

Review

Application of Natural Coagulants in Water Treatment: A Sustainable Alternative to Chemicals

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Abstract: Water treatment (WT) is currently among the major areas of research due to the depletion of water resources and fearmongering regarding environmental pollution, which has compelled the upgrading of conventional WT technology towards recycling and reuse. This review aims to provide the current state of natural coagulants and their application in the purification of surface water as sufficient clean water is required for household needs, health security, and environmental safety. A thorough and systematic review of the existing literature was performed, and the information related to water treatment using natural coagulants was compiled from 237 articles under various sections using a computerized bibliographic search via PubMed, Scopus, Web of Science, Google Scholar, CAB Abstracts, and several websites. The work provides explicit information related to natural coagulants and their merits and limitations, outlines methods to increase their coagulation performance, and highlights their coagulation mechanism, efficacy, valorization potential, and sustainability. From the information obtained, it can be concluded that although chemical coagulants are efficient in WT, they are usually expensive, toxic, associated with health issues, and thus non-sustainable. A sustainable alternative is the use of natural coagulants, which are readily available, economical, easy to use, biodegradable, non-toxic, eco-friendly, effective, and generate lower sludge volumes. They work via an adsorption process that involves polymeric bridging or neutralization of the charge. The WT efficiency of natural coagulants ranges from 50–500 nephelometric turbidity units (NTUs), which is similar to chemicals. Thus, they can be deployed in WT regimes and can contribute to the health security of rural populations in developing countries. It is unfortunate that, despite the known benefits of natural coagulants, their acceptance, commercialization, and widespread industrial application across the globe are still low. Therefore, there is a need for more exhaustive investigations regarding the mode of action, adoption, and commercialization of natural coagulants as a sustainable alternative to chemicals for a circular economy.

Keywords: biodegradable; sustainable; cost-effective; eco-friendly; natural coagulants



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1. Introduction

Water is an indispensable and precious substance for living beings on earth, as it is required for various domestic, agricultural, and industrial activities [1,2]. Approximately 1400 million cubic kilometers of water are available on earth, with 97.5% and 2.5% in marine and freshwater bodies, respectively [1]. Thousands of pollutants that are hazardous to man and ecosystems were identified in water bodies worldwide [3]. Water pollution is caused by several non-anthropogenic mechanisms such as hydrogeologic processes, changes in the climate, natural disasters like floods, droughts, earthquakes etc., as well as atmospheric

deposition, which may be slow or fast. Such natural forces frequently result in high amounts of dissolved organic material and unsustainable quantities of specific minerals or metals [4]. Fluoride is found in a variety of rocks and minerals in the Earth's crust, including fluorospar, cyyolite, and fluorapatite [5–7], which seep out through weathering processes and precipitation, contaminating surface water and groundwater, and consequently public water systems [8,9]. Anthropogenic influences include industrialization (nitrates, nitrites, lead, sulfur, mercury), R & D activities, urbanization, mining (metal wastes and sulphides), tourism, agricultural practices (the use of fertilizers, manures, herbicides, insecticides, fungicides, and crop residue), animal waste (from slaughterhouses, excreta), emerging contaminants (industrial chemicals, pesticides, pharmaceuticals, personal care products), radioactive wastes (uranium), marine dumping, accidental oil spills, microplastics, and the improper disposal of sewage from various sources such as restaurants, hospitals, households, and leakage from landfills and sewer lines. Additionally, various heavy metals such as zinc, lead, copper, etc., have also been reported to pollute the water, thereby making it unfit for human use [10–12]. The release of heavy metals into the atmosphere via fossil-fuel burning and other industrial activities of humans, which eventually enter streams through rainfall, as well as the release of industrial effluents and sewage water into streams and surface water bodies, constitute the anthropogenic origin of water pollutants. The most prominent heavy-metal pollutants of human sources include chemical element such as arsenic, cadmium, chromium, copper, nickel, lead, as well as mercury. Contaminant sources are described as either point (localized pollution) or nonpoint (pollution from different origins). Nonpoint pollutants represent the second category of pollution source, in which contaminants originate through widely scattered (and often difficult to detect) origins. Spontaneous weathering of ores and tiny metal particles in the air, water, and soils near coal-burning power plants via smokestacks is one example of localized metal contamination. The mining industry is the most common cause of metal pollution in waterways. Due to solubility, in acid solutions, they commonly utilize acid mine drainage systems to extract heavy metals from ores. They disseminate the acid solution, which contains high quantities of metals, into the groundwater after the drainage process [4,13,14]. In addition to heavy metals, water may be polluted by a range of contaminants due to the use of land for agriculture [15]. Of these pollutants, dissolved organic carbon [16], nutrients (nitrogen and phosphorus) [17], and pesticides [18,19] are the most important issues for certain land-owning UK water utilities due to the need to remove them from raw water to meet regulatory standards. Similarly, nutrient (nitrogen and phosphorus) loading in waterways from point and nonpoint origins is an environmental issue that influences surface water quality [20], as poorly managed agricultural activities can cause nutrient and insecticide pollution of surface and groundwater. Although nutrients are required for survival, excessive nutrient loading into water bodies can have an influence on the specified uses of water [21,22]. Nitrates in runoff can be leached or transferred [23]. Nitrates are tightly linked to agricultural land and grasslands [24], with concentrations peaking in the spring and during significant run-off occurrences. Water flow and pesticide dissemination are influenced by antecedent factors such as geography, soil type, agricultural method, and crop type [4].

These pollutants are associated with several water-borne diseases such as cholera, giardiasis, diarrhea, jaundice, typhoid, amoebic dysentery, Alzheimer's disease, and dementia [25,26]. Researchers have revealed that approximately 1.2 billion people are unable to obtain clean usable water and more than 6 million people die annually due to polluted-water-related issues [27–29], with 4 million people being affected by diarrhea-related issues, of which 2 million were reported to be children. Furthermore, fluoride, which is widely present in groundwater, leads to crippling skeletal fluorosis [26].

Currently, there is a scarcity of potable water in different parts of the globe due to several reasons including deficiency in resources and financial constraints associated with water treatment industries [30,31]. Moreover, the increasing human population and lack of water-harvesting technologies have caused vulnerability in the situation and have hiked the

cost of WT [32,33]. Therefore, it is crucial to provide clean and safe water for human use in an effort to mitigate the spread of water-related diseases and also promote health security.

Due to complication of the waste water-treatment process and the enormous number of parameters that must be established, it is important to establish the characteristic variables relevant to quality of water as auxiliary variables. The four types of data listed below can be used to evaluate the quality or effect of water quality in water treatment plants.

- Physical data: These are water that need to be monitored during the treatment process, which include total suspended particles, temperature, conductivity, clarity, total dissolved solids, etc.
- Chemical data: Chemical water-quality metrics of the national comprehensive discharge standard for contaminants in water, such as pH, biochemical oxygen demand, biochemical oxygen consumption, heavy metals, nitrates, etc.
- Biological data: Waterborne microorganisms such as *E. coli*, mayflies, and other microbes are examples of biomarkers.
- Environmental data: Environmental data encompass the entire water supply process, including weather, hydrology, soil, and ecological indices [34,35].

The aforementioned chemical, physical, or biological pollutants (from soil erosion, runoff, or due to high microbial count) discharged in water may be deposited on the waterbed or may remain suspended in water, causing turbidity, which is among the most common characteristics of polluted water [36–40]. One NTU has been recommended by the WHO as the highest point of water turbidity for human use [41].

Many scientific researches have been carried out on the treatment of polluted water using various strategies such as chemical precipitation, lime coagulation, ion exchange, reverse osmosis, and solvent extraction [42]. Although the chemical method is the most effective and commonly used technique, a transition from chemical to natural coagulants has been observed due to various limitations posed by chemical-based coagulants [40,43].

The natural coagulants deployed in WT are of plant, animal, or microbial origin [44,45]. These include starch, cellulose derivatives, gelatin, galactomannans, chitosan, alginate, glues, and microbial polysaccharides, which are all non-toxic. Molecule bridging, adsorption, and charge neutralization constitute the stages of the treatment process using natural coagulants. The major benefits of natural coagulants in waste water purification are that they are renewable, non-toxic, biodegradable, and cost-effective, and can efficiently remove turbidity [46,47]. The benefits of natural coagulants are presented in Figure 1. Therefore, natural coagulants (especially of plant origin) have drawn the attention of scientists in recent years [48].

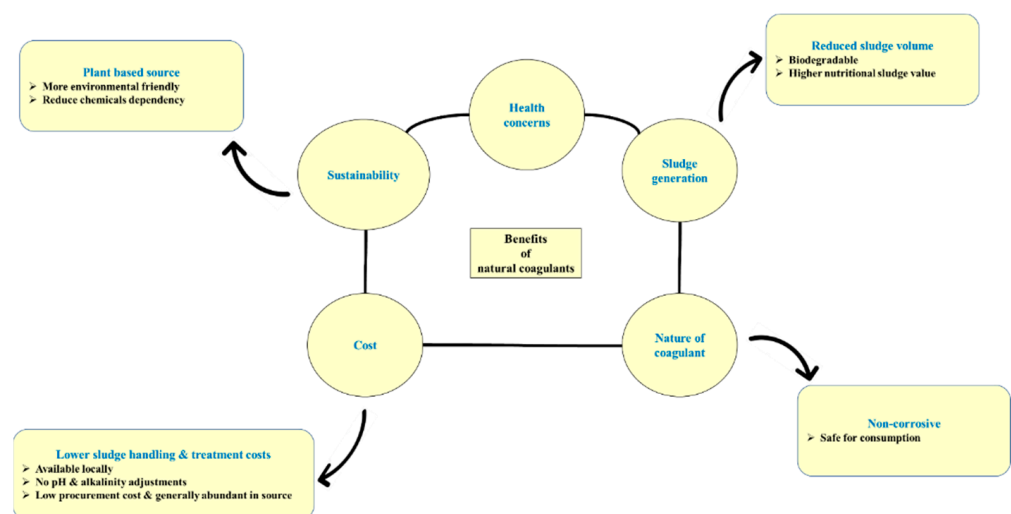


Figure 1. Advantages of natural coagulants over chemical-based coagulants.

This review encompasses reports on the use of natural coagulants as a sustainable alternative to chemical coagulants, in which the potential/success rate of plant-based and non-plant-based products have been discussed in relation to WT. The benefits, limitations, future prospects, and economic aspects of natural coagulants have also been included in this review.

1.1. The Concept of Water Treatment

Water treatment (WT) is the act of removing contaminants from polluted water. Such pollutants may include colloidal/dust particles, pathogens, suspended molecules, and various other toxic materials that are noxious and harmful to human health. Water purification/treatment can be achieved using primary and secondary stages. In the primary stage, sedimentation and filtration processes are deployed to remove solid particles from the water using a mechanical method, whereas, in the secondary stage, biological agents (anaerobic or aerobic microorganisms) are used for the breakdown and removal of the remaining waste, as well as other minute particles from the water. Presently, there are no suitable low-cost sustainable water treatment procedures available, especially for the rural population. Water treatment is achieved using chemical, physical, and biological techniques.

Chemical techniques include coagulation, ion exchange, disinfection, catalytic reduction, oxidation, and softening processes [49,50]. Adsorption, UV processes, settling, and media and membrane filtration are some of the processes that constitute physical methods of WT [51,52]. Biological methods include phytoremediation, bioreactor processes, microbial biodegradation, and the use of wetlands [53]. In some cases, two or more processes are used in a hybrid manner to improve efficiency [48,54,55].

Synthetic coagulants are highly effective in small concentrations and are capable of removing 99% of turbidity, heavy metals, and organic and inorganic substances [56,57]. Despite their effectiveness, their use is associated with several drawbacks as they are costly and are associated with a number of environmental effects [57,58]. To overcome the problem associated with the use of chemical coagulants, it is necessary to promote the utilization of natural coagulants for the treatment of turbid water and wastewater. To treat polluted waters, several parameters are taken into consideration for an efficient purification process. Such parameters include the pH, initial turbidity, temperature of the water, rapid mixing and coagulant dosage, biological oxygen demand, total dissolved solids, total suspended solids, total hardness, conductivity, acidity, alkalinity, etc., of the polluted water. Table 1 summarizes these parameters and their significance in the water purification process.

1.2. Factors Affecting WT

Understanding the most suitable conditions in the process of coagulation (the interaction between the coagulant and the pollutant) is vital because it facilitates understanding the highest efficiency of the coagulant in addition to a reduction in the operational cost and sludge volume. The factors associated with coagulation procedures in WT are the form/type of coagulant, dosage, mixing procedure of the coagulant, and the nature of the water to be purified [59]. The three vital conditions used to assess the impact of coagulant dosage in eliminating the water contaminants are under dosage, optimum dosage, and over dosage. Under dosage is the situation in which the coagulant dosage is insufficient to remove the dirt in contaminated water. Thus, an additional quantity will be needed to achieve the required level of purity [50]. However, using more coagulant, beyond the required quantity, will increase the impurity of the water and the excess will lead to saturation of the colloid surface. This will cause destabilization of the particles, which ultimately forms a repulsion force between the contaminants and, as a result, hinders the floc formation.

Table 1. Parameters to be considered while treating water.

Parameter	Instrument Used	Units of Measurement	Importance
Turbidity	Turbidimeter	NTU	Measure of relative clarity of water.
pH	pH meter	H ⁺ conc.	Indicator of water quality & measure of acidity/basicity.
Temperature	Thermometer	°C	Impacts both biological & chemical characteristics of water.
Conductivity	Conductivity meter	Sm ⁻¹	Measure of water capability to pass electrical flow.
Alkalinity	Potentiometer	V m ⁻¹	Measure of capacity of water to neutralize acids.
Acidity	pH meter	H ⁺ conc.	Indicator of industrial pollution.
Total dissolved solids	Conductivity meter	Sm ⁻¹	Measure of combined organic & inorganic substances dissolved in water.
Total suspended solids	Suspended solids density meter	mgL ⁻¹	Includes all particles suspended in water which won't pass through a filter.
Total hardness	Potentiometer	mgL ⁻¹	Measure of amount of dissolved Ca & Mg.
Ca hardness	Potentiometer	mgL ⁻¹	Measure of amount of dissolved Ca.
Mg hardness	Potentiometer	mgL ⁻¹	Measure of amount of dissolved Mg.
Chlorides	Potentiometer	mgL ⁻¹	Indicator of pollution in water.
Sulphate	Spectrophotometer	mgL ⁻¹	Indicator of algal growth in water.
Ammonia	Spectrophotometer	mgL ⁻¹	Indicator of fecal pollution.
Nitrate	Spectrophotometer	mgL ⁻¹	Indicator of sewage pollution.
Nitrite	Spectrophotometer	mgL ⁻¹	Indicator of contamination from fertilizer run off.
Calcium	Spectrophotometer	mgL ⁻¹	Measure of water hardness.
Magnesium	Spectrophotometer	mgL ⁻¹	Measure of water hardness.
Iron	Spectrophotometer	mgL ⁻¹	Excess level indicates presence of contaminants.
Manganese	Spectrophotometer	mgL ⁻¹	Excess level results in undesirable taste.
Fluoride	Spectrophotometer	mgL ⁻¹	Reduces tooth decay.
Sodium	Spectrophotometer	mgL ⁻¹	Maintains blood pressure & osmotic pressure.
Potassium	Spectrophotometer	mgL ⁻¹	Maintains osmotic pressure.
Dissolved oxygen	Colorimeter	mgL ⁻¹	Indicator of water quality.
Biological oxygen demand (BOD)	BOD meter	mgL ⁻¹	Measure of water quality. Higher BOD signifies lower water quality.
Chemical oxygen demand (COD)	COD meter	mgL ⁻¹	Measure of water & waste water quality.
Pesticides	Spectrophotometer	mgL ⁻¹	Indicator of water toxicity.
<i>E. coli</i>	Paper strip method	per 100 mL	Indicator of fecal contamination.
Fecal coliform	Paper strip method	per 100 mL	Indicator of fecal contamination.
Velocity	Current meter	ms ⁻¹	The speed at which water flows.
Detergent	pH meter	H ⁺ conc.	Results in algal blooms & depletes oxygen in water leading to water pollution.

2. Strategies of Water Treatments

The most popular and conventional method for WT is the use of chemical-based coagulants including ferric chloride (FeCl₃), alum (AlCl₃), synthetic polymers (polyacrylamide), and poly aluminum [47,60]. However, the use of this approach is not sustainable as it leads to the production of a large volume of non-biodegradable sludge [61]. Natural coagulants, on the other hand, serve as an alternative sustainable strategy for the removal of turbidity and WT, as they are cheap, safe, and biodegradable. Natural coagulants are derived from three major sources, which include plants, animals, and microorganisms. A brief discussion on chemical coagulants and explicit information on natural coagulants are presented below.

2.1. Water Treatment Using Chemical Coagulants

The application of chemicals in removing colloidal impurities in water is referred to as chemical coagulation, whereas flocculation is the formation of flocs as a result of neutralizing the charge [62]. The most popular and generally used chemical coagulant in WT is 'alum', of which the chemical formula is KAl (SO₄)₂·12H₂O and the chemical name is potassium aluminum sulphate. At a pH of 8.0 and a concentration of 450 mgL⁻¹, alum eliminates 99% of the color from the turbid and water [63]. When combined with other coagulants, alum's potency increases. For instance, when aluminum sulphate was combined with poly ferric sulphate (PFS) and polyacrylamide (PAA), the chemical oxygen demand (COD) removal efficiency increased from 68% to 82% [64,65]. In a previous study, ferric chloride/iron (III) chloride at a pH of 6.0 was used as a coagulant to reduce the COD levels of water from the cosmetic industry by 63.9% [66]. Ferric chloride was also used to treat water from molasses and was reported to reduce the color and COD by 96% and 86%, respectively [67]. Chemical coagulants are effective at minimizing the chemical and biochemical oxygen demand, as well as oil and grease [68,69]. In another study, for the treatment of black liquor water, several chemical coagulants including aluminum chloride,

poly-aluminum chloride, and anionic PAA (polyacrylamide) were used. The composite could eliminate 95% of total dissolved solids (TDS), 88% of the color, and 80% of COD [70]. These methods utilize additives (chemicals) to segregate small-sized particles in large flocs prior to their removal via sedimentation [71,72]. Poly-aluminum chloride and poly-titanium chloride can reduce the water turbidity from 7.0 NTU to 1.2 NTU [36,38,73,74]. Poly-aluminum ferric chloride at a 5mg/L concentration and pH of 7.5 can eliminate the color and turbidity from water by 86 and 100%, respectively, at pH of 7.5. Similarly, water turbidity can be reduced from 9 NTU to less than 1.0 NTU in a short span of 15 min using polymeric zinc-iron-phosphate [75,76].

Thus, chemical coagulants have a number of benefits as they are easily obtainable and can operate over a broad pH spectrum. They can be used alongside additives for long-term storage and increased efficiency. Other benefits include higher efficiency at low concentrations, easy water dissolvability, and the removal of turbidity and microorganisms such as *E. coli* by 99%. However, it is also true that the use of chemical coagulants jeopardizes the environment and human health. They are known to persist in water until the coagulation process is over, are not biodegradable, and, as a result, treated water contains traces of these chemicals [77,78], which leads to various neurological disorders including Alzheimer's disease [79–81], dementia, encephalopathy, and Hippocampal neuron staining. Aluminum traces may cause diseases such as Down's syndrome and Parkinson's disease, convulsions, and even death [82,83]. Moreover, high operational costs, the large quantity of sludge, and the cost of disposal are some of the limitations of chemical coagulants [84].

2.2. Emerging Use of Natural Coagulants

Considering the global issues related to chemical WT (being expensive, toxicity to humans and the environment, corrosive and carcinogenic nature, altering the pH of treated water, producing hazardous and non-biodegradable sludge, high disposal costs, etc.) [57,58,85], there is a need to explore other possible measures so as to reduce the ill effects of such coagulants on the ecosystem [86–88]. Thus, there has been a recent paradigm shift in water and WT, which encouraged industries to improve the culture of water operators in adopting and implementing sustainable development in their activities. Among functional practices is the replacement of chemicals with natural substances in WT processes, which has led to decreased environmental effects in terms of production, consumption, and secondary waste management. Natural coagulants are polyelectrolytes, which can be anionic, cationic, or neutral polymers [46]. They are safe and cost-effective with a great capacity to maintain the pH of the water being treated. Unlike chemical coagulants, natural coagulants do not increase the metal load during treatment and are characterized by the generation of a low volume of sludge, thereby making the cost of disposal very low [89,90], due to which they are a sustainable alternative to chemicals. Earlier research proved the effectiveness of natural coagulants in WT applications [46,91–94].

2.2.1. Sustainability of Natural Coagulants

Sustainability is the mode of development that fulfills the needs of current and future generations [95]. Although the crucial factor in the treatment of water is performance efficiency, the reliability of technology is also essential as per the concept of sustainability asserted by the United Nations. Thus, the concept of sustainability involves a combination of social, environmental, and financial aspects [96].

The social aspect of the sustainability of natural coagulants involves industrial acceptance and public health improvement. Industrial acceptance encompasses the ability of natural coagulants to provide results similar to chemical coagulants and be used as an alternative. However, due to the lack of real or pilot-scale use of plant-based coagulants and the lack of approval and regulatory guidelines in the treatment of potable water, industries hesitate to adopt natural coagulants. WT using natural coagulants, especially in rural areas, may facilitate health and hygiene and improve the living standards of all individuals (Figure 2). The technical aspect of sustainability involves treatment efficiency, product

stability, availability of materials, and compatibility with other techniques. Several natural coagulants have been time-tested and proven to be very efficient in the treatment of water and wastewater. Due to their natural origin, natural coagulants are considered safe and non-toxic. However, the toxicity of organic coagulants in humans and the environment still remains unclear, and there is a need to confirm their environmental safety. Thus, meticulous selection and dose optimization of efficient natural coagulants could provide promising results in WT and may act as a substitute for chemical coagulants. As discussed previously, natural coagulants offer reliability and robustness, are easily available, and can be obtained from a wide range of sources such as plants, microorganisms, or animals [93,97,98]. However, their susceptibility to biodegradation by microbial or other environmental factors adversely affects their long-term storage (shelf-life) and commercialization [99,100]. Environmental sustainability criteria involve the utilization of biodegradable and plant-based coagulants that are eco-friendly and capable of generating biodegradable sludge [101], which can be used for several other purposes such as agricultural practices, landfills, and in civil engineering industries [102,103].

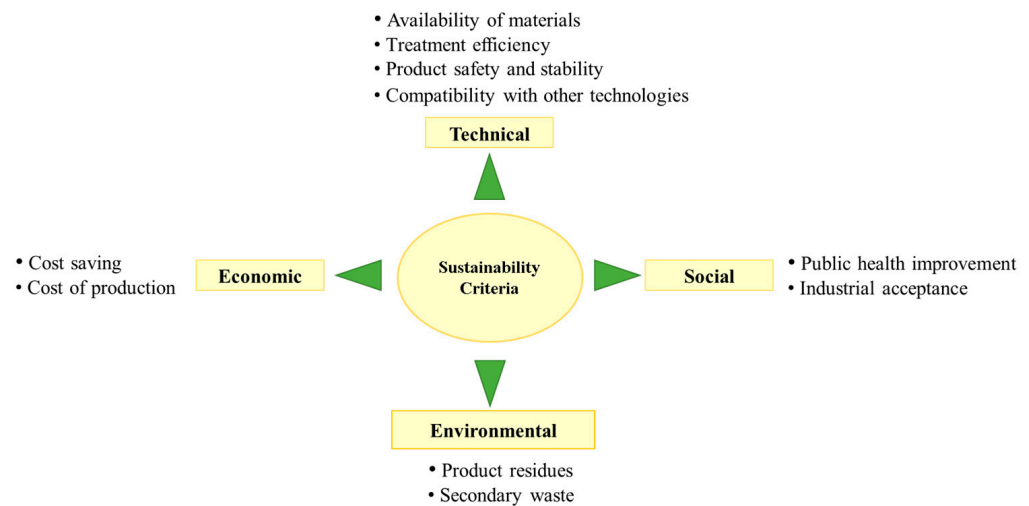


Figure 2. Sustainability criteria of natural coagulants.

The economic aspect of sustainability involves the use of cost-effective coagulants for WT. However, this claim is dissatisfactory as it does not address several other factors such as the cost of processing and maintenance or the variation due to geographical regions. Hence, the cost advantage of natural coagulants over chemical coagulants has not been well addressed. Therefore, one efficient strategy to increase the economic sustainability of natural coagulants is to use a combination of coagulants in order to compensate for the procurement cost and consumption demand along with the synergistic increase in clean-up efficiency [53]. Hybridizing natural coagulants with other coagulants to lower their utilization cost is an additional method to enhance the economics of WT. In contrast, the use of versatile natural coagulants may also result in a reduction in overall costs because many treatment functions can be carried out with a single substance in a single treatment unit. Moreover, as the majority of stated claims were based on lab-scale experimentation, it is challenging to translate the advantages of employing natural coagulants, such as the ‘creation of fewer sludge’ and ‘no need for pH adjustment’ into economic benefits. The ‘cost-effectiveness’ that is tangentially related to the advantages of employing natural coagulants must be more thoroughly evaluated in pilot studies. Because there has not been a thorough analysis of the total expenses of the coagulation process from extraction to the effects on other treatment units, we cannot draw conclusions on the economic aspect of natural coagulants [53].

2.2.2. Plant-Based Coagulants

Coagulants derived from plants are more readily available than those from either animals or microorganisms [92]. There are sufficient reports on the deployment of various plant-based products such as bagasse [104], banana peels [105,106], *Jatropha curcas* L. [107], and *Moringa oleifera* [105] for the treatment of polluted water. Macromolecules including proteins, polysaccharides, and some functional groups are known to promote the process of adsorption, polymer bridging, and charge neutralization [108]. They are usually effective in the treatment of water, with medium turbidity in the range of 50–500 NTU. However, the efficacy of natural coagulants can be enhanced through the optimization of the extraction and purification processes of coagulants [109]. Therefore, the proper extraction process using plant-based materials can exhibit better performance in the coagulation process, leading to greater/higher waste removal efficiency.

Some of the most widely used plant-based coagulants include *Moringa oleifera* Lam. (seeds), *Cicer arietinum* L. (seeds), *Azadiracta indica* A. Juss. (leaves), *Cactus latifolia* L. (leaves), *Pisum sativum* L. (seeds), *Vigna mungo* L. (Hepper) (seeds), *Arachis hypogea* L. (seeds), *Zea mays* L. (seeds), *Dolichos lablab* Linn. (fruits), *Phaseolus vulgaris* L. (seeds), etc. [110–112]. In addition to these, seeds of nirmali are also the source of anionic polyelectrolytes, and their carboxylic acid (–COOH) and hydroxyl (–OH) groups boost the effectiveness of coagulation. Galactomannan and galactan, polysaccharides from the seeds of *Strychno spotatorum* L.fil., are potent enough to decrease water turbidity by 80%. Moreover, plants such as Acacia, Catenae, and Schinopsis can also be used to remove contaminants from water as they possess naturally occurring tannins [113]. Species of cactus such as *Opuntia latifaria* L. are also used as natural coagulants as they contain certain compounds, such as d-galactose, d-rhamanosei, d-xylose, l-arabinose, and galacturonic acid, which are responsible for the bridging action with contaminants in water during the coagulation process [114]. The fruit of *Prunus armeniaca* L. has also been reported for its use as a purifying agent in ancient periods in countries such as Egypt and China, historically dating back to the 1100s [115]. The form of natural coagulants used in the coagulation process can also influence turbidity removal efficiency. For example, when the seeds of *Tamarindus indica* L. were blended in water [116], it was more effective than the water extract derived from ground seeds [117]. In addition to turbidity removal, the common bacteria present in surface water (e.g., *Escherichia coli*) can also be eliminated [47]. The wastes of many fruits possess coagulation properties, whereas some exhibit antimicrobial activity due to the presence of saponins, phenols, and flavonoids [47,118,119]. It was reported by [120] and [121] that the colloids present in the leaves of *H. undatus* Haw. resemble those present in the seeds of *M. oleifera* Lam. and are cationic in nature. The processes of colloid adsorption and neutralization of the charge are considered feasible means of coagulation, which results in the formation of flocs. Table 2 summarizes the reports on the use of natural coagulants in WT and their relative pollutant removal efficiency.

The most commonly utilized natural coagulants derived from plants are explained below.

2.2.3. Tannins

Tannin is a polyphenol compound used in the leather industry and is obtained from the wood and bark of trees such as Castanea, Acacia, and Schinopsis [151,152]. The use of tannin extracted from valonia/Asia minor oak has been investigated by several researchers as an effective coagulant in WT [95,96,153–155]. They concluded that tannin is a superior alternative to chemical coagulants. Tannin can be used in the removal of dyes such as indigo, azo, triphenylmethanones, and anthraquinonic [156]. The chemical structure of tannins derived from plants and the degree of tannin modification affect their efficiency in water treatment. Tannin contains phenolic groups and is a strong anionic hydrogen donor. The phenolic groups form phenoxide due to rapid deprotonation and are balanced by resonance. It is also considered an amphoteric compound, which not only reduces the turbidity and heavy metals but also the color of water and thus functions as an alternative to chemical coagulants. Thus, it can be concluded that the higher the number of phenolics, the greater the capacity to coagulate will be.

Table 2. Plant-based natural coagulants used in the treatment of polluted water.

Coagulant	Part Used	State of Coagulant	Opt. Dosage	Removal Efficiency (%)	References
<i>M.oleifera</i> Lam. (Drumstick)	Seeds	Powder	0.2 gL ⁻¹	Turbidity removal (61.60%); COD removal (65.00%)	[122]
<i>M.oleifera</i> Lam. (Drumstick)	Seeds	Deoiled powder	6000 mgL ⁻¹	TSS removal (95%)	[123]
<i>M.oleifera</i> Lam. (Drumstick)	Seeds	Powder	500 mgL ⁻¹	Turbidity removal (96.80%)	[124]
<i>M.oleifera</i> Lam. (Drumstick)	Seeds	Powder	0.6 gL ⁻¹	Turbidity removal (82%), COD removal (83%)	[125]
<i>M.oleifera</i> Lam. (Drumstick)	Seeds	Stock solution	0.3 mgL ⁻¹	Cu and Cd removal (98%); Pb removal (78%)	[126]
<i>M.oleifera</i> Lam. (Drumstick) and alum	Seeds	Solution	70 and 80 mgL ⁻¹	COD removal (50.41%); Turbidity removal (86.14%); TSS removal (81.52%)	[127]
<i>M.oleifera</i> Lam. (Drumstick) and alum	Seeds	Stock solution	50 mgL ⁻¹ each	BOD removal (80.67%); COD removal (66.73%)	[128]
<i>M.oleifera</i> Lam. (Drumstick)	Seeds	Powder	50 mgL ⁻¹	Turbidity removal <i>M.oleifera</i> (82.2%); Chickpea (81.2%); Cactus (78.54%)	[129]
<i>Cicer arietinum</i> L. (Chickpea)	Seeds	Powder	50 mgL ⁻¹	Pb removal (81%); Ni removal (74%); Cd removal (97%)	[12]
<i>M.oleifera</i> Lam. (Drumstick) and <i>Musa acuminata</i> L. (Banana peel)	Seeds	Powder	200 and 400 mgL ⁻¹		
<i>Malus sylvestris</i> L. (European crab apple) and aluminum sulphate	Seeds	Stock solution	62.5 mgL ⁻¹	Turbidity removal (66%)	[130]
<i>Cyamopsis tetragonoloba</i> L. (Taub.) (Guar)	Gum	Powder	300 mgL ⁻¹	Turbidity removal (67.82%)	[130]
<i>P.ovata</i> (Psyllium) and PAC	Husk	Powder	0.4 and 7.2 gL ⁻¹	Color removal (90%); COD removal (96%)	[131]
<i>Parkia biglobosa</i> Jacq. (Locust bean)	Gum	Powder	300 mgL ⁻¹	Turbidity removal (67.82%)	[130]
<i>Ocimum basilicum</i> L. (Basil plant)	Seeds	Stock solution	1.6 mgL ⁻¹	Color removal (68.50%); COD removal (61.60%)	[132]
<i>Opuntia ficus</i> Linn. (Miller) (Cactus)	Mucilage	Powder	150 mgL ⁻¹	Turbidity removal (49.56%)	[130]
<i>Plantago major</i> Linn. (Carls) (Broadleaf plantain)	Mucilage	Powder	297.6 mgL ⁻¹	Color removal (92.4%); COD removal (81.60%)	[133]
<i>Hibiscus rosasinensis</i> Linn.(Carls) (China rose) and alum	Seeds	Powder	500 and 400 mgL ⁻¹	Turbidity removal; Hibiscus (60%); Alum (100%)	[134]
<i>Trigonella foenum</i> L. (Fenugreek)	Seeds	Powder	0.1 gL ⁻¹	Turbidity removal (58%); COD removal (63%)	[122]
<i>Abelmoschus esculentus</i> L. (Okra)	Mucilage	Powder	3.2 mgL ⁻¹	Turbidity removal (97.24%); COD removal (85.69%)	[135]
<i>Abelmoschus esculentus</i> L. (Okra) and Iron (III) chloride hexa hydrate	Mucilage	Stock solution	2.5 mgL ⁻¹	Turbidity removal (74%)	[130]
<i>Cicer arietinum</i> L. (Chickpea)	Seeds	Powder	2 gL ⁻¹	TDS removal (82%); COD removal (84%); BOD removal (83%)	[136]
<i>Cicer arietinum</i> L. (Chickpea)	Seeds	Powder	0.1 gL ⁻¹	Turbidity removal (78.33%); COD removal (83%)	[122]
<i>Dolichos lablab</i> Linn (Hyacinth bean)	Fruits	Powder	0.2 gL ⁻¹	Turbidity removal (71.74%); COD removal (75%)	[122]
<i>Tamarindus indica</i> L. (Tamarind)	Seed	Powder	400 mgL ⁻¹	Turbidity removal (97.72%)	[137]
<i>Hibiscus sabdariffa</i> L. (Roselle)	Seed	Powder	60 mgL ⁻¹	Turbidity removal (87.18%)	[138]
<i>Opuntia indica</i> L. (Mill.) (Cactus)	Mucilage	Powder	0.4 gL ⁻¹	Turbidity removal (78.54%); COD removal (75%)	[128]
<i>Momordica charantia</i> L. (Bitter gourd)	Seed	Solution	400 ppm	Turbidity removal (61.03%)	[139]
<i>Gossypium barbadense</i> L. (Cotton)	Seed	Oil	30 mL ⁻¹	TSS removal (66.27%)	[140]
<i>Ricinus communis</i> L. (Castor)	Seed	Oil	40 mL ⁻¹	TSS removal (66.67%)	[136]
<i>S.potatorum</i> L. f.(Nirmali)	Seed	Stock solution	60 mL ⁻¹	BOD removal (65.23%), COD removal (72.71%), Turbidity removal (75.20%)	[140]
<i>Musa acuminata</i> L. (Banana)	Peel	Juice	90 mL ⁻¹	Turbidity removal (98.50%)	[60]
<i>Zea mays</i> L. (Maize)	Seed	Powder	30 gL ⁻¹	COD removal (68.82%); Color removal (47.03%)	[141]
<i>Plantago ovata</i> Forssk(Isabgol)	Seed	Powder	1.5 mgL ⁻¹	COD removal (89.30%)	[142]
<i>P.vulgaris</i> Gustav Hauser (French bean)	Seed	Powder	1.5 mgL ⁻¹	Color removal (73%)	[143]
<i>Cassia fistula</i> L. (Golden shower)	Seed	Powder	0.5 gL ⁻¹	Color removal (71.3%)	[144]
<i>Vitis vinifera</i> L. (Grape vine)	Seed	Powder	1.5 mgL ⁻¹	Color removal (80%)	[145]
<i>Citrus sinensis</i> L. (Osbeck) (Orange)	Peel	Powder	0.2 gL ⁻¹	Turbidity removal (97%)	[146]
<i>Artocarpus heterophyllus</i> Lam. (Jackfruit)	Seed	Powder	60 mgL ⁻¹	Turbidity removal (43%)	[147]
<i>Jatropha curcas</i> L. Britton and Mills.(Barbados nut)	Seed	Powder	14 mgL ⁻¹	Turbidity removal (93%)	[148]
<i>Strychnos potatorum</i> L. f. (Nirmali)	Seed	Powder	1.5 mgL ⁻¹	Turbidity removal (90%)	[149]
<i>Carica papaya</i> Linn. (Papaw)	Seed	Powder	0.4 g per 200 mL	Turbidity removal (90%)	[150]
<i>Juliflora Prosopis</i> var. <i>juliflora</i> (Sw.) DC (Mesquite bean)	Seed	Powder	1.5 mgL ⁻¹	Turbidity removal (96%)	[151]
<i>Acacia mearnsii</i> De Wild. (Black wattle)	Bark	Powder	14 mgL ⁻¹	Turbidity removal (75%)	[151]

2.2.4. Nirmali Seeds

Nirmali is a medium-sized tree mainly used as a traditional medicinal plant native to Sri Lanka, southern and central India, and Burma [157]. The seeds of this plant have been reported to have been used over 4000 years ago as a natural coagulant in the treatment of water [158,159]. The majority of research on its usage as a natural coagulant is confined to the Indian sub-continent [117,158,160,161]. Extracts of nirmali seeds are anionic poly-electrolytes that use coagulation (charge neutralization and bridging) for the removal of contaminants in water [162]. It has been observed that the coagulation efficiency can be increased due to the presence of alkaloids, lipids, and carbohydrates containing $-\text{COOH}$ and free $-\text{OH}$ groups present on the surface. In a previous study, galactomannan and galactan obtained from nirmali seeds reduced the turbidity of a kaolin solution by approximately 80% [163]. The presence of $-\text{OH}$ groups, along with galactan and galactomannan chains, is mainly responsible for the water-cleaning property.

2.2.5. Moringa Seeds

Moringa oleifera Lam. (horseradish or drumstick tree) is a versatile, medium-sized tree that usually grows in semi-arid, tropical, or sub-tropical areas and in various parts of Africa, Asia, Northwest India, and South America [164] belonging to the Moringaceae family. *M. oleifera* Lam. is non-toxic and is the most frequently identified natural coagulant utilized for WT [165]. In addition to its use as a source of food, fodder, and medicine, each part of the Moringa plant, including the leaves, seeds, flowers, roots, and bark, can be used as a coagulant for WT [166–168]. The seeds of Moringa, in addition to containing edible oil, also contain substances that are soluble in water [169]. Muller et al. [170] and Jahn et al. [171] were among the pioneer researchers who reported the use of *M. oleifera* Lam. seeds for the coagulation process in WT. Muyibi et al. [172] also reported it to be a minimal or low-cost natural coagulant that can be feasibly deployed in WT, at least in rural or semi-urban areas. The work of Ndabigengesere et al. [169], which reported the coagulation property of Moringa seeds in the treatment of water, ignited interest among environmental scientists. The active coagulating agents present in Moringa seeds are reported to be cationic proteins, which are dimeric in nature, possess an isoelectric point between 10 and 11 and a molecular weight of 12–14 kDa, and function via charge neutralization and adsorption mechanisms.

Gassenschmidt et al. [173] also reported that protein is the active ingredient of coagulation in Moringa seeds, possessing a mass of 6.5 kDa and an isoelectric point higher than 10. In a similar report, other researchers [174] stated that the protein has an isoelectric point greater than 9.6 and a mass of 6.5 kDa. In contrast to the aforementioned reports, [175] reported that the coagulating agent is an organic polyelectrolyte rather than a polysaccharide, protein, or lipid, with a molecular mass of 3.0 kDa.

As per the aforementioned reports, the most active coagulating agent is cationic proteins; however, Moringa may contain several other coagulating agents, which need to be studied further. Cationic proteins in the seeds of Moringa work via various electrostatic mechanisms such as neutralization, charge reversal, and adsorption [176]. These bind with impurity particles, which are usually negatively charged. Okuda et al. reported that the coagulation performance of Moringa extracts can be improved by using bivalent cations (Mg^{2+} and Ca^{2+}) [175]. Sulaiman et al. [177] used the seeds of Moringa in WT and observed excellent results. Dotto et al. [178] used the same method in the treatment of textile mill water and a significant COD reduction was reported. For the treatment of water from the dairy industry, seed powder was able to remove ~100% of the turbidity and 99.50–100% of the fecal coliform count. In a similar report, the seed powder could remove ~ 83.63% of the turbidity from laundry water [179]. Hence, *M. oleifera* is among the most versatile and reliable natural coagulants and has been proven to be a promising sustainable alternative to chemical coagulants.

2.2.6. Chickpea Seeds

Cicer arietinum L. (Fabaceae), commonly known as chickpea, is an important pulse crop that is grown in arid and semi-arid zones with good soil moisture and adequate rainfall. Hiremath et al. [180] and Jaseela et al. [181] used chickpea seeds to treat water released from the dairy industry and reported the removal of turbidity of 86.29%. Another group reported a more than 95% reduction of preliminary turbidity, which was similar to that of alum [182]. Choy et al. (2015) reported that the presence of high sugar and protein contents in chickpea were mainly responsible for the removal/coagulation of particles [92].

2.2.7. Peanut Seeds

The seeds of *Arachis hypogea* L. (peanut) are an important source of protein and are well known for their high lipid content. Traditionally, the seeds have been used against inflammation while the seed oil has been used as an ointment [183]. The lipid part of the peanut constitutes approximately half of its dry weight, but the lipid does not contribute to its purification properties. As a result, the relative percentage of the active agent is markedly decreased, leading to lower efficiency in the removal of raw surface-water turbidity. Mataka et al. [184] reported that peanut seeds exhibit a similar effect as that of moringa seeds, and delipidated cakes were found to be more effective (in the removal of heavy metals from wastewater) than the crude seed extract. Therefore, the effectiveness of peanut coagulation activity can be increased by removing the lipidic portion from the seeds.

2.2.8. Soybean Seeds

The seed extracts of *Glycine max* L. (soybean) can be used as the main coagulant or a coagulant aid to alum for cleaning contaminated raw surface water [185,186]. The coagulation efficiency of soybean seeds was excellent in the clarification of surface water beyond 450 NTU, whereas, as a coagulant aid to alum, ~96% turbidity removal was reported [186]. Although soybean seeds have a large lipid fraction that is considered second to groundnut, the lipid part does not contribute to coagulation activities. Therefore, seed delipidation (the removal of lipids) improves the coagulation potential. Moreover, deoiled seeds of soybeans have been reported to be less expensive bioadsorbents for the treatment of water contaminated by different dyes [93,187,188]. Soybean seeds possess palmitic acid, whereas stearic acid found in *Hibiscus esculentus* has been associated with bactericidal activities. Thus, their extracts have potential anti-bacterial properties and can treat raw surface water [183,189].

2.2.9. Cacti Mucilage

Opuntia ficus indica (OFI) (L.) P. Mill is the most common species of cactus, which has been used mainly for its medicinal properties and as a dietary source, as well as for treating water. *Cactus latifaria* has also been used as an organic coagulant [150]. Opuntia possesses a high coagulation capacity due to the presence of mucilage, which is a complex carbohydrate, and, hence, it is used as a potential coagulant for treating water [190,191]. Thus, it is eco-friendly and can be used as an alternative to aluminum and iron salts due to its abundance, renewability, adaptability, and biodegradability [66]. Rebah et al. also reported the potential of Opuntia in the treatment of wastewater [192]. Opuntia's mucilage has different sugar molecules including d-galactose, d-xylose, l-rhamnose, l-arabinose, and galacturonic acid [190]. However, the most active coagulating agent is galacturonic acid [193], which contributes to the removal of 50% turbidity. Galacturonic acid may also exist in a polymeric form as polygalacturonic acid. It is anionic due to the presence of carboxylic functional groups, which undergo partial deprotonation in an aqueous solution [194]. The mucilage in Opuntia species functions mainly via adsorption and the bridging coagulation process in which dirt particles bind to the mucilage and facilitates the removal of turbidity.

2.2.10. Okra Seed Extract

The coagulation potential of okra (*Abelmoschus esculentus* (L.) Moench) seed extracts has been examined by [195] who asserted that seeds extracted using distilled water and a sodium chloride solution can be used as organic coagulants for the removal of turbidity from water with an efficiency of 54.5% and 92% for distilled water and a NaCl (1.0 N) solution, respectively. Raji et al. [196] discovered that okra seed extracts were very effective in the removal of turbidity from 580 to 5 NTU at a dose of 300 mg/L and pH 7.0, which is within the standard limit recommended by WHO. Similar work conducted by Thakur et al. [197] and Mishra et al. [198] revealed that okra seed extracts can effectively remove dirt from the water even at a dose of 200 mg/L. The work of [174] also shows the effectiveness of okra seed extracts in WT with more than 69% and 95% removal efficiencies for dissolved and suspended solids (from the effluents), respectively.

2.3. Animal-Based Coagulants

Animals may also act as a crucial source of coagulating agents. They are usually extracted from shellfish exoskeletons, the shells of lobsters, shrimps, insects, crabs, diatoms, and molluscs, and freshwater and marine sponges. Chitosan is a high-molecular-weight biopolymer developed via the deacetylation of chitin. It occurs naturally as a complex sugar (polysaccharide) and is water-loving (hydrophilic), biodegradable, environmentally safe, and capable of absorbing several metal ions efficiently as it contains amino groups in its polymeric chain [199]. The deployment of chitosan as a natural coagulant in the treatment of polluted water in different sectors, including agriculture, textile, detergent, and food industries, as well as paper mills, has been well reported [200,201]. The characteristic feature of chitosan regarding cleaning water is its ability to react and generate a positive charge that destabilizes the negative charge of the colloidal particles [202]. Actinobacteria are also effective in the treatment of contaminated water [203]. Efficient flocculating activity was observed when *Cellulomonas* and *Streptomyces* spp. were deployed as flocculants in the treatment of kaolin-contaminated water. The substantial effects of these species were shown to be due to the presence of several molecules including proteins, natural sugar, polysaccharides, and uric acid, as indicated by chemical analysis [199]. Chitin is a naturally occurring and the second-most abundant polysaccharide (after cellulose). Every year, at least 10 gigatons of chitin are synthesized and destroyed throughout the biosphere. Chitin is a renewable resource found mainly in complexes with other polysaccharides and proteins. Chitin and its metabolites are utilized as chelating agents in the purification of water by segregating organic substances and heavy metals, as well as in the treatment of sewage via the precipitation of certain anionic wastes and the collection of contaminants such as PCBs (polychlorobenzene). The utilization of 10 mg per liter of chitosan in water has already received approval from the Environmental Protection Agency (EPA) [204].

3. Extraction of Natural Coagulants

The process of extraction of plant-based natural coagulants involves three steps: Primary, secondary, and tertiary extraction. In primary extraction, seeds are first collected and then dried conventionally. This is then followed by manual/mechanical pulverization and seed grinding (into a fine powder). During this process, settling tank is used to keep water temporarily where heavier solids sink to the bottom and lighter ones float to the surface. These materials are then held back once settled, while the rest of the liquid is discharged to the more rigorous secondary phase of water treatment process. The tanks are sometimes equipped with mechanical scrapers that drive the continually collected sludge at the base of the tank to a hopper, which pushes it to sludge-treatment facilities. This is most commonly used by local communities in rural areas. However, it is associated with a major disadvantage in that the coagulating agent constitutes a small portion of the seed powder, hence leading to an organic load in treated water. This drawback is overcome by secondary and tertiary stages. This is accomplished by removing their active components and purifying them to remove unwanted organic matter. In the secondary stage, active

coagulants are extracted via the solvent method, which includes salt solution extraction, water extraction, or alcohol/organic solvent extraction methods. Out of these, the water extraction method is the most popular as it is abundant and cost effective [174,175]. The tertiary stage is a rather uncommon method because it involves high costs and is mostly limited to academic research [174,175]. The major objective of tertiary water treatment is to improve the water quality to attain the standard domestically, industrially, and the specific needs regarding the safe discharge of water. Tertiary treatment of water also deals with the removal of microbes/pathogens, which ensures that water is safe for drinking purposes in the case of water treated by municipalities [174,175]. The natural coagulant extraction process is shown in Figure 3.

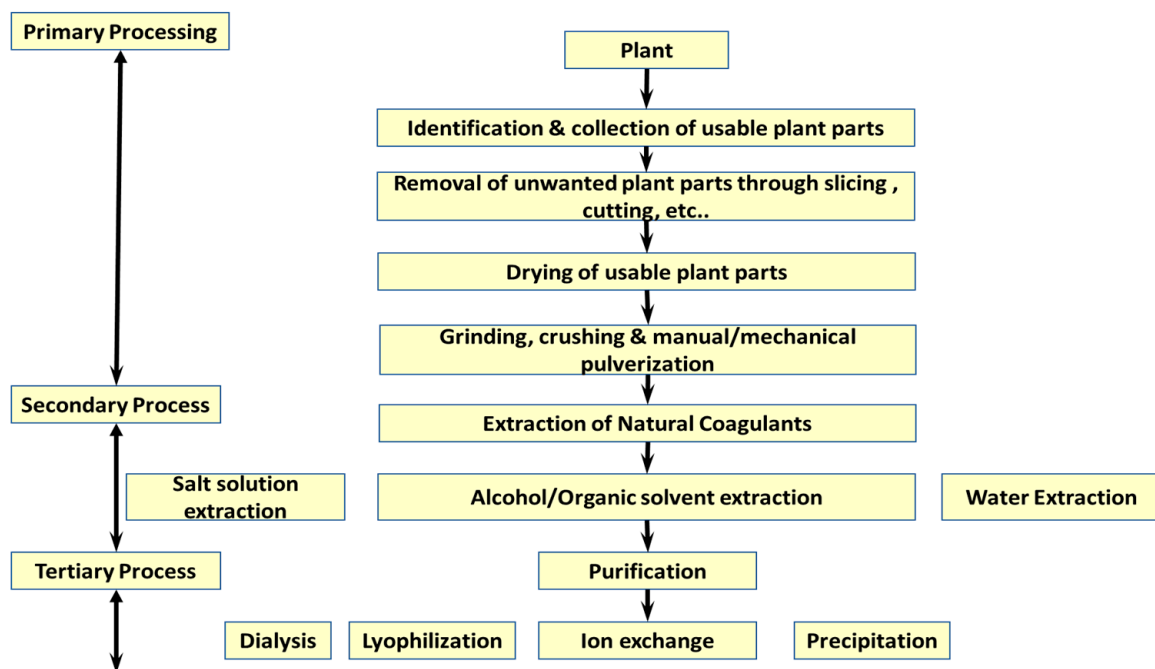


Figure 3. Schematic representation of natural coagulants' extraction process.

4. Mechanism of Action of Natural Coagulants

Coagulation is a method of destabilizing unsettled/slow-settling tiny particles with a size of 0.001 to 1.0 μm and above by adding a coagulating agent, which increases the floc size and the settling velocity [59]. This approach is crucial for water pretreatment as it removes dispersed/suspended contaminants and grants purified water the requisite quality for further processing. The global market for coagulants is expected to exceed USD 6.01 billion by the end of 2022, representing a 5.9% compound annual increase from 2017 to 2022 (4.35 billion in 2016) [59].

The mechanism of action of natural coagulants is similar to that of polyelectrolytes and contains a variety of functional groups such as $-\text{OH}$, $-\text{COOH}$, and $-\text{NH}_2$. The schematic representation of coagulation mechanisms as revealed through FT-IR and SEM is shown in Figure 4. The green circles represent the positively charged coagulants and the red colored circles represent the negatively charged colloidal particles. The mode of action of natural coagulants can be grouped into sweep flocculation, charge neutralization, double-layer compression, and antiparticle bridging. Sweep flocculation/coagulation is seen as a technique that eliminates colloids via entrapment/enmeshment using a net-like structure containing amorphous metal hydroxide precipitates that are formed by the hydrolysis process [205]. Several analyses including initial floc-aggregation, the relative settling factor, and the flocculation index revealed that the flocs developed through this method are relatively smaller in size with good settling capacity but are characterized by a slow rate of floc formation [205]. The floc produced via sweep flocculation has a high fractal dimension,

which shows that flocs are complex [206]. Stronger flocs that are tolerant of breakage are theoretical features of the high fractal dimension. However, the flocs of sweep flocculation are large with greater floc production but are easily broken. Charge neutralization occurs via the adsorption process between the coagulants and the colloid surface, which are oppositely charged [193]. Before reacting with the colloids, chemical coagulants undergo a hydrolysis process to form several cationic species. A patch-wise medium referred to as the electrostatic patch mechanism is deployed for the charge neutralization process. Various cationic species will patch on the surface of the colloids resulting in particle surfaces with positive and negative charges. Colloids' surfaces with a mixed charge will reduce the repulsive forces and increase van der Waals forces between particles [207]. Flocs produced via the charge neutralization method are stronger than those developed by sweep flocculation but weaker than those formed through inter particle bridging, despite their globular-shaped smaller particles [205], and also have a greater fractal dimension [193]. Double-layer compression is an approach that employs ions with the opposite charge of the colloidal particles to infiltrate the double layer that surrounds the colloids. Counter ions will make the double layer thinner and smaller in volume [208]. Continuous electrolyte compression reduces the electrostatic repulsion and increases van der Waals forces, facilitating the joining of the two destabilized colloids [205]. The flocs produced via this mechanism are larger in size because the rate of aggregation of the particles is high. However, they have a low level of sedimentation due to certain friction forces unnecessarily forming between the larger flocs. Moreover, the floc strength of the coagulants depends on the ionic charge of the coagulant. Monovalent ions that are weakly charged will produce large but loose flocs that require a longer time to settle. Nevertheless, the double layer is significantly charged with a high repulsive force towards weakly charged ions, which lessens the likelihood of agglomeration. The inter particle bridging relies on polymers that exist as a long chain with a highly reactive group dangling in the water. A particular region of the polymeric chain will attach itself to colloids, while the unattached portions of the polymeric chain attach to other colloidal particles to form a complex colloid–polymer–colloid structure in which the polymer serves as a bridge. The flocs that form are flaky with irregular void space between the network structures. Furthermore, the fractal dimension of the formed floc is low, which indicates it is not as complex as flocs formed by other mechanisms [48]. Theoretically, a low fractal dimension leads to the formation of weak flocs that are prone to breakage, but they are very strong and not easily broken into smaller clusters due to polymers that serve as bridges [52]. A study shows that the use of natural coagulants with the inter-particle bridging mechanism enhances the growth of the floc by at least thrice compared to the use of synthetic chemicals due to the ability of polymeric chains to stretch and get attached to as several colloids as possible [59].

This mechanism has been observed in the case of cactus mucilage and *Cassia obtusifolia* seed gum via adsorption and bridging wherein the long chains of polymers destabilize the charged impurities, forming bridges between them and culminating in macro flocs, which tend to settle faster (sedimentation). The adsorption process involves hydrogen bonds or dipole interactions. Proteins, lipids, and carbohydrates are the most common components of natural/bio-coagulants. Contaminant particles are anionic, whereas coagulating particles are cationic, which results in an electrostatic attraction between them and causes adsorption, charge reversal, and the neutralization of contaminant particles. Later on, flocs are generated and start settling (sedimentation) and are easily removed from the water, thus treating the water [176]. Polyelectrolytes possessing a high charge density show maximum flocculation ability.

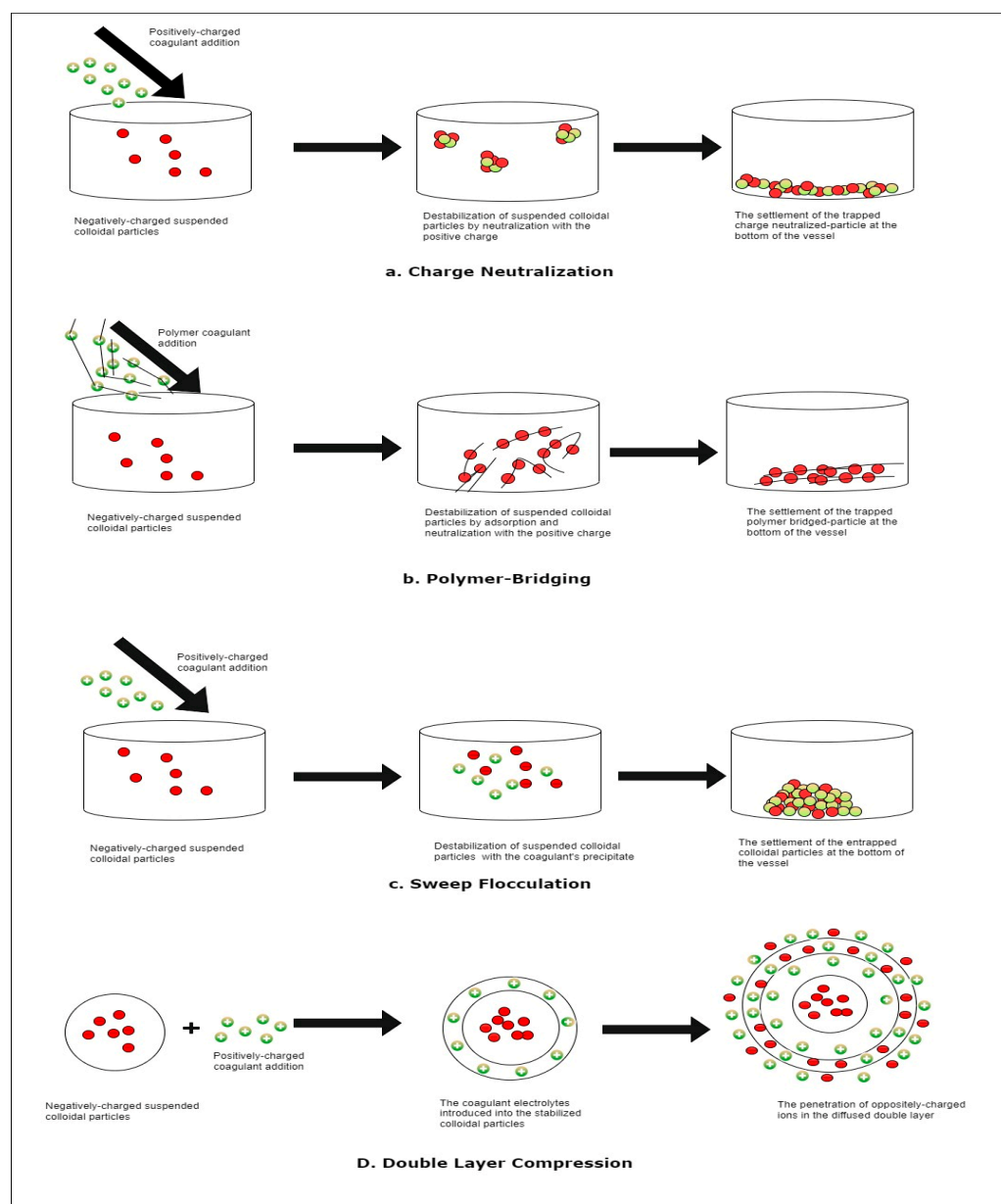


Figure 4. Schematic representation of the mechanisms involved in natural coagulation process.

5. Natural Coagulants—Constraints in Commercialization

Several extracts from natural sources have proven their coagulation potential in removing total dissolved solids (TDSs), total suspended solids (TSSs), chemical oxygen demand (COD), biological oxygen demand (BOD), turbidity, hardness, algae, and total coliforms. However, many such coagulants are yet to be accepted and commercialized. The major barriers that hinder their commercialization include monetary requirements, market awareness, research and development, and regulatory approvals [47]. Furthermore, Saleem et al. [93] demonstrated the ionic nature (cationic, anionic, and non-ionic) of coagulants and explored the barriers to their application and commercialization. The outcomes of the current studies are mostly restricted to the laboratory scale, having less relevance in real industrial applications. For natural coagulants to be successfully commercialized, there is a need for economically feasible extraction procedures and in-depth knowledge regarding their mode of action. Moreover, for any new products to be commercialized, approval from the relevant authority and other regulatory authorities must be granted, which is somewhat difficult as it requires ensuring product compliance with the respective

standards. Strong motivation for green chemistry and cleaner production are crucial for the development of natural coagulants and their applications. Figure 5 outlines the constraints hindering the successful industrialization of natural coagulants. Among the major issues, the raw materials required to generate natural coagulants must be available at a large scale for successful and realistic applications. Furthermore, the technical support of experts and new equipment are required to sustainably implement the deployment of natural coagulants [108].

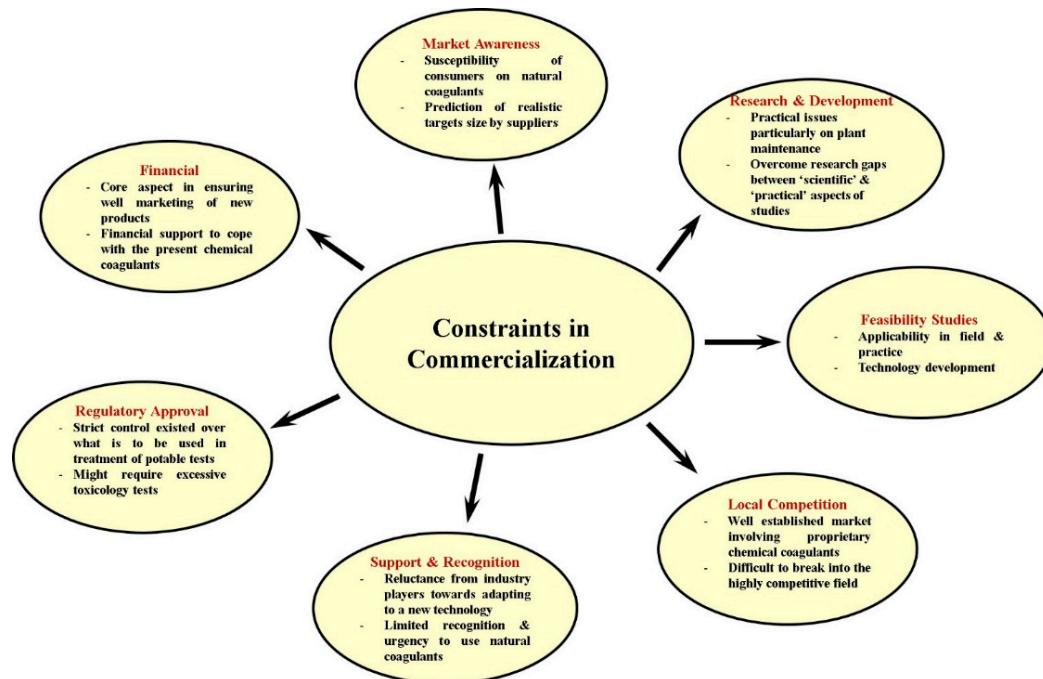


Figure 5. Various constraints in commercialization of natural coagulants.

6. Future Prospects of Natural Coagulants

Over the past few decades, the implementation of pollution-prevention strategies and environmental impact assessment (EIA) has become increasingly important and have resulted in changes in water, waste, and energy management systems through the reduction, reuse, and recycling approaches of the circular economy, with the aim of achieving sustainability [209,210]. Natural coagulants derived from plants have the ability to replace synthetic coagulants for the treatment of wastewater from the textile industry [92]. There is a plethora of reports that highlight the performance of natural coagulants based on their flocculation or coagulation potential in WT but all have their own limitations that need to be investigated, improved, or overcome for fine-tuning and process improvement.

As aforementioned, the use of crude extracts of natural agents has been associated with mixing certain organic matter (dissolved organic carbon (DOC) and total organic carbon (TOC)) into clean water [203]. The significant increase in the organic residues of treated water is unacceptable and can lead to an increase in microbial activity, which further results in changes in color appearance, bad odors, and bad tastes [203]. Moreover, crude plant-based products have several inorganic and organic molecules such as fats and oils that do not contribute to the process of active coagulation [174]. Therefore, the meticulous optimization of physico-chemical parameters prior to its implementation on a large scale is crucial. Unfortunately, this requires more time and effort because the physical-chemical characteristics of water change over time. The major obstacle in introducing plant-based natural coagulants in industries is their lack of year-round availability in order to meet industrial requirements. Moreover, the processing technology required and the cost of processing/production are also limitations. In addition to this, there are several other issues including the choice/possibility and source/availability of the plants

to be used. For instance, the use of *Jatropha curcas* seeds in WT has been associated with safety issues [148]. Among the issues related to the use of plant products (e.g., cereal crops) is the escalation in market prices of unprocessed grain due to increases in the human population and decreases in agricultural production [211]. Direct comparisons of coagulant forms, production stages, and costs involved in different geographical regions are extremely difficult due to differences in exchange rates, inflation factors, and cost value accuracies [46]. The needs and responses of markets will also influence the demand for natural coagulants as an alternative to chemicals. Although natural coagulants will not be commercialized immediately, these small initiatives will help to bridge the gaps and overcome the limitations (Figure 4). An appraisal of the social, economic, and ecological aspects can be maintained as a sustainability metric with the use of a pilot plant analysis. The approval of local authorities should be simple for the effective marketing of any new product [212]. As a remedy to the problem, policy framers should implement tax rebates, subsidy programs, tax reductions, incentives, and environmental laws and regulations so as to encourage reliance on plant-based natural coagulants.

Understanding the essence of water is important for developing a suitable water treatment process, implementing a protocol, determining appropriate residue requirements, determining the level of assessment necessary to verify the procedure, and deciding which residues to evaluate on the basis of toxicity [213]. Research on the active chemicals discovered in plant-based natural coagulants is another factor to consider when employing natural coagulants to improve coagulation–flocculation (CF) performance. The processes for identifying and isolating active coagulating compounds are too challenging due to the probability of interdependent effects among the components present [47,203]. Moreover, the coagulant type and dosage, total dissolved solids in the contaminated water, the distribution and size of colloidal particles of the water, the temperature of the water, ionic strength of the coagulants, pH of the water, and the configuration and percentage of organic matter in water are some of the factors that influence the coagulation CF effectiveness of the polluted water [214,215].

Active ingredients must be extracted, purified, isolated, and characterized in order to identify the chemicals that cause CF. The extraction of these compounds may directly increase the gross CF efficiency due to an increase in the relative amount of the agents [47]. Plant-based coagulants must thus be chosen for large-scale application based on the availability of annual species. Thus, meticulous research, tests, and trials are needed to find the most efficient natural coagulant or a hybrid of natural and chemical coagulants or natural coagulants alone, for the treatment of water [216].

Another important recently recognized cleaning agent for contaminated water is ‘biochar’. It is a carbonaceous porous substance generated through the thermochemical decomposition of feedstock biomass with or without oxygen (pyrolysis). Any natural/organic waste material could be used as feedstock biomass for the production of biochar. Examples of such organic wastes are plant residues, litter, algae, manures, wood chips, municipal solid wastes, and sewage sludge [217–219]. Biochar technology is considered a novel, cost-effective, and environmentally friendly resource that can be used as reliable technology in water treatment [220–222]. Biochar has been widely utilized as an adsorptive agent to eliminate metal contaminants, biological pollutants, and nutrients from the water. Thus, it can play a key role in environmental cleaning and reflects the concept of “treating wastes with wastes”, for a sustainable, circular economy. There are two substantial benefits behind the application of biochar for WT viz; (i) its production prevents the release of greenhouse gasses into the atmosphere, thus decreasing the impact of global warming in our environment [223], and (ii) biochar has a large surface area with numerous surface functional groups. These features have made it an effective, cheap, and ecologically safer adsorbent for WT [224–226]. ‘Engineered biochar’ has also been developed, which possesses a larger surface area, higher adsorption ability, and more widely available surface functional groups (SFG) than the normal biochar/activated carbon, indicating that it is a new type of carbon-based material with promising potential in treating water contaminated with

agro-chemicals, pharmaceuticals, and metalloids/metals generated from agro-industrial activities and municipal waste [219,222,227,228]. It may also be used for the improvement of soil fertility, crop growth, and production [218], and for the production of clean energy, so as to partially replace fossil fuels [229]. It is also applied as an adsorbent and catalyst to different contaminants and reduces the emissions of greenhouse gases [218,230,231]. Rajapaksha et al. [232] mentioned that sulfamethazine can be removed effectively from the contaminated water using steam-activated biochar but the rate of such contaminant removal is pH dependent. Heavy metals from agricultural wastewater such as As, Cu, and Pb constitute another pervasive issue. The potential of biochar to remove these pollutants was reported to be 69.4 mg/g and 34.1 mg/g for Cu^{2+} and As^{5+} , respectively, while the quantities for Cd^{2+} and Pb^{2+} were found to be 0.4 mg/g to 12.3 mg/g and 36 mg/g to 35 mg/g, respectively [233–235]. Biochar is also important for the treatment of polluted stormwater. Thus, the use of organic coagulants from plants and animal sources, including the application of biochar technology, represents a new, cost-effective, and environmentally friendly solution for WT. A recent study by Liu et al. [236] demonstrates that biochar impregnated with aluminum can significantly eliminate contaminants such as Zn^{2+} , Cu^{2+} , As^{5+} , and other runoff contaminants in urban runoff water. Removal rates greater than 85% and 95% were determined when a biochar-based filtration medium was deployed to remove copper and zinc in polluted runoff stormwater. However, there is a need for careful testing and designing of the filtration media to achieve success in stormwater treatment systems [237].

7. Conclusions

Due to the growing population, long-lasting periods of drought, soil and water pollution, and other factors, safe drinking water has now become a challenging resource in many regions across the globe. In the current age of scarcity of water, efficient water treatment is a basic requirement for survival, health development, and economic growth, and has been one of the most important sustainable development goals (SDG) proposed by the UN that is to be achieved by the year 2030. It is vital to establish and incorporate improved water treatment techniques with increased performance and reduced financial inputs. The present review has unraveled the potential of natural coagulants in the purification of waters and has proven effective, as evidenced by various research reports published in the past two decades. They are biodegradable and thus eco-friendly in nature and their performance is similar to that of chemicals. However, a number of factors need to be considered when utilizing natural coagulants. This is because the process of extraction and purification is complex, there is a limited supply of raw materials, and the water is variable in its composition (contaminants). Additionally, some of these products can work against surface water microbes such as *Escherichia coli* while some can serve to remove heavy metals too. In order to achieve commercialization, natural coagulants must have such qualities/attributes (biodegradability, eco-friendliness, low costs, easily available, reduce–reuse–recycle, etc.) so as to make space in the well-established markets where chemical coagulants occupy the stage. Even though organic coagulants are less effective compared to chemicals, a strategic blending of certain coagulants can synergistically enhance the efficacy of the water treatment regime. A high dosage, a lengthy contact period, and the maintenance of pH below 7 of plant-based coagulants can also reduce turbidity to a certain extent. Considering human health and environmental issues associated with the use of chemicals, there is an urgent need for meticulous and in-depth studies to analyze the mechanism of action of natural coagulants and develop strategies for the year-round availability of coagulants so as to deploy them for large-scale treatment of water and increase the sustainability of WT regimes.

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Abbreviations

NTU, nephelometric turbidity units; WT, water treatment; kDa, kilo-dalton; WHO, World Health Organization; UNESCO, United Nations Educational, Scientific and Cultural Organization; PFS, poly ferric sulphate; PAA, polyacrylamide; COD, chemical oxygen demand.

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