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REVIEW

Toward green technology: a review on some efficient model plant-based coagulants/flocculants for freshwater and wastewater remediation

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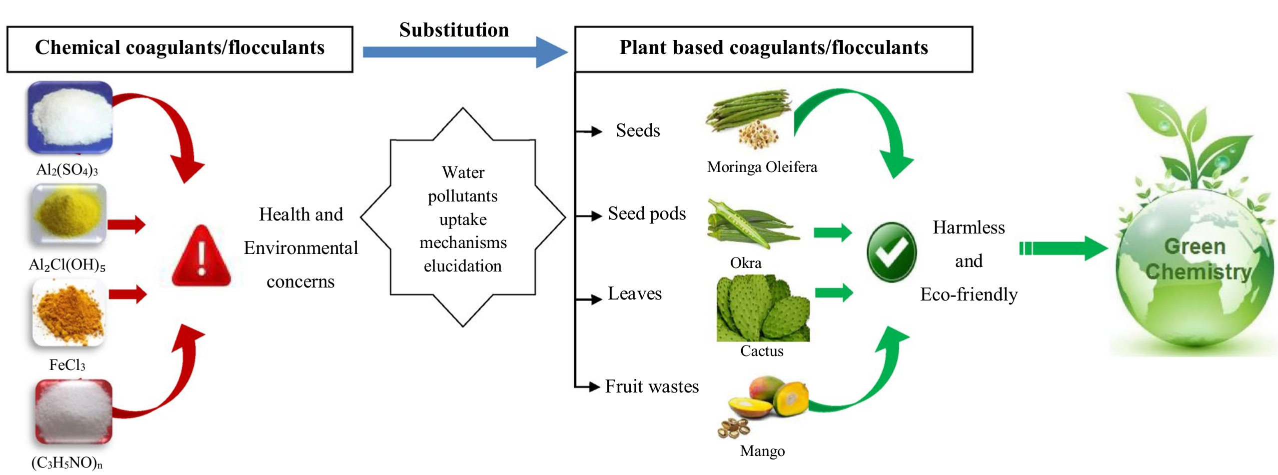
Abstract
Plant-based coagulants/flocculants are foreseen to be a major progress in water treatment technology owing to their safety, biodegradability and availability, unlike synthetic chemical water refiners such as Al, Fe salts and organic polymers claimed to cause threats to our ecosystem either via their residues in the treated waters or due to their generated toxic sludge. Further, the increasing global awareness about environmental issues is acting as a driving force behind the interest toward the use of green resources as valuable products for water treatment. Substitution of synthetic coagulants/flocculants by such natural materials can not only minimize ecosystem damages and threats, but would also foster the way toward an era of clean technology and a sustainable environment. The present paper reviews works on the most efficient model plant-based coagulants/flocculants, moringa seeds, cactus pads, okra seed pods and mango kernels, via highlighting their effectiveness in treating a variety of waters. This review focuses also on the extracting processes used for their preparation, on the type of their active compounds as well as on water pollutant removal mechanisms. Among the four known coagulation–flocculation phenomenon, both polymer bridging and charge neutralization were assumed to be the main predominant mechanisms of bio-coagulants/bio-flocculants toward water contaminant removal. Further, this paper sheds light on where future works should head aiming to stress on the exploitation of green materials in water remediation. We believe that this review can provide an immediate platform for scientists to intensify their research on more efficient natural products to be used in water processing for the sake of a safer environment.

Graphic abstract



Keywords Plant-based coagulants/flocculants · Water treatment · Bio-flocculants · Coagulation–flocculation · Clean technology

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Introduction

Along these later years, researchers assume that there is no doubt on the serious threats caused by the intensive dissemination of chemical reagents to remediate freshwater and wastewater using the so-called physicochemical process, more known as the coagulation–flocculation technique.

Commonly, the chemicals widely used in the conventional coagulation–flocculation process are coagulants such as alum salts ($Al_2(SO_4)_3$, $AlCl_3$), ferrous salts ($FeCl_3$, $FeSO_4$) (Sahu and Chaudhari 2013), organic polymers as well as flocculants like acrylamide, acrylic acid and polyacrylamide (Irfan et al. 2017). In general, the former chemicals are added to waters to destabilize the colloidal particles by neutralizing their negative surface charges (Gregory and O'Melia 1989). As a result, particles may collide through a rapid mixing and form larger and settleable aggregates. Otherwise, further addition of a flocculant is needed to bring the non-settled aggregates together into larger and heavier decantable flocs (Ives 1978).

Though their great efficacy, besides the resulting residues in the treated waters, synthetic coagulants and flocculants generate usually a secondary solid pollution called a sludge, known by a low or no biodegradability tendency and toxic effects (Mallevialle et al. 1984). Consequently, this chemical family of water refiners is accused to have adverse impacts on human beings (Martyn et al. 1989) and ecosystems (Mallevialle et al. 1984). Within this frame, it was reported that Alzheimer's disease could be ascribed to the remaining traces of Al in the treated wastewater finding its ways to water tables or marine ecosystems (Zhang et al. 2015). As well, Tietz et al. (2019) demonstrated that Al traces found in foodstuff sparked acute carcinogenic and genotoxic diseases. On the other hand, synthetic organic flocculant drawbacks and threats on human health have been also well documented, since the majority of commercial polymers have petroleum origin using a chemical processing that is not always safe or environmentally friendly (Dearfield et al. 1988). Bearing all these facts in mind, substitution of synthetic chemical coagulants/flocculants by effective and harmless natural products should be seen as an urgent need.

It is within this context that several natural products resulting from seeds, fruits and leaves such as *lime* seeds (Seghosime et al. 2017), pods seeds of *tamarind* (Buenaño et al. 2019), leaves of *acorn* (Benalia et al. 2019), pads from *cactus* (Kumar and Istalingamurthy 2017), fruits peels of *banana* (Zaidi 2019) and *hyacinth bean* (Shilpaa et al. 2012), have been receiving a great interest as promising coagulant/flocculant alternatives to synthetic reagents owing to their safeness, natural abundance and cost-effectiveness (Muruganandam et al. 2017). Despite the thorough investigations and recommendations on plant-based coagulants/flocculants, massive usage of chemical products in the field of freshwater and wastewater treatment is still predominant. Intensive efforts have therefore to be made to emphasize the importance of green coagulants/flocculants in water processing to ensure their wide exploitation and foster the way toward a green water treatment technology era.

In the literature, numerous papers provide a comprehensive summary on bio-products renowned by their coagulation–flocculation properties (Hussain and Hydar 2019).

For instance, in an exhaustive study on 14 plants as coagulant labeled vegetables and legumes elaborated by Choy et al. (2013), the existing research gaps in the use of bio-based coagulants were discussed aiming to provide a platform toward the necessity of further investigations for effective green coagulant discovery. More so, a consortium of 16 plants as coagulants was cited by Jayalakshmi et al. (2017) as efficient bio-resources. Kristiano (2017) investigated a wide range of fruit wastes and inedible legumes and claimed their potential use to treat various water types. As well, in his work upon a synopsis of 18 plants, Abiola (2018) underlined detailed viewpoints on the gap that accounted for the poor transition of laboratory findings to real-life applications of plant-based coagulant. Moreover, Amran et al. (2018) provided a mini review on researches achieved over the span of ten years and pointed out ultimately the necessity of carrying out more studies to fulfill a thorough grasp on the coagulation ability of plant-based materials. Lately, in a contemporary review conducted by Saleem and Bachmann (2019) unrolling the efficacy of diverse plants as cationic, anionic and nonionic coagulants, it was indicated the snags that hinder their wide application and commercialization. More recently, Mohd-Salleh et al. (2019) have reviewed more than 16 plants and outlined the future prospects of natural materials as aids and their potential as sustainable composite coagulants.

As it can be noticed, although the intensive laboratory works exploring plant-based coagulants/flocculants, there will be still a long way to overcome till their use in the actual field can be really convincing and hence widely spread. One reason of this hindrance might be due to the fact that finding efficient plants is still not yet an easy task for environmentalists and water practitioners. In fact, most of the reviewed papers either highlight the strength of a sole popular plant like moringa (Sulaiman et al. 2017) and cactus (Karanja et al., 2017), or investigate on a quite large number of bio-resources such as the survey conducted by Jayalakshmi et al. (2017) who reported an consortium of 16 plants, Mathuram et al. (2018) discussing 8 plants and Jones (2018) investigating on 5 plants. Nonetheless, these later works and many other alike, appear more likely as a kind of listing and no comparison was made among the studied plants making the choice of that efficient one seeming a bit convoluted.

To the best of our knowledge, rare the papers that are dealing with a reasonable number of efficient plants, established a comparison of their contaminant removal strength and elucidated the coagulation/flocculation mechanism with the plant's extract to ease the choice of the best biomaterials, such as in the case of the work conducted by Yin (2010), focusing mainly on nirmali seeds, *moringa oleifera*, tannin and cactus.

Keeping this in mind, the present paper focuses on four screened main efficient plant species, moringa, cactus, okra and mango, belonging to different plant families, thoroughly studied and exploited to remediate contaminated waters. This limitation of plant number is to provide more visibility for water practitioners and stakeholders as well as to withdraw researchers' attention to pay more care for plants alike.

The related relevance of the selected plants in treating freshwater and wastewater, the active agents extracting procedure and pollutant uptake mechanisms are highlighted and discussed. Further, some recommendations on where future works should head are also pointed out.

Overview of plant-based coagulants/flocculants and their preparation procedure

Since long time ago, bio-coagulants and bio-flocculants derived from plants have been investigated and often exploited in freshwater purification (Diaz et al. 1999). Additionally, over the recent years, further researchers have been exploring the coagulating–flocculating activity of various plant parts and extracts derived from seeds (Ngbolua et al. 2016), leaves (Sellami et al. 2014), stems (Alwi et al. 2013), fruits shells and kernels waste (Jayalakshmi et al. 2016) to remediate a variety of freshwaters (Jones and Bridgeman 2016) and wastewaters (Torres et al. 2014).

Regardless the origin of these green extracts and their active agent content, plant-based coagulants/flocculants have been prepared as follows (Fig. 1): (i) Plant parts were firstly cut, sliced or peeled, then oven or sun-dried and grinded to obtain a readily usable fine powder. (ii) For further usage, the dried powder can be either dissolved in water, in salt solutions (NaCl, KCl, $MgCl_2$, etc.) (Okuda et al. 2001) or in organic solvents (acetone, alcohol) (Pichler et al. 2012) to form a mucilaginous filtrate. From the ecological standpoint and environmental benefits, extraction using only water or salt solutions should be recommended over the use of organic solvents to minimize environmental burdens. In some cases, since filtrates and powders may enclose other plant tissues rich in inorganic constituents that may increase the organic load in waters, additional purification process is therefore required. This further processing is usually performed via a precipitation (Bouaouine et al. 2019) or a lyophilization (Khadhraoui et al. 2019) schemes.

Nowadays, the exploration of plant extracts for water treatment should be regarded as a great path toward a green and sustainable technology aiming for an ecological behavior and human health preservation. As a matter of fact, several plants have therefore been tested under seeds or other forms (husk, pith, kernels and leaves) for their coagulating/flocculating performance toward suspended matter (SM), chemical oxygen demand (COD) and turbidity (TN) removal from various types of freshwaters (Gaikwad and Munavalli 2019) and wastewaters (Adwuyi and Adwumi 2018).

Among the so many worldwide investigated plants and to provide a more concise discussion, emphasis will be hereafter placed on the most efficient plant model: *moringa oleifera*, cactus, okra and mango, commonly used for turbid, freshwater and wastewater processing, either as mucilage or powder extracts. Each of them belongs to a family classified

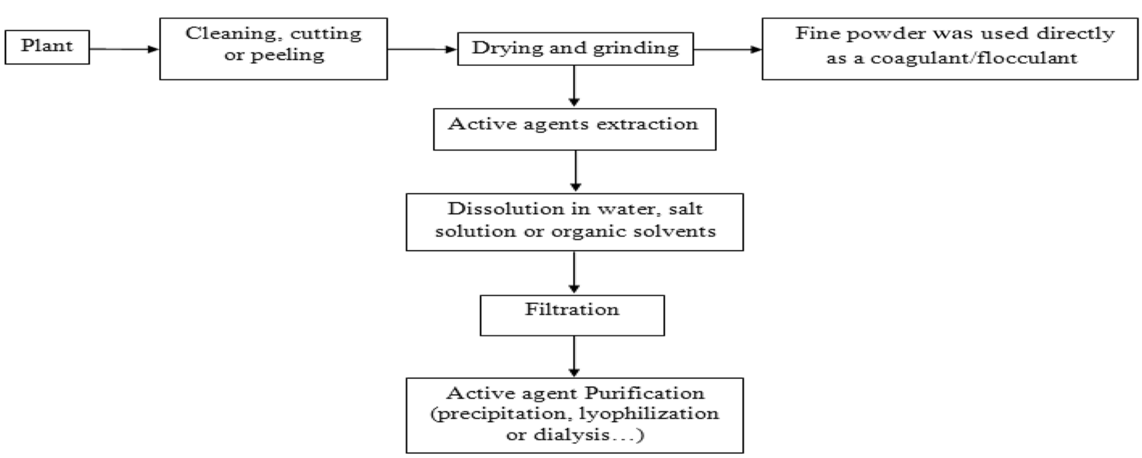


Fig. 1 General scheme of bio-coagulant/bio-flocculant preparation

either according to the form of the extract used as a reagent or according to the active agent content in the extract. Likewise, the coagulation–flocculation mechanism attributed to each plant is elucidated and a comparison of their pollutant removal power is discussed.

Most popular investigated bio-coagulants/bio-flocculants

Moringa oleifera (MO)

Moringa oleifera (MO) is a tropical plant originating from India and belonging to the Moringaceae family (Fig. 2). It is commonly known as the drumstick tree or ben oil tree for its beneficial vegetable and medicinal properties (Anwar et al. 2007).

Since long time ago, MO seeds are receiving a great interest in freshwater and wastewater remediation. They have been exploited as an effective bio-coagulant/bio-flocculent in some developing countries where synthetic chemicals are not affordable (Bhatia et al. 2007). According to Olayemi and Alabi 1994, these seeds contain around 34% of proteins, 15% of carbohydrates and 15.5% of lipids. This high protein amount ushered that these agents can be one of the most active coagulating compounds.

Generally, to prepare MO-based coagulant/flocculant extract, the matured seeds are removed from their pods, shelled, grinded and sieved. Once a wet powder is formed, two methods can be carried out as a further processing: either drying the previously obtained powder and its direct utilization as a coagulant (Sethupathy 2015) or extracting the active protein by dissolving the powder in water, in salt solutions (NaCl, KCl, $MgCl_2$, etc.) or in organic solvents (acetone, alcohol) (Santos et al. 2012). As the MO's active agents are proteins, using salty extractions is evidently the most accepted choice due to salting-in process known as the common ion effect, thus promoting the protein–protein dissociation. We recall that protein solubility increases as the solution ionic strength is increased by salt addition (Okuda et al., 2001). Consequently, the higher is protein solubility, the higher is the coagulating capacity of MO toward pollutants.

Fig. 2 Moringa tree and its seed extract as a powder

In fact, the ability of MO seeds as coagulant/flocculant was proved in myriad investigations. In the light of that, we display some typical examples of wastewaters treated by this bio-resource extract. For instance, in a study related to the treatment of a laundry wastewater having a COD of 450 mg/L and a turbidity of 68.1 NTU, Al-Gheethi et al. (2017) found that *Moringa oleifera* showed a high turbidity removal efficiency (84%) exceeding by far the rate found in the case of $FeSO_4$ (59%). However, COD removal (47.2%) using MO seems to be comparable to that obtained using $FeSO_4$ (54%). Further, bearing in mind the low sludge volume produced using natural coagulants, MO exhibited also better settling characteristics than $FeSO_4$.

Moreover, Al-Gheethi et al. (2017) reported that increasing the time of the coagulation phase using such natural extract had led to an enhancement of the coagulation–flocculation efficiency.

Based on some other researchers' findings, the coagulation–flocculation activity of plant-based material can be affected by the extract's particle size. As a matter of fact, while treating a dairy wastewater using MO powder with various particle sizes of 150 µm, 212 µm and 425 µm, Pallavi and Mahesh (2013) found that the finest particle showed the highest removal drop in terms of COD (from 2240 mg/L to 800 mg/L) and turbidity (from 230 to 32 NTU). Likewise, MO has demonstrated a very significant removal ability for azo dyes such in case of Chicago Sky Blue 6B (Beltrán-Heredia and Sánchez-Martín 2009) and for anthraquinonic dyes such as Alizarin Violet 3R, where nearly 95% of color abatement was achieved (Beltrán-Heredia et al. 2009). As well, in treating a palm oil mill effluent with an initial SM of 17.9 mg/L and a COD of 40.2 mg/L, Bhatia et al. (2007) showed that MO seeds are an effective coagulant and allowed a SM and a COD removal of 92% and 52.2%, respectively. Further, they noticed that the combination of MO with a commercial flocculant (NALCO 7751) enhanced significantly SM and COD abatements.

To be more concrete, Table 1 displays some typical examples of wastewaters treated by this bio-resource extract. The monitored parameters were COD, suspended matter (SM) and turbidity. Despite the variety of the contaminated water quality (laundry, dairy or oily effluent) and the biomaterial preparation procedures (different drying temperatures, particle sizes, aqueous or salty extractions, disseminated doses), it can be said that MO showed an effective coagulating/flocculating activity toward a wide range of water pollutants. Moreover, in all depicted cases in Table 1, removal rates were higher than 83% for turbidity, nearly 96% for SM and around 50% for COD (Idris 2016).

Table 1 Typical examples of wastewaters treated by the MO seed extract

Effluents	MO preparation procedure	Optimum conditions	Removal (%)	References
Laundry wastewater: Turbidity: 57.8–68.1 NTU COD: 423–450 mg/L pH: 7.5	MO seeds were dried at 100 °C for 24 h, milled and sieved through 300 μm sieve	0.12 g/L of MO at pH 5.7	Turbidity: 83.6 COD: 42.7	Al-Gheethi et al. (2017)
Tannery effluent: Turbidity: 66-96NTU COD: 28,000 mg/L pH: 5.5	MO seeds were sun-dried, ground and sieved to 600 μm particle size	0.6 g/L of MO at pH 4.5	Turbidity: 82.0 COD: 83.3	Kazi and Virupakshi (2013)
Dairy wastewater: Turbidity: 230 NTU COD: 2240 mg/L	MO seeds were dried and sieved to 212 μm particle size	500 mg/L of MO at pH 7	Turbidity: 86 COD: 64.2	Pallavi and Mahesh (2013)
Dye (Alizarin Violet 3R) removal	Salt extraction: 5 g of MO powder in 100 mL + NaCl	62.9 mg/L of MO at pH 7	Color: 95.0	Beltrán-Heredia et al. (2009)
Palm oil mill effluent: SM: 17.92 mg/L COD: 40 mg/L pH: 4.5	Aqueous extraction: 5 g of dried MO seeds after oil extraction in 100 mL distilled water	4 g/L of MO at pH 5	SM: 95.0 COD: 52.2	Bhatia et al. (2007)

Overall, these literature findings confirm the coagulation–flocculation effectiveness of MO seeds and promote their application as viable replacement for synthetic chemicals. Further, as the coagulating–flocculating activity of this bio-extract is attributed to its protein content, it is worth pointing that identification of this protein nature has been a source of debate.

In fact, Gassenschmidt et al. (1995) reported that the flocculant proteinaceous specimen extracted from MO seeds is a cationic protein with molecular weight of 6.5 KDa and an isoelectric pH above 10. This protein is well recognized as $MO_{2.1}$ which is composed of eight positively charged amino acids (7 arginines and 1 histidine) and only one negatively charged residue (aspartic acid).

In contrast, Okuda et al. (2001) pointed out that the coagulation active reagent produced via a saline extraction was far to be a protein; it was rather an organic polyelectrolyte possessing a low molecular weight of 3KDa. However, albeit the saline extraction Luz et al. (2013) found that the active MO seed extract is a lectin, namely cMol. The latter is a basic protein with high molecular weight of 30KDa and an isoelectric pH above 11. The cMol protein is composed of glutamine, alanine, proline and 17 amino acids (15 arginines and 2 histidine). We presume that the distinction in scholars' identification of the purified MO seed active agent nature can be ascribed to the diverse extraction procedures having been applied.

Nevertheless, whatever the nature and the properties of the extracted protein-based coagulant/flocculant, most of scientists stress on the efficiency of MO and agree that MO coagulation/flocculation activity is carried out mainly through adsorption and charge neutralization mechanism, where the short chain of the protein and its significant cationic surface charge adsorb and neutralize the negatively charged colloids present in waters (Bolto and Gregory 2007).

Cactus (Opuntia ficus-indica)

Cactus is a plant belonging to the Cactaceae family and commonly known as prickly pear, tuna, nopal or Opuntia ficus-indica (Prakash and Manikandan 2012). It is indigenous in the arid and semiarid regions of Mexico and it was later introduced into North Africa (Khatabi et al. 2016).

Cactus is characterized by its high water capacity retention (92%) in its pads renowned as cladodes making them very juicy (Fig. 3). The cladode composition is well documented in terms of their nutritional and environmental value. They contain mainly water (80–95%), fiber (1–2%), proteins (0.5–1%) and carbohydrates (3–7%) (Ginestra et al. 2009).

Further, cactus cladodes are well recognized by their slimy liquid called mucilage (Fig. 3). This liquid is worthy of attention as it is believed to be responsible for the coagulation–flocculation behavior of cactus cladodes. Indeed, it

Fig. 3 Cactus cladodes and their extracted raw juice

was demonstrated that the mucilage is composed of various carbohydrates with a molecular weight ranging from 2.3.10^4 to 3.10^6 g/mol (Bouatay and Mhenni 2014). Therefore, the considerable amount of carbohydrates offers to the cactus plant a coagulating and/or flocculating ability to deal with a variety of freshwater and wastewater. Interestingly, the activity of mucilage as bio-coagulant/bio-flocculant depends on its preparation method.

Within this context, one way of cactus extract preparation method is to rinse thoroughly the fresh cladodes with water to remove spines and dirty particles. Then, cleaned cladodes are cut into small pieces and crushed in a domestic mixer followed by a filtration to come up finally with a viscous pulp ready to be used as coagulant and/or flocculant (Sellami et al. 2014). Otherwise, the mucilage is refrigerated at a temperature below 4 °C. Under these conditions, cactus juice storage can be extended for up to two weeks to which its coagulating efficiency is well preserved (Miller et al. 2008). On the other hand, a cactus powder formula can be merely obtained by an oven-drying of the diced cladodes, followed by a grinding (Sellami et al. 2014). Nonetheless, it was reported that an oven-drying (above 120 °C) or using only cladode skin generally leads to an insignificant coagulation ability of the cactus plant (Miller et al. 2008).

Moreover, coagulant/flocculant compounds derived from cactus can be prepared from the raw cladode slices as mucilage or powder that will be mixed with different solutions (only water, salty water or organic solvents). From the environmental point of view and if it gives reasonable pollutant removal rates, water should be the most recommended solvent to be used since it is ecologically safer. However, in some cases, and for the sake of active reagent isolation, the gummy extract can be filtered and then precipitated by alcohol. The formed precipitate will be then oven-dried (Lassoued et al. 2018) or lyophilized (Bouaouine et al. 2019).

Over the recent years, the use of cactus extracts as coagulant/flocculant has been extensively investigated to treat various types of wastewaters. Within this frame, Souza et al. (2014) showed that cactus juice allowed nearly 64.8% of COD removal and about 91.3% of turbidity elimination in treating a jeans laundry effluent. Likewise, using cactus mucilage, these scholars studied the dye removal from a fabric dyeing mesh effluent and obtained a maximum turbidity and a COD reduction of 93.6% and 87.2%, respectively. Their mucilage was prepared via dissolving 1 g of cactus powder in different salt solutions (NaCl, KCl, $NaNO_3$). Surprisingly, NaCl extraction exhibited the most promising coagulant activity and removed 95% of the turbidity from synthetic turbid water. However, with KCl and $NaNO_3$ solutions, the turbidity reduction was, respectively, 91.2% and 93.6%. Further, in a study related to the treatment of textile effluent using cactus juice as bio-flocculant in combination with alum, Bouatay and Mhenni (2014) found removal rates of 88.76%, 91.66% and 99.84%, respectively, for COD, turbidity and dye.

Moreover, in a study conducted by Sellami et al (2014) to treat a glue industry effluent using cactus juice as bio-flocculant, removal rates of 83.3% and 67.5% for SM and COD, respectively, were observed. While testing polyacrylamide (PAM), SM and COD removals of 79.6% and 59.1%, respectively, were achieved. As regards the food industry effluent, Sellami et al. (2014) showed that cactus juice exhibited similar removal rates as PAM for the two above-mentioned parameters. The removal efficiencies were up to 91% for the suspended matter and nearly 69% for that of COD, indeed. As well and with regard to a tannery effluent treated with cactus juice-based coagulant, Kazi and Virupakshi (2013) reported an effective removal of 78.5% and 80.6% for turbidity and COD, respectively.

Cactus extract with its various formulations (raw juice, sun-dried powder or lyophilized powder) was also applied to eliminate heavy metals. For wastewater with high zinc and suspended matter (SM) load, Belbahloul et al. (2014) obtained an important zinc removal of around 96% and a SM removal of about 99%. Further, in treating an industrial effluent, Abid et al. (2008) showed that cactus juice allowed 99.5% of Cr (VI) removal and 98% of turbidity reduction.

Likewise, in treating a pseudo-industrial solution of Cr (VI) (100 ppm), cactus flocculating effectiveness was found to be comparable to an industrial flocculant (PPRQESTOL 2515) where Cr (VI) elimination was about 99.7% (Abid et al. 2008). Similar conclusions were also withdrawn by Taa et al. (2016) for Cr removal from synthetic chromium sulfate water.

In the light of these literature results, cactus mucilage seems revealing a good coagulating/flocculating performance in treating wastewaters. Typical examples of this removal power in terms of COD, SM and turbidity of some effluents purified by cactus extract are summarized in Table 2. It can be concluded that average turbidity, SM and COD reductions are, respectively, 90, 90 and 75% for all types of treated waters.

As mentioned earlier, cactus juice effectiveness in wastewater remediation can be traced back to cladode composition which are mainly L-arabinose (24.6–42%), D-galactose (21–40,1%), L-rhamnose (7–13.1%), D-xylose (22–22.2%) and galacturonic acid (8–12.7%) (Nobel et al. 1992). It was generally reported that galacturonic acid, known as polygalacturonic, might be the main active compound which contributes alone to a high coagulating activity with more than 50% for turbidity in comparison with the cladode polysaccharide content (arabinose, galactose, rhamnose and xylose), which enabled only 30% of turbidity reduction (Miller et al. 2008).

Figure 4 pinpoints a schematic illustration of different molecular interactions between polygalacturonic acid and

Table 2 Typical examples of effluents treated by cactus extracts

Effluents	Cactus preparation	Optimum conditions	Removal (%)	References
Jeans laundry effluent: Turbidity: 104 NTU COD: 1094 mg/L	Salt extraction: 1 g of cactus gum dissolved in 100 mL of a salt solution (KCl, NaCl and $NaNO_3$) After filtration, the suspension was lyophilized	2.6 mg/L of cactus juice at pH 5	Turbidity: 91.25 COD: 64.8	Souza et al. (2014)
Fabric dyeing mesh effluent: Turbidity: 31.5 NTU COD: 1264 mg/L		160 mg/L of cactus juice at pH 6	Turbidity: 93.62 COD: 87.19	
Textile effluent: Turbidity: 38 NTU COD: 2350 mg/L	Cactus pieces sun-dried for 3 h were dried at 60 °C for 24 h	40 mg/L of cactus juice at pH 7	Turbidity: 91.66 COD: 88.76 Color: 99.84	Bouatay and Mhenni (2014)
Glue industry: SM: 270 mg/L COD: 99,200 mg/L pH: 6.7	Cactus stripes ground, filtered through a gauze compress followed by the addition of distilled water (10%) to obtain the cactus juice	0.61 g/L of cactus juice at pH 4.21	SM: 83.3 COD: 59.1	Sellami et al. (2014)
Food industry: SM: 230 mg/L COD: 2376 mg/L pH: 4.94		0.056 g/L of cactus juice at pH 3.92	SM: 88.7 COD: 69.1	
Synthetic wastewater with [Zn] = 100 mg/L	Cactus cladodes grinded and supplemented with distilled water, filtered and the filtrate supernatant was dried using evaporator	1.2 mL of cactus juice at pH 11.5	SM: 99 Zn: 96	Belbahloul et al. (2014)
Tannery effluent: Turbidity: 66–960 NTU COD: 28,000 mg/L	Cactus powder with a particle size of 600 µm was obtained by drying the small cladode pieces of 1 cm at 60 °C/24 h	0.4 g/L of cactus powder at pH 5.5	Turbidity: 78.54 COD: 80.65	Kazi and Virupakshi (2013)
Industrial effluent: Turbidity: 100 NTU [Cr(VI)] = 97 ppm	Cactus cladodes grinded, filtered and diluted with distilled water (10%)	2 mL of cactus juice at pH 9	Turbidity: 98 Cr: 99.5	Abid et al. (2008)

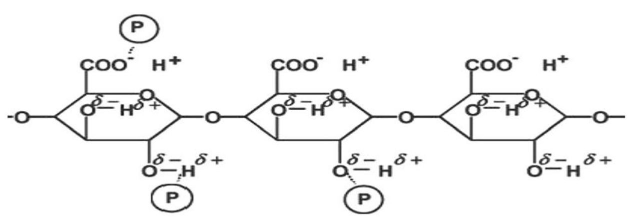


Fig. 4 Polygalacturonic acid molecular chemical structure and its interaction with anionic or cationic pollutants (P) (Theodoro et al. 2013)

Fig. 5 Okra pods and seeds

anionic or cationic pollutants. The polygalacturonic acid is composed of a long anionic chain, including carboxyl (–COOH), carbonyl (–C=O) and hydroxyl (–OH) groups. Hence, it is assumed that the deprotonation of the hydroxyl group enhances the adsorption of anionic pollutants and therefore their agglomeration. Additionally, we agree with Theodoro et al. (2013) that the oxidation of the terminal carbon hydroxyl edge to a carboxylic group leads to the cationic pollutant adsorption and their coagulation–flocculation subsequently. This also leads to suggest that, in treating various contaminated waters, pollutant removal is presumed taking place mainly through adsorption and bridging phenomenon. This same mechanism was also opinied by Nharingo et al. (2015).

Okra (*Abelmoschus esculentus*)

Okra is a plant belonging to the Malvaceae family and well known as ladies finger or gumbo (Fig. 5). It is native to Africa and now is growing all over the world. While okra is widely used as foodstuff owing to its edible fruits with high nutritional contents (Freitas et al. 2015), its unused fruits are being investigated for their effective coagulation capability. The coagulation–flocculation ability of this bio-resource is claimed to be attributed to the anionic gum, a polysaccharide made up of D-galactose, L-rhamnose and L-galacturonic acid (Anastasakis et al. 2009).

To explore okra gum for freshwater and wastewater treatment, various extraction techniques were carried out. Generally, okra mucilage was prepared via aqueous extraction with water after removing the upper crown head and the seeds. Then, the extraction is followed by a filtration step to obtain a mucilage which is subsequently precipitated with alcohol and finally dried up (Agarwal et al. 2003). On the other hand, okra gum can be obtained merely by drying the cleaned pods at 60 °C/1 h (Okolo et al. 2015), 105 °C/24 h (Lee et al. 2015) or 110 °C/6 h (Anastasakis et al. 2009) depending on author thoughts and aims. The dried materials are then grinded, sieved and stored for further usage.

The efficacy of okra gum has been demonstrated in several exhaustive studies. In this respect, Freitas et al. (2015) reported that the utilization of this natural resource as flocculant enhanced the coagulation–flocculation process. Indeed, in treating a laundry wastewater with okra based-flocculant, these authors found a significant turbidity drop, a COD and a color abatements of 97.2%, 85.7% and 93.6%, respectively. However, while using only an inorganic coagulant ($FeCl_3$) the same scholars proved turbidity and COD abatements of 93.7% and 49.9%, respectively. Further, on a study conducted by Anastasakis et al. (2009) for the remediation of a synthetic wastewater with an initial turbidity up to 7.3 NTU, a high turbidity removal of 93, 96 and 97.3%, respectively, for all sedimentation times 10, 20 and 30 min was observed, unlike alum which exhibited a lower turbidity reduction close to 25%. Also, in this work, Anastasakis et al. (2009) reported that the flocculating efficacy of okra was better than that of a commercial flocculant (NaLCO610).

Furthermore, in treating a textile wastewater with an initial SM ranging from 300 to 398 mg/L, Srinivasan and Mishra (2008) demonstrated that okra gum led to a SM removal rate of 98%. Similarly, using the okra gum to purify a tannary effluent, Agarwal et al. (2003) registered a SM reduction of 98.26%.

It is also worth noting that okra parts exhibited a varying coagulating or flocculating ability. In fact, research conducted by Fahmi et al. (2014) on the coagulating properties of okra parts (seeds, leaf, pods and stems) demonstrated that in treating synthetic water with an initial turbidity of 55 NTU at pH 7, okra seed extract showed a turbidity abatement of 64.5%, whereas okra leaf and pods presented a turbidity removal of 54.5% and 49%, respectively. However, okra stem showed an insignificant turbidity reduction even if it was used at a higher dosage.

Keeping in view all of these results, okra seeds are deemed to be the potential active part as a coagulant and/or flocculant for turbidity removal in comparison to the other plant parts. Within this line, according to Fahmi et al. (2014), the high turbidity reduction using seeds can be traced back to their relatively high protein content which ranged between 24 and 26%, whereas the protein content in okra's leaf and stems were 23% and 6.5%, respectively. Therefore, the correlation between protein content in the seeds and the removal efficiency denotes that proteins promote the coagulation–flocculation process.

Similarly to MO and cactus, the different literature findings clearly show the significant coagulating/flocculating potential of okra extract in treating various kinds

Table 3 Typical examples of effluents treated by okra extracts

Effluents	Okra preparation	Optimum conditions	Removal (%)	References
Laundry wastewater: pH: 5–6	Extraction of okra pods' gum using 100 mL of NaCl, KCl, $NaNO_3$ and distilled water	3.2 mg/L of okra at pH 6	Turbidity: 97.2 COD: 85.6 Color: 93.5	Freitas et al. (2015)
Synthetic turbid water: Turbidity: 55 NTU pH: 7	Okra seeds were air-dried at first and then oven-dried at 40 °C for 24 h. Okra mucilage extraction using NaCl and distilled water	25 mg/L of okra at pH 7	Turbidity: 92	Fahmi et al. (2014)
Synthetic wastewater: Turbidity: 6.3–7.3 NTU pH: 5.9	Okra seed pods were dried at 110 °C for 6 h ground and sieved to 500 µm particle size	5 mg/L of okra at pH 6	Turbidity: 97.3	Anastasakis et al. (2009)
Textile effluent: SM: 300–398 mg/L pH: 9.5–10.5	Okra seedpods were extracted with distilled water, precipitated with isopropanol, washed with acetone and dried	0.8 mg/L of okra at pH 4	SM: 98 Color: 53.4	Srinivasan and Mishra (2008)
Tannery effluent: SM: 2213 mg/L pH: 8.27	Aqueous extraction of seedpods of okra, precipitated with alcohol followed by washing with acetone and dried	0.4 mg/L of okra at pH 9.2	SM: 98, 26	Agarwal et al. (2003)

of wastewaters. Table 3 compiles typical examples of the optimal experimental conditions, leading to a significant treatment performance of this natural extract. As depicted in this table, the obtained removal efficiencies were in general higher than 80% for COD and SM and somehow significant for color removal (53%) while dealing with textile wastewater.

It is also worth noting that okra mucilage has a composition similar to that of cactus. As shown in Fig. 6, okra polysaccharide structure is composed foremost by a repeating unit with alternating rhamnose and galacturonic acid residues and contains disaccharide side chains made up of galactose attached to rhamnose residues (Sengkhamparn et al. 2009). Unexpectedly, galactose and rhamnose showed no coagulation activity (Freitas et al. 2015), stressing the fact that galacturonic acid might be the main active agent in the coagulation–flocculation process. Thus, it is possible to consider that the coagulation–flocculation mechanism of okra gum is the same as that of cactus extract. It operates mainly via adsorption and bridging mechanism in which colloids are bound to the polymeric polysaccharide chain via the active hydroxyl (OH) and carboxyl (COO^-) sites, and then settle down together (Freitas et al. 2015).

Mango seeds (*Mangifera indica*)

Mango (Fig. 7) is a tropical fruit belonging to the Anacardiaceae family. It is mainly used as a foodstuff, while its seeds are discarded as a fruit waste. In order to valorize the thrown mango seeds, various studies have been testing their ability to remove pollutants from synthetic (Ali et al. 2008) and real wastewaters (Ullah and Rathnasiri 2015).

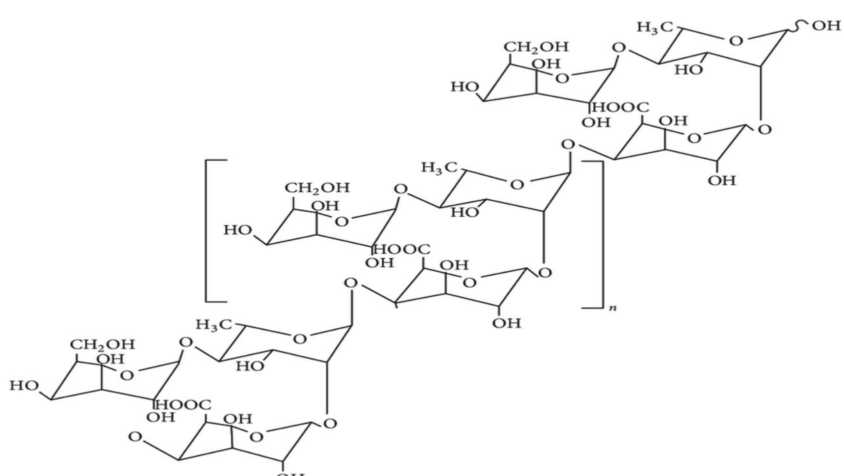
As a result of the many studies alike, mango seeds exhibited an important coagulating activity ascribed to its kernel content. The kernel, commonly known as pit or embryo, represents about 70% of the seed weight (Abdalla et al. 2007), and the coagulant active components are believed to be proteins with a percentage of 58% of mango seed content (Qureshi et al. 2011).

Fig. 7 Mango fruit and its seed powder

In order to assess the coagulation–flocculation properties of the mango fruit, the seed kernels are generally firstly removed, then cleaned with distilled water and sliced. Thereafter, they are dried at different temperatures, 105 °C/24 h (Dange and Lad 2015a, b) and 120 °C/1 h (Qureshi et al. 2011), or sun-dried for one week and then grinded to acquire a powder material (Seghosime et al. 2017).

Regarding their use in water treatment, mango seeds were exploited to purify several types of waters. For instance, Seghosime et al. (2017) used mango extract to clarify a turbid water, resulting from a mixture of river water and kaolin. For the different initial turbidity labeled as low (50 NTU), medium (100 NTU) and high (150 NTU), the abatement was found to be 74%, 87.5% and 92%, respectively. As well, Ullah and Rathnasiri (2015) showed that 1.2 mL/L of mango seed powder allowed an effective removal of 96%, 89% and 97%, respectively, for suspended matter (SM), COD and color in the treatment of a palm oil mill effluent. In addition, in cleaning a sewage water, Dange and Lad (2015a, b) investigated the coagulating ability of mango kernels and demonstrated that mango applied as a sole coagulant allowed, a SM and a COD removal efficiencies of 31.6% and

Fig. 6 Okra gum polysaccharide molecular structure (Zaharuddin et al. 2014)



33.4%, respectively. However, the combination of alum with mango increased the removal rates to 66.8% for SM and to 69.2% for COD.

Other scholars had found a turbidity removal efficiency of 98% while dealing with a synthetic turbid water using 0.5 mL/L of mango and even demonstrated that for low turbid water (17.5 NTU), this bio-coagulant exhibited high turbidity reduction rate of about 70% (Qureshi et al. 2011).

From these literature withdrawn results, it appears that mango extract has gained a considerable importance as coagulant. As a matter of fact, Table 4 unrolls more examples about the optimal experimental conditions promoting the activity of mango seed kernel-based coagulant and their removal capability in terms of COD, SM and turbidity.

However, although it is thoroughly investigated as a bio-adsorbant (Nijuguna 2016), mango seems revealing a higher performance than other natural coagulants cited in the literature, and almost a similar power to that of aluminium salts in term of turbidity removal (Šćiban et al. 2009), suggesting hence, that this plant is deemed to be further explored as a bio-coagulant/bio-flocculant. As the active component in the coagulation–flocculation capability of mango kernels is assigned to the proteins, the main mechanism is assumed to be adsorption and charge neutralization.

Comparison of coagulating–flocculating ability of the four reviewed plants: moringa, cactus, okra and mango

The selected plants, moringa, cactus, okra and mango, in the current review belong to different families, and it seems that each of them has its own performance toward freshwater and wastewater remediation owing to its extracted parts exploited as coagulant/flocculant. As previously demonstrated, efficient plant parts are the seed content in case of moringa and mango, seed pods for okra and pad part when it comes to cactus. Their coagulating and flocculating ability is ascribed to their active agent content (polysaccharides and/or proteins), making them ideal to treat contaminated waters, with of course some discrepancy in their removal rate.

For instance, Thakur and Choubey (2014) showed that a maximum turbidity reduction of 80.7% was achieved using Moringa against 78.7% while using okra when treating a synthetic turbid water (Table 5). Besides, for a tannery effluent, Kazi and Virupakshi (2013) demonstrated that moringa seed allowed a higher turbidity and COD reduction of 82% and 83%, respectively, compared to that achieved using cactus mucilage (turbidity removal of 78.5% and a COD abatement of 80.6%; Table 5). Regarding mango, Qureshi et al. (2011) reported that the coagulation power of this plant (98%) toward turbidity was higher than that of moringa (86%). As well, in a study conducted by Ali et al. (2008), it was concluded that moango seed extract can be considered to be the best natural coagulant in removing turbidity from drinking water in comparison with moringa.

Based, on those gathered information, pollutant removal power of these 4 studied plants can be classified as follows: mango > moringa > cactus = okra.

These outstanding findings unroll that mango coagulating performance ought to be thoroughly investigated. In fact, albeit the previous studies which claimed that proteins are the sole active mango coagulating compound, the high carbohydrate content (76.73%) in the mango seed kernels surmised to interfere with its coagulating activity as recently noticed by Seghosime et al. (2017). Subsequently, for ango

Table 4 Typical examples of effluents treated by mango seed kernels

Effluents	Mango preparation	Optimum conditions	Removal (%)	References
Synthetic turbid water: Turbidity: 150 NTU pH: 7.2 SM: 64	Mango seeds sun-dried for 4 weeks, shelled and squeezed to obtain the seed kernels which were dried again for 1 week	25 mL/L of mango at pH 7	Turbidity: 92	Seghosime et al. (2017)
Palm oil mill effluent: COD: 890 mg/L SM: 700 mg/L	Mango kernels dried at 130 °C for 1 h and milled to 440 µm particle size. Then 5 g of sieved powder was dissolved in 100 mL of distilled water	1.2 mL/L of mango seeds at pH 4	COD: 89 SM: 96 Color: 97	Ullah and Rathnasiri (2015)
Sewage wastewater	Mango kernels dried at 105 °C for 24 h; then, 1 g of powder was mixed with 100 mL of distilled water	168 mg/L of mango seeds at pH 5.2	SM: 31.6 COD: 33.4	Dange and Lad (2015a, b)
		135.2 mg/L of mango with 33.8 mg/L of Acacia Nilotica	SM: 51.8 COD: 49.2	Jain et al. (2015)
Synthetic turbid water: Turbidity: 17.5–90 NTU	Mango kernels dried at 120 °C for 1 h, sieved to 400 µm particle size. To extract the coagulant, 5 g of mango powder was mixed with 100 mL of distilled water	0.5 mL/L of mango seeds at pH 13	Turbidity: 98.6	Qureshi et al. (2011)

Table 5 Comparison of turbidity removal efficiency between moringa, okra, cactus and mango

Effluents	Plant extracts	Turbidity removal (%)	References
Synthetic turbid water	Moringa	80.7	Thakur and Choubey (2014)
	Okra	78.7	
Tannery wastewater	Moringa	82.3	Kazi and Virupakshi (2013)
	Cactus	78.5	
Synthetic turbid water	Moringa	86.0	Qureshi et al. (2011)
	Mango	98.0	

seed kernels, we assume that the two coagulation–flocculation mechanisms, charge neutralization and bridging, may occur together and afford this natural resource to deal not only with turbidity uptake, but also with other different water contaminants.

On the other hand, the encouraged obtained results using these four natural plant-based coagulants/flocculants were comparable to those achieved while using chemical reagents such as Al, Fe salts and organic polymers. As an example, in a comparative study using moringa and aluminum sulfate, Abirami and Rohini (2017) showed that moringa allowed a turbidity removal of 75% versus 70% in case of alum.

Overall, and as given in Table 5, it can be said that a significant turbidity drop using all these plant extracts is noticed and this makes them ideal to be used chiefly as freshwater clarifiers due to their harmless and their local availability especially in remote areas where clean and drinkable water cannot be offered. The slight disparity between the presented values in Table 4 can be attributed to the experimental conditions, treated water characteristics and merely the specificity of each plant (molecular weight, charge density and intrinsic viscosity).

Point of view on plant-based coagulants/flocculants

For the four reviewed plants, it is clear that MO seeds, cactus pads, okra seed pods and mango seed kernels afford salient coagulation–flocculation ability in spite of the difference in their extract and active compounds nature. As well, it can be said that both proteins and carbohydrates are accountable for the high reactivity of these green coagulants/flocculants. Undeniably, the outstanding coagulating–flocculating activity of these plants promotes exploring further green resources with a typical identity and widely cultivated. Additionally and with reference to okra, valorization of fruit wastes as sustainable coagulants/flocculants in water processing is economically and environmentally feasible.

Further, providing a comprehensive understanding for the interactions occurring between plant-based coagulants/flocculants and water pollutants could be the corner stone to help screening more efficient plants among the claimed bioresources and therefore their valuable application in water processing.

As a hint, we depicted in Table 6 the so far main believed coagulation–flocculation mechanism for the currently investigated plants and other plants encompassing the same active agents.

Generally, the biopolymer active coagulating/flocculating agents can be mainly proteins or polysaccharides. As per Table 6 and owing to their efficiency, it can be deduced that mango or MO seed extracts could be typical models for other plant seed holding proteins as active coagulating–flocculating compounds like hyacinth bean (Unnisa et al. 2010) and cowpea (Hussain and Hydar 2019). Therefore, regardless of their protein content and their coagulating/flocculating performance, the predominant pollutant uptake mechanism of these plants can be considered to be adsorption and charge neutralization. However, for the other plant seeds like nirmali or fenugreek, their active coagulating agents are identified to be polysaccharides. These compounds are commonly known as galactomannans which are composed of galactose and mannose chain with 1:2 ratios as presented in Fig. 8.

These biopolymers are highly hydrophilic that deemed to be excellent hydrogen donors (Oladoja et al. 2017). Moreover, the long polymeric chain may also enable the seed extract-based polysaccharides to be used as a flocculant besides their coagulating proprieties (Lyklema, 1978).

Thus, and based on Table 6, alike cactus and okra, the coagulation–flocculation mechanism of all plants having active compounds as polysaccharides can be considered to be adsorption and bridging.

Further, as demonstrated by some scholars, several natural extracts could exhibit at the same time coagulating (Oladoja, 2015) and flocculating (Bouaouine et al. 2019) activities. In this frame, besides the conventional coagulation–flocculation process, arises another water treatment technology known as the direct flocculation. This new technique seems to be straightforward and gained a considerable importance owing to its eco-friendly features and low cost (Chong 2012).

According to Lee et al. (2014), the direct flocculation, usually conducted by amphoteric bio-resource-based

Table 6 Relationship between active coagulating agents and occurring mechanisms

Coagulation–flocculation mechanism	Plant	Plant parts	Assumed coagulating active agent	Other plant-based coagulants–flocculants	References
Adsorption and charge neutralization	*Moringa oleifera*	Seeds	Proteins: cationic polymeric chain of positively charged amino acids	Hyacinth bean (*Dolichos lablab*)	Unnisa et al. (2010)
	Mango (*Mangifera indica*)	Seed kernels	Cationic protein Interference of tannin (amphoteric behavior)	Cowpea (*Vigna unguiculata*)	Hussain and Hydar (2019)
Adsorption and interparticle bridging	Cactus (*Opuntia ficus-indica*)	Cladodes	Polysaccharides: anionic long chain of monosaccharides: arabinose, galactose, rhamnose, xylose and polygalacturonic acid	Coccinia indica fruit mucilage	Patale and Pandya (2012)
	Okra (*Abelmoschus esculentus*)	Seed pods	Polysaccharides: galactose, rhamnose, polygalacturonic acid	Isabgol (*Plantago psyllium*) seed husk	Mishra and Bajpai (2005)

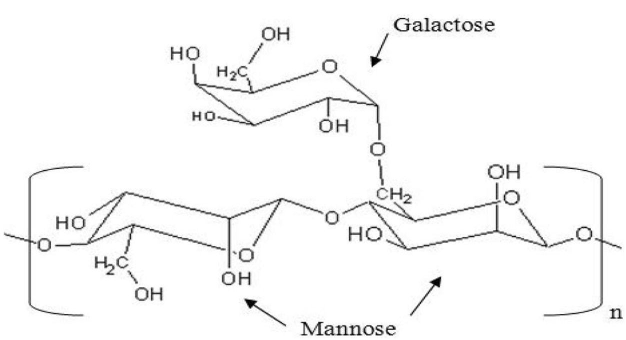


Fig. 8 Galactomannan molecular structure

coagulants/flocculants, revealed an efficient flocculating activity compared to the conventional coagulation–flocculation method. In addition, virtue of the strong charge density of the amphoteric plant-based coagulants/flocculants, pH adjustment might not frequently required since these biopolymers are ultimately able to bridge merely colloids mostly at any pH condition owing to the dual presence of anionic and cationic active compounds (Mukhtar et al. 2015). In the direct flocculation process, charge neutralization and bridging mechanisms may occur concurrently, indeed. Consequently, researchers should pay closer attention to provide more understanding on the direct flocculation process while using effective plant extracts.

To sum up, the valorization of efficient biomaterials in water and wastewater remediation can be regarded as a new gateway for the application of green chemistry and clean technology. In fact, unlike synthetic chemicals, natural coagulants/flocculants are freely available, safe, highly biodegradable and leading to a more compacted and non-corrosive sludge which might be treated by biological process or disposed as a soil conditioner or fertilizers under certain circumstances (Zhang et al. 2018).

Lastly, in treating water, it is interesting to point out that plant-based coagulants/flocculants do not alter the final pH of the treated effluent unlike mineral coagulants such as alum and iron salts which cause appreciable pH dropping. Moreover, using natural resources, pH variation is only recommended in case of ionic plant-based coagulants/flocculants with the aim of promoting the electrostatic interactions between effluents contents and the ionized biopolymer active compounds, such as amine (NH_2), carboxyl (COO) and hydroxyl groups (OH), while pH adjustment is not really needed in case of amphoteric bio-flocculant.

Bearing all the aforementioned benefits in mind, efficient plant-based coagulants/flocculants should be seen as a real and an effective surrogate to synthetic chemical reagents.

Conclusion

Plant-based coagulants/flocculants are being widely investigated for their effectiveness in removing various water pollutants. These green products are known by their eco-friendliness unlike synthetic chemicals which have been questioned for their detrimental impacts on environment and human health.

Outcomes gained from the present review reveal the efficacy of the four screened natural products (moringa seeds, cactus cladodes, okra seedpods and mango seed kernels) toward freshwater and wastewater remediation, though mango depicted the highest turbidity removal power, making it ideal for freshwater clarification.

To gain more understanding in the reactivity of the bio-resource material, the four presently investigated plants should be regarded as typical models to elucidate the relationship between active agents and their relative coagulation–flocculation mechanism. Within this line, it was concluded that the water contaminant uptake is mainly governed by adsorption and charge neutralization when the plant active agents are cationic proteins such in case of moringa or mango. However, for plants encompassing mucilaginous carbohydrates as active coagulating/flocculating compounds, pollutant removal mechanism is believed to be adsorption and bridging alike the case of cactus and okra.

Recommendations

In addition to their safety and efficiency, certainly, the biodegradability is one of the interesting features that can promote the application of green products in water treatment as alternatives to recalcitrant chemicals. Therefore, the shelf life of natural extracts (mostly proteins or polysaccharides) needs to be suitably investigated since they are highly biodegradable. As well, the influence of storage conditions and the efficient extract formulation which will be afterward available for commercial use should be closely looked up.

Lastly, to boost up the wide use of bio-based coagulant, it is recommended that laboratory studies should be scaled up on pilot plant feasibility using the most efficient plant extracts such as mango. Researchers should also continue digging to look for new coagulants/flocculants derived from other plants with similar characteristics to the currently reviewed bio-resources, and this surely will help giving up with chemical reagent input to waters, which in turn would pave the way toward a sustainable environment and a clean technology.

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