 *Tamarindus indica* mucilage and its acrylamide-grafted copolymer as flocculants for removal of dyes. *Colloid and Polymer Science* **285** (2), 161–168.
Mohd-Salleh, S. N. A., Mohd-Zin, N. S. & Othman, N. 2019 A review of wastewater treatment using natural material and its potential as aid and composite coagulant. *Sains Malaysiana* **48** (1), 155–164.
Morrissey, K. L., Fairbanks, B. D., Bull, D. S., Stoykovich, M. P. & Bowman, C. N. 2020 Flocculation behavior and mechanisms of block copolymer architectures on silica microparticle and *Chlorella vulgaris* systems. *Journal of Colloid and Interface Science* **567**, 316–327.
Mu, J., Zhou, H., Chen, Y., Yang, G. & Cui, X. 2018 Revealing a novel natural bioflocculant resource from *Ruditapes philippinarum*: effective polysaccharides and synergistic flocculation. *Carbohydrate Polymers* **186**, 17–24.
Mukhtar, A., Ali, W. & Hussain, G. 2015 A preliminary study of *Opuntia stricta* as a coagulant for turbidity removal in surface waters. *Proceedings of the Pakistan Academy of Sciences* **52** (2), 117–124.
Muruganandam, L. M. P., Kumar, S., Jena, A., Gulla, S. & Godhwani, B. 2017 Treatment of waste water by coagulation and flocculation using biomaterials. *IOP Conference Series: Materials Science and Engineering* **263** (3), 032006.
Ngbolua, K. T. N., Pambu, A. L., Mbutuku, L. S., Nzapo, H. K., Bongo, G. N., Muamba, N. B., Falanga, C. M., Gbolo, Z. B. & Mpiana, P. T. 2016 Etude Comparée de l'activité Floculante de *Moringa oleifera* et Vetivera Zizanoides dans la Clarification des Eaux de Mare Au Plateau de Batéké, République Démocratique Du Congo. *International Journal of Innovation and Scientific Research* **24** (2), 379–387.
Nharingo, T., Zivurawa, M. T. & Guyo, U. 2015 Exploring the use of cactus *Opuntia ficus indica* in the biocoagulation–flocculation of Pb(II) ions from wastewaters. *International Journal of Environmental Science and Technology* **12** (12), 3791–3802.
Nobel, P. S., Cavelier, J. & Andrade, J. L. 1992 Mucilage in cacti: its apoplastic capacitance, associated solutes, and influence on tissue 5. *Journal of Experimental Botany* **43** (5), 641–648.

Ortiz-Oliveros, H. B. & Flores-Espinosa, R. M. 2019 Simultaneous removal of oil, total Co and ^{60}Co from radioactive liquid waste by dissolved air flotation. *International Journal of Environmental Science and Technology* **16**, 3679–3686.

Pallavi, N. & Mahesh, S. 2013 Feasibility study of *Moringa oleifera* as a natural coagulant for the treatment of dairy wastewater. *International Journal of Engineering Research* **2** (3), 200–202.

Pham, T. L. & Bui, M. H. 2020 Removal of nutrients from fertilizer plant wastewater using *Scenedesmus* sp.: formation of bioflocculation and enhancement of removal efficiency. *Journal of Chemistry* **2020** (Article ID 8094272), 1–9.

Porwal, H. J., Mane, A. V. & Velhal, S. G. 2015 Biodegradation of dairy effluent by using microbial isolates obtained from activated sludge. *Water Resources and Industry* **9**, 1–15.

Qureshi, K., Bhatti, I. & Shaikh, M. S. 2011 Development of bio-coagulant from mango pit for the purification of turbid water. *Sindh University Research Journal – SURJ (Science Series)* **43** (1), 105–110.

Rachdi, R., Srarfi, F. & Shimi, N. S. 2017 Cactus *Opuntia* as natural flocculant for urban wastewater treatment. *Water Science and Technology* **76** (7), 1875–1883.

Saleem, M. & Bachmann, R. T. 2019 A contemporary review on plant-based coagulants for applications in water treatment. *Journal of Industrial and Engineering Chemistry* **72**, 281–297.

Seghosime, A., Awudza, J. A. M., Richard, B. & Kwarteng, S. O. 2017 Comparative studies on proximate composition and phytochemical screening of mango, key lime, African star apple and African pear seeds as possible coagulant aids for water treatment. *American Journal of Environmental Sciences* **13** (4), 325–333.

Sellami, M., Zarai, Z., Khadhraoui, M., Jdidi, N., Leduc, R. & Ben Rebah, F. 2014 Cactus juice as bioflocculant in the coagulation–flocculation process for industrial wastewater treatment: a comparative study with polyacrylamide. *Water Science and Technology* **70** (7), 1175–1181.

Sethupathy, A. 2015 An experimental investigation of alum and *Moringa oleifera* seed in water treatment. *International Journal of Advanced Research* **3** (1), 515–518.

Shah, N. S., Khan, J. A., Sayed, M., Khan, Z. U., Iqbal, J., Arshad, S., Junaid, M. & Khan, H. M. 2020 Synergistic effects of H_2O_2 and $S_2O_8^{2-}$ in the gamma radiation induced degradation of congo-red dye: kinetics and toxicities evaluation. *Separation and Purification Technology* **233**, 115966.

Shahadat, M., Teng, T. T., Rafatullah, M., Shaikh, Z. A., Sreekrishnan, T. R. & Ali, S. W. 2017 Bacterial bioflocculants: a review of recent advances and perspectives. *Chemical Engineering Journal* **328**, 1139–1152.

Sharma, D., Kumar, V., Nautiyal, R. & Sharma, P. 2020 Synthesis and characterization of quaternized *Cassia tora* gum using Taguchi L'16 approach. *Carbohydrate Polymers* **232**, 115731.

Srarfi, F., Rachdi, R., Khela, K. & Shimi, N. S. 2018 Emerging plant-based flocculation treatment of phosphate clay: case study from Metlaoui-Gafsa (southwest Tunisia). *Water Science and Technology* **17** (3), 743–751.

Swelam, A. A., Sherif, S. S. & Hafez, A. I. 2019 Removal comparative study for Cd(II) ions from polluted solutions by adsorption and coagulation techniques using *Moringa oleifera* seeds. *Egyptian Journal of Chemistry* **62** (8), 1499–1517.

Tawakkoly, B., Alizadehdakhel, A. & Dorosti, F. 2019 Evaluation of COD and turbidity removal from compost leachate wastewater using *Salvia hispanica* as a natural coagulant. *Industrial Crops and Products* **137**, 323–331.

Tawila, Z. M. A., Ismail, S., Dadrasnia, A. & Usman, M. M. 2018 Production and characterization of a bioflocculant produced by *Bacillus salmalaya* 139SI-7 and its applications in wastewater treatment. *Molecules* **23** (10), 2689.

Theodoro, J. D. P., Lenz, G. F., Zara, R. F. & Bergamasco, R. 2013 Coagulants and natural polymers: perspectives for the treatment of water. *Plastic and Polymer Technology* **2** (3), 55–62.

Tietz, T., Lenzner, A., Kolbaum, A. E., Zellmer, S., Riebeling, C., Gürtler, R., Jung, C., Kappenstein, O., Tentschert, J., Giulbudagian, M., Merkel, S., Pirow, R., Lindtner, O., Tralau, T., Schäfer, B., Laux, P., Greiner, M., Lampen, A., Luch, A., Wittkowski, R. & Hense, A. 2019 Aggregated aluminium exposure: risk assessment for the general population. *Archives of Toxicology* **93**, 3503–3521.

Torres, L. G., Cadena, G., Carpinteyro-Urbán, S. & Corzo, L. J. 2014 New galactomannans and mucilages with coagulant–flocculant activity for an environment-friendly treatment of wastewaters. *Current Advances in Environmental Science* **2** (2), 52–58.

Ullah, A. & Rathnasiri, R. P. G. 2015 Optimization of adsorption-coagulation process for treatment of palm oil mill effluent (pome) using alternative coagulant. *International Research Symposium on Engineering Advancements (RSEA)* **2015**, 68–70.

Vimala, R. T. V., Escaline, J. L. & Sivaramakrishnan, S. 2020 Characterization of self-assembled bioflocculant from the microbial consortium and its applications. *Journal of Environmental Management* **258**, 110000.

WHO Guidelines for Drinking-Water Quality 2008 *Volume 1: Recommendations*. World Health Organization, Geneva.

Zaid, A. Q., Ghazali, S. B., Mutamim, N. S. A. & Olalere, O. A. 2019 Experimental optimization of *Moringa oleifera* seed powder as bio-coagulants in water treatment process. *SN Applied Sciences* **1**, 504.

Zaidi, N. S., Muda, K., Loan, L. W., Sgawi, M. S. & Rahman, M. A. A. 2019 Potential of fruit peels in becoming natural coagulant for water treatment. *International Journal of Integrated Engineering* **11** (1), 140–150.

Zhang, Q., Zhang, F., Ni, Y. & Kokot, S. 2015 Effects of aluminium on amyloid-beta aggregation in the context of Alzheimer's disease. *Arabian Journal of Chemistry* **12** (8), 2897–2904.

Zhou, Y., Zheng, H., Wang, Y., Zhao, R., Liu, H., Ding, W. & An, Y. 2020 Enhanced municipal sludge dewaterability using an amphiphilic microblocked cationic polyacrylamide synthesized through ultrasonic-initiation: copolymerization and flocculation mechanisms. *Colloids and Surfaces A* **594**, 124645.

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