Water Treatment by Green Coagulants—Nature at Rescue

Manoj Kumar Karnena and Vara Saritha

Abstract Water purification and treatment are significant challenges for environmental engineers to several extents, continually increasing pressure to provide safe drinking water to the consumers. Nevertheless, water treatment with chemicals is minimized as they are toxic to the environment and humans. One of the fundamental steps for water treatment for consumption is water clarification by Coagulation and flocculation, eliminating colloidal particles, impurities, algae, etc. The sustainability in water treatment is only possible by the usage of natural materials combined with innate technologies. In these lines, the present book chapter will appraise the efficiency of natural coagulants' in treating surface water. Several researchers tested the natural coagulants for water treatment; but, various natural coagulants and their coagulation efficiency were not presented. Thus, an attempt is made to achieve a comprehensive account of widely used different plant-based coagulants, to understand their properties and efficiencies in treating water. The approaches mentioned in the book chapter will develop a knowledge database on available coagulants and their best utility techniques that can be adopted at any given time.

Keywords Natural coagulants · Water treatment · Turbidity · Coagulation · Flocculation

1 Introduction

Water is a vital resource for biological activities and human beings; strengthening the profiles of lakes, rivers, oceans, and forests becomes a segment in the hydrological cycle marking its significance in developing social life and ecosystems [126]. In recent years be wary about the resources are becoming a crucial mission progressively. The global community alerted the WHO (World Health Organization) and the UN (United Nations) regarding water scarcity and unrestrained disposal of pollutants into surface water. When measured as a parameter affecting human lives, it is

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A. Vaseashta and C. Maftei (eds.), *Water Safety, Security and Sustainability*, Advanced Sciences and Technologies for Security Applications,
https://doi.org/10.1007/978-3-030-76008-3_9

undoubtedly one of humankind's intricate development's critical factors. It is crucial to connect human health, education, and poverty, whose implications are essential for humans' development [14]. Also, in the challenge of Millennium Development Goals, water is an intersecting aspect. Availability and reuse of water are not a new concern; the human community is worried about water stress for many years. [32]. Intended essential for life, present inappropriate water use, including industrialization, urbanization, increasing population, and climate change, enhanced these reserves [15]. The scarcity of water affects all community sectors, including social and economic, resulting in a threat to natural resource sustainability [60]. The water's purity level for consumption is crucial as it directly affects individual health [75]. Sowmeyan et al. [124], reported that about two million people use unhealthy aqua, resulting in diarrhea besides several diseases caused due to water, specifically in developing countries. Regardless of advancements in water treatment technology, one of the significant challenges confronted by many developing countries is the deficiency of safe and clean water for their citizens [104]. Safe and pure water requirements triggered detailed and comprehensive research for treating water and wastewater [77]. Treating water to make it potable is a question of essential significance, together for emerging and advanced countries. The trait of aqua in developing countries generally is unsatisfactory and unsafe for wellbeing. In contrast, in developed countries, water decontamination involves the application of chemicals, knowing it is not safe on health upon long-standing vulnerability is questionable [82, 87].

Many communities in most of the third world countries depend to some extent on surface water sources for domestic water supply. Water drawn from rivers contains high turbidity, particularly in the rainy season. Many studies have reported that seasonal variation in turbidity is a significant problem in treating surface water [85, 93]. Generally, surface waters contain fine suspended particles, sediments, and natural organic materials, which cause turbidity. Chemically turbidity of water is due to the existence of negatively charged elements in the colloidal structure, hence positively charged agents called coagulants are used to form complexes in a process called Coagulation [88, 101]. Coagulation is a crucial process in treating surface water, removing dissolved chemical species, and turbidity. Coagulation is simple, economical, and dependable consuming less energy, which is practiced commonly. Furthermore, it is a proven process for removing suspended, colloidal, and dissolved particles by aggregating macro and micro particulates to large size, which eventually settle by sedimentation. Coagulation is also applicable for removing many other impurities like micro-pollutants, organic compounds, color and oils, and fats [67].

Conventional Coagulation was achieved using chemical-based coagulants like alum (Aluminium sulfate) and ferric chloride. Alum is a broadly used coagulant accepted by many treatment units worldwide due to its performance, availability, management, and cost-effectiveness. The efficiencies of chemical-coagulants [47], nevertheless, drawbacks related to their usage include their ineffectiveness in waters with lower-temperature [63], costs associated with the acquisition, generation of larger volumes of sludge, alteration of treated water pH and harmful impacts on humans. Shreds of proof relating to alum coagulants to Alzheimer's disorder in humans are also reported [51].

Thus, it becomes necessary to search for feasible alternatives that are safe for humans and the environment [62]. Many studies have explored and evaluated cleaner and eco-friendly coagulants originating from natural sources like plants, animals, and microorganisms. Advantages of natural coagulants over chemical coagulants include the production of lower sludge volume (20–30% than that of alum), biodegradability, economical, does not alter pH of treated water, require lower doses, and safe for humans [58, 130]. Technologies using natural, locally available coagulants entails water treatment practices that are environmentally friendly, culturally suitable, and reduce the external dependency of mechanisms and equipment.

Natural coagulants from plant origin are derived from leaves, seeds, bark or sap, fruit extract, and roots [67]. Some widely used natural coagulants that have reported to have good coagulation ability include *Moringa olefiera* [74, 97], starch [106], *Strychnos potatorum* [114] and cactus [3, 8]. Accepted coagulants like tannins have also been applied for water treatment, along with wastes of fruits and numerous legumes and vegetables [29, 30].

Studies to improve Coagulation and flocculation are being carried out during recent years, focusing on developing novel coagulants incorporating natural organic polymers and metal coagulants [137]. The greater efficiency of natural organic polymers is attributed to their greater molecular weight conferring better aggregation properties. Hence, growth in the size and molecular weight of coagulants can be a means towards additional improvement, which could be achieved by composite coagulants [92]. Thus, these novel composite coagulants will combine the positive aspects of respective individual components resulting in enhanced coagulant stability, being effective in broader pH range, requiring low dosage, and forming denser flocs virtuous settling properties. Furthermore, these coagulants simplify treatment procedure and improve overall cost-effectiveness and reduce the extent of toxicity because of non-reacted monomers in polymer due to interactions among metal species and natural coagulant complexation reactions [128].

2 History of Coagulation—Development of Coagulation technology over the years

The history of coagulation tracks back to three thousand years. Around 2000 BC Egyptians, used smeared almond as coagulant for clarification of river water. During 77 AD Romans got familiar with alum which was used as a coagulant. Alum as coagulant for water treatment was used in England by 1757. During 1881 alum was adopted as coagulant formally for treating public water supplies [49, 70, 132].

Use of Ferric Chloride and Aluminium Sulphate for modern water treatment was initiated about a decade ago after which scientific studies on Coagulation were started. Widely known rule of Schulze-Hardy rule was put forth explaining mechanisms of

Coagulation [10, 49, 64]. Concept of particle collision that formed the foundation for knowing variations in particle number during the course of flocculation was developed by Smoluchowski [123]. Mattson was the first to report aluminium and iron hydrolysis products, which was widely accepted after 30 years [84]. Influence of pH and different anions on formation of floc was studied by Black and Colleague. Afterwards, the focus of research in Coagulation was directed towards better floc production through mechanical methods, along with exploration for superior coagulant aids like silicates, limestone and bentonite [18]. In 1940s, Derjaguin, Landau, Verwey and Overbeek independently proposed the DLVO theory of colloid stability colloid interaction based on Vander Waals attraction and electrical interaction that result in repulsion or attraction which depends on surroundings [40, 129].

Novel coagulation concept was later established by Langelier and Ludwig [78], illustrating two mechanisms for exclusion of colloidal particles as:

- Compression of the double layer, procedure that allows particles to overcome the repulsive forces resulting in agglomeration and precipitation, and
- Enmeshment of precipitate during which physical enmeshment of small particles takes place by metal precipitate as they form and settle.

Both of these mechanisms were theoretically explained by LaMar and Healey 1963 and they have also suggested terms Coagulation and flocculation. Micro- electrophoresis has introduced in developing concept of Coagulation for studying destabilization of colloidal that has allowed quantification of zeta potential on particles of colloid during 1960s, along with study of stoichiometric relationship among coagulant dosages which are mandatory for neutralizing colloids along with concentration of these particles in water. Additionally, these studies too indicated influence of chemical factors like ionic strength, properties of colloids for the removal and pH on colloidal particle surface potential.

Investigations during 1970s highlighted that mechanism of Coagulation and performance of Coagulation are always related to water quality standards. After the identification of organic compounds like haloforms and halogenated natural organic matter (NOM) in water after treated, principal focus of coagulation/flocculation was to be focussed on to remove NOM. Stringent drinking water standards have made many water treatment plants to adopt granular activated carbon (GAC) filter for effective removal of organic compounds from water [111].

During 1980s scientific studies on Coagulation for removal of NOM assessed on various surface water sources focussed on various factors like coagulant type, coagulant dosage, addition of chemicals, pH, re-stabilization zones and water quality characteristics [61]. Apart from NOM other precursors like humic substances, microorganisms like Legionella, Giardia, Cryptosporidium and different viruses have still remained as the main impurities of concern during 1990s. Removal of these impurities can be achieved by treatment process which includes Coagulation, filtration and disinfection. To achieve anticipated results coagulation is an indispensable process to be adopted [39, 89].

During the later years it is understood that Coagulation is also influenced by upstream and downstream treatment processes like pre-oxidation process in upstream and settling, filtration, adsorption (activated carbon) and disinfection at downstream respectively. At the same time pre-ozonation effect on attainment and efficiency of Coagulation on different raw surface waters with diverse amounts of NOM along with the effect of Coagulation on adsorption with activated carbon have been taken up which still are under research and development.

Moreover, studies focussing on improved Coagulation through adding excess coagulant dose, reducing pH or through preparation of effective coagulants are being evaluated targeting elimination of NOM and enhancing performance of Coagulation. Mathematical models are also developed for studying coagulant dose, removal efficiency of NOM and also for describing coagulation physical aspects [26, 34].

3 Theory of Coagulation

Coagulation essentially consists of destabilization and aggregation of stable small colloidal contaminations to larger particles known as floc, that are effectively removed by physico-chemical processes like rapid mixing, slow mixing, separation of solid-liquid stages in sedimentation and filtration. Colloidal impurities generally include viruses, bacteria, protozoans, color producing substances, inorganic solids and organic matter.

Conventional method of removing these impurities include a combination of the following steps:

- **Coagulation**—includes addition of coagulants and rapid stirring in raw water for complete mixing of the coagulant;
- **Flocculation**—includes slow stirring of water after rapid stirring in order to assist the particles to flocculate, so that they gain weight and settle down;
- **Sedimentation**—after coagulation and flocculation certain time is provided for the flocs to settle;
- **Filtration**—includes passing the treated water through the filters for removal of all the suspended matters.

In principle, Coagulation de-stabilizes the colloidal particles by charge neutralization after dosing of the coagulant, whereas in flocculation aggregation the colloids (with sizes μm and sub-μm) take place to form floc with mm size. Although coagulation and initial stage of flocculation happen quite swiftly, in actual process, there is a slight dissimilarity. Hence, terms either "coagulation" or "flocculation" can be adopted for describing complete process.

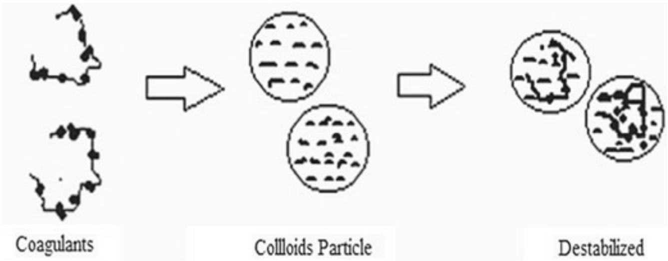

Fig. 1 Double layer compression (Redrawn with permission from Duan et al. [46])

4 Coagulation Mechanisms

As identified by O'Melia and Dempsey, Coagulation of colloidal particles can be attained in four mechanisms, which are: compression of double layer, neutralization of charge, sweep Coagulation and inter-particle bridging [33].

4.1 Double Layer Compression

Classical mechanism used to define particle destabilization; double layer compression is accomplished through addition of an electrolyte (coagulant) into the suspension of colloids. Under stable conditions the colloidal particles exist in high stability where they are unable to get close to each other owing to the presence of thick double layer. The added electrolyte brings about the change in the ionic strength of the double layer around the colloid resulting in destabilization of the colloid [46, 125]. Compression of double layer has been established to be significant destabilization mechanism (Fig. 1).

4.2 Charge Neutralization

Colloids under normal conditions possess negative charge. The charge neutralization process includes adding of metals/polymers having cations that neutralize negative charges of colloids subsequently resulting in surface charge neutralization. Thus, efficiency of the mechanism is intensely reliant on coagulant dosage, since restabilization of particle can be evident after exceeding optimum dosage (Fig. 2) [20, 29, 125].

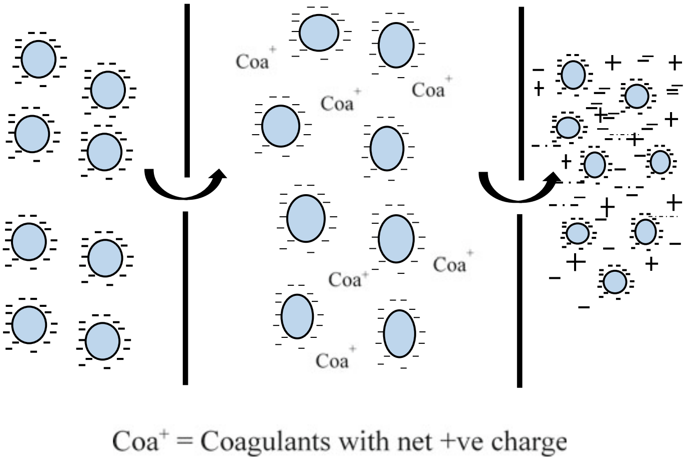

Fig. 2 Charge neutralization (Redrawn with permission from Choy et al. [29])

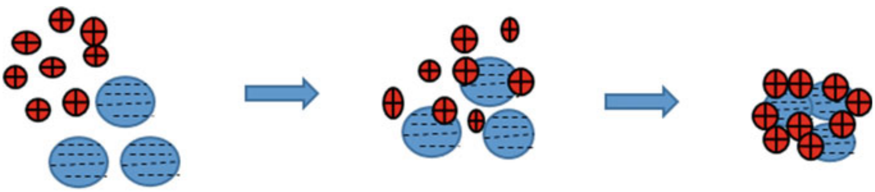

Fig. 3 Sweep Flocculation (Redrawn with permission from Duan and Gregory [45])

4.3 Sweep Coagulation

The process of forming coagulant precipitates through addition of huge quantity of coagulant is the chief process leading to sweep Coagulation (Fig. 3). The formed coagulant precipitates enmesh the colloidal particles thus successfully removing the colloidal suspension [122]. In comparison to charge neutralization sweep coagulation could result in enhanced Coagulation [45].

4.4 Inter-particle Bridging

The long-chain polymers or polyelectrolytes form bridge between them binding numerous colloids together. Higher bridging competence is achieved with larger molecular weight coagulants owing to their extended polymeric chains. The governing factor for bridging is availability of adequate vacant surface on particles so that polymer chain segments can attach [20, 29, 108].

Polymer branches that are formed may cause restabilization due to particle destabilization. In the process of inter-particle bridging, elements contour mesh-like matrix comprising of colloids that are destabilized and branches of polymer when they come together. This floc may entrain smaller particles as it begins to settle, which is stated as sweep floc that include other coagulation mechanisms in order to form the initial floc. Sweep floc is predominate mechanism of Coagulation in most of the conventional water treatment plants [7].

The mechanism of sweep coagulation is associated with metal coagulants like Al^{3+} and Fe^{3+}. These metals tend to produce huge quantity of metal hydroxide when added in larger concentrations. The formed amorphous hydroxide sweeps particles of colloid on its way towards downward for settling [109]. Significant particle removal is observed with sweep coagulation in comparison to only destabilization by charge neutralization. This can be partly owed to significantly enhanced aggregation rate, due to the increased concentration of solid. The open structure of hydroxide precipitates results in a large efficient volume concentration even with a small mass improving the possibility of adsorbing additional particles. Amirthatajah et al. 1990 illustrated that coagulation mechanisms are governed by pH and coagulant dose.

5 Factors Governing Coagulation

Factors that govern Coagulation include temperature, pH, alkalinity, concentration and composition of ions, type of coagulant used and dosage of coagulant [57, 102]. Characteristics of floc obtained from chemical coagulation using alum was compiled from different studies and anticipated that these principles are same for natural coagulants owing to the similar mechanisms of Coagulation. Depending on type and nature of natural coagulant, chief mechanisms of Coagulation important for turbidity removal is neutralization of charge and bridging, which are also governed by pH and coagulant dose. Hence, in order to understand complete prospective of natural coagulants optimization studies are required.

5.1 Temperature

One of the important parameters that governs water treated by coagulation-flocculation is temperature. At lower temperatures the process of Coagulation and flocculation is affected due to alteration in solubility of coagulant, increase in viscosity of water and obstructing the kinetics of hydrolysis reactions and particle flocculation. Furthermore, lower temperatures require elevated coagulant dosage, inclusion of filter aids, extended flocculation durations and lesser flotation, sedimentation and filtration rates, precisely upon use of alum.

5.2 pH

The most significant parameter affecting the surface charge of colloids, charge on functional groups of natural organic matter, charge on the coagulant species in dissolved phase, solubility of coagulant and surface charge of floc particles is pH, which is important for effective Coagulation.

5.3 Alkalinity

pH reduction during the coagulation process is limited by the alkalinity of raw water. At lower alkalinity concentrations coagulants tend to consume entire alkalinity available resulting in reduction of pH for efficient treatment. On the other hand, presence of high concentration alkalinity might require higher coagulant dose in order to reduce the pH towards favourable coagulation conditions. pH adjustment can be taken up by adding acid in case of high alkalinity, but this process might prove difficult for water services which deal with variations in influent of both alkalinity and coagulant dosage.

5.4 Type of Coagulants

Owing to the numerous interrelated parameters in the coagulation and flocculation process it is utmost vital to choose the precise coagulant, considering the quality of water entering the water treatment plant. Usually when common coagulants do not meet the required standards of treatment then coagulant aids are used. Hence, the type of coagulant selected should fit its use under critical conditions, so that the treated water can be of high quality for the user. Moreover, increase in sludge volume and inappropriate dewatering of sludge is due to the type of coagulant applied, which can be reduced by using a suitable coagulant [50].

5.5 Coagulant Dosage

Coagulant dosage is a significant parameter for the process of Coagulation and flocculation since it requires utmost care and appropriate control for achieving desired quality of treated water. In general, scanty coagulant dosage will result in substandard quality of treated water, due to the lack of process optimization. Diverse quality of impurities possessed by diverse water samples will need optimization of coagulant dose along with the type of coagulant used. Therefore, determining optimum coagulant dosage is required for achieving utmost turbidity removal at least cost of treatment. Charge neutralization and bridging are the only two mechanisms of the four coagulation mechanisms which are negatively affected by dosage of coagulant because of the stoichiometric relationship.

Coagulant dosages can fall under three categories like under dosing, optimum dosing and over dosing. Upon exceeding optimum coagulant dosage particle restabilization resulting in reduced turbidity removal efficacies. While surplus cationic coagulant adsorption in colloidal particles of neutral nature would result in reversing of charge which consequently mark repulsion of particles hampering the agglomeration of particles. Moreover, crowded coagulants of solution might further limit availability of adsorption sites for bridging of particles since the surface of the coagulant will be completely covered [20]. Hence, adding higher coagulant doses might not improve the coagulant process. On the other hand, coagulant dose lesser than required would result in inadequate and inefficient coagulation process as majority of colloidal particles are left in suspension.

6 Types of Coagulants

Coagulants can be either natural or chemical-based, natural coagulants are recognized for use in conventional water purification as apparent from citation in numerous ancient records [22, 44]. While alum has globally spread as coagulant intended to treat public water supply during 19th century [68]. China is world's primitive user of alum in treatment of water, which was adopted by other nations. Metal coagulants like ferric salts have reigned since 1880 in the United States but efficiency of chemical coagulants was doubtful under some working conditions like low temperature which caused admittance of polymerized aluminium coagulants (PACl). PACl reaped mounting market due to its greater efficiency and lower alkalinity consumption [20].

Infringement of chemical coagulants have ceased down the traditional water treatment methods employing natural coagulants which were restricted to rural and developing countries. This was marked as the commencement of paradigm change towards dependency on chemical coagulants for treatment of turbid waters. This shift has gradually resulted to dormancy in development of natural coagulants, making traditional methods obsolete.

6.1 Chemical Coagulants

The most widely accepted coagulants in water and wastewater treatment are Aluminum [Al (III)] and Iron [Fe (III)] based coagulants and organic coagulants (Fig. 4). Furthermore, composite coagulants are being explored in the recent years [25]. Metal salts of aluminium and iron form the metal coagulants which include Aluminum sulfate (Alum), Aluminum chloride, Acidified Aluminum Sulfate (Acid

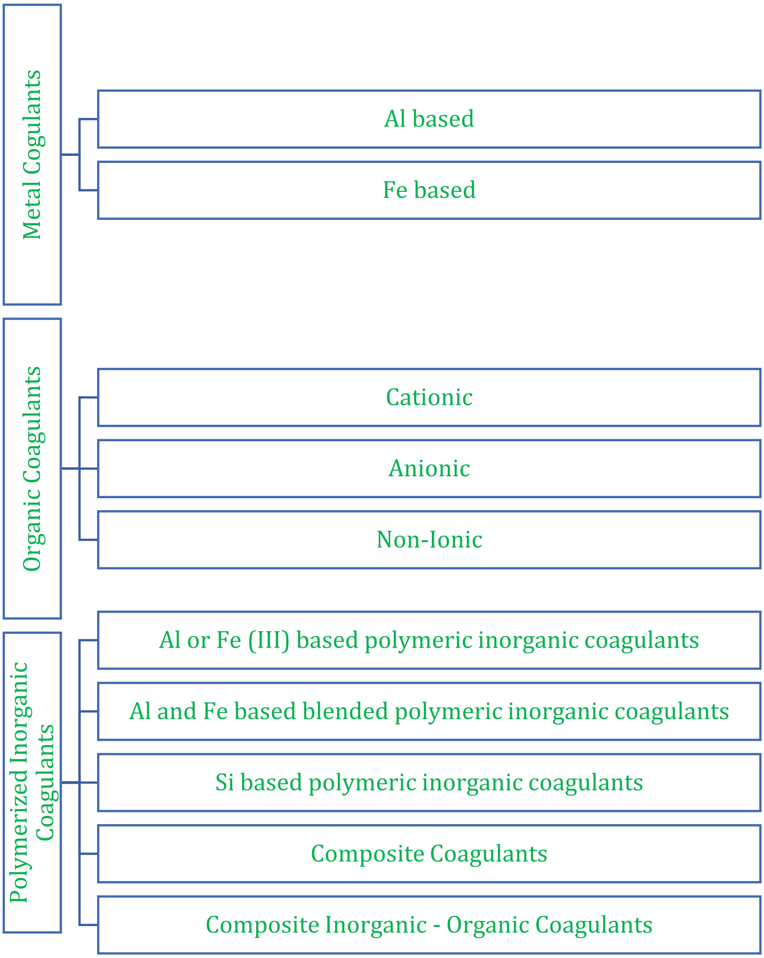

Fig. 4 Flow chart of different coagulants

Alum), Sodium Aluminate, Ferrous Sulfate, Ferric Sulfate (Alum), Ferric Chloride and Chlorinated Ferrous Sulfate. Coagulants are described as follows:

6.1.1 Aluminium Based Metal Coagulants

Widely adopted aluminium based metal coagulant is alum ($Al_2(SO_4)_3$), others being aluminium chloride ($AlCl_3$). Use of alum, however, can leave comparatively high aluminium residues in the treated water precisely under conditions of low temperatures and low pH causing health effects and other difficulties in distribution system like impulsive flocculation. Though it can be prevented by controlling pH, but then natural organic matter and turbidity will also be affected [28, 51].

6.1.2 Ferric-Based Metal Coagulants

Commonly used ferric salt-based coagulants are ferric chloride ($FeCl_3$) and ferric sulphate ($Fe_2 (SO_4)_3$) [23]. Ideal pH range for ferric-based coagulants is reported to be 4.5 to 6 with 29 to 70% removal of NOM in terms of dissolved organic carbon [1, 21, 23].

6.2 Organic Coagulants

Utilized as primary coagulants or coagulant aids for drinking water treatment, organic polymers are polymers with high-molecular-weight generally known as polyelectrolytes. When only organic polymers are used these may be effective in particle destabilization but result in production of poor-quality floc. Furthermore, organic polymers do not possess any disinfection by-product precursor removal [48]. Polymer preparations can contain contaminants originating from manufacturing process which include residual monomers, reaction by-products, which might have a potential negative impact on health of humans. Furthermore, polymers containing such contaminants can undergo reaction with chemicals added for treating water forming undesirable secondary products. These concerns have made countries like Switzerland and Japan to adopt legislations barring utilization of such organic (synthetic) polymers in treatment of potable water. Stringent limits on use of organic polymers have been established by United States, France and Germany [2, 81].

Classification of organic polymers is based on the electric charge on their macro ions that dissociate in water. These are Cationic, Anionic and Non-ionic. Among these cationic polymers are generally used as primary coagulants, whereas anionic and non-ionic polymers are stated as coagulant aids or flocculants.

6.2.1 Cationic Polyelectrolyte

Example of this class include Poly dimethyldiallyl ammonium chloride produces ions which are positively charged upon dissolution in water. These are generally utilized as primary coagulants/coagulant aids to metal coagulants owing to the fact that suspended and colloidal particles present in water are usually negatively charged.

6.2.2 Anionic Polyelectrolyte

Example of this class include Copolymer acrylamide/Trimethylaminoethyl acrylate, which produce negatively charged ions upon dissolution in water. These polymers are generally used in blend with metal coagulants for removing positively charged solids.

6.2.3 Non-ionic Polyelectrolyte

Example of this class include Non-ionic polyacrylamide, which have balanced ions, releasing both cations and anions upon dissolution in water.

6.3 Polymerized Inorganic Coagulants

The development and use of polymerized inorganic coagulants trace back to 1980s. Since than these coagulants have proved to be promising with their greater performance in treating water and wastewater. Their performance can be owed to their wider working pH range, lower sensitivity towards water temperature and moreover these require quite small dosages to attain the same treatment competence and also result in very fewer residual concentrations of metal-ions. These characteristics are accredited to the existence of wide range of polymeric species consisting great molecular weights. In general polymers possess higher cationic charge that increase their surface activity and capacity of charge-neutralization, making them more competitive, enhancing the rate of colloidal charge neutralization, floc development and settlement, cost reduction, production of less sludge over conventional coagulants [56, 71]. Generally used inorganic polymeric coagulants are Al and Fe (III) based, a few examples are as follows:

6.3.1 Aluminum Chlorohydrate

Probably the first ploymerized coagulant that was developed in United Kingdom during 1950s. Used both as sludge conditioner and as primary coagulant it has a theoretical formula $Al_2(OH)_5Cl$.

6.3.2 Al or Fe (III) Based Polymeric Inorganic Coagulants

Having wield application for water and wastewater treatment these include Polyferric chloride and Polyaluminum chloride, which are hydrolysed products of Al and Fe (III) [38, 79]. Other hydrolysed products from aluminum chloride with aluminum sulphate and ferric sulphate are Polyaluminum chloride Sulphate and Polyferric Sulphate respectively [56, 71].

6.3.3 Al and Fe (III) Based Blended Polymeric Inorganic Coagulants

Blended inorganic coagulant prepared from solution of Al and Fe which is hydrolyzed with alkaline materials is polyaluminumferric chloride [54].

6.3.4 Si Based Polymeric Inorganic Coagulants

These are prepared from metal polymerized inorganic coagulants in combination with polysilicic acid [19, 54, 55, 91]. Ex: Polyferric Silicate chloride, Polyaluminum Silicate chloride, and Poly-aluminum-ferric Silicate chloride, etc.

6.3.5 Composites Coagulants

When cationic polyelectrolyte organic polymers are injected into metal polymerized inorganic coagulants, these form composite coagulants. Ex: PACl with PDMDAAC, PFCl with PDMDAAC [133].

6.3.6 Composite Inorganic-Organic Coagulants

Latest innovation in coagulants is preparing composite coagulants by combining organic polymers to inorganic polymerized flocculants forming inorganic-organic coagulants to possess the advantages of both components.

6.4 Drawbacks of Chemical Coagulants

It can be understood that 21st century have been driven by chemical coagulants for water and wastewater treatment primarily because of comfort of availability, management, use, storage and mixing, being economical etc. [83, 118, 125]. Though these have been widely applicable, their usage comes with limitations and is being probed upon imparting brown colour to equipment by iron salts and impacts on environment due to higher concentrations of residual aluminium is in treated water, wastewater and sludge [37, 83, 136].

Harmful effects of using chemical coagulants upon human health were published in 1960s [121]. Moreover biochemical, neuropathological and epidemiological studies advocate [13, 86, 103, 110, 131] and senile dementia due to residual aluminium in treated waters. Results from several epidemiological and clinical observation reported at least 70% positive correlation between Alzheimer's disease and aluminium residue in drinking water. Nevertheless, conflicting results were also reported owing to absence of significant indication in several cases [134]. Therefore, a decisive relation between aluminium and development of Alzheimer's disease cannot be determined. However, in order to minimize risk over sustained period of consumption, threshold concentration of aluminium in treated water was observed and stated to be 0.02 mg/L [42], 0.1 mg/L [86, 110] and proposed aluminium concentration by WHO is 0.2 mg/L [121].

Limitations of their usage include their ineffectiveness at lower temperatures, harmful human health effects, generation of large volumes of sludge and pH variations in treated water [30, 116]. Environments like extreme pH and temperature result in production of very sensitive and fragile flocs leading to poor sedimentation and these flocs might undergo rupture upon any physical force. Generation of huge hydrous oxide natured sludge that is non-biodegradable is additional chief drawback of using metal coagulants. It is also reported that sludge obtained from using alum is hard to dewater. Also, alum sludge can cause uptake of phosphorous from plants through absorption of inorganic phosphorus leading to aluminium phytotoxicity [41]. Owing to these drawbacks of chemical coagulants, an urgent need for considering other potential alternatives fitting in the objective of green chemistry is required for treating water, which can minimize environmental damages and protect wellbeing of human population.

6.5 Advantage of Using Natural Coagulants

Advantages of natural coagulants are evident; they are economical, will not change pH of treated water [135], highly biodegradable, require low dose of coagulant, omit pH adjustment as these will not consume water alkalinity like alum [98], low/negligible toxicological risk, harmless to human health and aquatic life, subsequently lesser impact on environment [73].

Additionally, these are renewable in nature and hence have negligible net impact on global warming [117], does not cause secondary pollution [109], generally possess several surface charges increasing the efficacy of coagulation process [52], they are non-corrosive, eliminating concern of pipe line corrosion [30]. Furthermore, they produce lesser sludge volumes, nearly five times lesser, which can be treated biologically and disposed of as soil conditioners as the sludge possess higher nutritional value and is non-toxic [94]. This will reduce treatment and handling costs of sludge, making natural coagulants better sustainable option [31].

All the above-mentioned advantages are specifically amplified if the coagulant derived plant is indigenous or if these plants are cultivated, harvested and processed locally [52]. Moreover, in the present stage of climate change, environmental degradation and depleting natural resources, adaptation of natural coagulants, satisfying the requirements of green technology is vital effort towards sustainable development initiatives [88, 135].

7 Natural Coagulants

Due to environmental concerns worldwide, searching for alternatives to the chemical coagulants for water treatment has become compulsory as they produce toxic substances during their extraction and purification process. Thus, several researchers are searching for and exploring natural coagulants to treat water [36, 37, 116]. Usage of natural coagulants for water purification and wastewater is well-known through human history from ancient times to date [94, 112]. Natural coagulant usage has been dated for more than 200 years in countries like Africa, India, and China for removing turbidity [9]. In India, it is apparent as the Sanskrit writing dating back to 400 AD, also from the Old Testament and Romans records dating back to 77 AD [44]. Natural coagulants can be categorized into cationic, anionic, or non-ionic, also called polyelectrolytes [135]. Examples include [109]:

- Natural Cationic polymers: cationic starches and chitosan
- Natural Anionic polymers: modified lignin sulfonates and sulphated polysaccharides
- Natural Non-ionic polymers: cellulose derivatives and starch [65].

The mechanisms exhibited by the natural coagulants include adsorption and charge neutralization, and adsorption, and inter particulate bridging. Adsorption and neutralization results while suspended particles adsorb on oppositely charged ions; interparticle bridging happens once chains of polysaccharide coagulants adhere to several particles making particles bound to a coagulant and this can be attributed to long-chained structures with great molecular weights, which greatly enhance the number of unoccupied adsorption sites. It is understood that precisely these two mechanisms provide fundamental principles to internal mechanisms of plant-based coagulants. Moreover, electrolytes in the aqueous conditions can enable these coagulants' coagulants, as the electrostatic repulsion between particles is low [66].

Natural coagulants based on the origin can be grouped into three categories: plant-derived, animal-derived microorganism derived (Fig. 5).

- **Animal-derived**: While examples of animal sources include chitosan from the shells of crustaceans [22] and isinglass from shredded fish bladders [16]. Chitosan, natural polysaccharide possesses beneficial features like the competence of adsorbing metal ions due to amino groups, biocompatibility, hydrophilicity, non-toxic, biodegradability, and linear cationic polymer with high molecular weight.
- **Microorganism derived**: examples of microorganisms derived are microbial polysaccharides and alginate. Alginates are known for their characteristics like gelling, keeping water viscosifying and stabilizing properties [80] and Oladoja,

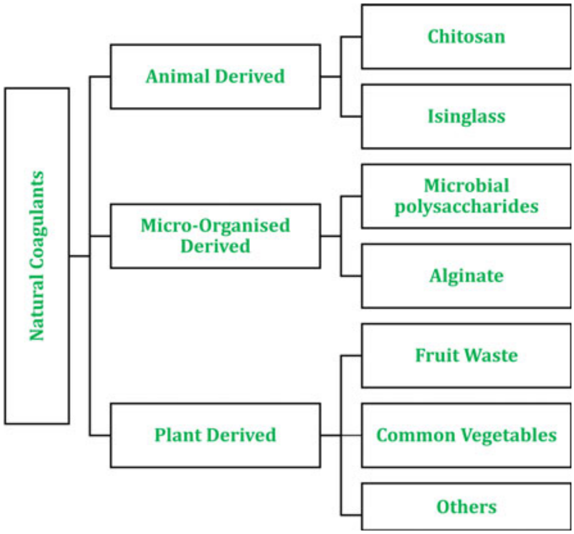

Fig. 5 Flow chart of different categories of natural coagulants

in his studies he reported that high flocculating activity due to the existence of polysaccharides and protein with neutral sugar, an amino sugar, and uranic acids [95, 98].

- **Plant-derived**: The availability of coagulants from plants is more significant than that from animal sources, suggesting that plant-derived coagulants are impending alternatives to chemical coagulants. Hence these have gained attention over the years [29]. These advantages are economical and efficient in treating waters with low to medium turbidity (50–500NTU).

7.1 Classification of Plant-Derived Coagulants

Sources of plant-derived coagulants are vast; these may be directly taken from various parts of plants or even waste after processing plant parts, leading to a disarray in the compilation. From the literature, plant-derived coagulants can be classified as originating from vegetables and legumes, fruit waste, and others (Fig. 6).

7.1.1 Common Vegetables

Moringa Olifera: is native north Indian species belonging to family Moringacea, it is a deciduous tree, and its coagulative properties were identified in the 18th century. The seeds contain positive ion proteins, which act as an antibacterial flocculant and used for water clarification. Amagloh and Benang [6], compared alum with moringa and revealed that 12 g/1000 ml of *Moringa olifera* powder showed analogous turbidity removal to alum. Pritchard et al. [105] reported that using *Moringa olifera*, 84%

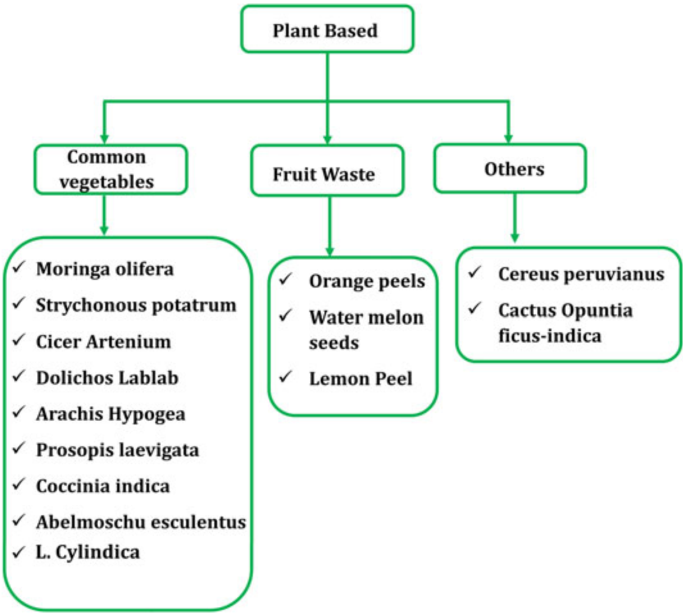

Fig. 6 Coagulants derived from plants

reduction in turbidity was achieved with an initial turbidity of 146 NTU. *Moringa oleifera* is a flocculant that is available naturally around regions like India, Africa, Madagascar, and Arabia; six of fourteen Moringa species are available abundantly. Developing countries like India and Sudan are using these seeds from generations for treating the water, and it is not new for them [59]. Also, comparison studies of these species were done with aluminum sulfate and ferric chloride in turbid water (160 NTU), and this species showed a reduction of 97% turbidity. In contrast, remaining coagulants like aluminum sulfate and ferric chloride showed 99 and 98%. Color removal using these coagulants was studied by Pritchard et al. [105], and achieved 83% reduction in colour by *Moringa oleifera*, 88% by alum and 93% by ferric sulfate. Gaikwad and Munavalli [53], reported that at lower turbidity (35NTU) by using lesser concentrations 50 mg/l, they achieved 90.6% removal. In comparison with other synthetic coagulants, the turbidity removal or performance is less. Hence, *Moringa oleifera* is competent enough and can be used as an alternative to the synthetic coagulants.

Strychnos potatorum: (Nirmali) is a traditional medicinal tree and available in central parts of India, Burma, and Srilanka. It is a medium-sized tree; materials form this tress produce alkaloids like acetyldaibolin and diabolin used for the treatment of epilepsy and urolithiasis. Niramli seeds contain alkaloids like nonvaccine, strychnine, etc., whereas bark has campest Erol, sitosterol, and isomotiol. The seed powder consists of polyelectrolytes of anionic charge and destabilizes the impurities in water by intermolecular bonding. Earlier studies revealed that extracts from the seed comprised of lipids, carbohydrates, and alkaloids containing carboxyl groups and alcohol groups. The carboxyl and alcohol groups are responsible for Coagulation and enhance the intermolecular bonding. Galactan and galactomannan are polysaccharides extracted from the seeds of *Strychnos potatorum* that have a capability in reducing the turbidity of the solution up to 80%. Galactomannans consist of 1,4-linked dmannopyranosyl with terminal ends d-galactopyranosyls. Even though many researchers used nirmali seeds for the removal of turbidity, the exact mechanism that has been associated with the Coagulation was not explained clearly. Some of the researchers summarised that presence of alcohol chains in the seeds is helping for adsorption. In bench-scale studies, *Strychnos potatorum* seeds showed good adsorption, and direct filtration is acting as an effective method for treating low turbidity water [53, 96].

Cicer arietinum: belongs to the family of Fabaceae and commonly called as chickpea, Choubey et al. [27], reported that 95.89% of turbidity reduction had been observed in comparison with alum in his study. These seeds have antibacterial properties and help in the removal of microorganisms from water. *Cicer arietinum* is more effective in removing high turbidity of water by maintaining the experimental setting three minutes high mixing, followed by twelve minutes slow mixing with 100 mg/l dosage. These plant materials are available cheaply in all parts of Asia. Contrarily, these are biodegradable and safe for consumption by humans [9].

Dolichos lablab: belongs to Fabaceae family and is commonly called Hyacinth bean. Shilpaa et al. [119], reported 99.14% of turbidity reduction with an initial NTU of 500 by treating with seeds of *Dolichos lablab.* Asrafuzzaman et al. [9], by their study stated that lower dosages of the coagulants showed high efficiency in removing the turbidity of the water, however increasing the dosage of the coagulants decreased turbidity removal of the solution. The most effective dose of hyacinth bean for turbidity removal is 200 mg/500 ml and showed a 68% removal at 60 min settling time [27].

Arachis hypogea: belong to the family of Fabaceae are known as peanut. Birima et al. [17], used the seeds of Arachis hypogea for turbidity removal and achieved 92% efficiency by extracting the peanuts with 6 mol/NaCl with initial NTU 200 turbidity using 200 mg/l dose. However, peanut extract with distilled water showed only a 31.5% reduction. The proteins present inside the peanut are responsible for the coagulation activity, and they found that KCl and $NaNO_3$ are also suitable solvents to extract the proteins in the coagulants, which are useful for coagulation activity.

Prosopis laevigata: commonly known as honey mesquite and belongs to Fabaceae, these trees can grow up to 13 meters. *Prosopis laevigata* have similar characteristics to the *Prosopis galactomannans* [24]. The gums of these seeds are generally used as a coagulant. Torres and Carpinteyro-Urban [127], used these seeds and concluded that galactomannans present in the seeds act as a coagulating agent for treating the water and wastewater.

Coccinia Indica: Patale and Pandya [99], used the *C. indica* fruit extract and stated that mucilage of these substances showed high efficiency in removing turbidity in comparison to low turbidity. Turbidity removal with an initial 100 NTU was obtained 94% removal, whereas the lowest turbidity 10 NTU got 82% removal.

Abelmoschus esculentus: Abelmoschus esculentus, commonly called Okra, belongs to the family Malvaceae. Mishra et al. [120], used okra seeds powder as a coagulant and compared the turbidity removal with alum. Okra seeds showed similar results to alum in clarification, 72% turbidity removal was achieved by using lesser concentrations of the coagulant in their study. Raji et al. [107], conducted similar investigations and used optimum dosage of 300 mg/l and reduced turbidity from 745 NTU to 11 NTU at neutral pH and concluded that using optimum dosage will give 99% efficiency.

Luffa cylindrica: also called sponge guard, consists of light-coloured black seeds. These plants are using traditional medicine as it contains triterpenoids and a-tocopherol acts as an anti-inflammatory drug [11]. *Luffa cylindrica* belongs to the family Cucurbitaceae. Sowmeyan et al. [124], used the extracts of the sponge guard and removed turbidity up to 85%. Extracts of the plant have not been specified by the authors and postulated that the plant extract might be seeds or whole fruits [4].

7.1.2 Fruit Waste as Coagulants

Citrus sinensis: are commonly called as oranges and belong to the family of Rutaceae. The peels of these fruits consist of carboxyl and hydroxyl functional groups along with substances like pectin and cellulose. The presence of these groups in the peels is acting as an adsorbent material in removing the organic substance from the water [115]. Pathak et al. [100], used the peel powder of orange and achieved 88.4% turbidity removal by using 60 mg/l dosage. They concluded that orange peel powder has the potential to reduce the turbidity. Klancnik et al. [76], used the peel powder and removed the colour up to 99.7%, and the efficiency of these substances is compared with the activated carbon treatment.

Citrus limon: belongs to Rutaceae family and locally called as lemon, and these fruits are available all over the world. Peels of these fruits have been used as a coagulant from ancient days and theses are non-corrosive and non-toxic to water treatment units and reduce sludge during the treatment. Dollah et al. [43], used the peel powder of Citrus limon and reduced the turbidity to 89% by using a coagulant dose of 60 mg/l.

Musa: The scientific name of the banana is Musa and is available around the word as edible fruit. The peels of this fruit are used for various purposes like composting and cosmetics preparation. These substances are biodegradable and will not cause any type of pollution. The Musa peels have high adsorption capacities due to the presence of lignocellulose. Mokhtar et al. [90], used banana peel powder as a coagulant and reduced the turbidity of water with 88% efficiency. Similar studies were conducted by John et al. [72], and achieved 95% turbidity removal. The stem juice of the Musa was used as a coagulant by Alwi et al. [5], and reported 98.5% efficiency in removing turbidity with 90 ml/l dosage.

Citrullus lanatus: is a plant species belonging to the Cucurbitaceae family and commonly known as watermelon. More than a thousand varieties are available and cultivable around the world. Datti et al. [35], used the seed powder of melon and achieved 75% turbidity removal with initial turbidity 58.7 NTU. Similar studies were conducted by Sekar et al. [113], and treated turbid waters with watermelon seed powder and obtained results within limits given by world health organization.

7.1.3 Others

Cactus Opuntia ficus-indica: belongs to cacti species, and it consists of mucilaginous cells and stores mucilage in both peel and pulp. These plant species are available in semi-arid regions of the world, and it can grow up to five meters. Many researchers [69, 88] studied the characteristics of opuntia, and mucilage is used as a coagulant. Owing to its availability and low cost, it has gained more importance in treating water and wastewater. Bandala et al. [12], converted the mucilage into powder and used as an adsorbent for coagulation technique and achieved excellent results.

Cereus peruvianus: belongs to the family of Cactaceae and commonly known as Peruvian Apple Cactus. These plants can grow up to 10 meters and is widely used as an ornamental plant. De Souza et al. [37], extracted mucilage from the Peruvian and used for the water clarification and achieved 95% efficiency in turbidity removal.

8 Conclusions

Nature's diversity and stability provide numerous solutions; so that species on the earth are distributed equally, and no other species are exhausted. Thus, knowledge of more materials having coagulating properties might provide options even in the abnormal or worst conditions leading to resilience, one of the primary sustainability principles. The present book chapter presents the properties and efficiency of natural coagulants classified under plant-based, fruit waste, etc. Furthermore, an in-depth understanding of the above said coagulants concerning their optimizing process parameters is essential in future research and exploring more natural resources having coagulating properties for treating water and wastewater.

References

1. Abbaszadegan M, Mayer BK, Ryu H, Nwachuku N (2007) Efficacy of removal of CCL viruses under enhanced coagulation conditions. Environ Sci Technol 41(3):971–977
2. Aizawa T (1990) Problems with introducing synthetic polyelectrolyte coagulants into the water purification process. Water supply 8:27–35
3. Al-Saati NHA, Hwaidi EH, Jassam SH (2016) Comparing cactus (Opuntia spp.) and alum as coagulants for water treatment at Al-Mashroo canal: a case study. Int J Environ Sci Technol 13(12):2875–2882
4. Altınışık A, Gür E, Seki Y (2010) A natural sorbent, Luffa cylindrica for the removal of a model basic dye. J Hazard Mater 179(1–3):658–664

5. Alwi H, Idris J, Musa M, Ku Hamid KH (2013) A preliminary study of banana stem juice as a plant-based coagulant for treatment of spent coolant wastewater. J Chem 2013
6. Amagloh FK, Benang A (2009) Effectiveness of Moringa oleifera seed as coagulant for water purification
7. Amirtharajah A, O'melia CR (1990) Coagulation processes: destabilization, mixing, and flocculation. McGraw-Hill, Inc., USA, p 1194
8. Antov MG, Šćiban MB, Prodanović JM (2012) Evaluation of the efficiency of natural coagulant obtained by ultrafiltration of common bean seed extract in water turbidity removal. Ecol Eng 49:48–52
9. Asrafuzzaman M, Fakhruddin ANM, Hossain M (2011) Reduction of turbidity of water using locally available natural coagulants. ISRN Microbiol 2011
10. Austen PT, Wilber FA (1884) Annual report of the State Geologist of New Jersey
11. Azeez MA, Bello OS, Adedeji AO (2013) Traditional and medicinal uses of Luffa cylindrica: a review. J Med Plants 1(5):102–111
12. Bandala ER, Tiro JB, Lujan M, Camargo FJ, Sanchez-Salas JL, Reyna S, Torres LG (2013) Petrochemical effluent treatment using natural coagulants and an aerobic biofilter. Adv Environ Res 2(3):229–243
13. Banks WA, Niehoff ML, Drago D, Zatta P (2006) Aluminum complexing enhances amyloid β protein penetration of blood–brain barrier. Brain Res 1116(1):215–221
14. Beltrán-Heredia J, Sánchez-Martín J, Gómez-Muñoz MC (2010) New coagulant agents from tannin extracts: preliminary optimisation studies. Chem Eng J 162(3):1019–1025
15. Benetti AD (2008) Water reuse: issues, technologies, and applications. Engenharia Sanitaria e Ambiental 13(3):247–248
16. Biggs S (2015) Polymeric flocculants. In: Encyclopedia of surface and colloid science. CRC Press, pp 5918–5934
17. Birima AH, Hammad HA, Desa MNM, Muda ZC (2013) Extraction of natural coagulant from peanut seeds for treatment of turbid water. In: IOP conference series: earth and environmental science, vol 1. IOP Publishing Ltd., pp 1–4
18. Black AP (1934) Coagulation with iron compounds. J (Am Water Works Assoc) 26(11):1713–1718
19. Boisvert JP, To TC, Berrak A, Jolicoeur C (1997) Phosphate adsorption in flocculation processes of aluminium sulphate and poly-aluminium-silicate-sulphate. Water Res 31(8):1939–1946
20. Bolto B, Gregory J (2007) Organic polyelectrolytes in water treatment. Water Res 41(11):2301–2324
21. Bond T, Goslan EH, Parsons SA, Jefferson B (2010) Disinfection by-product formation of natural organic matter surrogates and treatment by coagulation, MIEX® and nanofiltration. Water Res 44(5):1645–1653
22. Bratby J (2006) Coagulation and flocculation in water and wastewater treatment, 2nd edn. IWA Publishing, London
23. Budd GC, Hess AF, Shorney-Darby H, Neemann JJ, Spencer CM, Bellamy JD, Hargette PH (2004) Coagulation applications for new treatment goals. J-Am Water Works Assoc 96(2):102–113
24. Chaires-Martínez L, Salazar-Montoya JA, Ramos-Ramírez EG (2008) Physicochemical and functional characterization of the galactomannan obtained from mesquite seeds (Prosopis pallida). Eur Food Res Technol 227(6):1669
25. Chen Z, Fan B, Peng X, Zhang Z, Fan J, Luan Z (2006) Evaluation of Al30 polynuclear species in polyaluminum solutions as coagulant for water treatment. Chemosphere 64(6):912–918
26. Cheng RC, Krasner SW, Green JF, Wattier KL (1995) Enhanced coagulation: a preliminary evaluation. J-Am Water Works Assoc 87(2):91–103
27. Choubey S, Rajput SK, Bapat KN (2012) Comparison of efficiency of some natural coagulants-bioremediation. Int J Emerg Technol Adv Eng 2(10):429–434
28. Chow CW, van Leeuwen JA, Fabris R, Drikas M (2009) Optimised coagulation using aluminium sulfate for the removal of dissolved organic carbon. Desalination 245(1–3):120–134

29. Choy SY, Prasad KMN, Wu TY, Ramanan RN (2015) A review on common vegetables and legumes as promising plant-based natural coagulants in water clarification. Int J Environ Sci Technol 12(1):367–390
30. Choy SY, Prasad KMN, Wu TY, Raghunandan ME, Ramanan RN (2014) Utilization of plant-based natural coagulants as future alternatives towards sustainable water clarification. J Environ Sci 26(11):2178–2189
31. Choy SY, Prasad KN, Wu TY, Raghunandan ME, Ramanan RN (2016) Performance of conventional starches as natural coagulants for turbidity removal. Ecol Eng 94:352–364
32. Cisneros BJ, Rose JB (eds) (2009) Urban water security: managing risks: UNESCO-IHP, vol 5. CRC Press
33. Crittenden JC, Trussel RR, Hand DW, Howe KJ, Tchobanoglous G (2005) Coagulation, mixing and flocculation. Water Treat Princi Design 2
34. Crozes G, White P, Marshall M (1995) Enhanced coagulation: its effect on NOM removal and chemical costs. J-Am Water Works Assoc 87(1):78–89
35. Datti Y, Barau SS, Nura T (2019) Chemical compositions and the phytochemical constituents of the seed of Sesamum Indicum grown at Katsina State, Northern Nigeria. Fudma J Sci 3(4):201–205. Issn: 2616-1370
36. de Souza MTF, Ambrosio E, de Almeida CA, de Souza Freitas TKF, Santos LB, de Cinque Almeida V, Garcia JC (2014) The use of a natural coagulant (Opuntia ficus-indica) in the removal for organic materials of textile effluents. Environ Monit Assess 186(8):5261–5271
37. de Souza MTF, de Almeida CA, Ambrosio E, Santos LB, de Souza Freitas TKF, Manholer DD, …, Garcia JC (2016) Extraction and use of Cereus peruvianus cactus mucilage in the treatment of textile effluents. J Taiwan Inst Chem Eng 67:174–183
38. Dempsey BA, Ganho RM, O'Melia CR (1984) The coagulation of humic substances by means of aluminum salts. J-Am Water Works Assoc 76(4):141–150
39. Dempsey BA, Sheu H, Ahmed TT, Mentink J (1985) Polyaluminum chloride and alum coagulation of clay-fulvic acid suspensions. J-Am Water Works Assoc 77(3):74–80
40. Deryagin BV, Landau LD (1941) Theory of stability of strongly charged lyophobic sols and adhesion of strongly charged particles in electrolytic solutions. J Exp Theor Phys 11:802
41. Dharmappa HB, Hasia A, Hagare P (1997) Water treatment plant residuals management. Water Sci Technol 35(8):45–56
42. Doll R (1993) Alzheimer's disease and environmental aluminium. Age Ageing 22(2):138–153
43. Dollah Z, Abdullah ARC, Hashim NM, Albar A, Badrealam S, Zaki ZM (2019) Citrus fruit peel waste as a source of natural coagulant for water turbidity removal. In: Journal of physics: conference series, vol 1349, no 1. IOP Publishing, p 012011
44. Dorea CC (2006) Use of Moringa spp. seeds for coagulation: a review of a sustainable option. Water Sci Technol Water Supply 6(1):219–227
45. Duan J, Gregory J (2003) Coagulation by hydrolysing metal salts. Adv Coll Interface Sci 100:475–502
46. Duan J, Niu A, Shi D, Wilson F, Graham NJD (2009) Factors affecting the coagulation of seawater by ferric chloride. Desalinat Water Treat 11(1–3):173–183
47. Edzwald JK (1993) Coagulation in drinking water treatment: particles, organics and coagulants. Water Sci Technol 27(11):21–35
48. Edzwald JK, Becker WC, Wattier KL (1985) Surrogate parameters for monitoring organic matter and THM precursors. J-Am Water Works Assoc 77(4):122–132
49. Faust SD, Aly OM (1998) Chemistry of water treatment. CRC Press
50. Fazeli M, Safari M, Ghobaee T (2014) Selecting the optimal coagulant in turbidity removal and organic carbon of surface water using AHP. Bull Environ Pharmacol Life Sci 3:78–88
51. Flaten TP (2001) Aluminium as a risk factor in Alzheimer's disease, with emphasis on drinking water. Brain Res Bull 55(2):187–196
52. Freitas TKFS, Oliveira VM, De Souza MTF, Geraldino HCL, Almeida VC, Fávaro SL, Garcia JC (2015) Optimization of coagulation-flocculation process for treatment of industrial textile wastewater using okra (A. esculentus) mucilage as natural coagulant. Ind Crops Prod 76:538–544

53. Gaikwad VT, Munavalli GR (2019) Turbidity removal by conventional and ballasted coagulation with natural coagulants. Appl Water Sci 9(5):130
54. Gao BY, Hahn HH, Hoffmann E (2002) Evaluation of aluminum-silicate polymer composite as a coagulant for water treatment. Water Res 36(14):3573–3581
55. Gao BY, Yue QY, Wang BJ (2006) Properties and coagulation performance of coagulant poly-aluminum-ferric-silicate-chloride in water and wastewater treatment. J Environ Sci Health, Part A 41(7):1281–1292
56. Gao B, Yue Q (2005) Effect of SO_42-/Al^{3+} ratio and OH−/Al^{3+} value on the characterization of coagulant poly-aluminum-chloride-sulfate (PACS) and its coagulation performance in water treatment. Chemosphere 61(4):579–584
57. Geng Y (2006) Application of flocs analysis for coagulation optimization at the Split Lake water treatment plant
58. Ghebremichael KA, Gunaratna KR, Henriksson H, Brumer H, Dalhammar G (2005) A simple purification and activity assay of the coagulant protein from Moringa oleifera seed. Water Res 39(11):2338–2344
59. Gottsch E (1992) Purification of turbid surface water by plants in Ethiopia. Walia 1992(14):23–28
60. Guarino AS (2017) The economic implications of global water scarcity. Res Econom Manage 2(1):51
61. Gullick RW (2003) AWWA's source water protection committee outlines how to maintain the highest quality source water. J-Am Water Works Assoc 95(11):36–42
62. Gunaratna KR, Garcia B, Andersson S, Dalhammar G (2007) Screening and evaluation of natural coagulants for water treatment. Water Sci Technol Water Supply 7(5–6):19–25
63. Haaroff J, Cleasby JL (1988) Comparing aluminum and iron coagulants for in-line filtration of cold waters. J Am Water Works Assoc 80(4):168–175
64. Hardy WB (1900) A preliminary investigation of the conditions which determine the stability of irreversible hydrosols. Proc R Soc London 66(424–433):110–125
65. Hassan MAA, Hui LS, Noor ZZ (2009) Removal of boron from industrial wastewater by chitosan via chemical precipitation. J Chem Nat Res Eng 4:1–11
66. Huang C, Chen Y (1996) Coagulation of colloidal particles in water by chitosan. J Chem Technol Biotechnol Int Res Process Environ Clean Technol 66(3):227–232
67. Jadhav MV, Mahajan YS (2013) Investigation of the performance of chitosan as a coagulant for flocculation of local clay suspensions of different turbidities. KSCE J Civ Eng 17(2):328–334
68. Jahn SAA (2001) Drinking water from Chinese rivers: challenges of clarification. J Water Supply Res Technol—AQUA, 50(1):15–27
69. Jeon JR, Kim EJ, Kim YM, Murugesan K, Kim JH, Chang YS (2009) Use of grape seed and its natural polyphenol extracts as a natural organic coagulant for removal of cationic dyes. Chemosphere 77(8):1090–1098
70. Jiang JQ (2001) Development of coagulation theory and pre-polymerized coagulants for water treatment. Sep Purif Methods 30(1):127–141
71. Jiang JQ, Graham NJ (1998) Preparation and characterisation of an optimal polyferric sulphate (PFS) as a coagulant for water treatment. J Chem Technol Biotechnol Int Res Process Environ Clean Technol 73(4):351–358
72. John B, Baig U, Fathima N, Asthana S, Sirisha D (2017) Removal of turbidity of water by banana peel using adsorption technology. J Chem Pharmaceut Res 9(4):65–68
73. Kakoi B, Kaluli JW, Ndiba P, Thiong'o G (2016) Banana pith as a natural coagulant for polluted river water. Ecol Eng 95:699–705
74. Katayon S, Noor MMM, Asma M, Ghani LA, Thamer AM, Azni I, Suleyman AM (2006) Effects of storage conditions of Moringa oleifera seeds on its performance in coagulation. Biores Technol 97(13):1455–1460
75. Kingsely OJ, Nnaji JC, Ugwu BI (2017) Biodisinfection and coagulant properties of mixed Garcinia kola and Carica papaya seeds extract for water treatment. Chem Sci Int J 1–9
76. Klančnik M (2014) Coagulation and adsorption treatment of printing ink wastewater. Acta graphica: znanstveni časopis za tiskarstvo i grafičke komunikacije 25(3–4):73–82

77. Kumar P, Rouphael Y, Cardarelli M, Colla G (2017) Vegetable grafting as a tool to improve drought resistance and water use efficiency. Front Plant Sci 8:1130
78. Langelier WF, Ludwig HF (1949) Mechanism of flocculation in the clarification of turbid waters. J (Am Water Works Assoc) 41(2):163–181
79. Leprince A, Fiessinger F, Bottero JY (1984) Polymerized iron chloride: an improved inorganic coagulant. J-Am Water Works Assoc 76(10):93–97
80. Mabrouk ME (2014) Production of bioflocculant by the marine actinomycete Nocardiopsis aegyptia sp. nov. Life Sci J 11:27–35
81. Mallevialle J, Bruchet A, Fiessinger F (1984) How safe are organic polymers in water treatment? J-Am Water Works Assoc 76(6):87–93
82. Martyn CN, Coggon DN, Inskip H, Lacey RF, Young WF (1997) Aluminum concentrations in drinking water and risk of Alzheimer's disease. Epidemiology 281–286
83. Matilainen A, Vepsäläinen M, Sillanpää M (2010) Natural organic matter removal by coagulation during drinking water treatment: a review. Adv Coll Interface Sci 159(2):189–197
84. Mattson S (1928) The electrokinetic and chemical behavior of the alumino-silicates. Soil Sci 25(4):289–312
85. McConnachie GL, Folkard GK, Mtawali MA, Sutherland JP (1999) Field trials of appropriate hydraulic flocculation processes. Water Res 33(6):1425–1434
86. McLachlan DRC (1995) Aluminium and the risk for Alzheimer's disease. Environmetrics 6(3):233–275
87. Miller RG, Kopfler FC, Kelty KC, Stober JA, Ulmer NS (1984) The occurrence of aluminum in drinking water. J-Am Water Works Assoc 76(1):84–91
88. Miller SM, Fugate EJ, Craver VO, Smith JA, Zimmerman JB (2008) Toward understanding the efficacy and mechanism of Opuntia spp. as a natural coagulant for potential application in water treatment. Environ Sci Technol 42(12):4274–4279
89. Miltner MJ, Nolan SA, Summers RS (1994) Evolution of enhanced coagulation for DBP coagulation: critical issues in water and wastewater treatment. In: Proceeding of the 1994 national conference on environmental engineering, ASCE, Boulder, CO
90. Mokhtar NM, Priyatharishini M, Kristanti RA (2019) Study on the effectiveness of banana peel coagulant in turbidity reduction of synthetic wastewater. Int J Eng Technol Sci 6(1):82–90
91. Moussas PA, Zouboulis AI (2008) A study on the properties and coagulation behaviour of modified inorganic polymeric coagulant—polyferric silicate sulphate (PFSiS). Sep Purif Technol 63(2):475–483
92. Moussas PA, Tzoupanos ND, Zouboulis AI (2011) Advances in coagulation/flocculation field: Al-and Fe-based composite coagulation reagents. Desalinat Water Treat 33(1–3):140–146
93. Muyibi SA, Okuofu CA (1995) Coagulation of low turbidity surface waters with Moringa oleifera seeds. Int J Environ Stud 48(3–4):263–273
94. Ndabigengesere A, Narasiah KS, Talbot BG (1995) Active agents and mechanism of coagulation of turbid waters using Moringa oleifera. Water Res 29(2):703–710
95. Nwodo UU, Green E, Mabinya LV, Okaiyeto K, Rumbold K, Obi LC, Okoh AI (2014) Bioflocculant production by a consortium of Streptomyces and Cellulomonas species and media optimization via surface response model. Colloids Surf, B 116:257–264
96. Ociński D, Jacukowicz-Sobala I, Kociołek-Balawejder E (2016) Alginate beads containing water treatment residuals for arsenic removal from water—formation and adsorption studies. Environ Sci Pollut Res 23(24):24527–24539
97. Ogunsina BS, Radha C, Indrani D (2011) Quality characteristics of bread and cookies enriched with debittered Moringa oleifera seed flour. Int J Food Sci Nutr 62(2):185–194
98. Oladoja NA (2015) Headway on natural polymeric coagulants in water and wastewater treatment operations. J Water Process Eng 6:174–192
99. Patale V, Pandya J (2012) Mucilage extract of Coccinia indica fruit as coagulant-flocculent for turbid water treatment. Asian J Plant Sci Res 2(4):442–445
100. Pathak PD, Mandavgane SA, Kulkarni BD (2015) Fruit peel waste as a novel low-cost bio adsorbent. Rev Chem Eng 31(4):361–381

101. Peavy HS, Rowe DR, Techobanoglous G (1985) Water quality: defination, characteristics, and perspectives. Environ Eng 11–43
102. Pernitsky DJ, Edzwald JK (2006) Selection of alum and polyaluminum coagulants: principles and applications. J Water Supply Res Technol—AQUA 55(2):121–141
103. Polizzi S, Pira E, Ferrara M, Bugiani M, Papaleo A, Albera R, Palmi S (2002) Neurotoxic effects of aluminium among foundry workers and Alzheimer's disease. Neurotoxicology 23(6):761–774
104. Pritchard M, Craven T, Mkandawire T, Edmondson AS, O'neill JG (2010) A comparison between Moringa oleifera and chemical coagulants in the purification of drinking water–an alternative sustainable solution for developing countries. Phys Chem Earth, Parts A/B/C 35(13–14):798–805
105. Pritchard M, Mkandawire T, Edmondson A, O'neill JG, Kululanga G (2009) Potential of using plant extracts for purification of shallow well water in Malawi. Phys Chem Earth, Parts A/B/C 34(13–16):799–805
106. Qudsieh IY, l-Razi AF, Kabbashi NA, Mirghani MES, Fandi KG, Alam MZ, …, Nasef MM (2008) Preparation and characterization of a new coagulant based on the sago starch biopolymer and its application in water turbidity removal. J Appl Polymer Sci 109(5):3140–3147
107. Raji YO, Abubakar L, Giwa SO, Giwa A (2016) Assessment of coagulation efficiency of okra seed extract for surface water treatment. Int J Sci Eng Res 6(2):1–7
108. Rasteiro MG, Garcia FAP, Ferreira P, Blanco A, Negro C, Antunes E (2008) The use of LDS as a tool to evaluate flocculation mechanisms. Chem Eng Process 47(8):1323–1332
109. Renault F, Sancey B, Badot PM, Crini G (2009) Chitosan for coagulation/flocculation processes–an eco-friendly approach. Eur Polymer J 45(5):1337–1348
110. Rondeau V, Commenges D, Jacqmin-Gadda H, Dartigues JF (2000) Relation between aluminum concentrations in drinking water and Alzheimer's disease: an 8-year follow-up study. Am J Epidemiol 152(1):59–66
111. Rook JJ (1974) Formation of haloforms during chlorination of natural waters
112. Sanghi R, Bhatttacharya B, Singh V (2002) Cassia angustifolia seed gum as an effective natural coagulant for decolourisation of dye solutions. Green Chem 4(3):252–254
113. Sekar M, Sutharesan N, Mashi DA, Shaiful MH, Shazni M, Wei KM, Abdullah MS (2014) Comparative evaluation of antimicrobial properties of red and yellow watermelon seeds. Int J Curr Pharmaceut Res 6(3):35–37
114. Sen AK, Bulusu KR (1962) Effectiveness of nirmali seed as coagulant and coagulant aid. Indian J Environ Health 4:233–244
115. Shah PD, Kavathia S (2015) Development and application of hybrid materials in coagulation and flocculation of wastewater. J Environ Res Develop 9(4):1218–1224
116. Shamsnejati S, Chaibakhsh N, Pendashteh AR, Hayeripour S (2015) Mucilaginous seed of Ocimum basilicum as a natural coagulant for textile wastewater treatment. Ind Crops Prod 69:40–47
117. Sharma BR, Dhuldhoya NC, Merchant UC (2006) Flocculants—an ecofriendly approach. J Polym Environ 14(2):195–202
118. Sher F, Malik A, Liu H (2013) Industrial polymer effluent treatment by chemical coagulation and flocculation. J Environ Chem Eng 1(4):684–689
119. Shilpaa B, Akankshaa K, Girish P (2012) Evaluation of cactus and hyacinth bean peels as natural coagulants. Int J Chem Environ Eng 3(3)
120. Mishra S, Singh S, Srivastava R (2017) Okra seeds: an efficient coagulant. Int J Res Appl Sci Eng 5(VI)
121. Simate GS, Iyuke SE, Ndlovu S, Heydenrych M, Walubita LF (2012) Human health effects of residual carbon nanotubes and traditional water treatment chemicals in drinking water. Environ Int 39(1):38–49
122. Sincero AP, Sincero GA (2002) Physical-chemical treatment of water and wastewater. CRC Press

123. Smoluchowski MV (1917) An experiment on mathematical theorization of coagulation kinetics of the colloidal solutions. Zeitschrift fur physikalisch Chemie 92:129–168
124. Sowmeyan R, Santhosh J, Latha R (2011) Effectiveness of herbs in community water treatment. Int Res J Biochem Bioinf 1(11):297–303
125. Teh CY, Budiman PM, Shak KPY, Wu TY (2016) Recent advancement of coagulation–flocculation and its application in wastewater treatment. Ind Eng Chem Res 55(16):4363–4389
126. Theodoro JP, Lenz GF, Zara RF, Bergamasco R (2013) Coagulants and natural polymers: perspectives for the treatment of water. Plastic Polymer Technol 2(3):55–62
127. Torres LG, Carpinteyro-Urban SL (2012) Use of Prosopis laevigata seed gum and Opuntia ficus-indica mucilage for the treatment of municipal wastewaters by coagulation-flocculation. Nat Resour 3(2):2012
128. Tzoupanos ND, Zouboulis AI (2008) Coagulation-flocculation processes in water/wastewater treatment: the application of new generation of chemical reagents. In: 6th IASME/WSEAS international conference on heat transfer, thermal engineering and environment (HTE'08), August 20th–22nd, Rhodes, Greece, pp 309–317
129. Verwey EJW, Overbeek JTG, Van Nes K (1948) Theory of the stability of lyophobic colloids: the interaction of sol particles having an electric double layer. Elsevier Publishing Company
130. Vijayaraghavan G, Shanthakumar S (2015) Efficacy of alginate extracted from marine brown algae (Sargassum sp.) as a coagulant for removal of direct blue2 dye from aqueous solution. Global Nest J 17(4):716–726
131. Walton JR (2013) Aluminum involvement in the progression of Alzheimer's disease. J Alzheimers Dis 35(1):7–43
132. Wang Q, Yang Z (2016) Industrial water pollution, water environment treatment, and health risks in China. Environ Pollut 218:358–365
133. Wang Y, Gao BY, Yue QY, Wei JC, Zhou WZ (2006) Novel composite flocculent ployferric chloride-polydimethyldiallylammonium chloride (PFC-PDMDAAC): its characterization and flocculation efficiency. Water Pract Technol 1(3)
134. Wettstein A, Aeppli J, Gautschi K, Peters M (1991) Failure to find a relationship between mnestic skills of octogenarians and aluminum in drinking water. Int Arch Occup Environ Health 63(2):97–103
135. Yin CY (2010) Emerging usage of plant-based coagulants for water and wastewater treatment. Process Biochem 45(9):1437–1444
136. Zhu G, Zheng H, Zhang Z, Tshukudu T, Zhang P, Xiang X (2011) Characterization and coagulation–flocculation behavior of polymeric aluminum ferric sulfate (PAFS). Chem Eng J 178:50–59
137. Zouboulis AI, Moussas PA, Tzoupanos ND (2009) Coagulation-flocculation processes applied in water or wastewater treatment. In: Industrial waste: environmental impact, disposal and treatment. Nova Science Publishers, Inc., Hauppauge, pp 289–324

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