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Review Article

BIO-COAGULANTS, A SUBSTITUTE OF CHEMICAL COAGULANTS

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ABSTRACT

Coagulants are substances which cause particles in a liquid to curdle and clot together. Different types of common coagulants such as aluminium sulphate, polyaluminium chloride (PAC) and iron salts, are used in potable water treatment. These coagulants are usually very expensive and can affect human health. These chemical coagulants change the pH of the treated water and also produce non biodegradable sludge. Moreover, the increase of global awareness about environmental issues is acting as a driving force behind the interest toward the use of green resources as valuable products for water treatment. Natural coagulants have the potential to be effective in water treatment which is sustainable and environmentally appropriate. The bio-coagulation process proved to be efficient in turbidity and colour removal. A number of natural coagulants from plants or animal origin can be effectively used for the treatment of drinking water. Many plant materials have been used over the years, the seeds from *Moringa oleifera* have been shown to be one of the most effective coagulants for water treatment. Seeds of *Carica papaya* and leaves of *Cactus opuntia* showed effectiveness in coagulation. This review tries to find out readily available natural products which are cheap, appropriate and sustainable solutions for producing potable water in some developing nations. This paper also includes advantages and disadvantages of natural coagulants prior to identifying several potential research gaps to provide a platform towards the need of further study.

Keywords: Natural Coagulant, Biodegradable sludge, Water treatment, Turbidity, Coagulation, Flocculation.

1. INTRODUCTION

Water is a key substance in all natural and human activities. Humans totally depend on water for their survival. Water regenerates the shape of oceans, seas, rivers, lakes and forests, becoming part of the hydrological cycle that is important for the development of ecosystems and human life [1]. However, the problem of lacking access to quality water has major negative impacts on people's well-being such as; poverty, massive health effects, a shortage of safe drinking water, poor personal hygiene and a problem of sanitation. According to Rijsberman [2], it is estimated that a minimum of 7.5 litres of water is required for individual consumption, personal hygiene and preparing food. Hence; a safe, affordable, reliable and easily accessible, quality water supply is crucial for household activities, good health and agriculture. This will equally improve the individuals' life in terms of poverty reduction and other life comfortability matters. But insufficiencies in water supply affect health adversely both directly and indirectly. The need for clean water had triggered more comprehensive research in water and wastewater. Heavy metals released into environment that is active in development and rapid growth of industrialization that affect bio-network. Industries such as; metal coating, excavation, and battery production, consequence of profound metals to ecosystem [3]. Coagulation and flocculation processes are widely used in water and wastewater treatment. Its main objective is to remove suspended colloidal particles and to reduce turbidity in water the body [4]. The removal of suspended matter from water is one of the major goals of water treatment. Only disinfection is used more often or considered more important. In fact, effective clarification is really necessary for completely reliable disinfection because microorganisms are shielded by particles in the water. Clarification usually involves:coagulation, flocculation, settling, and filtration [5]. It has been discussed that in developing countries more than 1.6 million people are using unhygienic water & among them most of the people suffer from diarrhoea and other water related diseases [6]. It is described that developing countries are paying a high cost for water treatment by importing the chemicals [7]. The ultimate need in today's uprising world is to provide access to clean drinking water by cost effective means, particularly to the rural population who are not capable of affordable water treatment [8]. Chemical coagulants are widely used for turbidity removal in water and wastewater treatment resulting in dangerous health problems.It has been demonstrated in various studies that chemicals (such as aluminum salts, acrylamides, etc.) used in the coagulation-flocculation process remain in treated water and may induce health problems. As indicated above, various health effects such as neurotoxic, carcinogenic, genotoxic and cancerogenic properties were reported [9]. Moreover, artificial polymers and undesirable substances related to them could react with other materials throughout the treatment and make by-products with unknown health effects [10]. Many studies have been reported about the relationship between Alzheimer's disease and redundant aluminum [11-13]. As they are uneconomical, it is difficult to use these coagulants in developing countries. The Recent work aimed at comparative study of turbidity removal efficiency with various types of natural coagulants available [14]. Coagulation-flocculation physicochemical processes play a major role in surface water treatment by reducing turbidity, bacteria, algae, color, organic compounds, and clay particles [15]. Several natural products resulting from seeds, fruits and leaves such as lime seeds [16], pods seeds of tamarind [17], leaves of acorn [18], pads from cactus [19], fruits peels of banana [20] and hyacinth bean [21], have been receiving a great interest as promising coagulant/flocculant alternatives to synthetic reagents owing to their safeness, natural abundance and cost-effectiveness [22]. Some of the plants are able to be a coagulant because they are able to conduct some of the coagulation mechanisms which are neutralizing the charge in colloidal particles and perform polymer bridging [23]. Natural coagulants produce readily biodegradable and less voluminous sludge that amounts only 20-30% that of alum treated counterpart [8]. Natural coagulants must be safe for human health and their surrounding environment. In recent years, numerous studies on a variety of plant materials which can be used as natural coagulants have been reported [24].

2. CONVENTIONAL WATER TREATMENT PROCESS

Surface waters must be treated to remove turbidity, color and bacteria. The object of coagulation (and subsequently flocculation) is to turn the small particles of color, turbidity and bacteria into larger flocs, either as precipitates or suspended particles. These flocs are then conditioned so that they will be readily removed in subsequent processes. Technically, coagulation applies to the removal of colloidal particles. Coagulation is a method to alter the colloids so that they will be able to approach and adhere to each other to form larger floc particles. Generally drinking water treatment protocols consist of two major steps:coagulation/flocculation and disinfection (Fig.1).

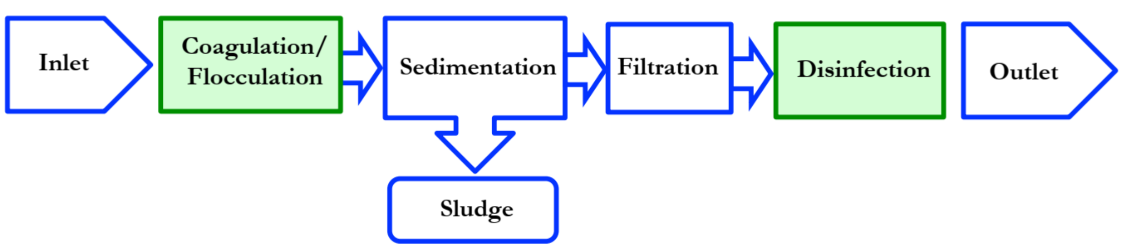


Fig. 1:Schematics over conventional water treatment process [25]

Turbidity is caused by suspended particles and natural organic matter (NOM) present in the water. Colloids are suspended in solution and cannot be removed by sedimentation or filtration. Very simply, the particles in the colloid range are too small to settle in a reasonable time period and too small to be trapped in the pores of a filter. Most colloids are stable because they possess a negative charge that repels other colloid particles before they collide with one another. Coagulants will destabilize the particles by neutralization of the negative charges. Flocculation is the agglomeration of these particles into large size particles known as flocs, which will settle by gravity, i.e. sedimentation. If the water is highly turbid, a flocculant aid is necessary in addition to the coagulant. Turbidity of water is most commonly measured by a turbidimeter and expressed in Nephelometric Turbidity Units (NTU).

3. CHARACTERISTICS OF COAGULANTS

Colloids are stable because of their surface charge. In order to destabilize the particles, we must neutralize this charge. Such neutralization can take place by addition of an ion of opposite charge to the colloid. Since most colloids found in water are negatively charged, the addition of positive charged ions should reduce the charge. It was found by Schulze and Hardy [26] that one mole of a trivalent ion can reduce the charge as much as 30 to 50 moles of a divalent ion and as much as 1,500 to 2,500 moles of a monovalent ion. The purpose of coagulation is to alter the colloids so that they can adhere to each other. During coagulation, a positive ion is added to water to reduce the surface charge to the point where the colloids are not repelled from each other. A coagulant is the substance (chemical) that is added to the water to accomplish coagulation. There are three key properties of a coagulant:

1. The colloids most commonly found in natural waters are negatively charged, hence a cation is required to neutralize the charge. A trivalent cation is the most efficient cation.
2. For the production of safe water, the coagulants must be nontoxic.
3. The coagulant that is added must precipitate out of solution i.e., must be insoluble in the neutral pH range so that high concentrations of the ion are not left in the water. Such precipitation greatly assists the colloid removal process.

Flocculation is the agglomeration of these particles into large size particles known as flocs, which will settle by gravity, i.e. sedimentation (Fig. 2).

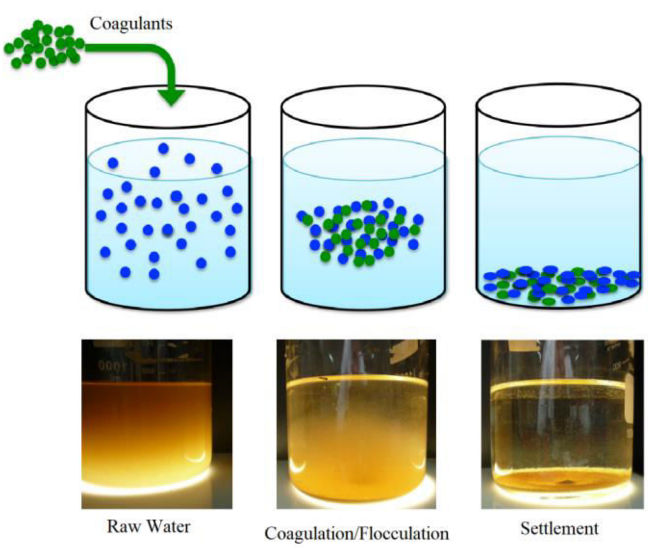


Fig. 2:Coagulation/Flocculation mechanism [25]

One of the most common methods to evaluate coagulation efficiency is to conduct a jar test. Jar tests are performed in an apparatus that is shown in Fig. 3. Jar test was conducted to determine the effective dosage of coagulant to reduce the turbidity of the sample. The standard procedure was 1 min of rapid mixing (120 rpm) followed by 15 minutes of slow mixing (30rpm) for flocculation and 60 minutes of settling.

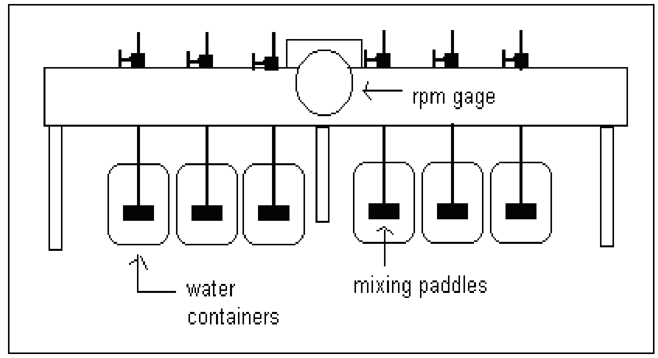


Fig. 3:Jar Test Apparatus

There are several factors which can affect the effective coagulation. The factors are (i) Kind of coagulant, (ii) Quantity of coagulant, (iii) Amount, character of colour and turbidity of water, (iv) pH value of water, (v) Time of mixing and flocculation, (vi) Temperature, (vii) Violence of agitation.

Coagulants are characterised according to the water properties that relate to enhance the efficiency of the treatment process to achieve required quality of water on standards [27]. For an effective coagulation, (i) Turbidity, (ii) Organic and inorganic matter, (iii) Colour, (iv) Harmful and other pathogenic bacteria, (v) Algae, planktons and other organisms, (vi) Taste and odour producing substances are to be removed. It should be noted that microorganisms are not completely removed by coagulation, they are removed via disinfection.

4. INORGANIC AND ORGANIC SYNTHETIC COAGULANTS

At present time, most of the large water treatment plants i.e., pilot scale production of water use these types of coagulants. Some inorganic coagulants are aluminum sulfate, polyaluminum chloride, ferric chloride, polyferric sulfate etc and organic synthetic coagulants are polyacrylamide derivatives and polyethyleneimine etc [28]. These coagulants are very costly, have adverse health effects but these are used due to lack of alternative source.

4.1. Disadvantages of Inorganic and/or Synthetic Coagulants

Chemical coagulants are widely used for turbidity removal in water and wastewater treatment resulting in dangerous health problems. Alum (aluminium sulphate), has been the most popular for treatment of water and widely used in treatment plants. It has been found to pose some health, economic and environmental problems upon usage, among which are neurological diseases such as percentile dementia and induction of Alzheimer's disease [9-13]. It also inhibits bone mineralisation [29]. Sludge produced is also voluminous and non-biodegradable after treatment, leading to increase in cost of treatment. The high cost of chemical importations results in loss of foreign exchange to nations. The effect of most chemical coagulants like Aluminum on the pH of the treated water attracts extra cost on lime which should be added to buffer its effect [30].

To overcome these disadvantages, we need substitution i.e., we have to find an alternative source. Natural coagulants are a very good alternative to chemical coagulants. Here we discuss plant based natural coagulants only.

5. NATURAL COAGULANTS

Because of cost savings, environmental issues of synthetic organic polymers and inorganic chemical products, it is of interest to use natural materials. One of the realistic practices is to replace the chemicals used in the treatment processes with "green" chemicals that cause lesser environmental impacts in terms of production, consumption, and secondary waste management. Numerous review articles have reportedly proved the effectiveness of plant based natural coagulants in various water treatment processes [14-22]. Different natural coagulants like Seeds of *Moringa Oleifera, Cactus Opuntia, Carica papaya* seeds etc have good potential towards coagulation of water.

5.1. Active Component Extraction

The green extract may have different origins, but their extraction procedure is almost the same. The seeds can be extracted by using different types of solvent like water, organic compounds or by using salt solution [31, 32]. Among the different solvents, water is the most popular choice due to its good polarity, availability and cost. But for the extraction of protein, salt is the second most widely used solvent. The active components have been prepared according to Fig. 4. In some cases, since filtrates and powders may enclose other plant tissues rich in inorganic constituents that may increase the organic load in waters, additional purification process is therefore required. This further processing is usually performed via precipitation or a lyophilization or an ion-exchange or dialysis schemes [33].

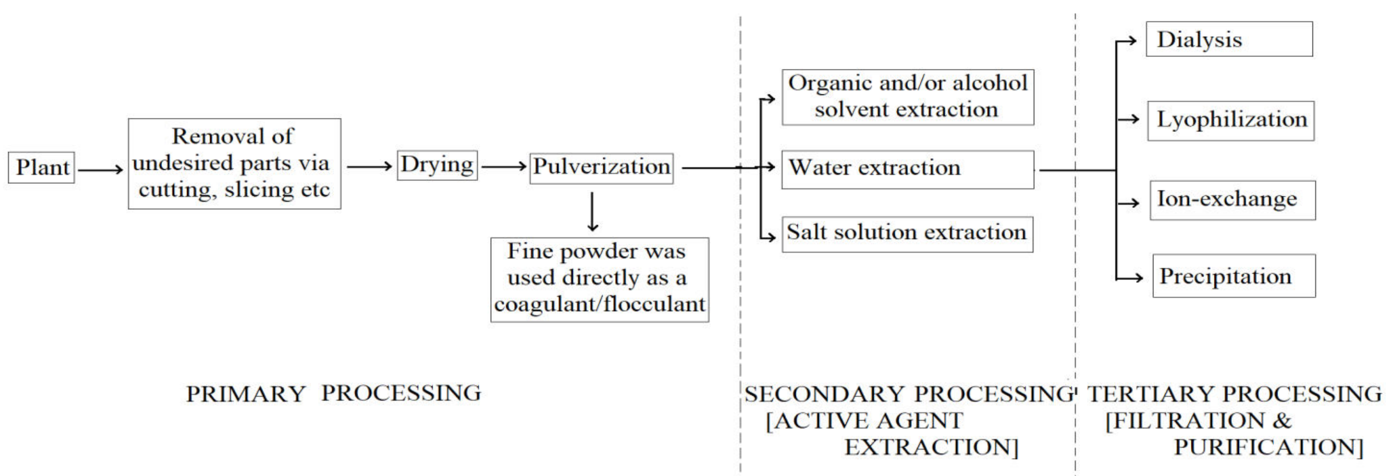


Fig. 4:General processing steps in preparation of plant based coagulants [33]

5.2. Mechanism of Natural Coagulant

There exist mainly four types of coagulation mechanisms. These are double layer compression, polymer bridging, charge neutralization and sweep coagulation. For plant based natural coagulants, only polymer bridging mechanism and charge neutralization mechanism are possible [34]. Long chain polymers [35] when added in small dosage to a suspension of colloidal particles, adsorb onto them in such a manner that an individual chain can become attached to two or more particles thus "bridging" them together (Fig. 5) [36]. The aggregates which are formed by bridging flocculation mechanisms are much stronger than charge neutralization mechanisms.

Raw water generally contains negatively charged colloidal particles. Charge neutralization mechanism uses ionisable polymers (polyelectrolytes) as coagulants to stabilize the colloidal particle. Therefore, polycation is used to stabilize the particles, gaining near to zero zeta potential. The basis of charge neutralization is that the adsorption of high charge density polyelectrolytes by low charge density colloidal particles occurs in a 'patch wise' manner. This is called the electrostatic patch mechanism (Fig. 6), meaning that the particle's surface has patches of positive and negative regions. These regions cause additional attraction between particles given that the opposite charged regions are properly aligned [36].

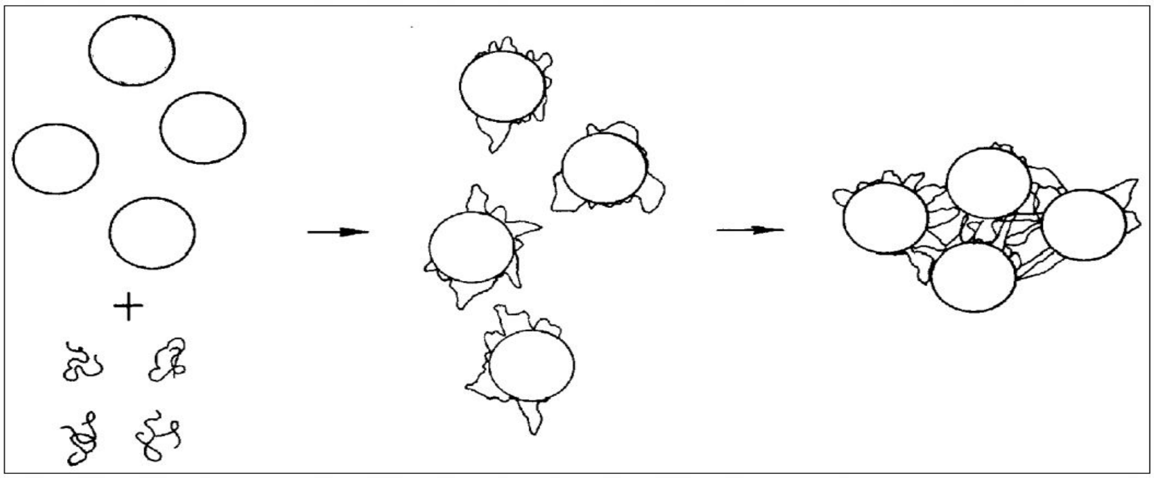


Fig. 5:Schematic illustration of bridging coagulation by adsorbed polymer [36]

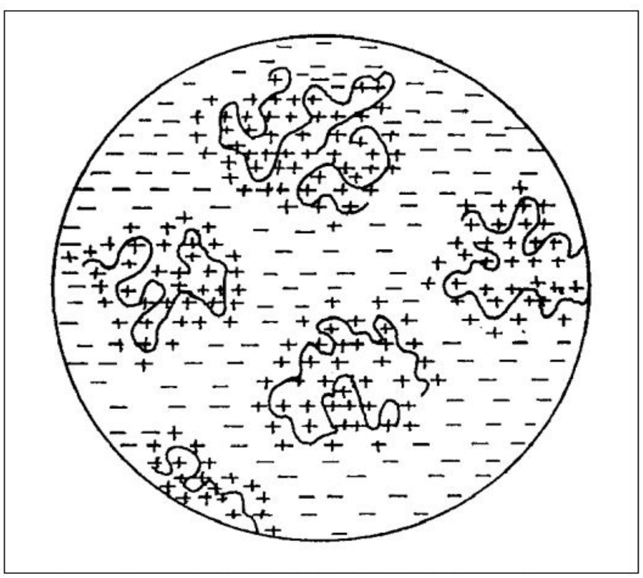


Fig. 6:Schematic illustration of electrostatic patch mechanism for charge neutralization [36]

5.3. Turbidity Control by Natural Coagulants

Coagulation efficiency of the different natural coagulants can be done by using Jar test. This efficiency depends on the nature of plant materials used. The percentage of removal turbidity by natural coagulants is represented in table 1.

The percentage of the turbidity removal is calculated by the following formula:

Prcentage of turbidity removal= $\{(T_i\text{-}T_f)/T_i)\}$ x 100

Where, T_i is the turbidity of the water sample before treatment and T_f is the turbidity of the water sample after treatment.

5.4. Review of some Natural Coagulants in Turbid Water

5.4.1. Seeds of Moringa Oleifera

Moringa oleifera is a tropical plant belonging to the family of Moringaceae. It is a fast-growing, deciduous tree that can reach a height of 32-40 ft and trunk diameter of 1.5 ft. [42]. The bark has a whitish-grey color and is surrounded by thick cork. Young shoots have purplish or greenish-white, hairy bark. The tree has an open crown of drooping, fragile branches, and the leaves build up feathery foliage of tripinnate leaves. Flowering begins within the first six months after planting. In seasonally cool regions, flowering only occurs once a year in late spring and early summer. The flowers are fragrant and hermaphroditic, surrounded by five unequal, thinly veined, yellowish-white petals. The fruit is a hanging, three-sided brown capsule of 20-45 cm size, which holds dark brown, globular seeds with a diameter around 1 cm (Fig. 7).

Table 1:Various types of natural coagulants and their turbidity removal efficiencies

Serial Number	Species Name	Plant part	Turbidity Removal Efficiency	Reference
1	Moringa Oleifera	Seed	99.3%	37
			86%	38
2	Cactus Opuntia	Entire species cut into strips, dried and powdered	99.74%	21
			78.5%	39
3	Carica papaya	Seed	90.29%	40
			96.19%	41

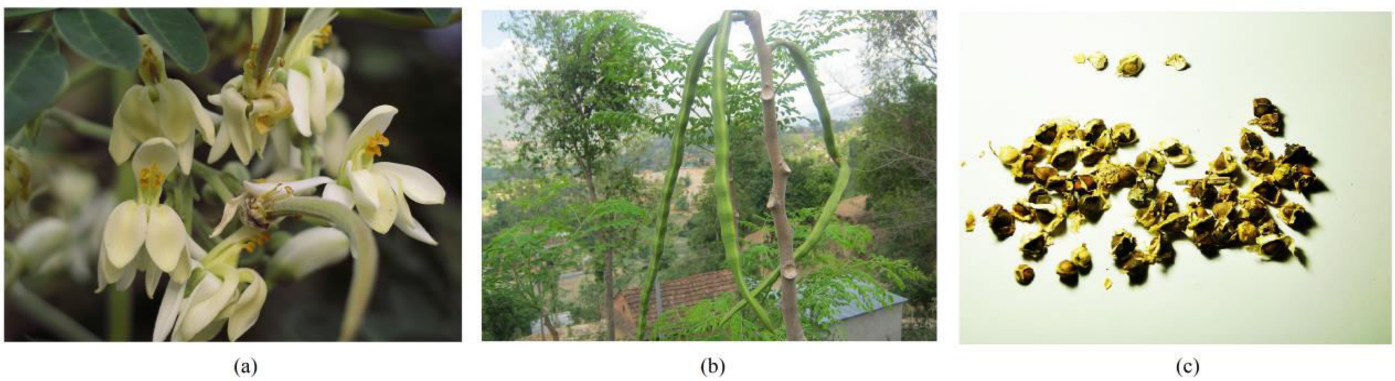


Fig. 7:(a) Moringa Oleifera flowers; (b) Pods of Moringa Oleifera; (c) Moringa Oleifera Seeds

The seeds have three whitish papery wings and are dispersed by wind and water. Its seeds have been shown to be one of the most effective main bio-coagulants for water treatment [37]. *Moringa oleifera* seeds are very useful in the water treatment process both as a coagulant and disinfectant. All samples of *M. oleifera* exhibited moisture levels varying from 5.7 to 8.9%, fat from 34.7 to 40.4%, fiber from 6.8 to 8.0%, ashes from 4.4 to 6.9%, protein from 29.4 to 33.3%, and carbohydrates from 16.5 to 19.8% [43]. Fig.8 represents the proposed flow chart for the extraction of the active ingredient of *moringa oleifera* seeds [44].

The main active ingredient which is responsible for coagulation include a polyelectrolyte. The seeds also have antibacterial activity against both of the gram positive and gram-negative bacteria [45].

The seeds of *Moringa Oleifera* combined with activated carbon derived from coconut shells where these natural materials were mixed in several ratios in a sachet. Activated carbon is commonly known as a water treatment agent and adsorbent. The seeds of *Moringa Oleifera* may act as a natural catalyst as substitute for chemical catalysts. Seeds of *Moringa Oleifera* also show excellent ability as a coagulant agent especially in highly turbid water [46].

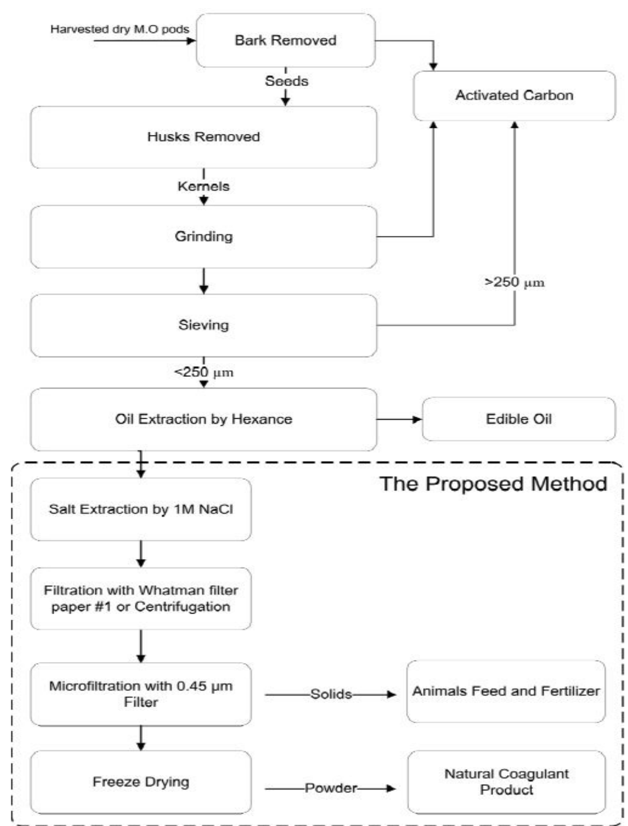


Fig. 8:Proposed production process flow chart [44]

5.4.2. Cactus Opuntia

Opuntia, commonly called prickly pear, is a genus of flowering plants in the cactus family Cactaceae. It is a large, trunk-forming, segmented cactus that may grow to 16-23 ft with a crown of over 10 ft in diameter and a trunk diameter of 1 m (Fig. 9).

Mature stem of opuntia contains approximately 11.2% moisture, 3.33% ash, 11.6% crude protein, 4.4% fat, 4.4% crude fibre and 64% carbohydrate [47]. Opuntia contains carbohydrates such as L-arabinose, D-galactose, L-rhamnose, D-xylose and galacturonic acid. Also report that galacturonic acid was possibly the active ingredient that affords the coagulation capability of Opuntia species though it should be noted that it only accounts for only 50% of turbidity removal. [48]. Opuntia mucilage contains polygalacturonic acid and five sugars. Cactus was used in the treatment of waters, using either the mucilage or the whole cladode powder. Water treatment without Fe/Al synthetic polymers would yield water with less toxicity [22, 49]. Cactus are being considered as alternatives to conventional synthetic chemical coagulants in aspects such as costs, health effects, non-biodegradability, altered pH in post-treatment water and corrosion and transmission problems. In addition, cactus appears to have no significant effects on the pH of treated water [50]. For highly turbid water, Opuntia can reduce the turbidity upto 89-93% and also it has colour reducing property [51]. If lime is combined with opuntia, the removal efficiency for turbidity and suspended solids reached over 95% while that for color was between 67% and 94% [50]. Daza et al. [52] reported that for cactus opuntia species, turbidity removal percentage was 88.56% and colour removal percentage was 97.67%.

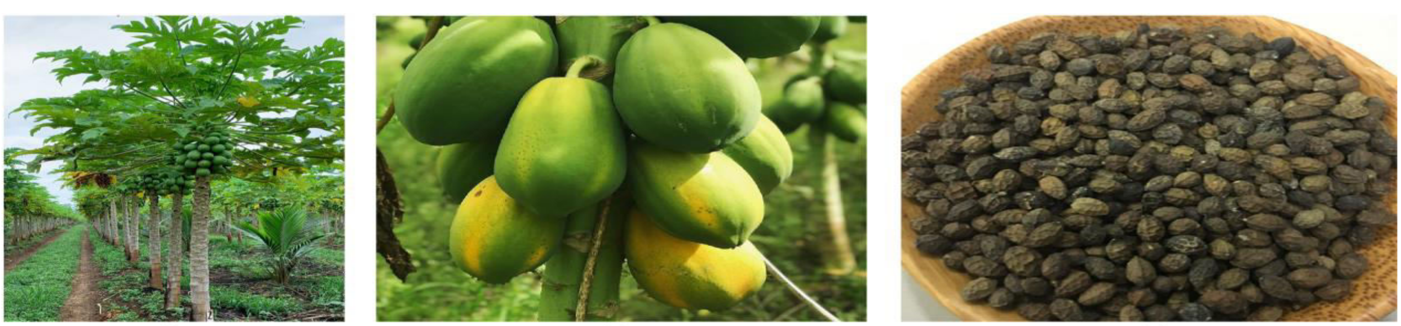
5.4.3. Carica papaya seeds

The papaya plant is considered a tree, though its palmlike trunk, up to 30 feet tall, is not as woody as the designation generally implies. The plant is crowned by deeply lobed leaves, sometimes 2 feet across, borne on hollow petioles (leaf stalks) 2 feet long. The life of a leaf is 4 to 6 months. Both the stem and leaves contain copious white milky latex [53].

Generally, the fruit is melon-like, oval to nearly round, somewhat pyriform, or elongated club-shaped, 6 to 20 inches long and 4 to 8 inches thick; weighing up to 9 kg [53]. It is a berry of thin skin, of a green yellowish and orange colour. The pulp is of a red, orange or yellow colour, sweet and very juicy. Papaya fruit contain a large number of small black color seeds. The seeds are about 5 mm long, each coated with a transparent, gelatinous aril (Fig. 10).

(a) (b) (c)

Fig. 9:(a) Opuntia flowers; (b) Opuntia plant; (c) puntia fruits



(a) (b) (c)

Fig. 10:(a) Papaya trees; (b) Papaya fruits; (c) Papaya seeds

The fruit and seeds contain large amounts of protein. Papaya seeds have anti-inflammatory properties, wound healing properties, suitable for digestion, prevention of cancer and kidney disorders, provide heart health and its use increase immunity because it contains vitamin A & C. Papaya seed is a rich source of proteins. *Carica papaya* seeds have high value of proteins and these proteins are the active coagulating agent for the treatment of raw water [40]. *Carica papaya* seeds of a tropical tree comprise water-soluble and positively charged protein known as cystine protease which emerged as a putative coagulant in both water and wastewater treatments. It is indispensable to optimize process variables such as pH, turbidity, total dissolved solids (TDS), *E. coli* and coliform counts accuracy to raise the efficiency of coagulation operation via employing *C. papaya* [54]. Fig.11 represents the proposed flow chart for the extraction of active ingredients of *Carica papaya* seeds.

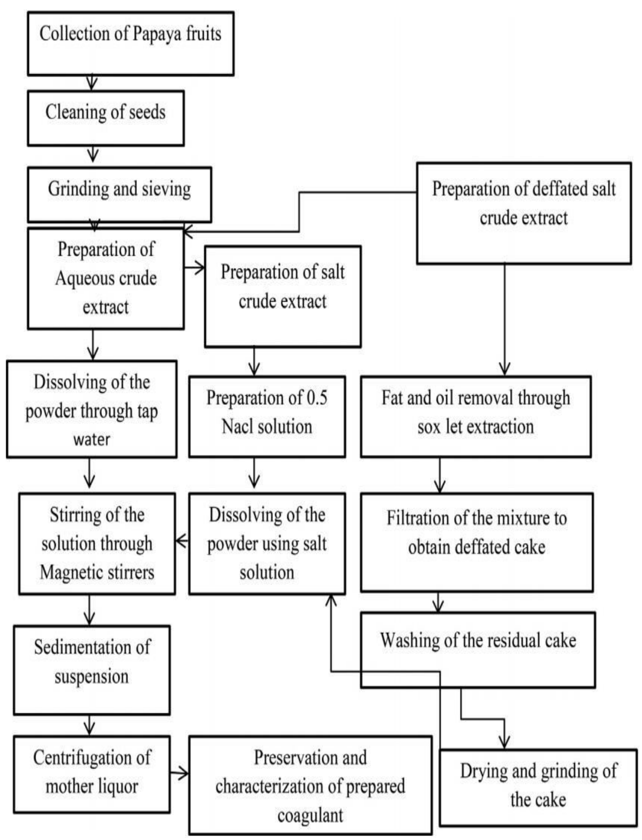


Fig. 11:Sequential coagulant extraction flowchart for Papaya seed extract [41]

Seed works as a coagulant due to the presence of positively charged proteins which bind with negatively charged particles (silt, clay, bacteria and toxins etc), allowing the resulting flocs to settle and obtain clear water (adsorption & charge neutralization) [40]. The seeds have turbidity removal efficiency from raw water upto 96.19% [41].

6. ADVANTAGES AND DISADVANTAGES OF NATURAL COAGULANTS FOR COMMERCIAL USE

Chemical coagulants are widely used for turbidity removal in water and wastewater treatment resulting in dangerous health problems.Various health effects such as neurotoxic, carcinogenic, genotoxic and cancerogenic properties were reported [9]. But natural coagulants are eco-friendly, non-toxic, non-hazardous, non-corrosive and therefore are safe to use [4, 55-56]. Natural coagulants produce readily biodegradable and less voluminous sludge [8]. Due to cost effectiveness, a number of developing countries have tendencies to use natural coagulants. Effective water treatment is beneficial to several individuals, governments and organizations. But there are some constraints to use natural coagulants in large scale water production for commercial use. The different types of challenges of the acceptance of natural coagulants by the water industry are summarized in Fig.12.

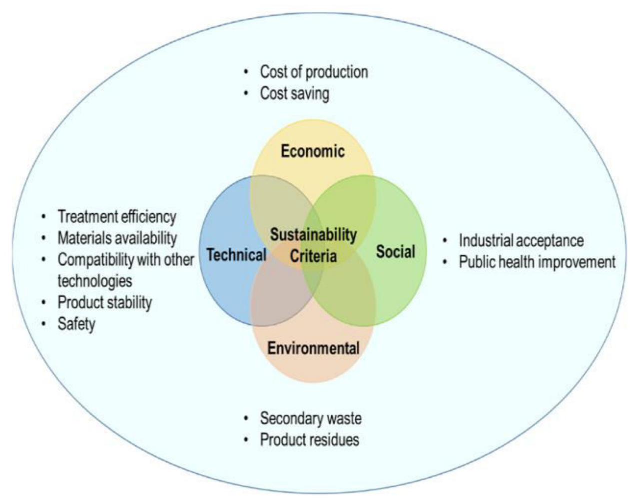


Fig. 12:Sustainability criteria commonly considered for water and wastewater treatment process [57]

There must have to maintain the steady supply of raw materials to coagulant production from their renewable sources. Therefore, it requires a lot of available land for cultivation of plants and also requires processing and storage which can add to the cost [8]. Abaliwano *et al.* [58] reported that in the crude coagulant preparations that are prepared from pre-processed materials will release unwanted carbon loads into treated waters, resulting in undesirable microbial growth. Breakthrough in acceptability by potential investors for plant-based coagulants as a new product is another challenge. Lack of regulatory approvals on plant-based coagulants is another challenge faced in commercializing this technology.

7. FUTURE STUDY

Further study is required for the complete knowledge of co-flocculation, co-precipitation, agglomeration etc involved in the turbidity removal steps by the use of natural coagulants. Therefore, SEM, FTIR etc studies are required to know the appropriate mechanism involved in coagulation/flocculation mechanism.

8. CONCLUSION

The high cost of water treatment has been overcome by the eco-friendly natural coagulants. Likewise, many significant problems posed by chemical coagulants have been overcome by these coagulants. Plant-based coagulants, mostly tested in extract forms, are excellent substitutes for conventional chemicals and produce WHO acceptable drinking waters. This review revealed that natural coagulants have the efficiency of being an alternative or a supplement to aluminium sulphate or other proprietary polyelectrolyte and can save cost. Outcomes gained from the present review reveal the efficacy of the three screened natural products (*Moringa oleifera* seeds, *Cactus opuntia* and *Carica papaya* seeds) towards the treatment of raw water. Among them, *Moringa oliefera* seeds could be employed at industrial scale because of its efficacy in water treatment.

9. ACKNOWLEDGEMENTS

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Conflict of Interest

There is no conflict of interest in submission of this manuscript.

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