



REVIEW PAPER

Effectiveness of natural coagulants in water and wastewater treatment

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ABSTRACT

Natural waterways are contaminated due to industrialization, urbanization, population growth etc., degrading their quality. Contaminated waterways cause numerous health and environmental hazards. Therefore, it is imperative to remove contaminants. Coagulation is one of the efficient primary chemical treatment methods that could be used to treat such contaminants. Natural coagulants have gained popularity in the water and wastewater treatment industry due to their advantage over chemical coagulants. Natural coagulants are derived from either plants, animals, or microorganisms. This study has elaborated on the nature and mechanisms, and types of natural coagulants. In this review work, many studies have proposed several types of natural coagulants. However, plant-based natural coagulants extracted from different plant components have been extensively discussed and compared based on their application and efficiency in water and waste treatment. The primary purpose of this review is to refine the knowledge on the potential use and optimization of the effectiveness of eco-friendly and sustainable natural coagulants. Besides, the development efforts and the barriers reported by recent findings for the commercialization of natural coagulants are also discussed. Further, few modified natural coagulants have also been presented for exploring the other possible approaches to promote their usage in water and wastewater treatment in the future studies.

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INTRODUCTION

Water is the fundamental requirement for all human activities and biological activities. It is the main component in the hydrological cycle. The water resources are continuously decreasing around the world due to various environmental degradation activities, climate change (Konapala et al., 2020), population growth (Zubaidi et al., 2020), and increasing standards of living and urbanization (Wu et al., 2013). Rapid population growth and haphazard waste disposal have resulted in the impending water crisis. In order to sustain the water requirements, various processes and technologies are being researched to improve the quality of water (Ullah et al., 2020). These technologies fall into three main categories, namely physical, chemical, and biological treatment methods. Physical methods include settling, media, and membrane filtration (Obotey Ezugbe and Rathilal, 2020), adsorption (Ali and Gupta, 2006), and UV processes (O'Malley et al., 2020). Coagulation (Alibeigi-Beni et al., 2021), disinfection (Collivignarelli et al., 2017), ion exchange (Ergunova et al., 2017), catalytic reduction (Guo et al., 2020; Sivakumar, 2015), oxidation (Gogate and Pandit, 2004), and softening processes (Brastad and He, 2013) are some of the chemical methods used in the wastewater treatment. Biological methods include microbial biodegradation (Huang et al., 2018), phytoremediation (Hu et al., 2020), bioreactor processes (Neoh et al., 2016), constructed wetlands (Wu et al., 2015) etc. Moreover, some processes are combined with others to improve efficiency (Ang and Mohammad, 2020). One of the most widely used processes in water and wastewater's primary treatment is coagulation for removing suspended particulate matter and colloids in wastewater (Staicu et al., 2015). Coagulation is considered one of the simple methods to remove suspended solids and impurities in water efficiently. Successful coagulation can be attained by using either chemical-based (inorganic and synthetic organic) coagulants or natural coagulants (de Paula et al., 2018). Natural coagulants have been recognized for their traditional local water purification (Choy et al., 2014; Dorea, 2006). Naturally occurring coagulants are sustainable, environmentally friendly, and less toxic than chemical coagulants (Teh et al., 2014). Natural coagulants have grasped the scientific community's attention in the past decades due to their significant health and environmental benefits, and it solves most of the common problems associated

with chemical coagulants. Natural coagulants are produced or extracted from different sources such as microorganisms, animals, or plants (non-plant-based and plant-based). Now, several effective coagulants which have plant origin are being identified. Some of the common ones include *Hibiscus sabdariffa* (Roselle seeds) (Mohd-Esa et al., 2010), *Dolichos lablab* (Hyacinth bean) (Daverey et al., 2019), *Moringa oliefera* (Nonfodji et al., 2020), Nirmali seeds (Prabhakaran et al., 2020) watermelon seeds (Bhattacharjee et al., 2020) and cactus species (Rebah and Siddeeg, 2017). The drawbacks of chemical coagulants have resulted in the search for eco-friendly and sustainable natural coagulants in their usage and production. The main advantages of natural coagulants are renewability, biodegradability, nontoxicity, and cost-effectiveness. These studies have already proved the effectiveness of natural coagulants in wastewater treatment applications (Choy et al., 2014; Yin, 2010). However, the industrial usage of natural coagulants in wastewater treatment applications is limited. This is mainly due to the processing cost and the performance consistency of the extracted compounds from natural sources. Due to this, researchers tend to focus on modifying natural coagulants to get the maximum benefits (Muruganandam et al., 2017; Ahmed et al., 2016). This study aims to identify potential research gaps to refine the knowledge on natural coagulants and summarize the optimization methods for coagulants for improving their efficiency in water and wastewater treatment. This study also showcases the application of these coagulants for large-scale commercial usages and may assist in future studies. These will be discussed in sections as follows: the need for natural coagulants, mechanisms of natural coagulants, types of natural coagulants, barriers in the commercialization of natural coagulants, and examples of modified or blended natural coagulants. This study has been carried out in Colombo, Sri Lanka, in 2021.

Need for natural coagulants

Chemical coagulant used has raised controversial issues due to its toxic nature for living organisms and can be categorized into three types: hydrolyzing metallic salts, pre-hydrolyzing metallic salts, and synthetic cationic polymers (Freitas et al., 2018; Verma et al., 2012). Due to the low cost, easy handling, storage, and high availability, chemical coagulants are more prevalent in wastewater treatment processes.

$\text{Al}_2(\text{SO}_4)_3$, $\text{Fe}_2(\text{SO}_4)_3$, AlCl_3 , and FeCl_3 are the most commonly used coagulant salts (Freitas *et al.*, 2018; Matilainen *et al.*, 2010; Sher *et al.*, 2013). Despite the availability, low cost etc.; chemical coagulants are far behind in green chemistry due to high residual concentrations of aluminum found in treated wastewater (Freitas *et al.*, 2018; Matilainen *et al.*, 2010). According to Freitas *et al.*, 2018; McLachlan 1995; Polizzi *et al.* 2002, Alzheimer's disease is linked with the neurotoxicity of aluminum. Synthetic polymer coagulants form hazardous secondary products such as acrylamide which is carcinogenic and neurotoxic, and also synthetic polymers have low biodegradability (Freitas *et al.*, 2018; Kurniawan *et al.*, 2020). Excessive concentrations of chemical coagulants such as aluminum reduce the pH of water tends and also, they can be accumulated to food chains (Kurniawan *et al.*, 2020). Improper disposal of toxic sludge pollutes the groundwater and soil. Accumulation of toxic sludge, such as aluminum, iron etc., in natural water bodies causes adverse effects on aquatic organisms and plant species (Kurniawan *et al.*, 2020). Hence there is a need for the efficient utilization of natural coagulants for water and wastewater treatment.

Mechanism of coagulation by natural coagulants

Coagulation occurs between the coagulant added, the impurities, and the alkalinity of the water, resulting in the formation of insoluble flocs. Flocs are the agglomerations of particulate suspended matter in the raw water, reaction products of the added chemicals, colloidal and dissolved matter from the water adsorbed by these reaction products. Unprocessed water from the reservoir contains organic and inorganic impurities, such as silt, rotten substance, alga, bacterium, etc. Hence coagulation is the essential step in water purification. In addition, coagulants make suspensions in water to gather and reduce the turbidity of water (Z. Song *et al.*, 2009). The successful coagulation of natural coagulants (Ang *et al.*, 2020) stands on these three pillars: characteristics of coagulant used, characteristics of water to be treated, characteristics of mixing process (Ang *et al.*, 2020; Kumar *et al.*, 2017). As Fig. 1 shows, these coagulation factors play a significant role in determining the most efficient coagulant required for the treatment. Coagulants' molecular weight (Ang *et al.*, 2020; Gautam and Saini, 2020), types of equipment and reagents used, chemical and physical properties of the pollutants such as zeta

potential (Ang *et al.*, 2020), color, the concentration of the colloidal particles, the presence or absence of impurities (trace elements and dissolved salts (ions and chemicals) also affect the coagulation process (Ang *et al.*, 2020; Kumar *et al.*, 2017; Muruganandam *et al.*, 2017). If the natural coagulant contain positive surface charge, its coagulation activity against negatively charged suspended particles will be higher and vice versa for negatively charged natural coagulants with positively charged suspended particles. Functional groups also contribute to surface charge (Ang *et al.*, 2020). Molecular weight of natural coagulant is very important in particle bridging. If the molecular weight of natural coagulant is higher, it can form strong bridges with the particles and it leads to the formation of strong flocs and improve settling (Ang *et al.*, 2020). Mixing is another critical step in the coagulation process. Fast mixing increases the interactions between coagulants and suspended particles and forms micro flocs. Slow mixing leads to the aggregation of micro flocs into large flocs (Kurniawan *et al.*, 2020). Coagulation also affects the other steps of the treatment process. An efficient and effective coagulation process favors the microbiological quality (Kumar *et al.*, 2017) of the end product and increases the lifetime of filters (Kumar *et al.*, 2017), reducing the total cost of treated water.

Natural coagulants are composed of carbohydrates, protein, and lipids. The primary building blocks are the polymer of polysaccharides and amino acids. According to the previous research, the main mechanisms governing coagulation activity are charge neutralization and polymer bridging.

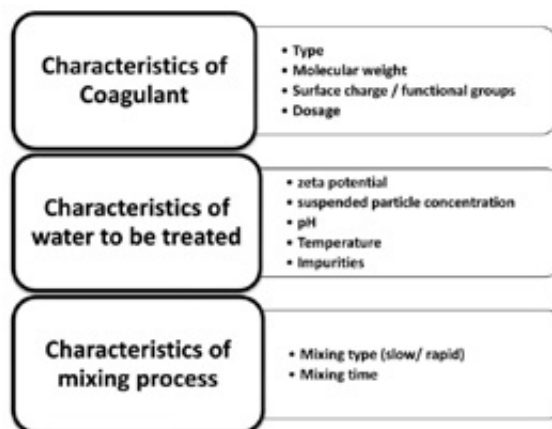


Fig. 1: Factors affecting coagulation (Ang *et al.*, 2020; Gautam and Saini, 2020; Kumar *et al.*, 2017)

Polymer bridging is preceded by polymer adsorption. Because of the affinity between long-chain polymers and colloidal particles, long-chained polymers can attach to the colloidal particle's surface. A part of the polymer is attached to the particle while the other parts form loops and tails. These loops and tails are the main structure of polymer bridging loops, and tails allow attaching to other colloidal particles and form larger flocs. The basis of charge neutralization is known as the electrostatic patch mechanism. The patches of positive and negative regions on the particle's surface cause the additional attraction between particles. Ionizable polymer (polyelectrolytes) is used as a coagulant in the charge neutralization mechanism. It stabilizes the negatively charged colloidal particles. Polycation is used to stabilize the particles, gaining near to zero zeta potential. The optimum dosage of polyelectrolyte needed will be determined by the charge density of the polyelectrolyte (Amran *et al.*, 2018; Yin, 2010). Natural coagulants have varied mechanisms of action. Let us consider some of the coagulation mechanisms of natural coagulants.

As shown in Table 1 Chitin is a cellulose-like biopolymer. It is found in fungi, marine invertebrates, yeasts, and insects. Chitosan is formed by the deacetylation of chitin (Hassan *et al.*, 2009; Saranya *et al.*, 2014). It is efficient in cold waters at low concentrations, producing less sludge and sludge degraded by microorganisms. Both charge

neutralization (has positively charged amino group) and bridging are the two coagulation mechanisms identified. Chitosan is a potential substitute for metallic salts and synthetic polyelectrolytes used in wastewater treatment (Nechita, 2017). Chitosan has a high content of amino groups. It provides a cationic charge at acidic pH, supports the destabilization of colloidal suspension, and promotes rapid-settling, large floc. Since Chitosan is a long-chain polymer with positive charges, it can also coagulate negatively charged particulate and colloidal materials via adsorption and hydrophobic flocculation (Roussy *et al.*, 2005; Saranya *et al.*, 2014; Karbassi and Heidari, 2015). Seed extracts of Nirmali (*Strychnos potatorum*) are anionic polyelectrolytes. It can destabilize the particles in water via inter-particle chemical bridges. Nirmali seed extract contains lipids, carbohydrates, and alkaloids. Hydroxyl groups found in the polymer chain provide adsorption sites for forming chemical bridges (Theodoro *et al.*, 2013; Yin, 2010) and -COOH and free -OH surface groups increase the coagulation competency (Yin, 2010). Polysaccharides mixtures of galactanii and galactomannani extracted from *Strychnos potatorum* seeds can reduce turbidity up to 80% (Adinolfi *et al.*, 1994). According to the study carried out by Ndabigengesere *et al.*, 1995, Moringa extracts consist of water-soluble cationic coagulant proteins. Coagulation activity is carried out via the mechanism of charge neutralization and adsorption (Ndabigengesere *et al.*, 1995; Theodoro *et al.*, 2013;

Table 1: Summary of functional groups and mechanism proposed for natural coagulants

Natural coagulant	Source of extraction	Mechanism proposed	Functional groups	Reference
<i>Moringa oleifera</i>	leaves, flowers, seeds, roots, bark	Adsorption and charge neutralization	Starch, cationic protein, fatty acids, phenolic compounds Amines, glucose, alcoholic compounds, and carboxylate groups	Kumar <i>et al.</i> , 2017; Kurniawan <i>et al.</i> , 2020; Yin, 2010
Nirmali (<i>Strychnos potatorum</i>)	seeds	Inter-particle bridging	galactan and galactomannan	Vijayaraghavan <i>et al.</i> , 2011
Cactus Musilage	Cactus pads	adsorption and bridging coagulation method	D-xylose, galacturonic acid, l-arabinose, l-rhamnose, and d-galactose.	Vijayaraghavan <i>et al.</i> , 2011; Yin, 2010
Chitosan	fungi, marine invertebrates, yeasts	Charge neutralization and bridging	N-acetyl-D-glucosamine (acetylated unit), β -(1-4)-linked D-glucosamine (deacetylated unit)	Saranya <i>et al.</i> , 2014
Tannins	Castanea, Acacia, or Schinopsis	Adsorption and charge neutralization	polyphenol compounds	Vijayaraghavan <i>et al.</i> , 2011; Yin, 2010

Yin, 2010). Tannins are secondary metabolites of plants, produced from the barks, leaves, fruits, seeds regarded as a potential natural coagulant for water and wastewater treatment (Yin, 2010; Grenda *et al.*, 2020). These polyphenol compounds are obtained from Castanea, Acacia, or Schinopsis plants. Tannins contain phenolic groups of anionic nature. These phenolic groups can be deprotonated, and form phenoxide stabilized resonance, allowing coagulation (Özacar and Sengil, 2003; Yin, 2010). The effectiveness of tannin as an eco-friendly coagulant depends on the chemical structure of the extracted tannins and their degree of modification. If more phenolic groups are available in a tannin structure, the coagulation capability will increase (Yin, 2010). The high coagulation ability of the cactus is due to the presence of mucilage. It assumes as sticky and complex carbohydrates. Surface cactus pads have high water retention capability. Cactus mucilage is made up of galacturonic acid, galactose, arabinose, xylose, and irhamnose. It is stored in internal and external parts of the cactus (Sáenz *et al.*, 2004; Theodoro *et al.*, 2013). According to Miller *et al.*, 2008, cactus mucilage coagulation occurs by forming chemical bridges via hydrogen bonds or dipole interactions. Polygalacturonic acid present in mucilage is responsible for forming chemical bridges (Miller *et al.*, 2008). Polygalacturonic acid structure consisted of an anionic chain, Chemisorption is involved between the charged particles and –OH and –COOH groups due to their partial de-protonation in aqueous solutions (Theodoro *et al.*, 2013; Yin, 2010).

Types of natural coagulants

The natural coagulants have characteristics that are not noxious to an aquatic environment. It includes microbial polysaccharides (Saleem and Bachmann, 2019), bio-wastes (Atchudan *et al.*, 2020), alginate, gelatin, cellulose-based materials, and Chitosan (Vigneshwaran *et al.*, 2020). Most of the natural coagulants are polysaccharides; hence they are also termed polymeric coagulants. According to the origin, natural coagulants can be divided into three categories, as shown in Table 2.

Animal-based natural coagulants

Chitosan (CS) is a linear copolymer produced by the deacetylation of chitin (Verma *et al.*, 2012). Chitosan offers several advantages over traditional compounds. For example, it is widely available (higher

after cellulose), sustainable, cost-effective, non-toxic, biodegradable, biocompatible, soluble in weak acids, pH-sensitive (Martău *et al.*, 2019; Pontius, 2016), better biosorption, no secondary pollution, sludge can reuse as agricultural fertilizer etc. (Abreu *et al.*, 2020; Huang *et al.*, 2000). In addition, Chitosan is efficient in reducing chemical oxygen demand, suspended solids, and turbidity (Abdullah and Jaeel, 2019).

Plant-based natural coagulants

The use of natural plant extracts dates back to 2000 BC, where Egyptians have inscribed the evidence of plant materials used for water treatment (Sivaranjani and Rakshit, 2016). According to Fatombi *et al.*, 2013, it is clear that nuts such as beans, almonds, and *Strychnos potatorum* were used in Sudan, Egypt, and India, respectively. These nuts are reported to stimulate the coagulation of turbid waters (Fatombi *et al.*, 2013). Since the late 1970s, various plant-based polyelectrolytes and polymers have been researched as coagulants. Plant-based coagulants are generally derived from the various parts of the plants and are organic, water-soluble, ionic, and non-ionic polymers in nature (Bodlund *et al.*, 2014; Dezfooli *et al.*, 2016; Fatombi *et al.*, 2013). In the colloid-free aqueous state and the colloidal particle solution consisting of restricted irreversible loop arrangements, they maintain random configurations and help in destabilization by forming micro or macro flocs through charge neutralization (Hameed *et al.*, 2016). Some plant-based materials may also behave as flocculant by strengthening the flocs for better settleability (Al-Hamadani *et al.*, 2011; Awang and Aziz, 2012). Several works of literature have reported applying plant-based coagulants for water and wastewater treatment (Kansal and Kumari, 2014; Kristianto, 2017; Oladoja *et al.*, 2017). Most of the investigated coagulants are from family *Fabacea*, primarily extracted from the leaves (Rak *et al.*, 2012)

Table 2: Types of natural coagulants

Plant-based	Animal-based	Microorganism-based
<i>Moringa Oleifera</i>	Chitosan	Xanthan gum
Cactus	Alginate	<i>Aspergillus sp.</i>
Nirmali seeds	Chitin	<i>Enterobacter</i>
Tannin		<i>Streptomonas</i>
Potato starch		
Banana peel		
Common beans		
Tamarind seeds		

and seeds (Jayalakshmi et al., 2017). One of the most popular and extensively researched plant-based coagulants is *Moringa oleifera* belonging to the family *Moringaceae* (Baptista et al., 2017; Camacho et al., 2017). The other common coagulants are Nirmali seeds, Tannins, Roselle seeds, Hyacinth bean etc, which have been studied for turbidity reduction (Saharudin et al., 2014; Fermino et al., 2017; Choubey et al., 2012). As in Table 3 is shown, they are low cost, non-toxic, locally available, readily implementable, and show great potential. Plant-based coagulants are advantageous because i) They are not dependent on chemicals; ii) they generate smaller amounts of sludge and biodegradable; and (iii) less toxic and not corrosive (Rocha et al., 2019).

Many wastewater treatments have substituted chemical coagulants with plant-based coagulants because of their low price, abundant source, multi-purpose, and biodegradability (Othmani et al., 2020).

Comparison of natural coagulants efficiency

The list of plant-based coagulants studied as natural coagulants are summarized in Table 4, with brief accounts of their optimal conditions, applications, and efficiencies. Roselle seeds (*Hibiscus sabdariffa*) were high in proteins (28 %) and soluble in water. When in solution, they carry an overall positive charge. These positively charged proteins bind to the turbidity causing negatively charged particles in the solution. According to the research, roselle seeds' highest turbidity removal efficiency is within 77 % - 87 % for synthetic wastewater at pH 10 and 81 % - 93 % at pH 4 (Saharudin and Nithyanandam, 2014). *Moringa oleifera* has been one of the best plant extracts for water purification. It is effective in removing Biological Oxygen Demand (BOD), turbidity, Chemical Oxygen Demand (COD), total coliforms removal, algal removal, Hardness, Total dissolved solids (TDS), and Total suspended solids (TSS) etc. According to the research carried out by (Choubey et al., 2012), *Moringa oleifera* removes turbidity from 100 NTU to 5.9 NTU and after dosing, and filtration to 5 NTU and total coliform remove by 96 % in synthetic raw water. Furthermore, in laundry, wastewater turbidity removed by 84 % and COD by 46% (Al-Gheethi et al., 2017). In Municipal wastewater, the reduction of turbidity, COD, BOD, hardness, TSS, and TDS are found to be 61 %, 65 %, 55 %, 25 %, 69 %, and 68%, respectively (Kumar Kaushal and Goyal, 2019). Hyacinth bean (*Dolichos lablab*) peels are

Table 3: Characteristics of natural coagulants

Parameters	Characteristics
Carbon footprint	Environmentally friendly
Toxicity	Less toxic
Heavy Metals	Settling will occur along with the coagulation process
Sludge	Sludge volume/amount reduction, Low sludge handling cost, and treatment cost with good biodegradability

characterized for usage as a protein source. Hyacinth bean peels have a moderate concentration of protein. Turbidity removal efficiency is 99% with the dosage of 20 mg/L in synthetic water (Bs and Papegowda, 2012). With the 200 mg/500 ml dosage, turbidity removes from 100 NTU (Nephelometric Turbidity Units) to 11.1 NTU and after dosing, filtration to 9.5 NTU, and total coliform removal 89% in synthetic water (Choubey et al., 2012). *Cactus* is another efficient natural coagulant. According to various research studies, cactus species prove efficient in removing turbidity, COD, and color. For example, in textile wastewaters, cactus removes turbidity by 92 %, COD by 89 %, and color by 99 % at the dosage of 40 mg/L and pH 7.25 (Bouatay and Mhenni, 2014). *Nirmali seeds* are another crucial natural coagulant used to remove turbidity and total suspended solids (TSS). It removes TSS by 76%, turbidity by 96% in laundry wastewater (Mohan, 2014). *Watermelon (Citrullus lanatus)* is the latest approach in developing an effective natural coagulant. The efficiency of turbidity removal was 88 % for the tannery effluent and 98% for synthetic wastewater. The other physicochemical parameters of tannery wastewater, such as TSS, BOD, and COD, were also reduced significantly. The COD removal efficiency was 50%, and the BOD of the wastewater was reduced by 55%. When employed as a coagulant, the watermelon seeds significantly decrease the synthetic wastewater's TSS, turbidity, BOD, and COD, and the tannery effluent (Sathish et al., 2018). Table 4 summarizes the facts on potential applications of plant-based materials that can be used as natural coagulants. This is significant in developing new mixed natural coagulants which deliver maximum efficiency.

Therefore, it can be said that natural coagulants have found their diverse application not only for physical and biological water and wastewater treatment but also as a disinfectant. From a

Table 4: Summary of the removal efficiencies of some natural coagulants for water and wastewater treatment

Natural coagulant	Wastewater source	Optimal Conditions		Performance		References
		Dosage	pH	Parameters	Removal Efficiencies	
Industrial wastewater						
<i>Hibiscus sabdariffa</i> (Roselle seed extract)	Glove manufacturing wastewater	60 mg/L	10 ≤	Turbidity	87%	Saharudin and Nithyanandam, 2014
<i>Moringa oleifera</i> (Drumstick Tree)	Laundry wastewater	120 mg/L	5.7	COD	43%	Al-Gheethi et al., 2017
				Turbidity	84%	
<i>Opuntia ficus indica</i> (Cactus species)	Textile wastewater	40 mg/L	7.25	Turbidity	92%	Bouatay and Mhenni, 2014
				COD	89%	
				Color	99%	
<i>Citrullus lanatus</i> (Seeds of Water Melon)	Tannery wastewater	2000 mg/L	-	Turbidity	87%	Sathish et al., 2018
				BOD	55%	
				COD	50%	
				TSS	69%	
<i>Strychnos potatorum</i> (Nirmali seeds)	Laundry wastewater	8000 mg/L	-	Turbidity	96%	Mohan, 2014
				TSS	76%	
<i>Ocimum basilicum</i> (Basil)	Textile wastewater	1600 mg/L	8.5	COD	62%	Shamsnejati et al., 2015
				Color	69%	
<i>Corchorus Olitorius L.</i> (Jute mallow)	Agricultural wastewater	3 mg/ L	-	Turbidity	95%	Altaher et al., 2016
				TOC (Total organic carbon)	100%	
				BOD	95 %	
				COD	88 %	
Bamboo (<i>Bambusa vulgaris</i>)	Electroplating industry wastewater	1500 mg/L	5.5	Cl-	90 %	Sivakumar et al., 2018
				Sulphate	93 %	
				TDS	97 %	
				Ni	99 %	
				TSS	87%	
<i>Cassia obtusifolia</i> (Sickle pod seed gum)	Raw pulp and paper mill effluent	750 mg/L	5	COD	36%	Subramonian et al., 2014
				Turbidity	90%	
Pectin of orange peel pith	Surfactant	8000 mg/L	-	TSS	82%	Mohan, 2014
				Turbidity	99%	
Dragon fruit foliage	Concentrated latex effluent	800 mg/L	10	COD	95%	Idris et al., 2012
				TSS	89%	
Papaya Seed	Textile wastewater	570 mg/L	2	Color	85%	Kristianto et al., 2018
	Olive mill wastewater	400 mg/L	-	TSS	81 %	Rizzo et al., 2008
Chitosan	Paper and pulp wastewater	1800 mg/L	-	Turbidity	84 %	
				BOD	90%	
				COD	93 %	
	Brewery wastewater	120 mg/L	-	Turbidity	95 %	Gautam and Saini, 2020
				COD	73%	
Textile wastewater	30 mg/L	-	Turbidity	95%	Hassan et al., 2009	
			COD	65%		
			Cr(VI)	96 %		
			TDS	92 %		
			TS	98 %		
<i>Aspergillus niger</i>	Tannery industry wastewater	4.0 g	3	COD	85 %	Sivakumar, 2016
				BOD	90 %	
				Sulphates	94 %	
				Turbidity	61%	
Municipal wastewater						
<i>Moringa oleifera</i> (Drumstick Tree)	sewage, gray water (water from sinks and showers)	-	-	Turbidity	61%	Kumar Kaushal and Goyal, 2019
				COD	65%	
				BOD	55%	
				TSS	69%	
				TDS	68%	
<i>Citrullus lanatus</i> (Seeds of Water Melon)	Sewage wastewater	72.3 mg/L	5	Hardness	25%	Joaquin et al., 2021
				BOD	92%	
				TSS	93%	
				COD	80%	
<i>Cucumis melo</i> (Cantaloupe seeds)	sewage, gray water (water from sinks and showers)	76.7 mg/L	7	TSS	88%	Kumar Kaushal and Goyal, 2019
				Turbidity	65%	
				BOD	56%	

Natural coagulants for water and wastewater

Continued Table 4: Summary of the removal efficiencies of some natural coagulants for water and wastewater treatment

	from sinks and showers)			COD	67%	
				TDS	69%	
				TSS	72%	
				Hardness	30%	
Synthetic water						
<i>Moringa oleifera</i> (Drumstick Tree)	Raw water synthetic	100 mg/L	10 ≤	Turbidity	94 %	Choubey <i>et al.</i> , 2012
				Total coliforms	96%	
<i>Dolichos lablab</i> (Hyacinth bean)	Turbid water (synthetic)	400 mg/L	-	Turbidity	89 %	Choubey <i>et al.</i> , 2012
				Total coliforms	89%	
<i>Cicer arietinum</i> (Chickpea)	River water (Synthetic)	20 mg/L	-	Turbidity	99%	Bs and Papegowda,
				Turbidity	96 %	
<i>Opuntia ficus indica</i> (Cactus species)	Turbid water (synthetic)	400 mg/L	-	Total coliforms	90%	Choubey <i>et al.</i> , 2012
Mango pith	River water (Synthetic)	20 mg/L	-	Turbidity	99%	Bs and Papegowda, 2012
<i>Trigonella foenum-graecum</i> (Fenugreek seeds)	Turbid water (Synthetic)	500 mg/L	13	Turbidity	98%	Qureshi <i>et al.</i> , 2011
<i>Citrullus lanatus</i> (Seeds of Water Melon)	Turbid water (Synthetic)	300 mg/L	8.0	Turbidity	98%	ELsayed <i>et al.</i> , 2020
<i>Jatropha curcas</i> seed	Turbid water (Synthetic)	2000 mg/L	-	Turbidity	98%	Sathish <i>et al.</i> , 2018
<i>Lens culinaris</i> (Red lentil)	Turbid water (Synthetic)	120 mg/L	3	Turbidity	96%	Abidin <i>et al.</i> , 2011
<i>Cassia fistula</i> (Golden shower)	Turbid water (Synthetic)	26.3 mg/L	4	Turbidity	99%	Chua <i>et al.</i> , 2019
<i>Alginate</i>	Synthetic paint industry wastewater	160 mg/L	8.4	Color	96%	Vishali <i>et al.</i> , 2020
				Turbidity	98%	
	Turbid water synthetic	10 mg/L	-	Turbidity	98%	Devrimci <i>et al.</i> , 2012
Surface water						
<i>Moringa oleifera</i> (Drumstick Tree)	Raw surface water	150 mg/L	7	Algal removal	65%	Ali <i>et al.</i> , 2008
	Surface water	150 mg/L	7.5	Turbidity	97%	Pritchard <i>et al.</i> , 2010
<i>Cassia alata</i> (Christmas candles)	Raw surface water	1000 mg/L	-	Turbidity	93%	Rak and Ismail, 2012
				Suspended solids	56%	
				Turbidity	93%	
<i>Opuntia dillenii</i> (Cactus species)	Highly turbid lake water	1000 mg/L	-	Color	15%	Nougbodé <i>et al.</i> , 2013
				Suspended solids	89%	
<i>Prunus armeniaca</i> (Apricot)	Raw surface water	30 mg/L	7	Algal Removal	55%	Ali <i>et al.</i> , 2008
<i>Mangifera indica</i> , (Mango)	Raw surface water	30 mg/L	7	Algal Removal	68%	Ali <i>et al.</i> , 2008
Tannin	Surface water	30 mg/L	9	Turbidity	80%	Sánchez-Martín <i>et al.</i> , 2010
				Turbidity	99%	
				COD	54%	
				Suspended Solids	96%	
				Sulphates	99%	Kakoi <i>et al.</i> , 2016; Karbassi and Pazoki, 2015
Banana pith	Polluted river water	100 mg/L	4	Nitrates	89%	
				Cu	100%	
				Cr	100%	
				Fe	92%	
				Zn	81%	
				Pb	100%	
				Mn	60%	

disinfection point of view, comparing the chemical and natural coagulants is interesting, considering their different parameters during usage. The natural coagulants are biodegradable and have no toxic effect on the receiving water bodies, which is a significant issue for chemically disinfected waters. Moreover, compared to chemical coagulants, the natural coagulants generally are readily available and generally sourced from the local areas making an attractive alternative as a disinfectant, which eliminates the need for storage in a controlled room. A study by Amran *et al.* (2018) has also emphasized on the need to conduct more detailed studies on the efficiency of plant-based coagulants for water and wastewater treatment. Therefore, detailed studies are required to explore its possibilities as a disinfectant for commercial purposes.

Natural coagulants - barriers for the commercialization

Most natural extracts have proven their coagulation capabilities in removing COD, BOD, TSS, turbidity, etc.; not many have accepted and reached commercialization. Four main barriers hinder commercialization: Financial capability, regulatory approval, market awareness, and research development (Choy *et al.*, 2014). Another study by Saleem and Bachmann (2019) has highlighted the coagulants’ cationic, anionic, and non-ionic nature and explored its application and commercialization constraints. The existing research outcomes are mostly confined to laboratory scale, lacking in real industrial applications. Lacking financial freedom and

understanding about the market hinder the investors from investing in a new product. Economically feasible extraction methods are essential for successful commercialization. Comprehensive studies on the coagulation mechanism are also limited. Approval from the local government authorities and other regulatory authorities must be granted to launch any new products successfully. Obtaining approval is not easy without ensuring product compliance to the respective standards. Strong motivation for green chemistry concepts and cleaner production of the investors are crucial for the natural coagulant development and their applications. Table 5 summarizes the barriers that affect the successful commercialization of natural coagulants.

The main barrier for the commercialization of natural coagulants is difficulty in bulk production of raw materials; plant species. Raw materials used to produce chemical coagulants such as aluminum, iron are abundant in nature. For a successful and realistic application, raw materials required to produce natural coagulant should be available in large scale. Technical support, expert support and new equipment are necessary in sustainable implementation of natural coagulants so that production cost will ultimately increase. In short run this is not very economical so that market acceptance will be less. Hence the absence of mass plantation of recourses hinders steady supply of raw materials and the long term applications (Kurniawan *et al.*, 2020). As Table 5 shows, natural coagulants are not readily available. Plant materials pass few stages before convert in to a plant-based

Table 5: Barriers in the commercialization of natural coagulants

Environmental and Technical Constraints	Economic and Social Constraints
<ul style="list-style-type: none"> • Complex extraction process • Absence of mass plantation for bulk processing • Due to the organic properties of natural coagulants, COD levels might increase. • Lack of toxicological studies for purified coagulants. • Seasonal variations in some plant resources. (Cactus grow in hot seasons) • Lack of research regarding the practical usage and issues occurring during the operations within the plant • Lack of proper arrangements for storage of the natural coagulants in stock. • Improper estimation of the quality characteristics of the treated water 	<ul style="list-style-type: none"> • Lack of money and time to invest in research and development. • Lack of maintaining a steady supply of raw materials. • Lack of meeting the minimum quality requirement. • Lack of regulatory approvals on plant-based coagulants. • Lack of awareness and market interest. • Well established, competitive market. • High initial establishment cost. • Industrial acceptance • Lack of knowledge on health improvements

natural coagulant. These stages includes: extraction of active compound, purification etc. Extraction processes are different form plant- based to animal-based coagulants. Hence these processes should be carefully analyzed and should produce simplified and economically feasible processing steps. So that further studies needed to be carried out to analyze the converting and handling of powdered forms of coagulants, storing and preservation as well as toxicity (Kurniawan et al., 2020). Therefore commercialization process is costly as well as may take more time than expected. As mentioned in Fig. 1 coagulation process depends on coagulant type and dosage, pH, temperature etc. Hence these parameters should be optimized before implementing to the industry and it is more time consuming so that it will be difficult to cope with the competitive market (Kurniawan et al., 2020). Government and non-government regulatory bodies should encourage the industry to use natural coagulants in wastewater treatment process by implementing environmental rules (Freitas et al., 2018; Kurniawan et al., 2020) and reducing tax payments (Kurniawan et al., 2020), introduce new loan schemes to cover the initial implementation costs, linking the connection between research organization and industry and give recognition for the use of natural coagulants. Due to the use of high optimal dosages of natural coagulants, high amounts of residual organic matter (Freitas et al., 2018; Oladoja, 2016) is found in treated wastewater hence the COD levels will increase. This will ultimately increase the microbial activity leading to change in color and emission of unpleasant odors (Kurniawan et al., 2020; Oladoja, 2015). When selecting a plant species as raw material for the production of natural coagulants, it is important to consider their seasonal variations and availability (Kurniawan et al., 2020). Hence research and development studies should be conducted to identify best, economically productive natural coagulants and also to identify feasible combinations of chemical and natural coagulants.

Modified natural coagulants

A breakthrough in the commercial profit-oriented market can be made by emphasizing the need for blended coagulants. Lately, Mohd-Salleh et al., (2019) indicated natural materials as aids for future coagulant production and discussed the potential to develop composite coagulants that are sustainable. Natural coagulants can be used in conjunction with modifying

agents. Table 6 shows some of the modifying agents used in combination with natural coagulants. Many of them are also active as flocculants when combined with a modifying agent. Chitosan is biodegradable and eco-friendly in comparison with traditional coagulants and acts as a bio-flocculant. It is used as a substitute for conventional coagulants in water treatment. Chitosan has primary amino groups; these amino groups can remove various contaminants. Previous studies have shown that Chitosan removed around 99% of *Microcystis aeruginosa* cells. However, its low production yield leading to a high cost of operation has hindered its application as a sole coagulant in practical scenarios (Ma et al., 2016). Due to its insolubility in neutral and alkaline conditions, the application of Chitosan is usually limited. Chemically modified chitosan materials have been manufactured to overcome this issue (Dharani and Balasubramanian, 2015; Zhang et al., 2014). According to the study (Vigneshwaran et al., 2020), Moringa seeds are considered natural coagulants/flocculents. It is mainly comprised of lipids and protein. The protein molecules of moringa seeds can bridge $-NH_2$ groups and $-OH$ groups present in the chitosan molecule. Bridging will leads to the destabilization and aggregation of the small stable colloidal impurities into larger particle units. Hence, it is known as floc. The floc can remove through several physicochemical processes such as solid-liquid separation, slow mixing, and rapid mixing (Vigneshwaran et al., 2020). Recently, natural polymeric coagulants, such as cellulose, Chitosan, starch, have drawn more attention due to their advantages of low price, biodegradability, vast resources, and low toxicity. In addition, attention was focused on starch-based coagulants because starch is one of the most abundant natural polymers globally and has been applied in various fields (Li et al., 2015).

According to Ma et al., 2016 dual coagulant prepared by using chitosan and aluminum chloride is efficient in removing toxic cyanobacteria *Microcystis aeruginosa* found in water bodies. This novel mixed coagulant shows a strong bridging ability and high adsorption. This study suggests that CTASC is efficient, cost-effective, specific, and safe in removing *Microcystis aeruginosa*. Zhang et al., 2014, mentioned chitosan-based flocculent N-carboxyethylated chitosan (CEC) is eco-friendly coagulant produced by chitosan and acrylic acid. It flocculates copper (II) and tetracycline (TC). The advantages are in the aspects of optimal dosage,

Table 6: Summary of the literary works on the modified natural coagulants

Natural coagulants	Modifying agent	Wastewater source	Parameter	Optimal conditions		Removal efficiency (%)	References
				Dosage (mg/L)	pH		
	Aluminum chloride (AC)	Drinking water treatment	(<i>Microcystis aeruginosa</i>) cyanobacteria removal	2.6 CS + 7.5 AC	-	98	Ma et al., 2016
Chitosan (CTS)	Acrylic acid	Livestock wastewater	Copper (II) removal	50	8	98	Zhang et al., 2014
	Mercaptoacetic acid	Turbid water (Synthetic)	Turbidity	50	5	100	Zhang et al., 2015
			Copper (III) removal	50	7.3	90	
Starch	N-methyl piperazinium chloride	Tannery effluent	BOD	5	6.7	86	Dharani and Balasubramanian, 2015
			COD	5	6.7	96	
	<i>Moringa oleifera</i>	Turbid water (Synthetic)	Turbidity	200	8	84	Vigneshwaran et al., 2020
	(2-hydroxypropyl) trimethylammonium chloride	Synthetic wastewater	Kaolin suspension removal	1000	4	93 %	Li et al., 2015

lowest residual concentrations, and floc properties. The charge neutralization mechanism is used for the removal of copper (II), and TC makes a coordination complex with copper(II) hydroxides and eliminates with copper (II) at pH 9. Mercapto - acetyl chitosan (MAC) is prepared by combining mercaptoacetic acid and chitosan. It has the ability of adsorption bridging so that water solubility is high (Zhang et al., 2015). Turbidity is removed by electrical neutralization of the turbidity substances. Heavy metal ions produce coordination complexes or chelate with MAC and form flocs (Zhang et al., 2015). According to Dharani and Balasubramanian, 2015 study, Chitosan-g-N-MPC is prepared by grafting N-methyl piperazinium chloride to chitosan. Flocculation mechanisms are charge neutralization and bridging. Due to the considerable molecular weight and high charge density, it requires low optimal dosages. According to Vigneshwaran et al., 2020, acid-treated carbonized chitosan-*Moringa oleifera* (ACCM) shows better adsorption ability, high coagulation capability, low sludge formation and low leaching level. Coagulation mechanisms are charge neutralization and adsorption. Li et al., 2015 discussed two kinds of starch-based flocculants. Though they have the same chemically modified functional groups, substitution degrees are different. CMS-CTA-P and CMS-CTA-N have opposite surface charge properties

in water. CMS-CTA-N is efficient in the hematite suspension at neutral conditions. CMS-CTA-P shows a better flocculation activity in the kaolin suspension at neutral and acidic conditions. Patching, bridging, and charge neutralization are found as flocculation mechanisms.

CONCLUSION

Coagulants obtained from many natural sources have found their place in the water and wastewater industry world and are widely being used as primary coagulants or coagulant aids. Natural coagulants are environmentally friendly, inexpensive, less hazardous to human beings, and viable alternatives to chemical coagulants. Plant-based, animal-based, and microorganism-based coagulants have been researched for ages and have become popular in developing countries. This review summarized the efficiencies of common natural coagulants such as Roselle seeds, *Moringa oleifera*, Hyacinth bean, Cactus, Nirmali, Chitosan, Tannins and Watermelon seeds etc., used in the water and wastewater treatment and suggested that plant-based species showed good efficiencies in removing turbidity, color, organic matters as well as pathogens. It was noticed that many studies had investigated the application of plant-based coagulants in the primary treatment

for turbidity removal and secondary treatment for organic pollutant (TSS, BOD, and COD) removal. However, its disinfection aspect is not well explored. Studying the plant-based coagulants or plant species for the tertiary treatment of water and wastewater could be an exciting area for future research. Further, plant-based coagulants are advantageous due to their low toxicity and eco-friendly sludge production. Despite having significant benefits, some crucial barriers to the commercialization of natural coagulants are identified in this review. The significant barriers are environmental, technical, economic, and social challenges. However, there have been efforts made to commercialize natural coagulants through modified natural coagulants. There are two modified coagulants summarized in this work: Chitosan and starch, which are considered an alternative way to enhance the efficiency of the coagulants and increase its market demand. The concept of modified or composite coagulants could be taken as an indirect example for tackling these constraints. However, there are limited studies on these barriers, and this review recommends that more investigations and assessment methods are required to find the origin of these constraints and solve it through more scientific approaches. Further, from a sustainability perspective, the demand for natural coagulants is destined to increase. Therefore, more researches in the modified coagulants hold promising prospects.

AUTHOR CONTRIBUTIONS

S. Nimesha has done most of the writing and preparing the manuscript. C. Hewawasam has done some part of writing, editing and supervision of works of first author. D.J. Jayasanka has done some part of writing, editing and supervision of works of first author. Y. Murakami is the advisor for the writing this review. N. Araki is the advisor for this research work. N. Maharjan has given significant intellectual inputs and supervised this work.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, ethical issues, including plagiarism, informed consent, misconduct, data fabrication and falsification, double publication and submission, and redundancy, have been entirely witnessed by the authors.

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ABBREVIATIONS

AC	Aluminum chloride
ACCM	Acid treated carbonized chitosan- <i>Moringa oleifera</i>
AD	Alzheimer's disease
BOD	Biological oxygen demand
CEC	Carboxy-ethyl Chitosan
<i>Chitosan-g-N</i>	N-methyl piperazinium chloride grafted Chitosan
<i>MPC</i>	(2 hydroxypropyls)
<i>CMS-CTA-P</i> /	trimethylammonium chloride
<i>CMS-CTA-N</i>	etherified carboxymethyl starch (two different substitution degrees)
COD	Chemical oxygen demand
-COOH	Carboxyl group
Cr	Chromium
CS	Chitosan
CTS	Chitosan to be modified
CTS- AC	Chitosan -aluminum chloride

Cu	Copper
Fe	Iron
g	Grams
MAC	Mercapto - acetyl chitosan
mg/L	Miligrams per liter
Mn	Manganese
-NH ₂	Amine group
Ni	Nickel
NTU	Nephelometric turbidity units
-OH	Hydroxyl group
Pb	Lead
pH	Potential (power) of hydrogen
TC	Tetracycline
TDS	Total dissolved solids
TOC	Total organic carbon
TS	Total solids
TSS	Total suspended solids
Zn	Zinc
%	Percentage sign

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