NATURAL COAGULANTS: ANALYSIS OF POTENTIAL USE FOR DRINKING WATER TREATMENT IN DEVELOPED AND DEVELOPING COUNTRIES

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Abstract:

The water intended for human consumption must be characterized by being colorless, odorless and tasteless and free of substances or micro-organisms that can cause disease. The process that allows obtaining this quality water is the purification, understood as the treatment of the water to make it suitable for human consumption. The purification is traditionally composed of a sequence of standard treatments including filtration, coagulation-flocculation, settling and disinfection. Different compounds named coagulants are used for coagulation-flocculation, which are able of reducing the suspended matter and clarify the water. These coagulants are synthetic in nature, therefore represent a high cost and are difficult to access in some areas, especially in developing countries. In addition, their use is being revised and restricted in different international standards, in order to control or prohibit its use because of the possible toxic and hazard effects that the remains of these products in treated water can cause to people. This paper shows natural coagulants as an alternative to chemical coagulants, conducting a comprehensive review of the most researched and their potential application in the treatment of drinking water.

Keywords: *drinking water, natural coagulants, water purification*

1. Introduction

Water is essential for the quality of life and the welfare of the human being. However, its consumption will be safe for people if it is drinkable, i.e., odourless, colourless and tasteless, and free of substances that could damage the health of consumers. Therefore, it is required to apply a certain sequence of purification treatments, which depend on water source that will produce water with a quality suitable for drinking.

Water purification consists in the application of the physical, chemical and/or microbiological treatments that allow obtaining water with good quality so it can be consumed by people without risk to their health. The result should be a drinking water that meets the quality requirements established by different regulations such as the existing European Directive 98/83/EC [1], regulation "Safe Drinking Water Act" of the American Environmental Protection Agency [2] or international recommendations such as the ones of the World Health Organization [3].

The sequence of purification treatments to apply will depend on the physico-chemical and microbiological quality of the water source, on the resources and infrastructure available for the treatment, on its cost, and on the type of supply and distribution of the final treated water.

The basic conventional purification treatment includes clarification of the water after intake through the stages of coagulation-flocculation, sedimentation, filtration and disinfection, as shown in Figure 1.

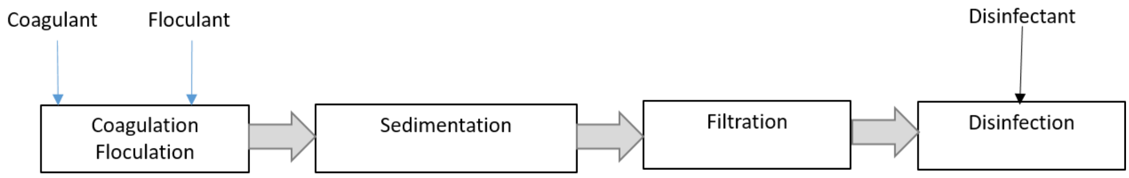

Figure 1. Treatment stages for water purification

Coagulation-flocculation aims to increase the size of suspended and dissolved particles in the water, helping to its separation through filtration or sedimentation. This stage requires the addition of coagulants and flocculants whose aim is, on the one hand, to neutralize the electrostatic charge of suspension solids to favor its aggregation, and, on the other hand, to increase the size of flocs formed in order to increase its sedimentation rate. The origin of these coagulants and flocculants can be synthetic or natural as it will be explain later. Occasionally, suspended solids, and in general water turbidity, masks the presence of microbiological contamination (viruses and bacteria) so coagulation-flocculation also reduces it. When suspended solids sediment, they drag with them much of microorganisms associated with the particles responsible for turbidity.

Once solids have settled, water is filtered through porous media (sand or a mixture of sand and anthracite, for example) in order to retain those flocs that have remained in suspension. Finally, water is disinfected (typically with chemicals such as chlorine) which removes pathogenic microorganisms from water.

The water thus treated is suitable for human consumption and can be distributed to final users. Occasionally, rechlorination dosage points along the water distribution network are installed to maintain its residual effect in the case of external water infiltrations which can contaminate treated water.

2. Water purification in developed and developing countries

Although technically the water purification is a resolved issue, today it remains being a major challenge for various reasons.

In developed countries, with appropriate technology and infrastructures to carry out the treatment, the challenge arises because resources are scarce and with poor quality. This shortage forces the use of water sources that had not been used in first stay, which requires more in-depth treatments to achieve the necessary quality. In addition, increased pollution of surface water and groundwater by industrial growth and agricultural activity, and the development of more accurate analytical techniques [4] have allowed detecting the so-called emerging contaminants. These contaminants, such as drugs and products of personal hygiene, hormones, pesticides, endocrine disruptors, or polychlorinated biphenyls, among others, cause serious effects on the environment but can also affect human beings due to their accumulation in the environment, and the conventional treatments of drinking water and wastewater are not able to eliminate them [5]. This fact represents a real challenge for researchers, which are developing and studying techniques that remove emerging contaminants from water and ensure that it is suitable for consumption.

On the other hand, in areas with limited economic resources and where access to technology is difficult, there is still population that has no access to drinking water. In the year 2000, United Nations raised the Millennium Development Goals [6], within which was "halving by 2015 the proportion of people without access to safe drinking water and basic sanitation services". The goal was to achieve access to improved water sources, which did not imply necessarily drinkable water resources. In 2010, this goal was achieved globally [7], however there are still 748 million people without access to safe drinking water, located in rural areas and in developing countries. Therefore, in 2015, raised Sustainable Development Goals [8] which intend, by the year 2030, improve water quality by reducing pollution, eliminating illegal discharges and minimizing the discharge of products and hazardous materials into the water, reducing the discharge of untreated wastewater and increasing recycling and reuse globally.

The access to drinking water in these areas is still a challenge due to political, social, economic and also technical causes. This makes conventional purification technologies not feasible or appropriate, due to, among other reasons, its high cost of investment and management, and to the maintenance that require, in addition to the need of qualified personnel for its operation and to the use of chemicals to their exploitation [9, 10]. Precisely this last feature represents a limiting factor in the application of conventional water treatment technology, since chemical reagents can achieve an economic considerable value if they are not produced locally, as they must be imported and paid in foreign currency, increasing their cost significantly.

In addition, in are rural areas where the population is dispersed, centralized drinking water purification systems can be not conveniently implemented due to lack of infrastructure suitable for water treatment and distribution, or due to the existence of obsolete systems, undersized poor maintained.

This circumstance forces people to directly consume surface water or groundwater without any treatment, or to implement decentralized systems for the treatment of the water both at the point of use (POU), usually at household level, and at a small scale (SSS) for small tows or villages [11].

Some of these POU systems are:

- *Boiling*: based on the use of heat at 100 ° C to inactivate pathogenic microorganisms present in the water.

- *Chlorination*: based on the addition of chlorine or its derivatives to inactivate or eliminate pathogenic microorganisms present in the water.

- *Filtration*: used to remove turbidity, cysts and protozoa from water, although it is not effective to remove bacteria or viruses. It is a simple and economic method that covers a broad spectrum of materials, from porous ceramic filters to textile, paper or fiber ones, which facilitates its application at household level.

- *Clarification with natural coagulants*: coagulation-flocculation with natural products extracted from seeds and plants that can be obtained locally allows reducing turbidity of the water and improves its physico-chemical quality.

Studies carried out in relation to water treatment and health had underestimated the potential of the application and the effectiveness of treatments at household level for decreasing the incidence of diarrhea. However, studies performed by some authors [12] have estimated that between 30-40% of cases of diarrhea can be avoided using only techniques that improve the quality of water at domestic POU, making this treatment more effective than improvements in the conventional system of purification in developing countries [13, 14].

Research and development carried out in recent decades, has resulted in innovative, effective, sustainable, economic, reliable and user-friendly technologies [15], which can be applied both in the POU and SSS, and provide drinking water to areas of scarce resources and limited infrastructure. Some of them are:

- *SODIS*: a simple technology that uses solar radiation to inactivate and destroy pathogenic microorganisms present in the water. It basically consists of exposing transparent plastic bottles filled with water to the sun for about six hours.

- *Pressure filtration systems*: based on membrane technology, such as micro and ultrafiltration. They use semipermeable polymeric membranes with a pore size able to retain macromolecules, bacteria and some viruses, improving the physical-chemical and microbiological quality of the water. In addition, these systems are characterized by its modularity, simplicity, adaptability to the pollutant and ease of handling. Its main advantage is that they allow obtaining high quality water and can be applied to the community supply as they can produce large volumes of water. Some examples of SSS based on ultrafiltration which have been implemented successfully at commercial level are: Aquapot [16], Aqualogix® [17], SkyHydrant®, AquaBoy® and SkyStation® [18] or Mobil watermaker [19].

- *Compact filtration and multilayer disinfection systems*: such as the marketed under the commercial names of Lifesaver® or LifeStraw® which are based on multilayer filters, which include meshes of different sizes, and active carbon filters or soaked in iodine which improve the physical-chemical and microbiological quality of the water. The LifeSaver®, for example, has an ultrafiltration membrane and an activated carbon filter, so it reduces the presence of chemical wastes such as pesticides, endocrine disruptors and heavy metals.

- *Clarification with natural coagulants*: systems on a small scale that prepare and condition natural coagulant previously to its addition to the water.

Thus, it is clear the need for further research in technologies that will allow universal access to drinking water, as well as adapt the existing ones to achieve greater efficiency, sustainability and safety. One of the most popular trends in recent years in this context is the use of natural coagulants, both for conventional and alternative water purification treatments. Description and review of this type of coagulants are subjects of the next section.

3. Coagulants and flocculants for water purification

The purification sequence includes a stage of coagulation-flocculation. As mentioned above, to carry out this stage the use of substances known as coagulants and flocculants is required, which have to be able to reduce the content of suspended matter and turbidity, clarifying the water. Coagulants can be classified in the two types:

- Inorganic coagulants
Belong to this group simple aluminum salts (such as alumina sulphate or sodium aluminate) or polymerized ones (polyaluminum chloride), salts of iron (such as sulphate or ferric chloride) or also the lime. Among all of them, the ones more frequently used are the polybases of aluminum also known as polyaluminium chloride (PAC). This includes all the coagulants produced by partial neutralization of a solution of aluminium chloride with basic solutions. These coagulants are characterized by: wide operation pH ranges, good sedimentation rate, with compact and easy-to-sediment flocs, and lower concentration of residual aluminum in treated water, with respect to other aluminum salts [20].

- Organic coagulants
This group consists of polymeric organic molecules, with high molecular weight, cationic, anionic and non-ionic nature, being the cationic ones the most frequently used. They can be natural or synthetic. The natural ones include microbiological origin polymers secreted by certain microorganisms and biological polymers extracted from plants, algae, or animals [21]. Synthetic coagulants are also known as polyelectrolytes, and are chains of monomer units that can contain only one kind of monomer in its molecular configuration, or up to two or three types of subunits, linked in linear or branched configuration. Some examples are the polyacrylamide or polyamines. In comparison to natural polymers, synthetic ones offer the advantages of having higher purity, more stable quality, and greater efficiency forming large and tenacious flocs with the addition of low amounts of substance.

- Flocculants

Occasionally, the coagulation process requires the addition of flocculants that increase the size of the formed floc and let it sediment faster. Flocculants are classified into natural and synthetic, and they can be:

- inorganic: products that act as support material and facilitate the agglomeration of the flocs. They form colloids of the same load than the particles to flocculate, and increase chances of collision between particles when they are added to the dispersion. Some examples are: quicklime, activated silica, clays (bentonite), salts of calcium carbonate or activated charcoal.
- organic: can be natural or synthetic. The natural ones are extracted from plant or animal matter as, for example, alginates, starches, gums, pectins, or xanthates. The synthetic flocculants are large macromolecular chains obtained by association of synthetic monomers, some of whom have charged or potentially ionizable groups. Some examples are anionic and non-ionic polyelectrolytes such as polyacrylamide, or partially hydrolyzed polyacrylamide.

3.1 Natural coagulants

Natural coagulants are defined as water soluble substances, coming from vegetable or animal materials [22, 23, 24, 25]. They act similarly to synthetic coagulants, agglomerating particles contained in the raw water, facilitating its sedimentation and reducing water turbidity. Some of these coagulants have also antimicrobial properties, so they also reduce or remove the content of pathogenic microorganisms that can cause diseases.

Developing countries have traditionally used techniques of clarification with natural coagulants to remove turbidity of the water in the domestic environment [26, 27, 28]. These techniques have been used since more than 2000 years ago by ancient civilizations and there are references in ancient texts of the use of plants and their derivatives for the treatment of water [29]. Already then, certain seeds and gems were believed to have "magical powers" with coagulant action. These properties can be explained by the presence of water soluble species containing di or trivalent cations [30]. Although the use of aluminum has been known since Hellenistic times, its use for water clarification was mentioned for the first time in China in the 17th century. However, already in those times, people with limited resources who did not have access to drinking water, used collagen material from bones of animals, apricot kernels (*Prunus armeniaca*), peach kernels (*Prunus persica*) or clarifying nuts (*Strychnos potatorum*) to clarify the raw water [31]. Despite this, it was a chemical compound, aluminium, which was chosen as coagulant to apply in emerging systems of treatment of drinking water at the beginning of the 19th century, probably due to the availability of the product at that time.

The main advantages of these coagulants are: local availability, origin from natural and renewable resources, ancient use which ensures safety for being human, and biodegradability of produced sludge that can be also used in agriculture. Its application not only can be directed to developing countries, but also to conventional purification and wastewater treatment [20]. Currently, the use of synthetic inorganic coagulants for water purification, such as aluminum sulphate, ferric chloride [32, 33, 34] or organic poly-electrolytes such as polyacrylamide [20], is being also questioned due to, among other reasons:

- environmental problems [35, 36], mainly due to generation of toxic sludge that cannot be used in agriculture;
- possible relationship with Alzheimer's disease [37, 38, 39];
- worsening of neurodegenerative diseases such as senile dementia [40, 41];
- possible relationship with cancer [42, 43].

In fact, some reports, linked for example the presence of residual concentrations of aluminum in drinking water with senile dementia, or the presence of monomers waste arising from the use of organic polymers (especially the cationic) with possible toxic effects to humans and the environment [20]. Some countries such as Japan or Switzerland have banned the use of organic polyelectrolyte in the treatment of drinking water [44], and others such as Germany and France have established strict limits for their use due to its potential toxicity and their high impact on aquatic organisms such as fish or algae [20].

It has to bear in mind that monomers are more toxic than polymers [45], which has led to adopt strict regulations in terms of the proportion of free monomer present in treated water, and in terms of the kind of polyelectrolytes to use, especially with products derived from acrylamide. The maximum free acrylamide content is limited to 0,0025% and the residue in drinking water about 0,5 micrograms per litre. In the case of Spain, until the year 2005, SCO-3719-2005 Ministerial Order limited in 0,02% the content of monomer free of acrylamide for cationic, ionic and non-ionic polyacrylamide, and the dose of active ingredient added to the water in 0,02 mg/L, as an average value, and 0,05 mg/L, as maximum concentration. From 2009, the use of polyacrylamide is completely prohibited. In the case of p-DADMAC, the monomer content limit is 0,5% in Europe, being 2% in United States. In Spain, the concentration should not exceed 10 mg/L of active principle (according to the order SCO/3719/2005), subsequently, the 2009 Order established that the parametric value (PV) of chloride must not exceed the concentration established in RD 140/2003.

For the production of drinking water, the NSF (*National Sanitation Foundation*) of USA has recommended a maximum dose for the most commonly used commercial polymers not exceeding 50 mg/L for p-DADMAC and 1 mg/L for any type of polyacrylamide.

In consequence, there is a global interest and a growing need to investigate the use of coagulants substitutes of current ones, which had been used for centuries, and are safer for humans and the environment.

3.1.1. *Traditional natural coagulants*

In 1988, Jahn published a list of natural coagulants with vegetable origin that had traditionally been used in sub-Saharan Africa, the India and South America. Among them, there were the seeds of almond, apricot and peach, very used in Egypt, Sudan's North or south of Tunisia; *Cactus Opuntia*, commonly used in native American tribes of Peru and Chile; legumes (various species of the genus *Phaseolus*), peas (*Pisum*), lentils (*Lens*), nuts (*Arachis*) or beans (*Vicia*), used by women in the rural area of the Sudanese Nile Valley; and seeds of guar (*Cyamopsis*, *Lens*, *Phaseolus* and *Cajanus* genus) applied in the mining industry in Africa or in water supply in the India. Some of them have been studied in greater depth, providing positive results in relation to its coagulant activity.

However, in view of the increasing importance of the use of natural, biodegradable and no toxic substances in the treatment of drinking water, research has increased in order to find new species with primary coagulant properties and to implement techniques for their application. The following are the species of natural coagulants which have been under research:

- *Strychnos potatorum*
- *Moringa oleifera*
- Okra
- Cassava
- Rice
- Starch
- *Cactus Latifaria* y *Prosopis juliflora*
- Tannins of Walonia
- Tamarind (*Tamarindus indica*)
- *Samanea saman*
- Seaweed
- White beans
- *Cactus opuntia*
- *Tuna Opuntia Cochinelifera*
- Sweet corn (*Zea mays*)
- *Vigna Unguiculata* and *Parkinsonia Aculeata*
- Peanut [46](Birina, Hammad, Desa y Muda, 2013)

Among all these coagulants, chitosan, starch and alginates are the most extended, authorized for use in the treatment of drinking water and used today, so it will be described in greater detail below.

A) Chitosan

Chitosan is the natural coagulant of animal origin most commonly used today. This coagulant is a deacetylated derivative of chitin that is located in the shell of mollusks, the exoskeleton of arthropods, and the cell wall of fungi, mushrooms and yeast.

Currently, it can be commercially found manufactured from the exoskeletons of crabs and prawns in countries such as Japan, China, Taiwan, the India and United States, where fishing and seafood processing are the main industries. Its cost of production is $2 (US $) per kg [22]. The commercial product has an average molecular weight close to 106, and a pH-dependent charge density. Its chemical structure is very similar to the cellulose. While cellulose is a polymer of D-glucose, chitosan is a polymer of glucosamine with a group NH_2 replacing the OH group on carbon 2 of d-glucose. It has been described as a cationic and non-toxic biodegradable polyelectrolyte, capable of eliminating up to 99% of the raw water turbidity [47]. Its use was authorized by the American Agency for Environmental Protection (USEPA) in 1981 for the treatment of drinking water up to a dose of 10 ppm.

Studies of the use of chitosan, alone or in combination with chemical coagulants such as aluminum or ferric chloride, have been widely documented since three decades ago [48, 49, 22]. Recent studies indicate its ability to remove organic matter [50] and its enormous potential for application alone or in combination with filtration sand bed for the treatment of textile industries effluents, effluents with high content of bentonite or kaolinitic clay, wastewater from agro-alimentary industries, removal of heavy metals, fats or phosphorus from water, and in sludge conditioning [51].

B) Starch

Starch is a natural polymeric coagulant occurring in two associated ways: amylose, which is a linear polymer soluble in water, and amylopectin, which is highly branched and is insoluble in water. Amylose is considered as the most effective variant of starch flocculant agent. Its use is documented in clarification of clays, wastewater effluents or as de-emulsifying agent of wastewater with high content of oil [20]. In Spain, it is recognized as a substance for the treatment of drinking water for human consumption, as it is reflected in the RD 140/2003.

C) Sodium alginate¡Error! Marcador no definido.

Sodium alginate is an extract from brown algae, widely used as an additive in food products such as ice cream, sauces and dairy derivatives. Its use in water treatment is documented since more than 40 years in Japan. Its use in drinking water was authorised by the Japanese Association of Water Treatment, which also set the standards of quality of this substance.

It is an anionic polymer resulting from the combination of alginic acid and sodium hydroxide or sodium carbonate. Major advantages include: cost; absence of adverse health effects, allowing it to be used in the treatment of drinking water; ability to dehydrate sludge; and efficiency as coagulant, as it increases the size and weight of the flocs formed with aluminum, allowing also savings in chemical products since it is very effective in small doses [22]. Like starch derivatives, it is a substance recognized as suitable for the treatment of drinking water in Spain, as it is reflected in the RD 140/2003.

3.1.2. Moringa oleifera as natural coagulant ***¡Error! Marcador no definido.***

Among all the natural coagulants indicated, the primary coagulant of vegetable origin more researched today because of its enormous potential is *Moringa oleifera* seed.

Moringaceae is the name of the family of plants that includes fourteen known species, endemic of African countries, Madagascar, Arabia and the India. Half of these species, namely *M. oleifera*, *M. peregrina*, *M. stenopetala*, *M. longituba*, *M. drouhardii*, *M. ovalifolia* y *M. concanensis*, are relatively common, although occasionally cultivated. All species of Moringa have coagulating ability [52, 26], although only the *Moringa oleifera* is cultivated throughout the tropical area.

Moringa oleifera was originally a tree native to the sub-Himalaya regions of India, which was moved to Sudan and planted on the banks of the River Nile, in public parks and gardens, with ornamental purposes. The tree (which can be seen in Figure 2) was extended to the American continents through the French West Indies from Cuba and Jamaica [53]. Its cultivation was spread not only in Central America but in such remote areas of the American continent as California, Brazil and Paraguay.

Figure 2. ***Moringa oleifera*** **tree**

This species has a great interest, due to its multiple uses and properties, which are described below.

A) Oil

The seeds of *Moringa oleifera* contain between 33-41% (w/w) of oil [54]. Profiles of its fatty reveals 73% oleic acid, a high content of tocopherols, and a high ratio of monounsaturated fatty acids versus saturated, what approaches its quality to olive oil and recommends its use as a cooking oil for human consumption [55].

It is also known as "Behen" oil or "Ben" by its content of behenic acid (docosanoic), it has a high resistance to oxidative degradation and numerous applications: cooking, soap and cosmetics manufacturing, fuel for heating and lighting, medicinal uses in rheumatism and gout, lubricant in watchmaking and precision machinery [56, 57]. Currently, *Moringa oleifera* oil is also being researched as a biofuel [58, 59].

B) Vegetable foodstuff and medicinal use

The pods, leaves and seeds of the plant (see Figure 3) are a complete vegetable food with a high nutritional power, making it an excellent product for those areas where the food supply is scarce. The leaves contain 27% of protein and significant amounts of calcium, iron and phosphorus, as well as vitamins A, B and C. The tender pods are cooked or used as condiment in the elaboration of some traditional dishes. It is also used as an additive to enrich commercial products, given its medicinal properties as well as its excellent nutritional properties [60].

All parts of the plant are used in a wide variety of traditional medicines, by its anti-inflammatory, antipyretic, antiepileptic, hepatoprotective, antihypertensive and antitumoral properties [61], and for the control of diabetes [60] properties. The ground seed is used as an ointment to treat common bacterial infections of the skin. Traditionally, in Philippines nursing women use leaves in the preparation of soups to promote the production of breast milk [61]. It has been used traditionally to treat rheumatism, poisonous bites and as a stimulant of the heart and circulatory system. Leaves, with high content of vitamins A and C, are considered useful in catarrhal illnesses; they possess purgative properties, promote digestion and are used as an external application to wounds. The flowers of the plants have stimulant, aphrodisiac and diuretic properties.

Figure 3. Detail of the dried husks and the seeds of *Moringa oleifera*

C) Desinfectant: antibacterial, fungicides and insecticides properties

The roots and flowers of Moringa also have antibacterial activity [62] due to its huge proportion of antimicrobial agents. They contain, among others, an antibiotic active substance, "pterygospermin", which has a significant fungicide and antibacterial effect [63].

Less known are the antifungal properties of Moringa, which are present both in the seeds and the essential oil extracted from the leaves, against species of fungi such as *Trichophyton rubrum*, *Trichophyton mentagrophytes*, *Epidermophyton Xoccosum*, *Microsporum canis* [64], *Fusarium solani* or *Rhizopus solani* [65].

Recently, the insecticidal properties of seeds against the *Aedes aegypti* mosquito have been researched [66].

D) Water treatment

The best-known use of the seed of *Moringa oleifera* is the treatment of water, thanks to coagulants, antibiotic [67, 68] and antifungal properties [64].

The extracts from seeds and leaves have antibacterial properties [68, 69] and are able to coagulate and eliminate gram positive and negative bacteria from water by sedimentation [70] such as *Escherichia coli*, *Pseudomonas aeruginosa* [71], *Staphylococcus aureus* [72], *Streptococcus pyogenes* [73] or *Salmonella typhi* [75], reaching rates of reduction of bacteria of 1-4 LRV (90-99, 99%). Sometimes, they also act directly on the growth of microorganisms, by inhibiting it. It is believed that antimicrobial peptides present in the extracts work destroying the cell membrane or by inhibiting key enzymes [76, 73]. Sutherland et al. (1990) also found that Moringa seeds can inhibit the replication of Bacteriophage [74]. The antimicrobial effects of seeds are specifically attributed to the compound 4(α-L-rhamnosiloxi)-bencil-isotiocianate [67].

The traditional method of clarification of raw water [77, 52] through the use of *Moringa oleifera* seeds consist of adding, to a pot containing the raw water to be treated, a cloth bag closed with finely crushed seeds of the plant. After a few hours, the suspension matter present in raw water sediments and the clarified supernatant is transferred to another vessel to be consumed directly.

The main advantages of the use of the seed include: low cost, biodegradability of sludge produced, and stability of the pH of the treated water. On the other hand, saline and aqueous crude extracts of Moringa have shown great efficiency as primary natural coagulant, achieving a high reduction of turbidity (between 92-99%) [27, 78], and a production of sewage sludge less than that produced by aluminum sulfate [79].

The main drawback of *Moringa oleifera* and other natural coagulants is that when they are added to the water in the form of powdered seeds, they increase significantly the organic load of water, adding up to 90% of organic substances which do not act as flocculating agents [27, 79, 80]. This fact prevents from storing treated water for more than 24-48 hours [27].

Researching about Moringa is focused on the optimization of the extraction of the active compound [81]; in its characterization, either protein [79] or polyelectrolyte [82]; in the purification of the active compound by simple methods [71]; or in the integration in conventional water treatment systems (such as combination in sand filter) allowing the use of coagulants extracts for the clarification of water.

In addition, the effectiveness of natural coagulants in comparison with aluminum sulphate [83] has also been studied and its application to pilot scale in systems of coagulation-flocculation - filtration [84], in community-based systems of drinking water in rural communities [26] and direct filtration [85] with dual bed filter consisting of *Strychnos potatorum* and *Moringa oleifera* [86, 57] or rice [87]. The expression of coagulant *Moringa oleifera* recombinant protein has also been studied [70] through microorganisms such as *Escherichia coli*, for its continuous production or fractionation [88].

In addition to its application in clarification and purification of raw water, it has been studied the application of natural coagulants in the removal of microalgae [89, in the treatment of waste water from textile industry [90], at olive oil extraction industries [91], in industrial water with high content of heavy metals such as cadmium, arsenic, zinc, or nickel [92] or in sludge dewatering [93], obtaining encouraging results.

The review of literature has shown that:

- The access to drinking water is a universal right of the human being which currently has not been achieved globally, so it is necessary to continue working to reach it.
- Water purification is based on a series of treatments that improve the physical-chemical and microbiological quality of the water making it suitable for human consumption.
- One of the treatments for water purification is the coagulation-flocculation, which requires the addition of coagulants to the water to reduce the suspended matter and settle it.

- There is a wide variety of substances of natural and synthetic type that can be added to the water as coagulant. Although synthetic ones are the most used, currently use is being questioned by their potential toxic effects to humans and the environment.
- There is a growing interest in the use of coagulants from natural origin, which are effective and safe in the treatment of the water.
- Among all of them, the natural coagulant extracted from *Moringa oleifera* seeds is the one that has a greater potential for water treatment and for other applications. In the treatment of the water it is able to effectively reduce the turbidity of the water, as well as the content of pathogenic microorganisms. In addition, the safety and biodegradability of sludge produced differentiate it exceptionally of coagulants used today, becoming a proper coagulant for use in conventional drinking water systems as well as in decentralised systems.

References

[1] EUR-Lex. (1998). Council Directive 98/83/CE of 3 november 1998 concerns the quality of water intended for human consumption. From: http://eur-lex.europa.eu/legal-content/ES/TXT/?uri=celex%3A31998L0083.

[2] USEPA. (1972). Clean Water Act. From: https://www.epa.gov/laws-regulations/summary-clean-water-act

[3] World Health Organization (2011). *Guidelines for Drinking Water Quality*. From: http://apps.who.int/iris/bitstream/10665/44584/1/9789241548151_eng.pdf

[4] Richardson, S.D. & Ternes, T.A. (2005). Water analysis: emerging contaminants and current issues. *Analytical Chemistry*, 77, 3807-3838.

[5] Petrie, B., Barden, R., & Kaspryzk-Hordem, B. (2015). A review on emerging contaminants in wastewaters and the environment: current knowledge, undestudied areas and recommendations for future monitoring. *Water research*, 72, 3-27.

[6] United Nations. (2000). Resolution of the General Assembly of the United Nations. A/55/L.2. *Millenium Declaration*. From: http://www.un.org/spanish/millenniumgoals/ares552.html

[7] World Health Organization (2010). From: http://www.who.int/mediacentre/news/releases/2012/drinking_water_20120306/es/

[8] United Nations (2015). Goals of Sustainable Development. From: http://www.un.org/sustainabledevelopment/es/objetivos-de-desarrollo-sostenible/

[9] Mara, D.D. (2003). Water, sanitation and hygiene for the health of developing nations. Public Health, 117, 452-456.

[10] Lee, E.J. & Schwab, K. J. (2005). Deficiencies in drinking water distribution systems in developing countries. *Journal of Water and Health*, 3 (2), 109-127.

[11] Peter-Varbanets, M., Zurbrugg, C., Swarts, C., & Pronk W. (2009). Decentralized systems for potable water and the potential of membrane technology. *Water Research*, 43, 245-265.

[12] Fewtrell, L., Kaufmann, R.B., Kay, D., Enanoria, W., Haller, L., & Colford, J.M. Jr., (2005). Water, sanitation, and hygiene interventions to reduce diarrhoea in less developed countries: a systematic review and meta-analysis. *Lancet Infectious Diseases*, 5, 42–52.

[13] World Health Organization (2007). *Combating waterborne disease at the household level.* From: http://www.who.int/household_water/advocacy/combating_disease_es.pdf

[14] Sobsey, M.D., Stauber, C.E., Casanova, L.M., Brown, J.M., & Elliot, M.A. (2008). Point of Use Household Drinking Water Filtration: A Practical, Effective Solution for Providing Sustained Access to Safe Drinking Water in the Developing World. *Environmental Science Technology*, 42, 4261–4267.

[15] Mintz, E., Bartram, J., Lochery, P., & Wegelin, M. (2001). Not just a drop in the bucket: expanding access to point-of-use water treatment systems. *American Journal of Public Health*, 91, 1565-1570.

[16] Arnal J.M., Sancho M., Verdú G., & Lora J. (2001). Design of a membrane facility for water potabilization and its application to third world countries. *Desalination*, 137, 63-69.

[17] Garrow, R.S. (2008, Mayo). *Two technologies which just need energy to clean drinking and waste waters.* Proceedings of International Seminar Water and Sanitation in international development and disaster relief, Edinburgh (United Kingdom).

[18] Butler, R. (2008, Mayo). *Skyjuice technology impact on the UN MDG outcomes for safe affordable potable water*. Proceedings of International Seminar on Water and Sanitation in international development and disaster relief, Edinburgh (United Kingdom).

[19] Groendjik, L., & de Vries H.E. (2008, Mayo). *Development of a Mobile water maker, a sustainable way to produce safe drinking water in developing countries*. Proceedings of International Seminar Water and Sanitation in international development and disaster relief, Edinburgh (United Kingdom).

[20] Bolto B. & Gregory J. (2007). Organic polyelectrolytes in water treatment. *Water Research*, 41, 2301– 2324.

[21] Bolto, B.A. (1995). Soluble polymers in water purification. *Progress in Polymer Science*, 20 (6), 987-1041.

[22] Kawuamura S. (1991) Effectiveness of natural polyelectrolytes in water treatment. *Journal of American Water Works Association,* October, 88-91.

[23] Lee, S.H., Lee, S.O., Jang, K. L., & Lee, T.H. (1995).Microbial flocculant from Arcuadendron SP-49. *Biotechnology Letters*, 17, 95-100.

[24] Ganjidoust, H., Tatsumi, K., Yamagishi, T., & Gholian, R. N. (1997). Effect of synthetic and natural coagulant on ligning removal from pulp and paper waste water. *Water Science Technology,* 35, 286-291.

[25] Broekaert, W.F., Cammue, B.P.A., De Bolle, M.F.C., Thevissen, K., De Samblanx, G. W., Osborn, R. W. (1997). Antimicrobial Peptides from plants. *Critical reviews in plant sciences,* 16, 297-323.

[26] Jahn, S.A.A. (1986). Proper use of African natural coagulants for rural water supplies-Research in the Sudan and a guide for new projects. Deutsche Gesellschaft für Technische Zusammenarheit (GTZ). Eschborn.

[27] Jahn, S.A.A. (1988). Using Moringa seeds as coagulants in developing countries. *Journal of American Water Works Association,* 80, 43–50.

[28] Dorea, CC. (2008). *Coagulant-based emergency water treatment.* Actas del International Workshop on Water and Sanitation in international development and disaster relief, Scotland (UK), 94-101.

[29] Baker, M. N. (1948). *The quest for pure water: The history of water purification from the earliest records to the Twentieth Century*. New York (USA): American Water Works Association.

[30] Jahn, S.A.A. (1999). From clarifying pearls and gems to water coagulation with alum-History, surviving practices and technical assessment. *Anthropos*, 94, 419-430.

[31] Jahn, S.A.A. (2001). Drinking water from Chinese rivers: challenges of clarification. *Journal of Water supply research and technology-Aqua,* 50, 15-27.

[32] Van Benchosten, E. (1990). Chemical aspects using aluminium salts. Hydrolytic reactions of alum and polyaluminium chloride. *Water Research*, 24, 1519-1526.

[33] Boisvert, J.P., To, T., Berrak, A., & Jolicoeur, C. (1997). Phosphate adsorption in flocculation processes of aluminium sulphate and poly-alumnium-silicate-sulphate. *Water Research,* 31, 1939-1946.

[34] Najm, I., Tate, C., & Selby, D. (1998). Optimizing enhanced coagulation with PAC. A case study. *Journal of American Water Works Association*, 90, 88-95.

[35] Christopher, H. M., Michael, P.S., & Doyle, G. G. (1995). In vitro acrylamide exposure alerts growth cone morphology. *Toxicology and Applied Pharmacology,* 131, 119-129.

[36] Kaggwa, R.C., Mulalelo, C.I., Denny, P., & Okurut T.O. (2001). The impact of alum discharges on a natural tropical wetland in Uganda. *Water Research*, 35, 795-807.

[37] Crapper, M.D.R., Krishnan, S.S., & Dalton, A.J. (1973). Brain aluminium distribution in Alzheimer’s disease and experimental neurofibrillary degenaeration. *Science*, 180, 511-513.

[38] Crapper, M.D.R. & Boni, D. (1980). Aluminum in human brains disease - an overview. *Neurotoxicology and Teratology,* 1, 3-16.

[39] Gauthier, E., Fortier, I., Courchesne F., Pepin, P., Mortimer, J. & Gauvreau, D. (2000). Forms in Drinking Water and Risk of Alzheimer's Disease. *Environmental Research*, 84 Section A, 234-246.

[40] Miller, R.G., Kopfler, F.C., Kelty, K.C., Stober, J.A., & Ulmer, N.S. (1984). The occurrence of aluminium in drinking water. *Journal American Water Works Association*, 76, 84-91.

[41] Rondeau, V., Jacqmin-Gadda, H., Commenges, D., & Dartigues J.F. (2001). Aluminum in drinking water and cognitive decline in elderly subjects: The Paquid Cohort. *American Journal of Epidemiology*, 154, 288-290.

[42] Dearfield, K.L., Abernathy, C.O., Ottley, S., Brantner, J.H., & Hayes P.F. (1924). Acrylamide: its metabolism, developmental and reproductive effects, genotoxicity, and carcinogenicity. *Mutation Research*, 195, 45-77.

[43] Mallevialle. J., Bruchet. A., & Fiessinger. F. (1984). How safe are organic polymers in water treatment. *Journal of American Water Works Association*, 76, 431-436.

[44] Letterman, R.D. & Pero, R.W. (1990). Contaminants in polyelectrolites used in water treatment. *Journal American Water Works Association,* 82, 87-97.

[45] Criddle, J. (1990). *A review of the mammalian and aquatic toxicity of polyelectrolites*. Report NR 2545, Foundation for Water Research, Medmenhan.

[46] Birina, A.H., Hammad, H.A., Desa, M.N.M., & Muda, Z.C. (2013). Extraction of natural coagulant from peanut sedes for treatment of turbid water. Actas del 4th International conference on Energy and Environment 2013. doi:10.1088/1755-1315/16/1/012065

[47] Divakaran, R. & Sivasankara, Pillai V.N. (2001). Flocculation of kaolinite suspensions in water by chitosan. *Water Research*, 35, 3904-3908.

[48] Bough, W.A. (1975). Coagulation with chitosan: an aid to recovery of by-products from egg breaking wastes. *Poultry Science*, 54, 1904-1912.

[49] Bough, W.A. (1975). Reduction of suspended solids in vegetable canning waste effluents by coagulation with chitosan. *Journal of Food Science,* 40, 297-301.

[50] Eikebrokk, B. & Saltnes, *T.* (2002). NOM removal from drinking water by chitosan coagulation and filtration through lightweight expanded clay aggregate filters. *Journal of Water supply research and technology-Aqua*, 51, 323–332.

[51] Pariser, E.R. & Lombard, D.P. (1989). *Chitin Sourcebook: A Guide to the Research Literature*. New York, USA: Wiley

[52] Jahn, S.A.A. (1984). Effectiveness of traditional flocculants as primary coagulants and coagulants aids for the treatment of tropical raw water with more than thousand-fold fluctuation in turbidity. *Water supply,* 2, 8-10.

[53] Brandis D. (1874). *The forest flora of North-West and Central India*. London: BHL.

[54] Sengupta, A., & Gupta, M.P. (1970). Studies on the seed fat composition of Moringaceae family. *Fette, Seifen, Anstrichmittel,* 72, 6–10.

[55] Tsaknis, J., Lalas, S., Grgis, V., Dourtoglou, V., & Spoliotis, V. (1999). Characterization of Moringa oleifera variety mbololo seed oil of Kenya. *Journal of Agricultural and Food Chemistry*, 47, 4495-4499.

[56] Ferrao, A.M.B., Ferrao, J.K. (1987). Fatty acids of the oil of Moringueiro (Moringa oleifera). *Agronomics* Angol, 30, 3-16.

[57] Chaudhri, R.D. (1996). *Herbal Drug Industry: A practical approach to industrial pharmacology*. New Delhi, India: Eastern Publishers

[58] Azam, M.M., Waris, A., & Nahar, N.M. (2005). Properties and potential of fatty acid methyl esters of some non-traditional seed oils for use as biodiesel in India. *Biomass Bioenergy,* 29, 293–302.

[59] Rashid, U., Anwar, F., Moser, B.R., & Knothe, G. (2008). Moringa oleifera oil: A possible source of biodiesel. *Bioresource Technology,* 99, 8175-8179.

[60] Gopalakrishnan, L., Doriya, K., & Kumar, D.S. (2016). Moringa oleifera: A review on nutritive importance and its medicinal application. *Food Science and Human Wellness*, 5, 49-46.

[61] Siddhuraju, P. & Becker, K. (2003). Antioxidant properties of various solvent extracts of total phenolic constituents from three different agroclimatic origins of drumstick tree (Moringa oleifera Lam). *Journal of agricultural food chemistry*, 15, 2144-2155.

[62] Rao, V.A., Devi, P.U., & Kamath, R. (2001). In vivo radioprotective effect of Moringa oleifera leaves. *Indian Journal of Experimental Biology,* 39, 858–863.

[63] Ruckmani, K., Kavimani, S., Anandan, R., & Jaykar, B. (1998). Effect of Moringa oleifera Lam on paracetamol-induced hepatoxicity. *Indian Journal of Pharmaceutical Science,* 60, 33–35.

[64] Chuang, P.H., Lee, C.W., Chou, J.Y. Murugan, M., Shieh, B.J., & Chen, H.M. (2007). Antifungal activity of crude extracts and essential oil of Moringa Oleifera Lam. *Bioresource technology,* 98, 232-236.

[65] Jabeen, R., Shahid, M., Jamil, A., & Ashraf M. (2008). Microscopic evaluation of the antimicrobial activity of seed extracts of Moringa oleifera. *Pakistan Journal of Botany,* 40, 1349-1358.

[66] Oliveira, A.P.S., Santana Silva, L.L., Albuquerque Lima, T. et al. (2016). Biotechnological value of Moringa oleifera seed cake as source of insecticidal lectin against *Aedes aegypti*. *Process Biochemistry*, 51 (10), 1683-1690.

[67] Eilert U., Wolters B., & Nahrstedt A. (1981).The antibiotic principle of seeds of Moringa oleifera and Moringa stenopetala. *Planta Medica*, 42, 55-61.

[68] Olsen, A. (1987). Low technology water purification by bentonite clay and Moringa oleifera seed flocculation as performed in Sudanese villages: effects on Schistosoma mansoni cercariae. *Water Research*, 21, 517–522.

[69] Madsen, M., Schlundt, J., & Omer, El-FE. (1987). Effect of water coagulation by seeds of Moringa oleifera on bacterial concentration. *American Journal of Tropical Medicine and Hygiene*, 90, 101–109.

[70] Broin, M., Santaella, C., Cuine, S., Kokou, K., Peltier, G., & Joët, T. (2002). Flocculent activity of a recombinant protein from Moringa oleifera Lam seeds. *Applied Microbiology Biotechnology*, 60, 114-119.

[71] Ghebremichael, K. A., Gunaratna, K.R., Henriksson, H., Brumer, H., & Dalhammar, G. (2005). A simple purification and activity assay of the coagulant protein from Moringa oleifera seed. *Water Research*, 39, 2338-2344.

[72] Caceres, A. & Lopez, S. (1991). Pharmacologic properties of Moringa oleifera: 3: Effect of seed extracts in the treatment of experimental Pyoderma. *Fitoterapia*, 62, 449–450.

[73] Suarez, M., Haenni, M., Canarelli, S., Fisch, F., Chodanowski, P., Servis, C., Michielin, O., Freitag, R., Moreillon, P., & Mermod, N. (2005). Structure function characterization and optimization of a plant derived antibacterial peptide. *Antimicrobial agents and chemotherapy*, 49, 3847-3857.

[74] Sutherland, J.P., Folkard, G., & Grant, W.D. (1990). Natural coagulants for appropriate water treatment: a novel approach. *Waterlines*, 8, 30–32.

[75] Doughari, J. H., Pukuma, M. S., & De, N. (2007). Antibacterial effects of Balanites aegyptiaca L. Drel. and Moringa oleifera Lam. on Salmonella typhi. *African Journal of Biotechnology*, 6, 2212-2215.

[76] Silvestro, L., Weiser, J.N., & Axelsen, P.H. (2000). Antibacterial and antimembrane activities of cecropin A in Escherichia coli. *Antimicrobial Agents and Chemotherapy*, 44, 602–607.

[77] Lund, E. & Jahn S.A.A. (1980). *Water Purification with Primitive Flocculation Methods as Done in Sudanese Villages*. Proceedings of the 2nd International Conference on the Impact of Viral Diseases on the Development of African and Middle East Countries.

[78] Muyibi, S.A. & Okuofu, C.A. (1995). Coagulation of low turbidity surface water with Moringa oleifera seeds. *International Journal of Environmental Studies*, 48, 263–273.

[79] Ndabigengesere, A., Narasiah, K.S., & Talbot, B.G. (1995). Active agents and mechanism of coagulation of turbid waters using Moringa oleifera. *Water Research,* 29, 703-710.

[80] Okuda, T., Baes, A. U., Nishijima, W., & Okada, M. (2001). Isolation and characterization of coagulant extracted from Moringa oleifera seed by salt solution. *Water Research,* 35, 405-410.

[81] Pereira Camacho, F., Serrao Sousa, V., Bergamasco, R., & Ribau Teixeir,a M. (2017). The use of Moringa oleifera as a natural coagulant in surface water treatment. *Chemical Engineering journal*, 313, 26-237.

[82] Okuda, T., Baes, A. U., Nishijima, W., & Okada, M. (2001). Coagulation mechanism of Salt solution: extracted active component in Moringa oleifera seed. Water Research, 35, 830-834.

[83] Garcia-Fayos, B., Arnal, J.M., Monforte-Monleon, L. & Sancho M. (2015). Alternatives to the use of synthetic organic coagulant aids in drinking water treatment: improvements in the application of the crude extract of Moringa oleífera seed. *Desalination and Water Treatment*, 55, 3635-3645.

[84] Raghuwanshi, P.K., Mandloi, M., Sharma, A. J., Malviya, H. S., & Chaudhari, S. (2002). Improving filtrate quality using agrobased materials as Coagulant Aid. *Water Quality Research Journal of Canada*, 37, 745-756.

[85] Al-Khalili, R.S., Sutherland, J.P., & Folkard, G. K. (1997). *Filtration with a natural coagulant*. Proceedings of 23rd WEDC Conference Water and sanitation for all: partnerships and innovations, Durban (Sudáfrica).

[86] Pollard, S.J.T., Thompson, F.E., & McConnachie, G.L. (1995). Microporous carbons from Moringa oleifera husks for water purification in less developed countries. *Water Research*, 29, 337-347.

[87] Barnes, D. & Mampitiyarachichi, T.R. (1983). Water Filtration Using Rice Hull Ash. *Waterlines*, 2, 21-23.

[88] Alves Baptista, A.T., Oliveira Silva, M., Guttierres Gomes, R., Bergamasco, R., Fernandes Vieira, M., & Salcedo Vieira, A.M. (2017). Protein fractionation of sedes of Moringa oleifera lam and its application in superficial water treatment. *Separation and Purification Technology,* 180, 114-124.

[89] Barrado-Moreno, M.M., Beltran Heredia, J., & Martin Gallardo, J. (2016). Microalgae removal with Moringa oleífera. *Toxicon*, 110, 68-73.

[90] Vasudevan, D. & Gopalakrishna, K. (2002). Coagulation using eco friendly natural coagulants. Proceedings of the 26th WEDC Conference: Sustainable Environmental Sanitation and Water Services, Calcuta (India).

[91] Meyssami B., & Kasaeian A.B. (2005). Use of coagulants in treatment of olive oil wastewater model solutions by induced air flotation. *Bioresource Technology,* 96, 303-307.

[92] Sharma P., Kumari, P., Srivastava, M.M., & Srivasatava S. (2006). Removal of cadmium from aqueous system by shelled Moringa oleifera Lam. seed power. *Bioresource technology,* 97, 299-305.

[93] Özacar, M. & Sengil, I. A. (2000). Effectiveness of tannins obtained from Valonia as a coagulant aid for dewatering of sludge. *Water Research*, 34, 1407-1412.