Universidade Federal do Mato Grosso Do Sul

Faculdade De Ciências Farmacêuticas, Alimentos e Nutrição (FACFAN)

Curso de Engenharia de Alimentos

Letícia Bernal Ferreira de Sousa

COAGULANTES À BASE DE PLANTAS PARA TRATAMENTO DE ÁGUAS RESIDUAIS EM INDÚSTRIAS DE ALIMENTOS

Campo Grande/2023

Letícia Bernal Ferreira de Sousa

COAGULANTES À BASE DE PLANTAS PARA TRATAMENTO DE ÁGUAS RESIDUAIS EM INDÚSTRIAS DE ALIMENTOS

Trabalho de Conclusão de Curso apresentado ao Curso de Engenharia de Alimentos da Universidade Federal do Mato Grosso do Sul como parte das exigências para a obtenção do título de Bacharel em Engenharia de Alimentos.

Profa. Orientadora: Dra. Thaisa Carvalho Volpe Balbinoti

Campo Grande/2023

Letícia Bernal Ferreira de Sousa

COAGULANTES À BASE DE PLANTAS PARA TRATAMENTO DE ÁGUAS RESIDUAIS EM INDÚSTRIAS DE ALIMENTOS

Para a elaboração do Trabalho de Conclusão de Curso utilizou-se o artigo intitulado "Plant-based coagulants for food industry wastewater treatment" publicado na revista Journal of Water Process Engineering, Volume 52, Página 103525, ISSN 2214-7144, em 27 de Fevereiro de 2023.

Campo Grande/2023

ABSTRACT: Sustainable effluent treatments are essential tools in lowering the environmental impact of industrial activities. The partial or complete replacement of synthetic coagulants by natural coagulants, especially plant-based ones, can reduce the footprint of the effluent treatment due to the higher biodegradability and non-toxicity. Natural coagulants are also generally cheaper. This review focuses on plant-based coagulants used in food industry effluent treatment. Extraction parameters of plant-based coagulants and effluent treatment conditions for different coagulants are presented. Based on an extensive assessment of peer-reviewed papers on food industry effluent treatment, the performance of plant-based coagulants is compared to that of traditional, synthetic coagulants, both alone and as coagulant aids. This review aims to guide researchers and industry professionals in optimizing and scaling up environment-friendlier effluent treatments using natural coagulants.

Keywords: Effluent Treatment; Wastewater Treatment; Coagulation; Natural Coagulants; Sustainability; Biocoagulants.

Table of Contents

1. INTRODUCTION

Industrial activities are one of the leading causes of water pollution due to the generation of significant amounts of effluents that contain toxic species or species that are difficult to degrade [1]. The production of liquid waste by the food industry is sizeable, especially in the animal-based food sector. Slaughtering operations and meat processing generate massive volumes of effluents rich in proteins, organic compounds, and fats [5]. For example, slaughtering a single pig or cow generates 330 L and 700 L of effluents, respectively [2–4]. Although drastically less than the animal sector, the production of plant-derived foods also uses significant amounts of water. For instance, the production of one ton of crude palm oil requires 5–7.5 tons of water [6,7]. Another example is the cassava starch industry, which generates 12–15 L of effluents for each m^3 of processed cassava. The effluents from cassava starch production exhibits high biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), total solids (TS), and cyanides [8].

Inadequate disposal of food industry effluents impacts the environment due to the high load of organic matter, heavy metals, alkalinity, and hardness, resulting in water pollution, odor generation, algal blooms, and mortality of aquatic and land animals [4,9]. The appropriate treatment of industrial effluents is carried out by Effluent Treatment Stations (ETSs) [10]. By diminishing the polluting load of effluents, ETSs enable their safe reuse or release into water bodies.

To treat effluents, ETSs utilize coagulation, flocculation, sedimentation, and other physical, chemical, and biological processes [11]. Eflluents treatments can be classified into physical (media filtration, settling, adsorption, membrane, and ultraviolet radiation), chemical (electrochemical, coagulation, oxidation, ion exchange, disinfection, catalytic reduction, and softening), and biological (microbial biodegradation, phytoremediation, constructed wetlands, and bioreactor digestion) [12,13]. Among these technologies, coagulation, and flocculation (CF) are the oldest and most widely employed in effluents treatment due to their simplicity, cost-efficiency, efficacy, and low energy demand [14].

Coagulation involves the addition of a coagulating agent to the effluent to reduce the forces that keep suspended particles apart from colloids. Flocculation agglomerates the resulting material to form larger particles that can be easily separated following sedimentation [13,15]. In effluents processing, CF has been used as a pretreatment to remove suspended impurities and improve the quality of the treated effluent for the subsequent stages [15]. CF operations are also utilized to remove compounds other than suspended particles and colloids, such as pigments, micro-pollutants, organic compounds, oils, and fats [14].

Due to the crucial role of CF in ensuring the safe discharge of effluents, the global coagulant and flocculant market is estimated to reach USD 6.01 billion by 2022, following an annual growth rate of 5.9% between 2017 and 2022 [16]. The economic relevance of CF warrants the study of plant-based coagulants as sustainable substitutes to synthetic-based coagulants [13]. Natural coagulants exhibit greater biodegradability, lower cost, and lesser toxicity and generate less sludge [9,17,18]. Given the alarming amplification of the climate and ecological crises, research applying natural coagulants has critical industrial relevance by promoting more environmentally friendly effluent treatments.

Research on natural coagulants is primarily focused on the treatment of drinking water [19,20]. However, studies that explore the application in industrial effluents, mainly from the food industry, have been growing in recent years. For example, Vieira et al. [9] and Real-Olvera et al. [4] investigated *Moringa oleifera* as plant-based coagulants in the treatment of dairy industry and slaughterhouse effluentss, respectively. Vieira et al. [9] reported a 90% removal of color and turbidity, while Real-Olvera et al. [4] obtained a 64% reduction in COD. Dozens of other research studies on the performance of plant-based coagulants to treat food industry effluents will be discussed in the following sections.

This review provides a database for industry professionals and scientists interested in applying plant-based coagulants in food industry effluents treatment.

2. COAGULATION AND FLOCCULATION

As colloidal particles present in effluents exhibit a low sedimentation velocity, ETSs use CF to reduce suspended and dissolved materials [21]. The use of coagulants during CF reduces the turbidity and color of effluents [9]. Turbidity, for instance, is reduced by 85–99% [22]. Coagulation and flocculation agglomerate the impurities from the effluent into larger and heavier particles (called flocs), which can be removed by sedimentation, filtration, or flotation [23] (Figure 1).

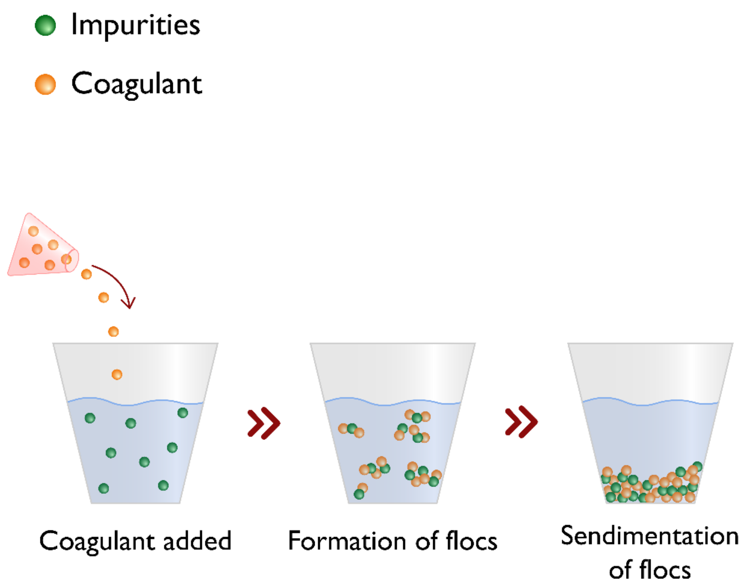

Figure 1 – Representation of the coagulation, flocculation, and sedimentation processes.

Coagulation is a process that affects the physical forces and chemical reactions between the effluent and the dissolved impurities. The coagulating agent generates positively charged ions in the water, which contains negatively charged colloids [21]. As a result, the repulsion between particles is reduced, causing the particles to collide and stick together to form flocs. From an electrostatic point of view, coagulation reduces the zeta potential (i.e., the electric potential between the liquid medium and the particles' surface) by adding specific ions. According to Okuda et al. [24], the zeta potential is an analytical method to evaluate the effect of coagulation, which helps select the optimal coagulant dosage and medium pH.

According to Pritchard et al. [19], the coagulant must be mixed vigorously for a few seconds immediately after addition into the effluent (Figure 2) to ensure uniform dispersion and high interaction between coagulant and effluent. Coagulation efficiency is a function of the contact between coagulant and suspended particles before the end of the mixing stage. In the subsequent step, flocculation occurs due to the neutralization between the coagulant's acidity and the effluent's alkalinity. Flocs form due to the electrostatic attraction between the positive charges resulting from the ionization of the coagulant and the negative charges of the particles. Flocs are larger, heavier particles exhibiting ionic bonds that tend to precipitate with decreased flow velocity [25].

During flocculation, proper time and stirring must be chosen to ensure floc formation, since the aggregation of suspended particles occurs due to the collision between the particles previously destabilized by the coagulant. Agglomeration is attributed to Van Der Waals forces [26]. The greater the velocity gradient, the faster the rate of particle agglutination. However, the flocs will grow to a maximum limit, as the high shear forces can cause the agglomerated particles to break [21]. As too intense agitation in the flocculation stage can hinder floc stability, lower mixing velocities must be selected at this stage (Figure 2) [27].

Worth noting, when coagulation and flocculation are not carried out properly, the performance of the treatment is compromised, jeopardizing the quality of the treated effluent.

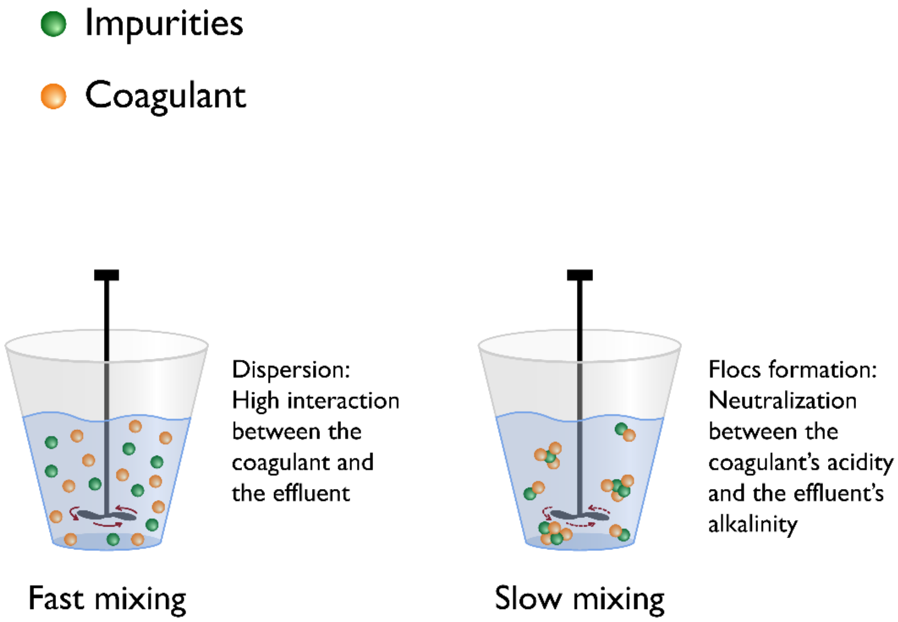

Figure 2 – Representation of the mixing procedure used in the coagulation and flocculation stage.

Table 1 presents the operational conditions (coagulation, flocculation, and sedimentation) used in the treatment of food industry effluents with plant-based coagulants.

Table 1[1] – Operational conditions used in the treatment of food industry effluents with plant-based coagulants.

Due to the wide range of industrial effluents, experiments must be conducted to define the optimal operational conditions for coagulation, flocculation, and sedimentation, aiming at high efficiency and low cost. An efficient way to determine the coagulant dosage and the coagulation pH is to conduct tests in static reactors, known as Jar-Tests (Figure 3). Jar testing entails subjecting the effluent added with specific coagulant dosages to vigorous agitation. The

[1] attached

stirring rate is then significantly reduced to promote floc formation, and the sample is let to sit for a defined period until the decanted water samples are collected (sedimentation) (Table 1).

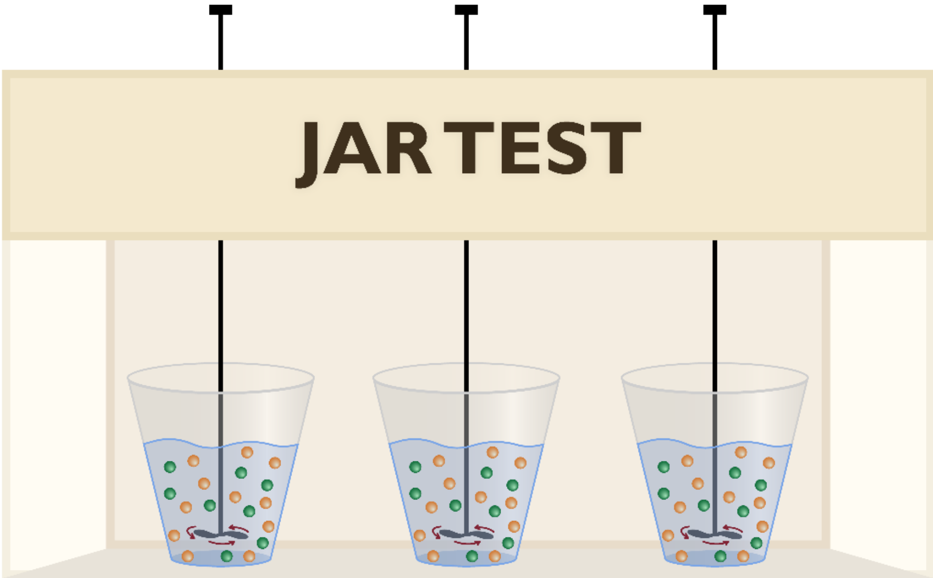

Figure 3 – Representation of a Jar-Test equipment for coagulation, flocculation, and sedimentation tests.

Coagulation efficiency depends not only on the chemical nature of the effluent and the coagulant, but also on such operational conditions as initial pH, coagulant dosage, particle size, temperature, stirring rate, and reaction time [49]. Research on effluent treatment often uses experimental design as a statistical tool to evaluate the ideal conditions in which coagulation, flocculation, and sedimentation occur [6,7,24,34,44]. The resulting data are plotted to illustrate changes in quality parameters (e.g., color, turbidity, COD, total solids, among others) as a function of operational conditions (e.g., medium pH and coagulant dosage). The objective is to derive the optimal conditions for maximum particle removal [50]. Such graphs are of prime importance in the design of Jar-Tests and interpretation of results [51].

3. COAGULANTS

The various coagulants used in effluent treatment can be classified into synthetic coagulants and natural coagulants [9,52]. The coagulant's efficiency depends on the affinity and specificity characteristics, which are a function of its physical-chemical properties [49]. Metal-based materials are the coagulants traditionally used in effluents treatment. Despite the broad applicability of metal-based coagulants, the challenges associated with their use have prompted research efforts to develop environment-friendly biobased coagulants [14].

3.1. SYNTHETIC COAGULANTS

The most used synthetic coagulants are trivalent iron and aluminum salts, with $AlCl_3$, $Al_2(SO_4)_3$, $FeCl_3$, and $Fe_2(SO_4)_3$ being the most frequent due to the low cost and proven coagulant capacity [53,54]. The choice depends primarily on the effluent's characteristics.

Coagulation performed with aluminum and iron salts involves chemical and physical phenomena. The hydrolysis promoted by the coagulant forming hydrolyzed and positive-charged species is a chemical process. The formation of hydrolyzed species depends on the coagulant dosage and the effluent's pH [55]. In general, these coagulants will only be effective if the effluent is alkaline; otherwise, coagulation will be limited due to excess protons released by the coagulant. In turn, the physical mechanisms encompass the contact of the hydrolyzed species with the impurities present in the effluent, forming flocs (flocculation stage) [21]. Particle aggregation results from the neutralization of negative charges of the effluent's solids by the hydrolyzed species, ensuring floc growth and hence rapid precipitation [55].

Despite the recognized efficacy of synthetic coagulants in effluents treatment, the use of agents based on aluminum salts requires strict control over the residual concentration of synthetic material in the treated effluent. According to Yin [23], after the salt is solubilized, the cation released will be adsorbed onto the solid suspended material. In other words, the sedimented material will have a high concentration of the cation (for example, Al^{3+}), which impedes the safe discharge of the treated effluent in the environment. Studies link high aluminum intake to potential health conditions, such as Alzheimer's disease [13].

3.2. NATURAL PLANT-BASED COAGULANTS

Plant-based coagulants play a prominent role in sustainable effluents treatment due to their high biodegradability, non-corrosive nature, and non-toxicity. An increasing number of research papers focus on the coagulation properties of seeds and bark resins from different plant species, but also of bone shell extracts, extracts from shellfish exoskeleton, and natural mineral soils [14,20,56,57].

Natural coagulants are composed of large molecular chains, either positively or negatively charged. These coagulants can be cationic, anionic, or non-ionic, with the former two collectively termed polyelectrolytes [58]. However, only cationic polyelectrolytes (those with positive charges) are ideal for use as coagulants [14].

Coagulants characterized as cationic polyelectrolytes are ideal in effluents treatment because the colloidal material present in the effluent generally exhibits a negative surface electrical charge. Cationic polyelectrolytes produce cationic species in the effluent, destabilizing the particles and thus contributing to coagulation. The resulting attraction between particles (Van der Waals force) enhances floc formation due to a reduction in the negative charge of the colloidal surface [52-54].

Natural coagulants have been studied as partial or total substitutes of synthetic coagulants to improve the CF stage – both the quality of the flocs produced and the reduction/elimination of metals in the final sludge [52]. The use of natural coagulants combines the properties expected for effluent treatment with abundant availability, low toxicity and cost, high biodegradability, and low rate of waste sludge production [52,59,60]. Nevertheless, the presence of natural organic polyelectrolytes in drinking water treatment can increase the organic matter content, prompting the onset of unpleasant tastes and odors. Therefore, the introduction of natural coagulants in water treatment must be carefully considered, as the treated water is likely to contain residues that will be ingested by consumers [19,20,56]. In effluent treatment, this aspect is still little explored [61-63].

Table 2 presents the main conclusions of several studies on plant-based coagulants in the treatment of food industry effluents.

Table 2[2] – Plant-based coagulants used to treat food industry effluents.

3.2.1 Parameters that influence the performance of plant-based coagulants

In view of the wide range of existing coagulants (both synthetic and natural), choosing the most efficient can be challenging. The process parameters (such as dosage, settling time, mixtures, temperature and pH) must be weighted appropriately to obtain the best performance in removing contaminants [64,65]. Tables 1 and 2 summarize the process parameters commonly used in food industry effluents treated with plant-based coagulants, as well as the main performance results.

Dosage

Inadequate coagulant dosing results in unsatisfactory water quality after treatment. Therefore, it is critical to determine the dosage range needed to obtain maximum pollutant removals at minimum cost. Among different coagulation mechanisms, only charge neutralization

[2] attached

and bridging are negatively affected by dosage, mainly due to the stoichiometric relationship [44,66].

When the effluent's turbidity is high (>100 NTU), the required coagulant dosage tends to be lower compared to low turbidity effluents (5–10 NTU) [59]. A likely cause is the collision frequency between particles and coagulant [15]. As high turbidity means a high concentration of impurities (and a high collision frequency), coagulation is more likely. In this case, lower dosages of coagulant are necessary. In contrast, low turbidity requires a high dosage of coagulant because the collision frequency between impurities and coagulants is lower [50]. Optimal dosage can be obtained by plotting the measured turbidity (or any other pollutant parameter) versus the applied dosage [60,61].

There is a decline in contaminant removal efficiency when the coagulant dosage is less than or exceeds the required dosage. Overdosing can lead to charge reversal and consequently result in particle repulsion, preventing floc formation. Overdosing restricts the number of available adsorption sites by covering the coagulant surface [44,51]. Therefore, adding more coagulant to the effluents treatment does not enhance the coagulation process. On the other hand, a dosage much lower than necessary results in incomplete and ineffective coagulation because most of the particles remain in suspension.

Real-Olvera et al. [4] tested a dosage of 3 to 7 g/L of *Moringa oleifera* for the treatment of slaughterhouse effluent and observed that 7 g/L yielded the best results. When *Cassia obtusifolia* at a dosage of 0 to 2.5 g/L was used in effluent coagulation, the optimal condition was 1.0 g/L to treat palm oil residue [6] and 2 g/L for rice starch [62].

pH

A solution's pH affects the efficiency of the coagulation process by changing the electrochemical nature of the solvent and ionic polymers used [44,63]. Therefore, the system's pH optimization is essential to ensure efficient coagulation [61,66]. The decrease in pH lowers the load of natural organic matter (NOM), reducing their water solubility. Consequently, coagulation efficiency is reduced, as the main coagulation mechanism is charge neutralization, in which cations or polyelectrolytes are required to bind to the negative charge of the NOM [44,61]. An optimal pH is also critical for forming metal poly-hydroxides and the precipitation of solid colloidal phases [61]. Cationic polyelectrolytes are the most widely used organic polyelectrolytes owing to their ability to bind with negatively charged NOMs. The positive charge in these polymers can increase at lower pH when the functional groups responsible for the cationic behavior are amines (primary, secondary, and tertiary) [61].

Some studies have optimized the pH of the treatment system when using plant-based coagulants, for example *Coccinia indica* [67], okra gum [68], cactus, and hyacinth bean peels [69]. The optimal pH for coagulation using okra gum was 9.2, with 98.3% SS removal [68]. At this pH, okra gum uncoiled further to achieve a flat, dangling form, which enhances the coagulation process. Most legumes used as natural coagulants perform better when coagulation occurs at an optimized pH of 7 to 8.5 [20,56]. However, the same coagulant can exhibit different optimal pHs depending on the effluent under treatment. For example, *Moringa oleifera* showed optimal performance at pH 9.0 for slaughterhouse effluent [4]; between 4.0 and 5.0 for the effluent from the pulp of coffee fruits [6], and 4.0 for distillery spent wash [46].

Temperature

Temperature is the factor that receives the least attention in effluent treatment. Few studies have reported the effect of temperature on treatment efficiency [6,36,40,70]. Temperature affects coagulation kinetics and impurity removal efficiency [71,72]. Coagulation efficiency improves with an increase in temperature due to the higher collision rate among particles. This results in lower viscosity, favoring the homogeneous dissipation of the coagulation species in the mixing stage [73].

At low temperatures, floc aggregation tends to be weak due to the lesser particle-particle collisions. Due to the formation of irregular flocs under low-temperature conditions, the coagulant dosage required to remove organic matter is higher [70,72].

In many cases, the effluent's temperature is a non-controllable parameter, which affects the removal of NOM by coagulation. Therefore, dosages must be reassessed across seasons to account for temperature changes [2,70].

For plant-based coagulants, the optimal process temperature is 25-30°C [6,36,40,70]. As the active agents in plant-based coagulants are generally proteins, system temperature must be increased carefully to avoid a reduction in the coagulation capacity due to protein denaturation. As denaturation causes changes in the protein's spatial configuration, its biological activity is reduced or lost.

Mixing

Mixing is crucial in coagulation processes, including both rapid mixing to promote the interaction of coagulants with suspended particles to form microflocs, and slow mixing to promote microfloc aggregation and large floc formation. Excessively low mixing speed and considerably short mixing time can decrease the rate of floc formation, while excessively fast mixing speed and considerably long mixing time can break the flocs, yielding poor settling efficiency [55,70,71]. As shown in Table 1, mixing parameters in the treatment of food industry effluents with plant-based coagulants vary from 100 to 300 rpm for 30 s to 5 min for fast mixing, and 20 to 100 rpm per 5-40 min for fast mixing, followed by sedimentation for 30 min to 24 h.

3.3 PLANT-BASED VERSUS SYNTHETIC COAGULANTS

In effluent treatment, the coagulation stage is conducted by adding a coagulating agent to reduce or remove suspended impurities and other types of pollutants, such as organic compounds [15,74]. The coagulating agent can be synthetic-based or naturally derived [75].

Currently, synthetic coagulants predominate in industrial settings due to their low cost, high efficiency, easy handling, and widespread market availability. $AlCl_3$, $Al_2(SO_4)_3$, $FeCl_3$, and $Fe_2(SO_4)_3$ are the most used coagulants [76]. Although these factors drive the choice of synthetic coagulants over plant-based alternatives, synthetic agents have raised controversial issues due to their low biodegradability and toxicity to living organisms. Residual aluminum concentrations are still detected after treatment [76,77]. Researchers warn that Alzheimer's disease is linked to aluminum neurotoxicity [78]. Furthermore, synthetic coagulants can form dangerous by-products such as acrylamide, a carcinogen and neurotoxic substance [79]. According to Kurniawan et al. [79], the disposal of toxic sludge pollutes water and soil, affecting plant and animal species negatively. As a result, synthetic coagulants are not considered sustainable and do not qualify as green options [76]. Therefore, it is necessary to promote sustainable and equally efficient coagulating agents.

Natural coagulants are biodegradable, non-toxic, non-corrosive, reduce the amount of sludge, and are environmentally friendly [9,17,18,37]. Natural coagulants are recognized in traditional water purification, especially in impoverished communities [20,56]. Based on these applications, natural coagulants have attracted the scientific community's attention due to their significant health and environmental benefits, and their efficiency in removing contaminants.

Several studies have proven the effectiveness of plant-based coagulants in effluents treatment applications: removal of 98% of color and turbidity from dairy industry effluents [9]; 64% COD removal from slaughterhouse effluents [4]; removal of 87% TSS and 55% COD from palm oil milling effluent [6]; reduction of 54% TSS, 100% COD, and 100% nitrate and nitrite from coffee fermentation effluents [44]; reduction of 95% suspended solids and 52.2% COD from palm oil milling effluent [35]; removal of 96.5% color, 87% COD, and 99.9% turbidity from distillery spent wash [46]; and removal of 90% of COD from palm oil milling effluent [39]. However, these studies are massively small-scale. The industrial application of natural coagulants in effluents treatment is still limited, especially owing to a lacking tax incentive and inconsistent information on performance, quality control, and production standards of plant extracts [20,76,80].

Although *Moringa oleifera* is the most commonly used and extensively researched plant-based coagulant [4,29,47,50,75], several others have been identified, such as: Nirmali seeds (*Strychnos potatorum*) [81]; Dolichos lablab or hyacinth bean (*Lablab purpureus*) [69], banana (*Musa paradisica*) [82]; Roselle seeds (*Hibiscus sabdariffa*) [83,84]; cactus species [85]; watermelon seeds [86]; sicklepod (*Cassia obtusifolia*) [6,7]; rice starch [37,38]; okra (*Abelmoschus esculentus*), sandpaper tree (*Ficus exasperate)*, and *Bridelia ferrugeneae* [41].

3.4. ASSOCIATION BETWEEN NATURAL AND SYNTHETIC COAGULANTS

Despite the environmental benefits of using natural coagulants in drinking water and effluents treatment, these might not be as effective as synthetic coagulants. To harness the advantages of natural coagulants without hindering the treatment efficiency, they can be combined with synthetic coagulants either as a composite coagulant or as coagulant aid [87,88]. The former is produced by combining two types of coagulants as a single coagulant, while coagulant aids are materials added some time after the dosing of the primary coagulant. These approaches minimize the consumption of non-renewable coagulants and their associated environmental impact [89].

There are few studies on the association of natural coagulants with synthetic coagulants, such as ferric chloride, polyaluminum chloride, and aluminum sulfate [62]. Shak and Wu [7] reported a 55% reduction in alum when gum extracted from *Cassia obtusifolia* seeds was employed as a coagulant aid. More impurities were adsorbed onto the coagulant aid due to the polymeric structure of the natural gum.

Ghebremichael et al. [90] concluded that dosing the moringa coagulant prior to alum resulted in better removal of turbidity and dissolved organic carbon from surface water (river). Furthermore, the use of the natural coagulant aid reduced the alum dosage by 50-75%. Similar benefits were achieved by dosing alum and moringa coagulants together in concrete effluents treatment [75]. Freitas et al. [91] also highlighted that dosing the moringa coagulant after alum results in greater turbidity removal than dosing them together to treat polluted stream water.

Natural materials that might not perform well as coagulants individually (e.g., lignin, starch, green algae, and cactus mucilage) can be used as coagulant aids or as part of a composite coagulant [13,89,91,92]. These coagulant aids strengthen the flocs through charge neutralization and bonding with the flocs formed by the primary coagulant. As the treatment becomes more effective and the dosage of the primary coagulant is reduced, the residual synthetic coagulant (e.g., aluminum) is also minimized.

4. ACTIVE PRINCIPLE UNDERLYING COAGULATION WITH NATURAL AGENTS

In broad terms, most natural coagulants can be categorized into two groups based on the main compounds with coagulation activity: polysaccharides and proteins. Examples of the first group are plant- or animal-derived materials such as chitosan, starch, and mucilage, while protein-based natural coagulants typically originate from plants [9,93]. The presence of hydroxyl and amino functional groups in these compounds contribute to their coagulation capacity [13].

Plant-based natural coagulants are mostly water-soluble organic molecules [94-96] extracted primarily from seeds [97,99] and mucilages [100,101]. For example, *Moringa oleifera* seeds are used as a coagulant due to the presence of soluble cationic proteins capable of reducing the turbidity of the treated liquid [4,102]. Nordmark et al. [103] isolated eight different cationic protein fractions from aqueous extracts of moringa seeds. In turn, Kwaambwa and Maikokera [104] hypothesized that the active agents responsible for coagulation in moringa are dimeric cationic proteins with a molecular mass in the range of 12–14 kDa and an isoelectric point of pH 10–11, which indicates a highly cationic nature.

Ghebremichael et al. [90] concluded that the active compound in moringa is not a single protein, but rather a mixture of proteins with similar physical characteristics. According to the authors, this protein mixture is heat-resistant and exhibits clotting activity after 5 h of heat treatment at 95°C. This property renders the active ingredient ideal for use in water and effluent treatment systems exposed to high temperatures (35-40°C), as frequently found in tropical countries.

Unlike conventional metallic synthetic coagulants, natural coagulants exhibit particularities that affect the quality control of the active ingredient, rendering it more challenging. This behavior results from the extraction method (which comprises various stages) and the coagulant source – especially plant-based coagulants, where the compounds vary according to the plant species and growth conditions [13].

Worth noting, non-coagulating impurities will inevitably be present in extracted natural coagulants if further purification is not conducted. The presence of impurities that possess no coagulation capacity affects the coagulation efficiency [102]. For example, the lipid fraction may inhibit the contact between coagulant and impurities, rendering the floc formation ineffective, while carbohydrates may increase the organic matter concentration in the solution [102,105]. Extensive purification can be conducted but will incur additional costs. According to the source and extraction approach, the variable performance of natural coagulants might hinder quality control [13].

5. PROCESSING TECHNIQUES OF NATURAL COAGULANTS

Whenever the objective is to utilize the isolated active ingredient (e.g., a particular protein), coagulant preparation must follow three main stages (Figure 4): a) primary – flour preparation; b) secondary – protein extraction; and c) tertiary – purification [18]. The characteristics of the coagulant and its performance in CF are directly influenced by the order of these stages [106].

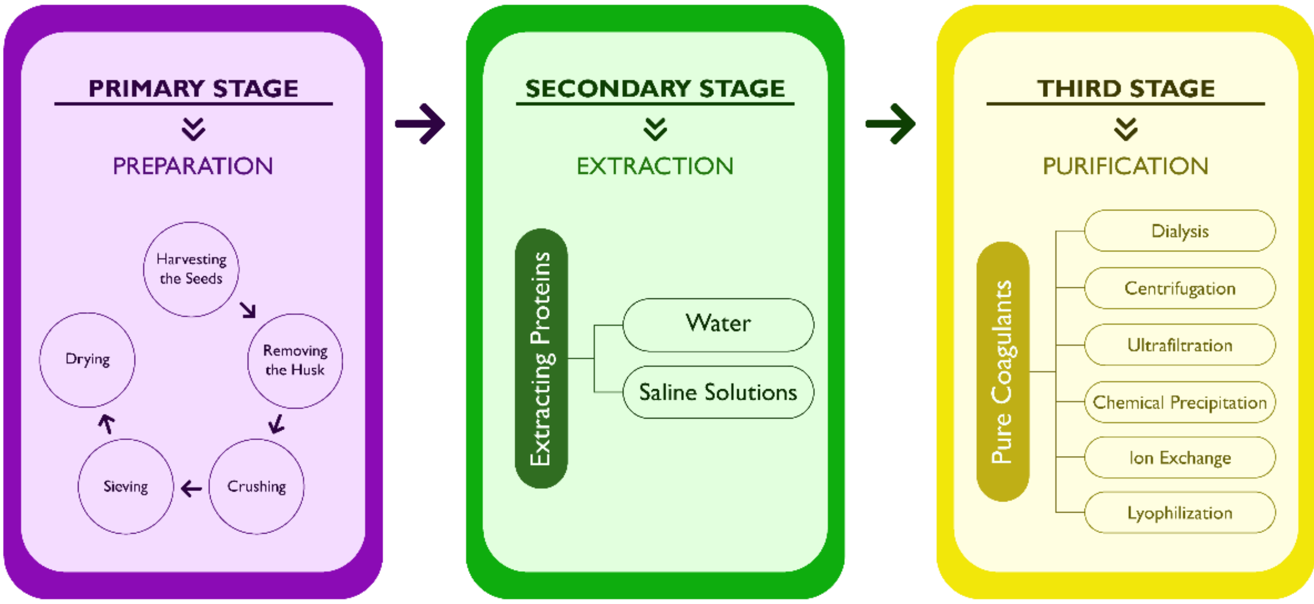

Figure 4 – Diagram of the main processing techniques of natural coagulants.

Using *Moringa oleifera* as a reference, the primary stage includes harvesting the seeds, removing the husk, crushing, sieving, and drying. The recommended particle size for water and effluent treatment varies between 0.25 mm and 1.25 mm [107]. Moringa seeds contain a wide variety of undesirable compounds in CF that increase the organic load in the treated effluents. Therefore, the material obtained in the primary stage is further treated.

The secondary stage consists of extracting proteins with water or saline solutions [18]. The simplest extraction method uses clean water. Jung et al. [108] mixed powdered moringa seeds with water and stirred the mixture to release the coagulating compounds, which are positively charged proteins. However, the coagulation efficiency of this extraction approach is often unsatisfactory due to the presence of non-coagulating impurities and a low number of coagulating compounds [13]. The main disadvantage of water extraction is the increased COD due to the dissolved organic carbon content, which discourages the use in drinking water treatment [109]. In effluents treatment, this method's efficiency is controversial: COD dropped in some studies and increased in others [33].

In the study by Oladoja [14], salt solution extraction exhibited better coagulant capacity than water extraction, which can be explained by the higher number of soluble proteins present in salt extracts. The author recommends using moringa seeds as a coagulant in the treatment of drinking water and wastewater effluents intended for water reuse only after purifying the active proteins.

The coagulation capacity of a particular substance can be improved by using salt solutions (e.g., NaCl and $CaCl_2$) during extraction [110,111]. The use of saline solution enhances protein extraction from natural coagulants due to the salt's capabilities of promoting protein-protein dissociation and increasing the solubility of proteins, which increases the efficiency of the coagulant ion [111]. Megersa et al. [111] demonstrated that the addition of salt solutions (NaCl, KNO_3, and NH_4Cl) enhances the coagulation performance compared to the water-extracted moringa coagulant. The optimum coagulant dosage extracted using 0.5 M NaCl was 2 mg/mL, yielding a 91% turbidity removal. Such improvement is attributed to the increased solubility of proteins from the natural coagulant in the presence of salt due to the breaking down of protein-protein bonds. Carvalho et al. [110] reported that moringa coagulant extracted with $CaCl_2$ resulted in higher turbidity removal compared to NaCl extraction, which could be explained by the participation of calcium ions in the coagulation process.

Apart from increasing the extraction yield (i.e., number of compounds with coagulation capacity), the purity of natural coagulants can be improved to reduce impurity concentration and coagulant dosage. Organic compounds present in natural coagulants that do not take part in coagulation will possibly end up in the treated water as dissolved organic matter [112]. The presence of organic residues is undesirable due to the increased risk of bacterial growth, which can serve as a substrate for the formation of hazardous by-products [113,114]. To avoid such adverse effects, natural coagulants must be purified after extraction.

Only those compounds that contribute to the coagulation properties are purified in the third stage. Regardless of the plant source, coagulating compounds are either polysaccharides or proteins [13]. To obtain pure coagulants, purification can be carried out using a combination of different methods, such as dialysis, centrifugation, ultrafiltration, chemical precipitation, ion exchange, and lyophilization [18,24]. The final extract may differ based on the purification method, and different solvent extractions and extract fractionation will yield different types of products [115].

Sánchez-Martín et al. [112] investigated the impact of ion exchange purification using NaCl solution on the coagulation performance of moringa extract. The optimum dosage of the single-step purified coagulant (elution with 0.6 M NaCl) was two times higher than the two-step purified coagulant (first elution with 0.3 M NaCl followed by a second elution with 0.6 M NaCl) in terms of turbidity removal. This led to the production of more purified coagulants containing active coagulant proteins after the second elution.

The purified coagulant produced via ion exchange performed at par with the conventional alum coagulant in terms of required dosage (1 mg/L) and turbidity removal efficiency (83%) for natural river water [111]. Without further purification, the moringa extract could only achieve 50% turbidity removal at a similar dosage and required fourfold the dosage for 83% removal efficiency. Therefore, the purification process can produce natural coagulants with a higher proportion of active coagulation compounds, which results in better performance even at lower optimal dosage [13].

Due to the high cost of ion-exchange chromatography, researchers have sought alternative purification approaches that are more economically viable at the industrial level [13]. Relatively simple chemical precipitation (using ammonium sulphate) of compounds possessing coagulation activity has been proposed for the purification of moringa extract [95,116]. Choudhary and Neogi [116] reported that the natural moringa extract isolated with saturated ammonium sulphate exhibited superior turbidity removal efficiency than alum. Hence, a simple but effective isolation and purification method for natural moringa extract may result in a natural coagulant with satisfactory coagulation capacity [13].

The methodology involved in the production of moringa coagulant can be replicated to extract and purify natural coagulants from other plant sources – e.g., seeds of plantago (*Plantago major L.*), beans (*Phaseolus vulgaris)*, chestnut (*Bertholletia excelsa*), basil (*Ocimum basilicum)*, oak (*Quercus robur*), hibiscus flower (*Hibiscus L.),* or cactus pads from *Opuntia* species [83,93,117-120]. In general, the extraction and purification of plant-based natural coagulants rely on three stages, as shown in Figure 3. Table 3 presents the manufacturing process of natural coagulants used to treat food industry effluents.

Table 3[3] – Natural coagulants used in the treatment of food industry effluents.

6. MECHANISMS OF COAGULATION WITH NATURAL AGENTS

Natural coagulants can be obtained from plants, animals, and microorganisms. The main mechanisms involved in the coagulation with plant-based coagulants are adsorption and charge neutralization or alternatively adsorption and bond formation. It is difficult to define the exact mechanism because the phenomena can co-occur [121,122]. Adsorption and charge neutralization refer to the sorption of two particulates with oppositely charged ions, while

[3] attached

interparticle bridging occurs when a coagulant provides a polymeric chain that sorbs particulates [101]. These two mechanisms are involved in the coagulant properties of plant-based coagulants [58,101].

Amran et al. [123] discuss in more detail how plant-based coagulants promote the coagulation of pollutants in effluents. In charge neutralization, an ionizable polymer (present in the coagulant) stabilizes the colloidal particles, which otherwise would repel one another for being negatively charged. In polymer adsorption, long-chain polymers partially attach themselves to the surface of the colloidal particles due to the affinity between them. The unattached parts, composed of loops and tails, are the main structure involved in bond formation or polymer bridging. Bridging ensures the formation of pollutant flocs by decreasing interparticle repulsion. Sufficient coagulant dosages will provide the required amount of polymer to promote strong polymer bridging.

In the study by Vieira et al. [9], the sorption potential of moringa coagulant was assessed for color, turbidity, and COD removal in dairy industry effluents (DIE). Maximum adsorption of DIW components from aqueous solutions occurred using 1 g/L of moringa seeds with a 60 min agitation time, which suggested an excellent affinity between DIE components and sorbent. Furthermore, the moringa coagulant showed sorption capacity within a pH range from 5 to 8.

Real-Olvera et al. [4] investigated the adsorption of organic pollutants from slaughterhouse effluents when *M. oleifera* was employed as a natural coagulant. The results indicated that 180 min is necessary for high adsorption. The maximum adsorption capacity of 0.523 g COD/g powder suggests a good affinity between organic pollutants and powdered seeds. Moringa seed powder exhibited sorption capacity over the pH range 5 to 9, but the best results were obtained at high pH levels.

7. BENEFITS OF USING NATURAL COAGULANTS

Natural coagulants overperform synthetic coagulants in terms of biodegradability, non-corrosivity, and lower or non-toxicity [24,54,106] (Figure 5). Such features are attractive for the development of "greener" water and effluents treatment, where processing costs are reduced while color and turbidity removal are improved [122]. In addition, the sludge produced during the coagulation process is innocuous and has a volume 4–5 times smaller than the sludge produced when aluminum sulfate or ferric chloride are used [9,124]. Furthermore, natural coagulants do not significantly change the pH of the medium [124] and do not cause corrosion problems [99].

Figure 5 – Advantages of natural coagulants over synthetic coagulants.

Although efficiency is one of the most critical factors in water and effluents treatment, other criteria are equally relevant for the long-term reliability of sustainable, plant-based coagulants. In effluent treatment, sustainability encompasses the integration of environmental, social, economic, and technical aspects [125] (Figure 6).

Figure 6 – Relevant sustainability criteria in effluent treatment.

8. ECONOMIC ASPECTS OF PLANT-BASED COAGULANTS

Synthetic coagulants are predominant in effluents treatment because of their low cost, easy handling, high availability, and market diversification. Nonetheless, synthetic coagulants do not fit into green chemistry due to the residual aluminum concentrations found after treatment [76,126].

Plant-based coagulants become economically attractive when such advantages as biodegradability, non-toxicity, non-corrosiveness, and easy implementation are factored in [127].

In addition, plant-based coagulants reduce the volume of sludge produced as well as handling and treatment costs due to the biodegradable nature of water treatment residuals [76]. According to Othmani et al. [128], effluents treatment systems will eventually replace synthetic coagulants with plant-based alternatives given their low price, multipurpose application, biodegradability, and abundant sources.

In a world facing a climate emergency, the depletion of natural resources, and widespread environmental degradation, plant-based coagulants are in line with global initiatives for sustainable development [62]. The necessary advance of environmentally sustainable technologies will contribute to the broader use of plant-sourced coagulants, reducing production costs due to economies of scale. Moreover, plant-based coagulants will become ever more competitive as the high performance in effluents treatment demonstrated in the scientific literature is translated into industrial practice [23].

Contrary to the abundant studies on the efficiency of plant-based coagulants to treat effluents (especially from the food industry), direct comparisons of production costs and end prices between natural-origin and synthetic coagulants are incredibly scarce. According to Vijayaraghavan et al. [62] and Yin [23], the attractive price of *M. oleifera* coagulant compared to other natural coagulants is the result of the plant's widely publicized advantages and the coagulant's broad application across different effluent treatment systems.

A single *M. oleifera* tree produces approximately 2,000 seeds per year. This amount can treat 6,000 L of water at a dosage of 50 mg/L [18]. If fully grown, the yield of a single tree can increase by up to 20,000 seeds, which would be enough to treat 60,000 L of water per year [18]. A study by the Water and Environmental Health at London and Loughborough (WELL) [129] shows that an *M. oleifera* tree producing an average of 3 kg of seeds can treat 30,000 L of effluent at a dose of 100 mg/ L. In about 1 ha, it would be possible to harvest 3,000 kg of seeds, which would be able to treat 30,000 m^3 of water [18].

The production costs of moringa seed depend on several factors, including harvest yield, climate, and agricultural practices. For perspective, the purchase price of moringa seed in Malawi was £7.5 per 1000 m^3 of treated water in 1994, while the cost of alum and soda ash was £49.8 [130]. In Malaysia, however, the production cost of 1 kg of *M. oleifera* in 2006 was approximately US$2, twice that of alum [59].

The main elements hindering the pricing and commercialization of plant-based coagulants include: (a) insufficient plantations to allow bulk processing of the source plants; (b) seasonal variations of the raw material; (c) little market demand; (d) incipient specialized knowledge and research development on economically viable extraction methods, storage and preservation conditions, and optimal effluents treatment parameters; (e) currently limited industrial use; (f) shortfall of market awareness and interest; (g) well-established and competitive market focused on synthetic coagulants; and (h) absence of production regulation and quality control for plant extracts [13,20,76,80].

While the cost-effectiveness of plant-based coagulants is currently restricted to small-scale use and academic research, exploration can be widened upon stakeholder endorsement [18,62]. Governmental and non-governmental regulatory bodies should implement environmental rules that encourage industries to use natural coagulants, including reduced tax payments and facilitated loans to subsidize implementation costs [77,80]. Furthermore, research should be advanced to identify the best natural coagulants with economic and industrial potential [76].

9. RECENT ADVANCES AND FUTURE PERSPECTIVES FOR PLANT-BASED COAGULANTS IN WATER TREATMENT

Plants with coagulation capacity in addition to Moringa oleifera

Othmani et al. [128] evaluated the performance of moringa, cactus, okra, and mango in freshwater and effluents remediation. According to the researchers, not only do the selected plants belong to different families, but also their coagulant capacity depends on the parts from which the coagulant agents are extracted. The parts of interest are seeds for moringa and mango, seed pods for okra, and pad for cactus. The coagulant capacity of these materials depends on the active agent extracted (polysaccharides, proteins, or both), which produces a variation in the removal rate of contaminants among plants.

Thakur and Choubey [131] observed an 80.7% turbidity reduction when treating synthetic cloudy water with moringa. When using okra, the removal rate was 78.7%. Kazi and Virupakshi [132] demonstrated that moringa seed reduced 82% and 83% of turbidity and COD, respectively, of tannery effluent, while cactus mucilage exhibited removal percentages of 78.5% and 80.6% for turbidity and COD, respectively. Qureshi et al. [133] reported that mango provided a 98% turbidity reduction versus 86% for moringa. In their study, Othmani et al. [128] found that the sequence mango > moringa > cactus = okra defines the plants' ability to remove turbidity. Given these results, mango should be further investigated for its coagulation capacity. In addition, active agents should be better explored. Although previous studies claim that proteins are the only active coagulant compound in plants, Seghosime et al. [134] hypothesize that the high carbohydrate content (76.73%) in mango seeds contributes to the plants' coagulant activity.

Therefore, the valorization and research of biomaterials for water and effluent remediation can be considered a gateway to applying green chemistry and clean technology in treatment systems [128].

Integrated process

An integrated process is a strategy to enhance the use of natural coagulants by combining coagulants with other treatment technologies. The treatment goals are met by utilizing the strength of each process while minimizing their shortcomings [13]. An example is integrating a coagulation stage (with natural coagulants) with a biological treatment. Hameed et al. [135] implemented a coagulation step (with a tannin-based coagulant) before the biological treatment. The integration reduced the organic load content, generating savings due to the lower need for aeration. Pavón-Silva et al. [136] demonstrated that combining coagulation and biological treatment resulted in COD removal rates of up to 99% in the treatment of food industry effluents. These results indicate that natural coagulants improve the performance of the subsequent treatment steps.

Another example is integrating coagulation with membrane systems, where the tendency towards membrane fouling is minimized [15]. Chitosan, utilized as a natural coagulant, improved nanofiltration membrane fouling in relation to organic matter by neutralizing the charges and weakening the antifouling capability (electrostatic repulsion) of the membrane [12]. Furthermore, integrating coagulation with electrolysis further refined the quality of the supernatant (by degrading the organic compounds in the effluent), making it suitable for discharge into water bodies [13]. Integrated processes involving coagulation and other treatments improve the quality of the treated effluent, allowing its safe disposal. Natural coagulants can be used in pre- or post-treatments combined with other technologies to benefit the entire process [13].

Improved extraction and purification of natural coagulants

Unlike synthetic coagulants, where quality characteristics are precisely controlled, the quality control of natural coagulants is challenging. Such peculiarity is associated with the synthesis of coagulants, which can be obtained through various processes and sources. To overcome this disadvantage and produce a pure natural coagulant, it is necessary to optimize the extraction process. The simplest extraction method uses water. However, the efficiency of this approach is usually unsatisfactory. This can be circumvented by optimizing the extraction method and including a purification step.

The coagulation capacity can be increased by using saline solutions (e.g., NaCl and $CaCl_2$) during the extraction step [110,111]. Megersa et al. [111] found that the use of saline solutions (NaCl, KNO_3 and NH_4Cl) increased the turbidity removal efficiency of moringa from 37% using water extraction to 91%. This result can be explained by the Debye-Huckel theory (increasing solvating power) [13]. Salts increase protein solubility by breaking down the protein-protein bonds of natural coagulants [111]. The number of active compounds with coagulation capacity in the coagulants extracted with saline solutions was higher, which reduces the required dosage for similar or superior performance [13,111].

The presence of organic residues in the extracted coagulant is undesirable because it promotes bacterial growth and serves as a substrate for by-product formation [113]. Therefore, the extracted natural coagulants must be purified. The most used purification method consists of removing the oil fraction in the coagulant extract through solvents, such as hexane and ethanol [111]. The decrease in lipid content enhances the removal of turbidity and COD, since oils hinder the coagulation process by adding impurities to the treated water [13]. Although this purification improves the performance of the natural coagulant, the extract can be further purified, yielding a final product with a high concentration of coagulation active compounds. To this end, several purification processes are being explored, such as chemical precipitation, dialysis, lyophilization, ultrafiltration, and ion exchange [95,105,112,116]. Sánchez-Martín et al. [112] investigated the effect of purification by ion exchange chromatography on the coagulation performance of moringa. The ideal coagulant dosage in a single elution step was twice that of the coagulant purified in two steps.

Due to the high cost of ion-exchange chromatography, researchers have been investigating alternative purification approaches [13]. An example is chemical precipitation with ammonium sulphate for extract purification [95,116]. Choudhary and Neogi [116] reported that moringa extract isolated with 30-60% and 60-80% saturated ammonium sulphate increased turbidity removal capacity compared to alum. This study suggests that a simple but effective purification method can produce natural coagulants with superior coagulation capacity.

Modification for improved performance

Natural coagulants exhibit certain limitations that can hinder their industrial application, including low solubility in water, undesirable isoelectric point, low stability (shelf life), narrow working window of pH range, weak surface charge, and moderate efficiency (translated to higher working dosage and cost) [70,131]. Reactive functional groups (e.g., hydroxyl, amino, and carboxyl) of natural coagulants promote modifications that can solve those concerns [13,76]. A study on the incorporation of valuable compounds through interaction with functional groups was carried out to remedy the negative points of natural coagulants. Generally, chemical reactions such as graft co-polymerization, crosslinking with aldehyde, esterification, etherification, amination, carboxyalkylation, hydroxyalkylation, and condensation have been adopted to improve the characteristics of natural coagulants [73].

Quarternized carboxymethyl chitosan is a modified chitosan used in coagulation processes. By including carboxymethyl and grafting of quarternary ammonium groups ((3-chloro-2-hydroxypropy)-trimethylammonium chloride) onto the chitosan backbone, the isoelectric point of chitosan is dislocated from 6,0-6,5 to beyond pH 9, which expands the use for alkaline effluents and effluents [137]. Furthermore, the water solubility of modified chitosan is improved. The combination of improved water solubility, high isoelectric point, and more positively charged functional groups enhances the performance of modified chitosan in removing contaminants compared to unmodified chitosan and synthetic coagulants.

Huang et al. [138] found that the positive charge of starch can be increased by introducing quaternary ammonium salt groups ((2-methacryloyloxyethyl) trimethylammonium chloride) onto the starch polymer backbone. A highly positive charge is desirable for coagulants because most suspended colloidal particles exhibit a negative surface charge. In the study by Huang et al. [138], a low dosage (0.5-0.7 ppm) of modified starch coagulant promoted removal rates as of 98%.

In addition to chemical modification and the search for efficient extraction and purification processes, natural coagulants can be combined with other coagulants (usually synthetic) [70,139]. More details on hybrid coagulants are presented in topic 3.4.

Multifunctional natural coagulants

As real effluents contain large amounts of impurities, various forms of treatment are necessary to remove the pollutants. The need for additional steps, sophisticated equipment, longer treatment time, and labor can increase the process costs [13]. Therefore, the use of multifunctional coagulants – capable of removing several types of pollutants in parallel – renders water and effluent treatment plants more compact and efficient [140].

The removal of suspended particles is the most common parameter to evaluate the performance of natural coagulants. However, the application of natural coagulants for the removal of pollutants such as heavy metals, organic compounds, and other contaminants should also be investigated. Typically, natural coagulants exhibit poor removal performance for dissolved contaminants [71,141]. Regardless of the coagulant used (natural or synthetic), coagulation has a low capacity to remove soluble heavy metals from effluents [70].

Consequently, coagulation must be integrated with other stages and unit operations to guarantee the complete removal of heavy metals and other dissolved pollutants [13,70].

To enhance the ability of the natural coagulant to remove multiple contaminants, functional groups with an affinity for such pollutants can be incorporated into the coagulant's molecular chain to produce modified coagulants. Yang et al. [142] introduced a dithiocarboxy group to the OH functional group on chitosan to produce xanthated chitosan. The researchers reported that copper (a heavy metal) and kaolin (responsible for turbidity) were successfully removed. Heavy metal chelating functional groups grafted onto natural coagulants allow the removal of coexisting copper, chromium, and nickel ions through the chelation process [143].

Viability of natural coagulants

Natural coagulants (modified, unmodified or hybrid) that demonstrate promising performance in laboratory tests should be scaled up to pilot and industrial-scale applications. The industrial viability of natural coagulants as to their performance, costs, and technical aspects should also be further investigated. Furthermore, the potential of plant-based coagulants in rural and impoverished areas should be explored, as natural coagulants can be locally sourced. Moreover, quality control tools can be used to standardize and optimize production [13,18,77].

Sustainability of natural coagulants

The investigation of sustainability criteria (environmental, economic, and social) will help elucidate uncertainties about natural coagulants. A constant supply of raw materials for the extraction and synthesis of natural coagulants might be necessary to ensure competition with conventional coagulants. A comprehensive cost study is needed to define the economic viability of natural coagulants, comprising the cost of raw materials, extraction, and purification.

Investigating the health and environmental impacts of natural coagulants (especially modified natural coagulants) can provide relevant results. In addition, a life cycle assessment may indicate opportunities for improvement in the coagulation process with natural agents. Finally, an analysis based on circular economy and bioeconomy concepts can contribute to the development and sustainability of natural coagulants used to treat effluents [13,18,74,77].

10. CONCLUSIONS

Despite the encouraging results reported on plant-based coagulants to treat food industry effluents, the number of information gaps identified in this research field is significant. For instance, only a few studies have elucidated the underlying coagulation mechanisms of natural coagulants, while the analysis of the coagulation efficiency of different plant-based materials for various types of effluents and coagulant combinations is still in its infancy. Studies reporting the economic aspects are exceptionally scarce. Despite the opportunities for improvement, the current scientific literature reports vital results:

- Plant-based coagulants are efficient for turbidity and pollutant removal;
- Although purification is not mandatory, it is frequently used to prevent an increase in organic matter concentration after the effluents treatment;
- High cost-efficiency and low toxicity are among the fundamental advantages of plant-based coagulants, both as sole coagulants and coagulation aid;
- The utilization of natural coagulants to treat food industry effluents has been limited to academic research. These studies must be scaled-up given the high potential of natural coagulants in sustainable environmental technologies;

- Among the factors hindering further use of natural coagulants are the limited cultivation of plants with coagulant properties and the lack of regulation specifying the quality of processed coagulant extracts;
- *M. oleifera* is the most researched source of plant-based coagulants. The results summarized herein for moringa can be employed to optimize process conditions for this species or can be used as a starting point for the investigation of other plant species.

The industrial acceptance of natural coagulants over conventional coagulants is still negligible. We trust this review paper will contribute to a more widespread application of natural coagulants by showcasing their properties.

ACKNOWLEDGMENTS

The authors would like to thank the following for their support: Federal University of Paraná, Federal University of Mato Grosso do Sul, SESI (Serviço Social da Indústria) of Mato Grosso do Sul, CAPES (Coordination for the Improvement of Higher Education Personnel), and CNPq (The National Council for Scientific and Technological Development).

REFERENCES

1. J.C. Garcia, J.L. Oliveira, A.E.C. Silva, C.C. Oliveira, J. Nozaki, N.E. de Souza, Comparative study of the degradation of real textile effluents by photocatalytic reactions involving UV/TiO2/H2O2 and UV/Fe2+/H2O2 systems, J. Hazard. 147 (2006) 105–110. https://doi.org/10.1016/j.jhazmat.2006.12.053.

2. M.I. Aguilar, J. Sáez, M. Lloréns, A. Soler, J.F. Ortuño, Microscopic observation of particle reduction in slaughterhouse wastewater by coagulation-flocculation using ferric sulphate as coagulant and different coagulant aids, Water Res. 37 (2003) 2233–2241. https://doi.org/10.1016/S0043-1354(02)00525-0.

3. S.A. Hosseiny, D. Huisingh, Evaluation of biogas capacity produced by slaughterhouse wastewater in Iran, 18th Greening of Industry Network Conference, Sweden, 2012.

4. J. del Real-Olvera, E. Rustrian-Portilla, E. Houbron, F.J. Landa-Huerta, Adsorption of organic pollutants from slaughterhouse wastewater using powder of Moringa oleifera seeds as a natural coagulant, Desalin. 57(21) (2015) 9971–9981. https://doi.org/10.1080/19443994.2015.1033479.

5. J.D.R. Olvera, A. Lopez-Lopez, Biogas production from anaerobic treatment of agro-industrial wastewater, in: S. Kumar (Ed.) Biogas, InTech, 2012, pp 91–112.

6. K.P.Y. Shak, T.Y. Wu, Coagulation-flocculation treatment of high-strength agro-industrial wastewater using natural Cassia obtusifolia seed gum: Treatment efficiencies and flocs characterization, Chem. Eng. J. 256 (2014) 293–305. https://doi.org/10.1016/j.cej.2014.06.093.

7. K.P.Y. Shak, T.Y. Wu, Optimized use of alum together with unmodified Cassia obtusifolia seed gum as a coagulant aid in treatment of palm oil mill effluent under natural pH of wastewater. Ind. Crops Prod. 76 (2015) 1169–1178. https://doi.org/10.1016/j.indcrop.2015.07.072.

8. S. Suhartini, N. Hidayat, E. Rosaliana, Influence of powdered Moringa oleifera seeds and natural filter media on the characteristics of tapioca starch wastewater, IJROWA, 2 (2013) 1–11. https://doi.org/10.1186/2251-7715-2-12.

9. A.M.S. Vieira, M.F. Vieira, G.F. Silva GF, A.A. Araújo, M.R. Fagundes-Klen, M.T. Veit, R. Bergamasco, Use of Moringa oleifera seed as a natural adsorbent for wastewater treatment. Water Air Soil Pollut. 206 (2010) 273–281. https://doi.org/10.1007/s11270-009-0104-y.

10. M. Khayet, A.Y. Zahrim, N. Hilal, Modelling and optimization of coagulation of highly concentrated industrial grade leather dye by response surface methodology. Chem. Eng. J. 167 (2011) 77–83. https://doi.org/10.1016/j.cej.2010.11.108.

11. P.R.F. da Costa, D.R. da Silva, C.A. Martínez-Huitle, S. Garcia-Segura, Fuel station effluent treatment by electrochemical technology. J. Electroanal. 763 (2016) 97–103. https://doi.org/10.1016/j.jelechem.2015.12.038.

12. W.L. Ang, A.W. Mohammad, Integrated and hybrid process technology, in: Sustainable Water and Wastewater Processing, Elsevier, 2019: pp. 279–328. https://doi.org/10.1016/B978-0-12-816170-8.00009-0.

13. W.L. Ang, A.W. Mohammad, State of the art and sustainability of natural coagulants in water and wastewater treatment, J. Clean. 262 (2020) 121267. https://doi.org/10.1016/j.jclepro.2020.121267.

14. N.A. Oladoja, Headway on natural polymeric coagulants in water and wastewater treatment operations. J. Water Process Eng. 6 (2015) 174–192. https://doi.org/10.1016/j.jwpe.2015.04.004.

15. J.-Q. Jiang, The role of coagulation in water treatment, Curr. Opin. Chem. Eng. 8 (2015) 36–44. https://doi.org/10.1016/j.coche.2015.01.008.

16. MarketsandMarkets, Flocculant and coagulant market by type (flocculant (anionic, cationic), organic coagulant, and inorganic coagulant), end-use industry (municipal water treatment, pulp & paper, textile, oil & gas, mining), and region-global forecast to 2022. Available at: https://www.marketsandmarkets.com/Market-Reports/flocculant-and-coagulant-market-243584994.html?gclid=CjwKCAjw2bmLBhBREiwAZ6ugo9IgYtgcKoy4tuXRD69hQ3gJMyZ5h0rS6QTpkWeYW8wA5evwJaiqqxoChyQQAvD_BwE. Accessed 18 Oct 2021.

17. H. Bhuptawat, G.K. Folkard GK, S. Chaudhari, Innovative physico-chemical treatment of wastewater incorporating Moringa oleifera seed coagulant. J. Hazard, 142 (2007) 477–482. https://doi.org/10.1016/j.jhazmat.2006.08.044.

18. Kansal SK, Kumari A (2014) Potential of M. oleifera for the treatment of water and wastewater. Chemical Reviews 114:4993–5010.

19. Pritchard M, Craven T, Mkandawire T, et al (2010) A comparison between Moringa oleifera and chemical coagulants in the purification of drinking water - An alternative sustainable solution for developing countries. Physics and Chemistry of the Earth 35:798–805. https://doi.org/10.1016/j.pce.2010.07.014

20. Choy SY, Prasad KMN, Wu TY, et al (2014) Utilization of plant-based natural coagulants as future alternatives towards sustainable water clarification. Journal of Environmental Sciences (China) 26:2178–2189

21. di Bernardo L, Dantas ÂDB (2005) Métodos e técnicas de tratamento de água, 2nd ed. RiMa, São Carlos, Brazil.

22. Jodi ML, Birnin-Yauri UA, Yahaya Y, Sokoto MA (2012) The use of some plants in water purification

23. Yin CY (2010) Emerging usage of plant-based coagulants for water and wastewater treatment. Process Biochemistry 45:1437–1444

24. Okuda T, Baes AU, Nishijima W, Okada M (2001) Coagulation mechanism of salt solution-extracted active component in Moringa oleifera seeds. Water Research 35(3):830–4.

25. Junior AT, Hasan SDM, Sebastien NY (2019) Optimization of Coagulation/Flocculation Treatment of Brewery Wastewater Employing Organic Flocculant Based of Vegetable Tannin. Water, Air, and Soil Pollution 230:. https://doi.org/10.1007/s11270-019-4251-5

26. Maximova N, Dahl O (2006) Environmental implications of aggregation phenomena: Current understanding. Current Opinion in Colloid and Interface Science 11:246–266

27. Pavanelli G, di Bernardo L (2001) Eficiência de diferentes tipos de coagulantes na coagulação, floculação e sedimentação de água com cor ou turbidez elevada. Master's dissertation, Escola de Engenharia de São Carlos (Brazil).

28. Junho AL, Santos IFS dos, Silva AML, et al (2021) Treatment of wastewater from the dairy industry with Moringa Oleifera using two different methods. Research, Society and Development 10:e21710716514. https://doi.org/10.33448/rsd-v10i7.16514.

29. George A, Roshan J, Emmanuel J (2016) Moringa oleifera- A Herbal Coagulant for Wastewater Treatment. International Journal of Science and Research (IJSR) 5:

30. dela Justina M, Rodrigues Bagnolin Muniz B, Mattge Bröring M, et al (2018) Using vegetable tannin and polyaluminium chloride as coagulants for dairy wastewater treatment: A comparative study. Journal of Water Process Engineering 25:173–181. https://doi.org/10.1016/J.JWPE.2018.08.001.

31. Mateus GAP, Formentini-Schmitt DM, Nishi L, et al (2017) Coagulation/Flocculation with Moringa oleifera and Membrane Filtration for Dairy Wastewater Treatment. Water, Air, and Soil Pollution 228:1–13. https://doi.org/10.1007/S11270-017-3509-Z/FIGURES/6.

32. Triques CC, Fagundes-Klen MR, Suzaki PYR, et al (2020) Influence evaluation of the functionalization of magnetic nanoparticles with a natural extract coagulant in the primary treatment of a dairy cleaning-in-place wastewater. Journal of Cleaner Production 243:118634. https://doi.org/10.1016/J.JCLEPRO.2019.118634.

33. Francisco JP, Silva JBG, Roque OCC, et al (2014) EVALUATION OF THE EFFECT OF THE SEED EXTRACT OF MORINGA OLEIFERA LAM OVER THE EFFICIENCY OF ORGANIC FILTERS IN WASTEWATER TREATMENT OF DAIRY CATTLE BREEDING. Eng Agríc 34:143–152.

34. Bhatia S, Othman Z, Ahmad AL (2007) Pretreatment of palm oil mill effluent (POME) using Moringa oleifera seeds as natural coagulant. Journal of Hazardous Materials 145:120–126. https://doi.org/10.1016/j.jhazmat.2006.11.003.

35. Bhatia S, Othman Z, Ahmad AL (2007) Coagulation-flocculation process for POME treatment using Moringa oleifera seeds extract: Optimization studies. Chemical Engineering Journal 133:205–212. https://doi.org/10.1016/j.cej.2007.01.034

36. Bhatia S, Othman Z, Ahmad AL (2006) Palm oil mill effluent pretreatment using

Moringa oleifera seeds as an environmentally friendly coagulant: laboratory and pilot plant studies. Journal of Chemical Technology & Biotechnology 81:1852–1858. https://doi.org/10.1002/JCTB.1619.

37. Teh CY, Wu TY, Juan JC (2014) Potential use of rice starch in coagulation–flocculation process of agro-industrial wastewater: Treatment performance and flocs characterization. Ecological Engineering 71:509–519. https://doi.org/10.1016/J.ECOLENG.2014.07.005.

38. Teh CY, Wu TY, Juan JC (2014) Optimization of agro-industrial wastewater treatment using unmodified rice starch as a natural coagulant. Industrial Crops and Products 56:17–26. https://doi.org/10.1016/J.INDCROP.2014.02.018

39. Mohamed Noor MH, Lee WJ, Mohd Azli MFZ, et al (2021) Microwave- vs Ultrasonic-synthesisof magnetic Moringa oleifera coagulant for the reduction of chemical oxygen demand in palm oil wastewater. Environmental Technology & Innovation 24:102069. https://doi.org/10.1016/J.ETI.2021.102069

40. A.H. Jagaba, S.R.M. Kutty, G. Hayder, A.A.A. Latiff, N.A.A. Aziz, I. Umaru, A.A.S. Ghaleb, S. Abubakar, I.M. Lawal, M.A. Nasara, Sustainable use of natural and chemical coagulants for contaminants removal from palm oil mill effluent: A comparative analysis, Ain Shams Engineering Journal. 11 (2020) 951–960. https://doi.org/10.1016/J.ASEJ.2020.01.018.

41. A.S. Adewuyi, J.R. Adewumi, Optimization of Coagulation-Flocculation Process for the Treatment of Wastewater Using Inorganic and Three Natural Coagulants, FUTA JOURNAL OF ENGINEERING AND ENGINEERING TECHNOLOGY. 12 (2018) 260–265. https://doi.org/10.22502/JLMC.

42. dos Santos JD, Veit MT, Juchen PT, et al (2018) Use of different coagulants for cassava processing wastewater treatment. Journal of Environmental Chemical Engineering 6:1821–1827. https://doi.org/10.1016/J.JECE.2018.02.039

43. Matos AT, Cabanellas CFG, Cecon PR, et al (2007) Effects from the concentration of coagulants and pH solution on the turbidity of the recirculating water used in the coffee cherry processing . Eng Agríc 27:544–551

44. Garde WK, Buchberger SG, Wendell D, Kupferle MJ (2017) Application of Moringa Oleifera seed extract to treat coffee fermentation wastewater. Journal of Hazardous Materials 329:102–109. https://doi.org/10.1016/j.jhazmat.2017.01.006

45. Prasad RK (2009) Color removal from distillery spent wash through coagulation using Moringa oleifera seeds: Use of optimum response surface methodology. Journal of Hazardous Materials 165:804–811. https://doi.org/10.1016/j.jhazmat.2008.10.068

46. David C, Narlawar R, Arivazhagan M (2016) Performance Evaluation of Moringa oleifera Seed Extract (MOSE) in Conjunction with Chemical Coagulants for Treating Distillery Spent Wash. Indian Chemical Engineer 58:189–200. https://doi.org/10.1080/00194506.2015.1006147

47. Lo Monaco PAV, de Matos AT, Ribeiro ICA, et al (2010) Use of extract of moringa seeds as coagulant agent in treatment of water supply and wastewater. Ambi-Agua 5:222–231. https://doi.org/10.4136/ambi-agua.164

48. Bindes MM, Reis MHM, Cardoso VL, Boffito DC (2019) Ultrasound-assisted extraction of bioactive compounds from green tea leaves and clarification with natural coagulants (chitosan and Moringa oleifera seeds). Ultrasonics Sonochemistry 51:111–119. https://doi.org/10.1016/J.ULTSONCH.2018.10.014

49. Altenor S, Gaspard S (2014) Biomass for water treatment: Biosorbent, coagulants and flocculants. RSC Green Chemistry.

50. Valverde KC, Moraes LCK, Bongiovani MC, et al (2013) Coagulation diagram using the Moringa oleifera Lam and the aluminium sulphate, aiming the removal of color and turbidity of water. Acta Scientiarum Technology 5:485–489. https://doi.org/10.4025/actascitechnol.v35i3.12268

51. Kim S-H, Moon B-H, Lee H-I (2001) Effects of pH and dosage on pollutant removal and floc structure during coagulation. Microchemical Journal 68:197–203

52. Madrona GS, Serpelloni GB, Salcedo Vieira AM, et al (2010) Study of the effect of Saline solution on the extraction of the Moringa oleifera seed's active component for water treatment. Water, Air, and Soil Pollution 211:. https://doi.org/10.1007/s11270-009-0309-0

53. Baghvand A, Zand AD, Mehrdadi N, Karbassi A (2010) Optimizing coagulation process for low to high turbidity waters using aluminum and iron salts. American Journal of Environmental Sciences 6:. https://doi.org/10.3844/ajessp.2010.442.448

54. Okuda T, Baes AU, Nishijima W, Okada M (1999) Improvement of extraction method of coagulation of active components from Moringa oleifera seed. Water Research 33:3373–3378. https://doi.org/10.1016/S0043-1354 (99)00046-9

55. Duan J, Gregory J (2003) Coagulation by hydrolysing metal salts. Advances in Colloid and Interface Science 100–102:475–502. https://doi.org/10.1016/S0001-8686(02)00067-2

56. Choy SY, Prasad KMN, Wu TY, Ramanan RN (2015) A review on common vegetables and legumes as promising plant-based natural coagulants in water clarification. International Journal of Environmental Science and Technology 12:367–390.

57. Ali GH, El-Taweel GE, Ali MA (2004) The cytotoxicity and antimicrobial efficiency of Moringa oleifera seeds extracts. International Journal of Environmental Studies 61:699–708. https://doi.org/10.1080/0020723042000189877

58. Bolto B, Gregory J (2007) Organic polyelectrolytes in water treatment. Water Research 41:230

59. Katayon S, Noor MJMM, Asma M, et al (2006) Effects of storage conditions of Moringa oleifera seeds on its performance in coagulation. Bioresource Technology 97: https://doi.org/10.1016/j.biortech.2005.07.031

60. Awad M, Wang H, Li F (2013) Preliminary Study on Combined Use of Moringa Seeds Extract and PAC for Water Treatment. Research Journal of Recent Sciences 2:

61. O.Sujana;, K.SyamalaDevi, M.Sreevalli (2019) Application of Natural Coagulants in Waste Water Treatment. Journal of Emerging Technologies and Innovative Research 6:

62. Vijayaraghavan, G.; Sivakumar T; VK (2011) Application of Plant Based Coagulants for Waste Water Treatment. International Journal of Advanced Engineering Research and Studies 1:

63. Prasad SVM, Rao BS (2016) Influence of Plant-Based Coagulants in Waste Water Treatment. Ijltemas V:

64. C.S. Lee, J. Robinson, M.F. Chong, A review on application of flocculants in wastewater treatment, Process Safety and Environmental Protection. 92 (2014) 489–508. https://doi.org/10.1016/j.psep.2014.04.010.1–2324.

65. A. Nath, A. Mishra, P.P. Pande, A review natural polymeric coagulants in wastewater treatment, Mater Today Proc. 46 (2021) 6113–6117. https://doi.org/10.1016/J.MATPR.2020.03.551.

66. V. Kumar, N. Othman, S. Asharuddin, Applications of Natural Coagulants to Treat Wastewater − A Review, MATEC Web of Conferences. 103 (2017) 06016. https://doi.org/10.1051/MATECCONF/201710306016.

67. V. Patale, J. Pandya, Mucilage extract of *Coccinia indica* fruit as coagulant-flocculent for turbid water treatment, Asian Journal of Plant Science and Research. 2 (2012) 442–445. www.pelagiaresearchlibrary.com.

68. M. Agarwal, S. Rajani, A. Mishra, J.S.P. Rai, Utilization of okra gum for treatment of tannery effluent, International Journal of Polymeric Materials and Polymeric Biomaterials. 52 (2003) 1049–1057. https://doi.org/10.1080/714975900.

69. B.S. Shilpa, P. Girish, Evaluation of Cactus and Hyacinth Bean Peels as Natural Coagulants, International Journal of Chemical and Environmental Engineering. 3 (2012). https://doi.org/10.13140/RG.2.2.31066.98247.

70. Lee KE, Morad N, Teng TT, Poh BT (2012) Development, characterization and the application of hybrid materials in coagulation/flocculation of wastewater: A review. Chemical Engineering Journal 203:370–386.

71. C.Y. Teh, P.M. Budiman, K.P.Y. Shak, T.Y. Wu, Recent Advancement of Coagulation–Flocculation and Its Application in Wastewater Treatment, Ind Eng Chem Res. 55 (2016) 4363–4389. https://doi.org/10.1021/ACS.IECR.5B04703.

72. G. Zhang, K. Huang, X. Jiang, D. Huang, Y. Yang, Acetylation of rice straw for thermoplastic applications, Carbohydr Polym. 96 (2013) 218–226. https://doi.org/10.1016/j.carbpol.2013.03.069.

73. B.Y. Gao, Q.Y. Yue, Y. Wang, Coagulation performance of polyaluminum silicate chloride (PASiC) for water and wastewater treatment, Sep Purif Technol. 56 (2007) 225–230. https://doi.org/10.1016/j.seppur.2007.02.003.

74. N.A. Oladoja, Perspectives on the use of equilibrium isotherm equations to elucidate coagulation–flocculation mechanisms in plant-based coagulants, Ind Crops Prod. 70 (2015) 211–212. https://doi.org/10.1016/J.INDCROP.2015.03.038.

75. de Paula HM, de Oliveira Ilha MS, Andrade LS (2014) Concrete plant wastewater treatment process by coagulation combining aluminum sulfate and Moringa oleifera powder. Journal of Cleaner Production 76:125–130. https://doi.org/10.1016/j.jclepro.2014.04.031.

76. S. Nimesha, C. Hewawasam, D. Jayasanka, Y. Murakami, N. Araki, N. Maharjan, Effectiveness of natural coagulants in water and wastewater treatment, Global Journal of Environmental Science and Management. 8 (2022).

77. K.F.S. Freitas, C.A. Almeida, D.D. Manholer, H.C.L. Geraldino, M.T.F. de Souza, J.C. Garcia, Review of Utilization Plant-Based Coagulants as Alternatives to Textile Wastewater Treatment, in: 2018: pp. 27–79. https://doi.org/10.1007/978-981-10-4780-0_2.

78. T.K.F.S. Freitas, V.M. Oliveira, M.T.F. de Souza, H.C.L. Geraldino, V.C. Almeida, S.L. Fávaro, J.C. Garcia, Optimization of coagulation-flocculation process for treatment of industrial

textile wastewater using okra (A. esculentus) mucilage as natural coagulant, Ind Crops Prod. 76 (2015) 538–544. https://doi.org/10.1016/j.indcrop.2015.06.027.

79. S.B. Kurniawan, S.R.S. Abdullah, M.F. Imron, N.S.M. Said, N. 'Izzati Ismail, H.A. Hasan, A.R. Othman, I.F. Purwanti, Challenges and opportunities of biocoagulant/bioflocculant application for drinking water and wastewater treatment and its potential for sludge recovery, Int J Environ Res Public Health. 17 (2020) 1–33. https://doi.org/10.3390/ijerph17249312.

80. A. Ahmad, S.B. Kurniawan, S.R.S. Abdullah, A.R. Othman, H.A. Hasan, Exploring the extraction methods for plant-based coagulants and their future approaches, Science of The Total Environment. (2021) 151668. https://doi.org/10.1016/J.SCITOTENV.2021.151668.

81. G. Prabhakaran, M. Manikandan, M. Boopathi, Treatment of textile effluents by using natural coagulants, in: Mater Today Proc, Elsevier Ltd, 2020: pp. 3000–3004. https://doi.org/10.1016/j.matpr.2020.03.029.

82. A. Daverey, N. Tiwari, K. Dutta, Utilization of extracts of Musa paradisica (banana) peels and Dolichos lablab (Indian bean) seeds as low-cost natural coagulants for turbidity removal from water, Environmental Science and Pollution Research. 26 (2019) 34177–34183. https://doi.org/10.1007/s11356-018-3850-9.

83. A.N. Jones, J. Bridgeman, A fluorescence-based assessment of the fate of organic matter in water treated using crude/purified Hibiscus seeds as coagulant in drinking water treatment, Science of the Total Environment. 646 (2019) 1–10. https://doi.org/10.1016/j.scitotenv.2018.07.266.

84. N. Mohd-Esa, F.S. Hern, A. Ismail, C.L. Yee, Antioxidant activity in different parts of roselle (Hibiscus sabdariffa L.) extracts and potential exploitation of the seeds, Food Chem. 122 (2010) 1055–1060. https://doi.org/10.1016/j.foodchem.2010.03.074.

85. F.B. Rebah, S.M. Siddeeg, Cactus an eco-friendly material for wastewater treatment: A review, Journal of Materialsand Environmental Sciences. 8 (2017) 1770–1782.

86. C. Bhattacharjee, S. Dutta, V.K. Saxena, A review on biosorptive removal of dyes and heavy metals from wastewater using watermelon rind as biosorbent, Environmental Advances. 2 (2020). https://doi.org/10.1016/j.envadv.2020.100007.

87. Oladoja NA (2016) Advances in the quest for substitute for synthetic organic polyelectrolytes as coagulant aid in water and wastewater treatment operations. Sustainable Chemistry and Pharmacy 3:47–58.

88. Mohd-Salleh SNA, Mohd-Zin NS, Othman N (2019) A review of wastewater treatment using natural material and its potential as aid and composite coagulant. Sains Malaysiana 48:155–164.

89. Rasool MA, Tavakoli B, Chaibakhsh N, et al (2016) Use of a plant-based coagulant in coagulation-ozonation combined treatment of leachate from a waste dumping site. Ecological Engineering 90:431–437. https://doi.org/10.1016/j.ecoleng.2016.01.057.

90. Ghebremichael K, Abaliwano J, Amy G (2009) Combined natural organic and synthetic inorganic coagulants for surface water treatment. Journal of Water Supply: Research and Technology - AQUA 58:267–276. https://doi.org/10.2166/aqua.2009.060.

91. Freitas JHES, de Santana KV, do Nascimento ACC, et al (2016) Evaluation of using aluminum sulfate and water-soluble Moringa oleifera seed lectin to reduce turbidity and toxicity of polluted stream water. Chemosphere 163:133–141. https://doi.org/10.1016/j.chemosphere.2016.08.019.

92. K.E. Lee, N. Morad, T.T. Teng, B.T. Poh, Development, characterization and the application of hybrid materials in coagulation/flocculation of wastewater: A review, Chemical Engineering Journal. 203 (2012) 370–386. https://doi.org/10.1016/j.cej.2012.06.109.

93. Shamsnejati S, Chaibakhsh N, Pendashteh AR, Hayeripour S (2015) Mucilaginous seed of Ocimum basilicum as a natural coagulant for textile wastewater treatment. Industrial Crops and Products 69:40–47. https://doi.org/10.1016/j.indcrop.2015.01.045.

94. Bodlund I, Pavankumar AR, Chelliah R, et al (2014) Coagulant proteins identified in Mustard: A potential water treatment agent. International Journal of Environmental Science and Technology 11:873–880. https://doi.org/10.1007/s13762-013-0282-4

95. Dezfooli SM, Uversky VN, Saleem M, et al (2016) A simplified method for the purification of an intrinsically disordered coagulant protein from defatted Moringa oleifera seeds. Process Biochemistry 51:1085–1091. https://doi.org/10.1016/j.procbio.2016.04.021

96. Adinolfi M, Michela Corsaro M, Lanzetta R, et al (1994) Composition of the coagulant polysaccharide fraction from Strychnos potatorum seeds. Carbohydrate Research 263:. https://doi.org/10.1016/0008-6215(94)00149-9.

97. Abidin ZZ, Mohd Shamsudin NS, Madehi N, Sobri S (2013) Optimisation of a method to extract the active coagulant agent from Jatropha curcas seeds for use in turbidity removal. Industrial Crops and Products 41:319–323. https://doi.org/10.1016/j.indcrop.2012.05.003

98. Marobhe NJ, Dalhammar G, Gunaratna KR (2007) Simple and rapid methods for purification and characterization of active coagulants from the seeds of vigna unguiculata and parkinsonia aculeata. Environmental Technology 28:671–681. https://doi.org/10.1080/09593332808618827.

99. Ghebremichael KA, Gunaratna KR, Henriksson H, et al (2005) A simple purification and activity assay of the coagulant protein from Moringa oleifera seed. Water Research 39:. https://doi.org/10.1016/j.watres.2005.04.012

100. Diaz A, Rincon N, Escorihuela A, et al A preliminary evaluation of turbidity removal by natural coagulants indigenous to Venezuela 1999. www.elsevier.com/locate/procbio.

101. Miller SM, Fugate EJ, Craver VO, et al (2008) Toward understanding the efficacy and mechanism of Opuntia spp. as a natural coagulant for potential application in water treatment. Environmental Science and Technology 42:4274–4279. https://doi.org/10.1021/es7025054.

102. Gidde Milind R, Bhalerao AR, Malusare CN (2012) Comparative Study of Different Forms of Moringa Oleifera Extracts for Turbidity Removal. International Journal of Engineering Research and Development 2.

103. Nordmark BA, Przybycien TM, Tilton RD (2016) Comparative coagulation performance study of Moringa oleifera cationic protein fractions with varying water hardness. Journal of Environmental Chemical Engineering 4:4690–4698. https://doi.org/10.1016/j.jece.2016.10.029.

104. Kwaambwa HM, Maikokera R (2007) A fluorescence spectroscopic study of a coagulating protein extracted from Moringa oleifera seeds. Colloids and Surfaces B: Biointerfaces 60:213–220. https://doi.org/10.1016/j.colsurfb.2007.06.015.

105. Baptista ATA, Silva MO, Gomes RG, et al (2017) Protein fractionation of seeds of Moringa oleifera lam and its application in superficial water treatment. Separation and Purification Technology 180:114–124. https://doi.org/10.1016/j.seppur.2017.02.040.

106. Villaseñor-Basulto DL, Astudillo-Sánchez PD, del Real-Olvera J, Bandala ER (2018) Wastewater treatment using Moringa oleifera Lam seeds: A review. Journal of Water Process Engineering 23:151–164.

107. Raj KR, Kardam A, Arora JK, et al (2013) Adsorption behavior of dyes from aqueous solution using agricultural waste: Modeling approach. Clean Technologies and Environmental Policy 15:73–80. https://doi.org/10.1007/s10098-012-0480-7.

108. Jung Y, Jung Y, Kwon M, et al (2018) Evaluation of Moringa oleifera seed extract by extraction time: Effect on coagulation efficiency and extract characteristic. Journal of Water and Health 16:904–913. https://doi.org/10.2166/wh.2018.078.

109. Ndabigengesere A, Subba Narasiah K (1998) Quality of water treated by coagulation using Moringa oleifera seeds. Water Res 32:781–791. https://doi.org/10.1016/S0043-1354(97)00295-9.

110. Carvalho MS, Alves BRR, Silva MF, et al (2016) CaCl2 applied to the extraction of Moringa oleifera seeds and the use for Microcystis aeruginosa removal. Chemical Engineering Journal 304:469–475. https://doi.org/10.1016/j.cej.2016.06.101.

111. Megersa M, Gach W, Beyene A, et al (2019) Effect of salt solutions on coagulation performance of Moringa stenopetala and Maerua subcordata for turbid water treatment. Separation and Purification Technology 221:319–324. https://doi.org/10.1016/j.seppur.2019.04.013.

112. Sánchez-Martín J, Ghebremichael K, Beltrán-Heredia J (2010) Comparison of single-step and two-step purified coagulants from Moringa oleifera seed for turbidity and DOC removal. Bioresource Technology 101:6259–6261. https://doi.org/10.1016/j.biortech.2010.02.072.

113. Jerri HA, Adolfsen KJ, McCullough LR, et al (2012) Antimicrobial sand via adsorption of cationic Moringa oleifera protein. Langmuir 28:2262–2268. https://doi.org/10.1021/la2038262.

114. Kalibbala HM, Wahlberg O, Hawumba TJ (2009) The impact of Moringa oleifera as a coagulant aid on the removal of trihalomethane (THM) precursors and iron from drinking water. Water Science and Technology: Water Supply 9:707–714. https://doi.org/10.2166/ws.2009.671.

115. Bhutada PR, Jadhav AJ, Pinjari D v., et al (2016) Solvent assisted extraction of oil from Moringa oleifera Lam. seeds. Industrial Crops and Products 82:74–80. https://doi.org/10.1016/j.indcrop.2015.12.004.

116. Choudhary M, Neogi S (2017) A natural coagulant protein from Moringa oleifera: Isolation, characterization, and potential use for water treatment. Materials Research Express 4:. https://doi.org/10.1088/2053-1591/aa8b8c.

117. Wan J, Chakraborty T, Xu C (Charles), Ray MB (2019) Treatment train for tailings pond water using Opuntia ficus-indica as coagulant. Separation and Purification Technology 211:448–455. https://doi.org/10.1016/j.seppur.2018.09.083.

118. Chaibakhsh N, Ahmadi N, Zanjanchi MA (2014) Use of Plantago major L. as a natural coagulant for optimized decolorization of dye-containing wastewater. Industrial Crops and Products 61:169–175. https://doi.org/10.1016/j.indcrop.2014.06.056.

119. Antov MG, Šćiban MB, Prodanović JM, et al (2018) Common oak (Quercus robur) acorn as a source of natural coagulants for water turbidity removal. Industrial Crops and Products 117:340–346. https://doi.org/10.1016/j.indcrop.2018.03.022

120. Sciban M, Klasnja M, Skrbic B (2009) Water Turbidity Removal by Natural Coagulants. Progress in Environmental Science and Technology, Vol Ii, Pts a and B.

121. Sapana MM, Sonal CG, D RP (2012) Use of Moringa Oleifera (Drumstick) seed as Natural Absorbent and an Antimicrobial agent for Ground water Treatment.

122. Poumaye N, Mabingui J, Lutgen P, Bigan M (2012) Contribution to the clarification of surface water from the Moringa oleifera: Case M'Poko River to Bangui, Central African Republic. Chemical Engineering Research and Design 90:2346–2352. https://doi.org/10.1016/j.cherd.2012.05.017.

123. Amir Hariz Amran, Nur Syamimi Zaidi, Khalida Muda, Liew Wai Loan. Effectiveness of Natural Coagulant in Coagulation Process: A Review. nternational Journal of Engineering & Technology, 7 (3.9) (2018) 34-37. https://doi.org/10.14419/ijet.v7i3.9.15269.

124. Abaliwano JK, Ghebremichael KA, Amy GL (2008) Application of the Purified Moringa Oleifera Coagulant for Surface Water Treatment. WaterMill Working Paper Series 1–19.

125. Kamali M, Suhas DP, Costa ME, et al (2019) Sustainability considerations in membrane-based technologies for industrial effluents treatment. Chemical Engineering Journal 368:474–494. https://doi.org/10.1016/j.cej.2019.02.075.

126. H.N.P. Dayarathne, M.J. Angove, R. Aryal, H. Abuel-Naga, B. Mainali, Removal of natural organic matter from source water: Review on coagulants, dual coagulation, alternative coagulants, and mechanisms, Journal of Water Process Engineering. 40 (2021). https://doi.org/10.1016/j.jwpe.2020.101820.

127. V.V.F. Rocha, I.F.S. dos Santos, A.M.L. Silva, D.O. Sant'Anna, A.L. Junho, R.M. Barros, Clarification of high-turbidity waters: a comparison of Moringa oleifera and virgin and recovered

aluminum sulfate-based coagulants, Environ Dev Sustain. 22 (2020) 4551–4562. https://doi.org/10.1007/s10668-019-00397-2.

128. B. Othmani, M.G. Rasteiro, M. Khadhraoui, Toward green technology: a review on some efficient model plant-based coagulants/flocculants for freshwater and wastewater remediation, Clean Technol Environ Policy. 22 (2020) 1025–1040. https://doi.org/10.1007/s10098-020-01858-3.

129. W.A.E.H.A.L.A.L. WELL, Water clarification using Moringa oleifera seed coagulant, 1999.

130. J.P. Sutherland, G.K. Folkard, M.A. Mtawali, W.D. Grant, Moringa oleifera as a natural coagulant, n.d.

131. S.S. Thakur, S. Choubey, Assessment of coagulation efficiency of Moringa oleifera and Okra for treatment of turbid water, Arch Appl Sci Res. 6 (2014) 24–30. www.scholarsresearchlibrary.com.

132. T. Kazi, A. Virupakshi, Treatment of Tannery Wastewater Using Natural Coagulants, 2007. www.ijirset.com.

133. K. Qureshi, I. Bhatti, M.S. Shaikh, Development Of Bio-Coagulant From Mango Pit For The Purification Of Turbid Water, Sindh Univ. Res. Jour. 43 (2011) 105–110.

134. A. Seghosime, J.A.M. Awudza, R. Buamah, S.O. Kwarteng, Comparative Studies on Proximate Composition and Phytochemical Screening of Mango, Key lime, African star apple and African pear Seeds as Possible Coagulant Aids for Water Treatment, Am J Environ Sci. 13 (2017) 325–333. https://doi.org/10.3844/ajessp.2017.325.333.

135. Y.T. Hameed, A. Idris, S.A. Hussain, N. Abdullah, A tannin-based agent for coagulation and flocculation of municipal wastewater: Chemical composition, performance assessment compared to Polyaluminum chloride, and application in a pilot plant, J Environ Manage. 184 (2016) 494–503. https://doi.org/10.1016/j.jenvman.2016.10.033.

136. T. Pavón-Silva, V. Pacheco-Salazar, J. Carlos Sánchez-Meza, G. Roa-Morales, A. Colín-Cruz, Physicochemical and biological combined treatment applied to a food industry

wastewater for reuse, J Environ Sci Health A Tox Hazard Subst Environ Eng. 44 (2009) 108–115. https://doi.org/10.1080/10934520802515467.

137. C. Dong, W. Chen, C. Liu, Flocculation of algal cells by amphoteric chitosan-based flocculant, Bioresour Technol. 170 (2014) 239–247. https://doi.org/10.1016/j.biortech.2014.07.108.1.

138. M. Huang, Z. Liu, A. Li, H. Yang, Dual functionality of a graft starch flocculant: Flocculation and antibacterial performance, J Environ Manage. 196 (2017) 63–71. https://doi.org/10.1016/j.jenvman.2017.02.078.

139. C. Zhao, J. Zhou, Y. Yan, L. Yang, G. Xing, H. Li, P. Wu, M. Wang, H. Zheng, Application of coagulation/flocculation in oily wastewater treatment: A review, Science of The Total Environment. 765 (2021) 142795. https://doi.org/10.1016/J.SCITOTENV.2020.142795.

140. Y. Tayalia, P. Vijaysai, Process Intensification in Water and Wastewater Treatment Systems, in: Computer Aided Chemical Engineering, Elsevier B.V., 2012: pp. 32–40. https://doi.org/10.1016/B978-0-444-59507-2.50005-6.

141. Y. Xue, Z. Liu, A. Li, H. Yang, Application of a green coagulant with PACl in efficient purification of turbid water and its mechanism study, J Environ Sci (China). 81 (2019) 168–180. https://doi.org/10.1016/j.jes.2019.01.015.

142. K. Yang, G. Wang, X. Chen, X. Wang, F. Liu, Treatment of wastewater containing Cu2+ using a novel macromolecular heavy metal chelating flocculant xanthated chitosan, Colloids Surf A Physicochem Eng Asp. 558 (2018) 384–391. https://doi.org/10.1016/j.colsurfa.2018.06.082.

143. Y. Sun, K.J. Shah, W. Sun, H. Zheng, Performance evaluation of chitosan-based flocculants with good pH resistance and high heavy metals removal capacity, Sep Purif Technol. 215 (2019) 208–216. https://doi.org/10.1016/j.seppur.2019.01.017.

Table 1 – Operational conditions used in the treatment of food industry effluents with plant-based coagulants.

Wastewater	Coagulant	Process temperature (°C)	Coagulant dosage	Effluent volume (L)	pH	Equipment	Coagulation	Flocculation	Sedimentation	Main findings	Reference
							Fast mixing	Slow mixing			
Dairy industry	*Moringa oleifera* seeds	25±1	0.15–0.25 g	0.2	5–10	Jar-Test	Different rotational speeds for up to 60 min		60 min	Up to 98% removal of color and turbidity.	[9]
Dairy industry	*Moringa oleifera* seeds	-	0.5–1.25 g/L	-	-	Jar-Test	200 rpm for 1 min	100 rpm for 40 min	40–60 min	Over 95% turbidity removal.	[28]
Dairy industry	*Moringa oleifera* seeds	-	100–400 mg/L	-	-	Jar-Test	60 s	15–20 rpm	30 min	Mean removal efficiency of 62.5% for turbidity, 51.9% for COD, and 80% for salinity.	[29]
Dairy industry	Vegetable tannin	-	100–600 mg/L	1.5	6.0–7.0	Jar-Test	120 rpm for 90 s	45 rpm for 30 min	60 min	48.1- 78.5% color removal at concentration 100-600 mg/L; maximum TS removal of 13.6% at 500 mg/L; maximum COD removal of 41.6% at 400 mg/L; maximum turbidity removal of 89.4% at 300 mg/L.	[30]
Dairy industry	*Moringa oleifera* seeds	-	3000 mg/L	-	no pH correction	Jar-Test	100 rpm for 2 min	20 rpm for 10 min	60 min	Removal efficiency of 96-99% for COD, turbidity, and color.	[31]
Dairy industry cleaning-in-place	Moringa seed extract and nanoparticles of iron oxide functionalized with moringa extract	-	800–2000 mg/L	0.25	natural pH and 9	-	100 rpm for 2 min	20 rpm for 10 min	60 min	Nearly 90% turbidity removal.	[32]

Dairy cattle breeding	*Moringa oleifera* seeds	-	60 mL	1	7.2	Organic filters made of thin coal, bamboo leaves, eucaliptus leaves, gliricidia branches, and wood sawdust	Manual stirring for 10 min		-	Removal efficiency up to 44.60% for TS, 49.23% for TSS, and 43.38% for COD.	[33]
Palm oil milling	*Cassia obtusifolia* seeds	20–90	0–2,5 g/L	0.3	2–4.7	Jar-Test	150 rpm for 5 min	0–50 rpm for 15 min	0–60 min	Maximum removal of 81-87% for TSS and 48-55% for COD.	[6,7]
Palm oil milling	*Moringa oleifera* seeds	50–70	500–6000 mg/L and 1000–5000 mg/L	0.5	4–9 and 3–7	Jar-Test	150 rpm for 5 min	30 rpm for 30 min	90 min	Removal efficiency up to 99.7% for TSS, 71.5% for COD, 68.2% for BOD, 100% for oil and grease, and 91% for total nitrogen.	[34–36]
Palm oil milling	Rice starch	25±1	0–3.0 g/L	0.3	2.5–9	Conventional jar or flocculator	150 rpm for 5 min	0–100 rpm for 15 min	0–90min	TSS removal up to 84.1% for rice starch alone and 88.4% for rice starch and 0.20-0.8 g/L of alum. COD removal up to 49.23% using rice starch and 0.38 g/L alum.	[37,38]
Palm oil milling	Magnetic *Moringa oleifera* seeds	-	0.5–2.5 g/L	1	5–9	Jar-Test	150 rpm for 2 min	30 rpm for 30 min	1–45 min	Approximately 90% COD removal.	[39]
Palm oil milling	*Moringa oleifera* seeds	27–30	500–5000 mg/L	0.5	4.51	Jar-Test	250 rpm and 3 min	30 rpm and 30 min	60 min	The optimal moringa dosage (2000 mg/L) provided removal	[40]

										percentages of 95.42% for TSS, 88.30% for turbidity, 90.15% for color, 89.81% for NH3-N, and 87.05% for oil and grease.	
Cassava processing	*Abelmoschus esculentus* (Okra), *Ficus exasperata* (Ipin tree), and *Bridelia ferrugeneae* (Iraodan tree)	-	0–100 mL	0.25	-	Jar-Test	30 s at 120 rpm	20 rpm for 15 min	60 min	Turbidity was reduced from 168 NTU to as low as 6 NTU.	[41]
Cassava processing	Natural commercial coagulants (Acquapol WW, Acquapol S5T, Tanfloc SL, and Tanfloc SG)	-	160–800 mg/L	0.5	natural pH	Jar-Test	120 rpm for 2 min	20 rpm for 15 min	5–20 min	Removal efficiency over 77.5% for color and 88.5% for turbidity.	[42]
Tapioca starch	*Moringa oleifera* seeds	26	110–150 mg/L	1	-	Clarifier tank	15–20 rpm for 5 min		3 h	Up to 99.6% removal for BOD, 99.7% for COD, and 91.5% for TSS. The effluent's pH increased from 5.8 to 6.7-7.8.	[8]
Coffee fruit pulping	*Moringa oleifera* seeds	25	0–60 mL/L	-	4–8	Jar-Test	160 rpm for 10 s	20 rpm for 5 min	90 min	Up to 84.27% TSS removal.	[43]

Coffee fermentation	*Moringa oleifera* seeds	-	0 –4 g/L	0.9	3–7	Jar-Test	2 min	5 min	24 h	Up to 54% removal efficiency for TSS, 100% for insoluble COD, 25% for total COD, and 100% for nitrates and nitrites.	[44]
Distillery spent wash	*Moringa oleifera* seeds	30	20–60 mL	0.5	4–9	Jar-Test	100 rpm for 2 min	40 rpm for 30 min	30 min	Maximum 56% and 67% color removal using moringa coagulant along with sodium chloride and potassium chloride, respectively.	[45]
Distillery spent wash	*Moringa oleifera* seeds	30	20–80 mL/L	1	2–6	Jar-Test	300 rpm for 2 min	30 rpm for 30 min	4 h	Removal efficiencies varied depending on the synthetic coagulants (ferric sulphate, aluminium sulphate, and calcium sulphate) used in conjunction with moringa. Aluminium sulphate along with moringa extract at pH 4.0 yielded the best removal rates: 97% for color, 87% for COD, and 100% for turbidity.	[46]
Swine farms	*Moringa oleifera* seeds	-	1–20 mL	0.5	4.2	Jar-Test	160 rpm for 30 s	15 rpm for 15 min	2–24 h	Total and thermotolerant coliforms were reduced by 96.5% and 94.8%, respectively, but turbidity was not affected.	[47]
Brewery	Vegetable tannin	25	0.12–0.18 mL/L	1.2	4.5–8	Jar-Test	120 rpm for 5 min	30 rpm for 30 min	10 min	99% turbidity removal.	[25]

Green tea extracts	*Moringa oleifera* seeds	-	100–10000 mg/L	-		Jar-Test	150 rpm for 5 min	30 rpm for 30 min	-	Removal of turbidity, TS, and polyphenols up to 96%, 16%, and 19%, respectively.	[48]
Slaughterhouse	*Moringa oleifera* seeds	25±1	3–7 g/L	1	5–9	Jar-Test	100 rpm for 5 min	20 rpm for 20 min	180 min	Up to 64% COD reduction.	[4]

Abbreviations: BOD = biochemical oxygen demand; COD = chemical oxygen demand; NH3-N = ammonia nitrogen; TS = total solids; TSS = total suspended solids.

Table 2 – Use of plant-based coagulants in the treatment of food industry effluents.

Coagulant source	**Effluent**	**Objectives**	**Main results**	**Reference**
Moringa oleifera seeds	Dairy industry wastewater	Investigate the sorption potential of moringa seeds in the removal of organic compounds from wastewater	Removal efficiencies of up to 98% for both color and turbidity using 0.2 g of moringa seed powder	[9]
Moringa oleifera seeds	Slaughterhouse wastewater	Investigate the adsorption of organic pollutants when moringa seed is employed as a natural coagulant	64% COD reduction using 7 g/L of powdered seed and pH 9	[4]
Cassia obtusifolia seeds	Palm oil mill effluent	Evaluate the potential of cassia seed gum as a natural coagulant alternative to alum	*C. obtusifolia* seed gum is as efficient to treat palm oil mill effluent as alum: at least 87% removal of TSS and 55% COD at concentration 1.0 g/L, initial wastewater pH 3, and settling time 45 min	[6]
Moringa oleifera seeds	Tapioca starch wastewater	Investigate the addition of powdered moringa seed in reducing physicochemical parameters and total coliform, as well as increasing pH	BOD, COD, and TSS were significantly reduced, while pH reached the acceptable range for effluent disposal	[8]
Moringa oleifera seeds	Coffee fruit pulping wastewater	Determine the best combination between pH and concentration of moringa seed extract	Greater removal of suspended solids from the wastewater at pH 4.0-5.0 and dosage 10 mL/L	[43]

Moringa oleifera seeds	Coffee fermentation wastewater	Investigate the efficacy of moringa seed extract in wastewater treatment	Significant reduction of TSS (from 8% to 54%), COD (from 26% to 100%), and nitrate and nitrite (from 20% to 100%)	[44]
Moringa oleifera seeds	Palm oil mill effluent	Evaluate the use of moringa seeds as natural coagulant in the coagulation-flocculation of effluent	95% removal of suspended solids and 52.2% reduction of COD, with best performance at 30°C	[34]
Moringa oleifera seeds	Distillery spent wash	Determine the effects of moringa-based coagulant dosage, pH, and salt concentration in effluent color removal	Partial color removal was obtained at optimal conditions, which were selected through the response surface methodology	[45]
Moringa oleifera seeds	Distillery spent wash	Evaluate the efficiency of moringa seed extract along with conventional coagulants for the removal of physical-chemical contaminants from effluent	Moringa seed extract exhibited significant color, COD, and turbidity reduction ability when used as a coagulant aid along with synthetic coagulants. Aluminum sulphate combined with moringa seed extract was most effective, removing 96.5% of color, 87% of COD, and 99.9% of turbidity at pH 4.0	[46]
Moringa oleifera seeds	Porcine wastewater	Evaluate the efficiency of moringa seed extract in the removal of turbidity and coliforms (total and thermotolerant) from wastewater	Turbidity remained unchanged, but total and thermotolerant coliforms were removed by 96.5% and 94.8%, respectively	[47]

Moringa oleifera seeds	Dairy cattle breeding wastewater	Evaluate the effect of moringa extract on the removal of TS, TSS, and COD from wastewater	Moringa seed extract increased the efficiency of organic filters in TS and COD removal, but TSS removal efficiency remained unchanged	[33]
Moringa oleifera seeds	Dairy industry wastewater	Evaluate the environmental and economic potential of the application of moringa seed powder in wastewater treatment	Turbidity was reduced by over 95%, enabling water reuse. The cultivation of moringa can be an extra source of revenue for dairy farmers	[28]
Moringa oleifera seeds	Dairy industry wastewater	Study moringa seed powder as a natural adsorbent for wastewater treatment	Successful removal of turbidity, COD, and salinity	[29]
Vegetable tannins	Brewery wastewater	Evaluate and optimize coagulation-flocculation conditions	99% removal of turbidity and reduction of apparent color using 0.23 mL/L of vegetable tannin at pH 4.9. Tanfloc SL aided flocculation, enabling water reuse	[25]
Moringa oleifera seeds	Green tea extracts	Compare the isolated effect of centrifugation to the impact of natural coagulants (chitosan and moringa) followed by centrifugation in green tea extract clarification	Optimized clarification conditions provided turbidity, TS, and polyphenols reductions of 96%, 16%, and 19% using moringa extract versus 95%, 16%, and 18% using chitosan. The extraction and clarification with moringa followed by centrifugation produced clear green tea extracts enriched with bioactive compounds at a lower cost	[48]
Rice starch	Palm oil mill effluent	Evaluate the performance of unmodified starches as natural	Rice starch and alum yielded similar TSS removal at dosage 2 g/L, pH 3, settling time 5 min, and stirring rate 10 rpm	[37]

		coagulants in coagulation-flocculation treatment compared to alum coagulant		
Moringa oleifera seeds	Dairy wastewater	Evaluate the use of moringa as a natural coagulant in CFS followed by MF or NF in wastewater treatment	The association of CFS-MF-NF treatments showed removal efficiencies between 96% and 99% for COD, turbidity, and color, meeting water reuse standards. The treated wastewater exhibited better quality compared to the conventional method	[31]
Abelmoschus esculentus (Okra) pods, *Ficus exasperata* (Ipin tree) bark, and *Bridelia ferrugeneae* (Iraodan tree) bark	Cassava processing	Determine the coagulant efficiency of three organic coagulants compared to an synthetic coagulant (aluminum sulphate)	For wastewater with initial turbidity of 168 NTU, the lowest turbidity attained upon applying the three organic coagulants was 6 NTU. The organic coagulants employed can partially replace the synthetic coagulant	[41]
Moringa oleifera seeds	Palm oil wastewater	Determine the COD removal efficiency of magnetic moringa obtained from moringa seeds and synthetic magnetic nanoparticles	The microwave synthesis of magnetic moringa is a cost-efficient process in the production of a highly effective coagulant (90% COD removal)	[39]
Moringa oleifera seeds	Palm oil mill effluent	Determine the optimum coagulant dose for contaminant removal	The optimum dosage was 2000 mg of powdered seeds per L of effluent	[40]
Vegetable tannins	Dairy wastewater	Compare the coagulation performance of tannin and polyaluminum chloride	No statistical differences between the performance of the two coagulants for the removal of COD, color, turbidity, and TS. Tannins were effective within a wide pH range (5.0 to 10.0)	[30]

Natural commercial coagulants (Acquapol WW, Acquapol S5T, Tanfloc SL, and Tanfloc SG)	Cassava processing wastewater	Investigate the efficiency of different coagulants in color and turbidity removal from effluent	Natural commercial coagulants yielded a higher color and turbidity removal than the synthetic coagulant. The best concentration of natural coagulant (Acquapol S5T and Tanfloc SL) was 320 mg/l, removing color by ≥77.5 % and turbidity by ≥88.5 %	[42]
Moringa oleifera seeds extract and nanoparticles of iron oxide functionalized with moringa extract	Dairy cleaning-in-place wastewater	Test two organic coagulants derived from moringa as alternatives for wastewater treatment	Mo-NP provides approximately 90% of turbidity removal in a reduced sedimentation time (7 min compared to 60 min when Mo is used alone)	[32]

Abbreviations: BOD = biochemical oxygen demand; CFS = coagulation/flocculation/sedimentation; COD = chemical oxygen demand; MF = microfiltration; Mo = moringa seeds extract; NF = nanofiltration; NP = nanoparticles; TS = total solids; TSS = total suspended solids

Table 3 – Natural coagulants used in the treatment of food industry effluents.

Effluent	Coagulant source	Coagulant preparation	Reference
Dairy industry wastewater	*Moringa oleifera* seeds	The biomass was cut, ground, and sieved. Then, the fraction with a mesh size <0.42 mm was selected. The seeds did not undergo drying or any other processing.	[9]
Slaughterhouse wastewater	*Moringa oleifera* seeds	The seeds were shelled, reduced to a fine powder, and sieved. White seed kernels with mesh size 0.51 mm were selected.	[4]
Palm oil mill effluent	*Cassia obtusifolia* seeds	Whole seeds were ground (0.5-1 mm), and 2.5 g samples were mixed with 100 mL of distilled water for 30 s. The mixture was filtered and the filtrate was used as gum.	[6,7]
Tapioca starch wastewater	*Moringa oleifera* seeds	Seeds were peeled, dried, and milled. Fine powders that passed through a 50-mesh sieve were used as a coagulant.	[8]
Coffee fruit pulping wastewater	*Moringa oleifera* seeds	The extract was obtained by mixing macerated seeds with water (10 mL of water for each seed).	[43]
Coffee fermentation wastewater	*Moringa oleifera* seeds	0.1 g of ground seeds were mixed with 25 mL of distilled water for 1 min before filtering the resulting solution.	[44]
Palm oil mill effluent	*Moringa oleifera* seeds	Seeds were ground to a fine powder, and the oil was extracted for 8 h with n-hexane. After drying the resulting cake, 5 g were mixed with 100 mL distilled water for 2 min. The resulting suspension was filtered.	[34–36]
Distillery spent wash	*Moringa oleifera* seeds	The seeds were ground to a fine powder, and the active component was extracted using 100 mL of NaCl or KCl solution for each 4 g of powder. The mixture was stirred for 30 min at room temperature, and the suspension was filtered.	[45]

Distillery spent wash	*Moringa oleifera* seeds	The active components from the seeds were extracted using 100 mL of 1 M NaCl solution for each 5 g of powdered seed mixed at 500 rpm for 15 min at room temperature. Then, the coagulant salt suspension was filtered.	[46]
Porcine wastewater	*Moringa oleifera* seeds	1 g, 2 g, and 3 g samples of ground seeds were extracted with 100 mL of distilled water. The mixture was then filtered.	[47]
Dairy cattle breeding	*Moringa oleifera* seeds	Seeds were removed from the pods and dried in an oven at 45 ºC for 24 h. The moringa extract was prepared by grinding 50 g of seeds in 1 L of distilled water followed by filtration.	[33]
Dairy industry wastewater	*Moringa oleifera* seeds	Two methods were employed: (1) 2 g of ground seeds were extracted with 1 mol/L NaCl solution (11.89 g in 200 mL water); and (2) seed pods were peeled and crushed, and the resulting powder was sieved through a 42 mm sieve.	[28]
Dairy industry wastewater	*Moringa oleifera* seeds	2 g of dried and finely ground seeds were extracted with 100 mL water. The suspension was stirred for 30 min and then passed through a filter paper.	[29]
Brewery wastewater	Vegetable tannin from *Acacia mearnsii*	Tanfloc was used as a flocculant. This is a commercial vegetable tannin derived from the leaching of black acacia bark.	[25]
Green tea extracts	*Moringa oleifera* seeds	Peeled and ground seeds were Soxhlet-extracted with 98% purity n-hexane for 8 h. After removal of the oil fraction, the powdered seeds were mixed with water at a ratio of 20 mL/g, and the mixture was filtered with an 11 µm paper filter.	[48]
Palm oil mill effluent	Rice starch	6 g of starch were added to 200 mL of distilled water. This mixture was firstly autoclaved at 121 ºC and 117 kPa for 20 min, and then kept at a constant temperature of 80 ºC with an agitation of 400 rpm.	[37,38]

Dairy wastewater	*Moringa oleifera* seeds	The coagulant solution was prepared by mixing 50 g of ground seeds with 1 L of 1 M potassium chloride solution. The mixture was stirred for 30 min and the solution was vacuum filtered.	[31]
Cassava processing wastewater	*Abelmoschus esculentus* (Okra), *Ficus exasperata* (Ipin tree), and *Bridelia ferrugeneae* (Iraodan tree)	Okra: pods were ground to produce the gum, and 1.0 g of gum was soaked in 100.0 mL of water under stirring for 1 h. The mucilage used as coagulant was thereafter obtained by filtering the resulting solution. Ipin tree and Iraodan tree: 200 g of the barks of both plants were soaked in 2 L of water for 3 days.	[41]
Palm oil wastewater	Magnetic *Moringa oleifera*	3 g of seed powder were mixed with 0.1 L of 0.5 M calcium chloride under 100 ºC for 30 min at 100 rpm. Then, the extracted solution was microwaved and sonicated. The microwave power and ultrasonic intensity varied within 200 to 800 W and 20 to 100%, respectively. The mixture was filtered and oven-dried at 60 ºC.	[39]
Dairy wastewater	Vegetable tannin from *Acacia mearnsii*	Tannin based coagulant was extracted from *Acacia mearnsii* (TANAC, 25% w/w concentration).	[30]
Cassava processing wastewater	Natural commercial coagulants from *Acacia mearnsii*	Several commercial natural coagulants extracted from the husk of Acacia-negra were tested, namely: Acquapol WW and Acquapol S5T (Acqua Química), and Tanfloc SL and Tanfloc SG (Tanac S.A). The Tanfloc SL and SG solutions were prepared by dissolving 2 g of coagulant in 50 mL water. Solutions of Acquapol WW and S5T coagulants consisted of 5 mL of coagulant for every 50 mL of water.	[42]
Dairy cleaning-in-place wastewater	Moringa seeds extract and nanoparticles of iron oxide functionalized with moringa extract	Extract: moringa coagulant was prepared by grinding 5 g of shelled seeds in a mill and then stirring this powder with 100 mL of a saline solution for 30 min. Then, the coagulant was obtained by vacuum filtering the extract.	[32]

		Magnetic coagulant: predetermined quantities of nanoparticles (50 or 100 mg) were added to 20 mL of moringa extract and sonicated for 5 min followed by 1 h of stirring.	

Methodology employed

Portuguese
English

Data base

"GoogleScholar", "ScienceDirect" and "Scopus"

Descriptors

"Coagulation in the food industry; Plant-based coagulants; and Natural coagulants"

Year of research

Since 1994

Studies found

108 studies related to descriptor words

Journal of Water Process Engineering 52 (2023) 103525

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Plant-based coagulants for food industry wastewater treatment

Jonas Raul Balbinoti [a,b], Ricardo Egídio dos Santos Junior [b], Letícia Bernal Ferreira de Sousa [c], Fatima de Jesus Bassetti [d], Thaisa Carvalho Volpe Balbinoti [c], Regina Maria Matos Jorge [a,*], Luiz Mário de Matos Jorge [e]

[a] Graduate Program in Chemical Engineering, Federal University of Paraná (UFPR), Av. Francisco Hoffman dos Santos, s.n., CEP 81530-900 Curitiba, PR, Brazil
[b] Serviço Social da Indústria (SESI), Av. Afonso Pena, 1206, CEP 79002-070 Campo Grande, MS, Brazil
[c] Faculty of Pharmaceutical Sciences, Food and Nutrition (FACFAN), Federal University of Mato Grosso do Sul (UFMS), Av. Costa e Silva, s/n, Bairro Universitário, CEP 79070-900 Campo Grande, MS, Brazil
[d] Graduate Program in Environmental Science and Technology, Federal University of Technology – Paraná (UTFPR), R. Deputado Heitor Alencar Furtado, 5000 – CIC, CEP 81280-340 Curitiba, PR, Brazil
[e] Chemical Engineering Department, State University of Maringá (UEM), Colombo Avenue, 5790, CEP 87020-900, Maringá, PR, Brazil

A R T I C L E I N F O

Keywords:
Effluent treatment
Wastewater treatment
Coagulation
Natural coagulants
Sustainability
Biocoagulants

A B S T R A C T

Sustainable effluent treatments are essential tools in lowering the environmental impact of industrial activities. The partial or complete replacement of synthetic coagulants by natural coagulants, especially plant-based ones, can reduce the footprint of the effluent treatment due to the higher biodegradability and non-toxicity. Natural coagulants are also generally cheaper. This review focuses on plant-based coagulants used in food industry wastewater treatment. Extraction parameters of plant-based coagulants and effluent treatment conditions for different coagulants are presented. Based on an extensive assessment of peer-reviewed papers on food industry wastewater treatment, the performance of plant-based coagulants is compared to that of traditional, synthetic coagulants, both alone and as coagulant aids. This review aims to guide researchers and industry professionals in optimizing and scaling up environment-friendlier wastewater treatments using natural coagulants.

1. Introduction

Industrial activities are one of the leading causes of water pollution due to the generation of significant amounts of wastewater that contain toxic species or species that are difficult to degrade [1]. The production of liquid waste by the food industry is sizeable, especially in the animal-based food sector. Slaughtering operations and meat processing generate massive volumes of wastewater rich in proteins, organic compounds, and fats [5]. For example, slaughtering a single pig or cow generates 330 L and 700 L of wastewater, respectively [2–4]. Although drastically less than the animal sector, the production of plant-derived foods also uses significant amounts of water. For instance, the production of one ton of crude palm oil requires 5–7.5 tons of water [6,7]. Another example is the cassava starch industry, which generates 12–15 L of wastewater for each m^3 of processed cassava. The wastewater from cassava starch production exhibits high biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), total solids (TS), and cyanides [8].

Inadequate disposal of food industry effluents impacts the environment due to the high load of organic matter, heavy metals, alkalinity, and hardness, resulting in water pollution, odor generation, algal blooms, and mortality of aquatic and land animals [4,9]. The appropriate treatment of industrial wastewater is carried out by Effluent Treatment Stations (ETSs) [10]. By diminishing the polluting load of effluents, ETSs enable their safe reuse or release into water bodies.

To treat wastewater, ETSs utilize coagulation, flocculation, sedimentation, and other physical, chemical, and biological processes [11]. Wastewater treatments can be classified into physical (media filtration, settling, adsorption, membrane, and ultraviolet radiation), chemical (electrochemical, coagulation, oxidation, ion exchange, disinfection, catalytic reduction, and softening), and biological (microbial biodegradation, phytoremediation, constructed wetlands, and bioreactor digestion) [12,13]. Among these technologies, coagulation, and flocculation (CF) are the oldest and most widely employed in wastewater

* Corresponding author.
E-mail addresses: jonas.balbinoti@sesims.com.br (J.R. Balbinoti), ricardo.junior@sesims.com.br (R.E. dos Santos Junior), leticia_bernal@ufms.br (L.B.F. de Sousa), bassetti@utfpr.edu.br (F. de Jesus Bassetti), thaisa.balbinoti@ufms.br (T.C.V. Balbinoti), rjorge@ufpr.br (R.M.M. Jorge).

https://doi.org/10.1016/j.jwpe.2023.103525
Received 23 August 2022; Received in revised form 13 January 2023; Accepted 18 January 2023
Available online 27 February 2023