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Natural coagulants: analysis of potential use for drinking water treatment in developed and developing countries

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ABSTRACT

Drinking water must be colorless, odorless and tasteless, and free of substances or micro-organisms that can cause disease. The process to obtain drinking water is purification, understood as the treatment to make water suitable for human consumption. Water purification is traditionally composed of a sequence of standard treatments including coagulation-flocculation, sedimentation, filtration, and disinfection. Some compounds named coagulants are used for coagulation-flocculation, which is the stage in which the suspended matter is removed and the water is clarified. Coagulants are usually synthetic, therefore represent a high cost and are difficult to gets 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 hazard effects that the remains of these products in treated water can cause to people. In contrast to synthetic coagulants, there are several natural compounds with coagulation properties which have some advantages such as easy availability and safety. At present, scientific community is paying more attention to natural coagulants as legal restrictions to synthetic ones are becoming more severe. This paper is a review of natural coagulants as an alternative to chemical coagulants, in which their potential application in the treatment of drinking water is analysed.

Keywords: Drinking water; Natural coagulants; Water purification

1. Introduction

Water is essential for the quality of life and the welfare of the human being. For some uses it must accomplish certain properties. In case of human consumption it must be drinkable, i.e., odourless, colourless and tasteless, and free of substances that could damage the health of consumers. A certain sequence of purification treatments, depending on water source, is required to make water drinkable.

Generally, water purification consists in the application of physical, chemical and/or microbiological treatments that allow obtaining water with good quality for being consumed by people without any health hazard. The result of purification must 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 depends on the physico-chemical and microbiological quality of the raw water, on the resources and infrastructure available for the treatment, on its cost, and on the characteristics of the distribution system of the final treated water.

The basic conventional purification treatment includes the stages of coagulation-flocculation, sedimentation, filtration and disinfection, as shown in Fig. 1.

Coagulation-flocculation aims to increase the size of suspended and dissolved particles in the water, thus making easier their separation through filtration or sedimentation. This stage requires the addition of coagulants and floccu-

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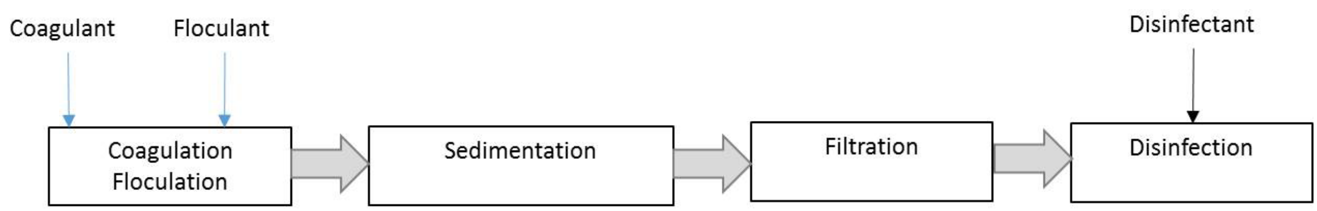

Fig. 1. Treatment stages for water purification.

lants to neutralize the electrostatic charge of suspension solids favouring its aggregation (flocs), and to increase the size of flocs to achieve higher sedimentation rates. The origin of these coagulants and flocculants can be synthetic or natural as it will be explained later. Occasionally, these clarification stages also reduce microbiological contamination (viruses and bacteria) since suspended solids sweep much of microorganisms associated with the particles responsible for turbidity, when they sediment.

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

The water thus treated is suitable for human consumption and can be supplied 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.

1.2. Coagulants and flocculants for water purification

The purification sequence includes a stage of coagulation-flocculation. As mentioned before, 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 two types:

1.2.1. 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), which include all the coagulants produced by partial neutralization of an aluminium chloride solution with basic solutions. These coagulants are characterized by: wide operation pH ranges, good sedimentation rate and lower concentration of residual aluminum in treated water, in comparison to other aluminum salts [4].

1.2.2. 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 polymers secreted by certain microorganisms and biological polymers extracted from plants, algae or animals [5]. Synthetic organic coagulants are also known as polyelectrolytes, and they 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.

1.2.3. Flocculants

Occasionally, the coagulation process requires the addition of flocculants that increase the size of the formed floc to sediment faster. Flocculants are also 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 flocculates 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.

1.3. Restrictions and limitations to synthetic coagulants

Currently, the use of synthetic inorganic coagulants for water purification, such as aluminum sulphate, ferric chloride [6–8] or organic poly-electrolytes such as poly acrylamide [4], is being also questioned due to, among other reasons:

- environmental problems [9,10], mainly due to generation of toxic sludge that cannot be used in agriculture;
- possible relationship with Alzheimer's disease [11–13];
- worsening of neurodegenerative diseases such as senile dementia [14,15];
- possible relationship with cancer [16,17].

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 [4]. Some countries such as Japan or Switzerland have banned the use of organic polyelectrolyte in the treatment of drinking water [18], 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 [4].

It has to bear in mind that monomers are more toxic than polymers [19], 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 µg/L. In the case of Spain, until the year 2005, SCO-3719-2005 Ministerial Order limited the content of monomer free of acrylamide for cationic, ionic and non-ionic polyacrylamide in 0.02%, 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 free residual chloride must not exceed the concentration established in RD 140/2003, which is 1 mg/L.

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 synthetic current ones. Natural coagulants and their potential use have increased scientific community interest as alternative to synthetic coagulants. This paper reviews traditional natural coagulants and their potential use for water purification, focusing on *Moringa oleifera*, one of the natural coagulants with more potential applications.

2. Natural coagulants

Natural coagulants are defined as water soluble substances, coming from vegetable or animal materials [20–23]. 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 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 [24–26]. 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 [27]. 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 [28]. 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 [29]. 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 natural coagulants are: local availability, origin from natural and renewable resources, safety for human being, and biodegradability of sludge that can be also used in agriculture. Its application can be directed not only to developing countries, but also to conventional purification and wastewater treatment [4].

2.1. Traditional natural coagulants

In 1988, Jahn [25] 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 India. Some of them have been studied in greater depth, providing positive results of 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 natural coagulants that have been under research are the following:

- Strychnos potatorum
- Moringa oleifera

- Okra
- Cassava
- Rice
- Starch
- Cactus Latifaria and 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 [30]

In addition to application in clarification and purification of raw water, natural coagulants have been studied for application in the removal of microalgae [31], in the treatment of waste water from textile industry [32], at olive oil extraction industries [33], in industrial water with high content of heavy metals such as cadmium, arsenic, zinc, or nickel [34] or in sludge dewatering [35], obtaining encouraging results.

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

2.1.1. Chitosan

Chitosan is the animal-origin natural coagulant most commonly used at present. This coagulant is a deacetylated derivative of chitin that can be found 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, India and United States, where fishing and seafood processing are the main industries. Its cost of production is US $2/kg [20]. 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, but 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, able to remove up to 99% of the raw water turbidity [36]. 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 [20,37,38]. Recent studies have shown its ability to remove organic matter [39] and its high potential for application alone or in combination with filtration sand bed for the treatment of textile 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 [40].

2.1.2. Starch

Starch is a natural polymeric coagulant with two possible molecular structures: 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 coagulant of both structures. Its use is documented in clarification of clays, wastewater effluents or as de-emulsifying agent of wastewater with high content of oil [4]. In Spain, it is recognized as a substance for the treatment of drinking water for human consumption, as it is indicated in the Royal legislative decree 140/2003.

2.1.3. Sodium alginate

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 authorized 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, 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 [20]. Like starch derivatives, it is a substance recognized as suitable for the treatment of drinking water in Spain, as it is indicated in the Royal legislative decree 140/2003.

2.2. Moringa oleifera as natural coagulant

Among all the natural coagulants mentioned previously, the vegetable-origin primary coagulant of more researched at present 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 India. Half of these species, namely *M. oleifera*, *M. peregrina*, *M. stenopetala*, *M. longituba*, *M. drouhardii*, *M. ovalifolia* and *M. concanensis*, are relatively common, although occasionally cultivated. All species of Moringa have coagulating ability [24,41], although only the *Moringa oleifera* is cultivated throughout the tropical area.

Moringa oleifera was originally a native tree of 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 Fig. 2) was extended to the American continents through the French West Indies from Cuba and Jamaica [42]. Its cultivation was spread not only in Central America but also in remote areas of the American continent such as California, Brazil and Paraguay.

Moringa oleifera has a great interest at present, thanks to its multiple uses and properties, which are described below.

Fig. 2. Moringa oleifera tree.

2.2.1. Oil

The seeds of *Moringa oleifera* contain between 33–41% (w/w) of oil [43]. It has a quality similar to olive oil, since it has 73% of oleic acid, a high content of tocopherols, and a high ratio of monounsaturated fatty acids versus saturated, so it is recommended as a cooking oil for human consumption [44].

Furthermore, oil from *Moringa oleifera* has numerous applications in different fields such as cosmetic or medicinal uses. [45,46], and it is also being researched as biofuel [47,48].

2.2.2. Vegetable foodstuff and medicinal use

The pods, leaves and seeds of the plant (see Fig. 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 have shown medicinal properties as well as excellent nutritional properties [49].

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

All parts of the plant are used in a wide variety of traditional medicines as they have anti-inflammatory, antipyretic, antiepileptic, hepatoprotective, antihypertensive and antitumoral properties [50], as well as properties for the control of diabetes [49].

2.2.3. Antibacterial, fungicide and insecticide properties

The roots and flowers of Moringa have antibacterial activity [51] thanks to its high proportion of antimicrobial agents. They contain, among others, an antibiotic active substance, "pterygospermin", which has a significant fungicide and antibacterial effect [52].

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* [53], *Fusarium solani* or *Rhizopus solani* [54].

Recently, the insecticidal properties of seeds against the *Aedes aegypti* mosquito have been also researched [55].

2.2.4. Water treatment

The best-known use of *Moringa oleifera* seed is the treatment of water, thanks to coagulants, antibiotic [56,57] and antifungal properties [53].

The extracts from seeds and leaves have antibacterial properties [57,58] and are able to coagulate and remove gram positive and negative bacteria from water by sedimentation [59] such as *Escherichia coli*, *Pseudomonas aeruginosa* [60], *Staphylococcus aureus* [61], *Streptococcus pyogenes* [62] or *Salmonella typhi* [64], reaching rates of bacteria reduction 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 act destroying the cell membrane or by inhibiting key enzymes [62,65]. Sutherland et al. (1990) also found that Moringa seeds can inhibit the replication of Bacteriophage [63]. The antimicrobial effects of seeds are specifically attributed to the compound 4(α-L-rhamnosiloxi)-bencil-isotiocianate [56].

The traditional method of raw water clarification [41,66] 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 pH stability 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%) [25,67], and a production of sewage sludge less than that produced by aluminum sulfate [68].

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 [25,68,69]. This fact prevents from storing treated water for more than 24–48 h [25].

Research about Moringa is focused on the following topics:

- optimization of the extraction of the active compound [70]
- characterization of the active compound, either protein [68] or polyelectrolyte [71]
- purification of the active compound by simple methods [60]
- 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 *Moringa oleifera* as coagulant in comparison with aluminum sulphate [72] has also been studied and its application to pilot scale in systems of coagulation-flocculation - filtration [73], in community-based systems of drinking water in rural communities [24], and direct filtration [74] with dual bed filter consisting of *Strychnos potatorum* and *Moringa oleifera* [46,75] or rice [76]. The sequence of coagulant *Moringa oleifera* recombinant protein has also been studied [59] through microorganisms such as *Escherichia coli*, for its continuous production or fractionation [77].

The review of literature about application of coagulants on water purification has shown that:

- 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 water treatment *Moringa oleifera* 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 potential coagulant for use in conventional drinking water systems as well as in decentralised systems.

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