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Article

Comparison of Four Plant-Based Bio-Coagulants Performances against Alum and Ferric Chloride in the Turbidity Improvement of Bentonite Synthetic Water

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Abstract: Due to their cost-effectiveness, low toxicity, and naturally renewable properties, bio-coagulants for water treatment are gaining popularity. In this context, four bio-coagulants were tested for their ability in turbidity reduction in a synthetic bentonite suspension: *Opuntia ficus indica* (OFI) and *Moringa oleifera* (MO) seeds, two well-established vegetal materials in the domain of bio-coagulation, and Algerian *Aloe vera* (AV) and *Pinus halepensis seeds* (PHS), both of which have received very few or no study in turbidity reduction in water and wastewater. A comparative study was conducted with regard to two well-known inorganic coagulants, ferric chloride and Alum. Extraction salts, pH, coagulant dose, initial turbidity, and aging effects were examined. When compared to chemical coagulants, the four bio-coagulants worked very well, removing approximately 100% of the turbidity at natural pH of 7.5 using optimal doses of 1.5, 3, 3, 3.5, 1.5, and 1.5 mL/200 mL for AV, OFI, MO, PHS, Alum, and FeCl₃, respectively, at a broad range of pH (2 to 8), and causing no pH alteration of the treated water. Polysaccharides and proteins involved in inter-particle bridging and charge neutralization may be the active coagulation-flocculation molecules. The four bio-coagulants produced less sludge amount than Alum and Ferric chloride. This research not only shows that MO and OFI are effective in eliminating turbidity, but it also highlights the great potential of Algerian AV and PHS seeds as promising bio-coagulants in the treatment of polluted water.

Keywords: Algerian *Aloe vera* and *Pinus halepensis* Mill.; bio-coagulants; turbidity reduction; sludge amount; pH alteration



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1. Introduction

Water is one of the fundamental components of sustainable development, and its conservation is essential to the preservation of biodiversity on our planet. Urbanization, changing production, and consumption patterns in industrialized and developing nations provide new issues for managing the water cycle; wastewater sanitation is one of them [1], it poses a dual threat to public health and environmental protection. Wastewater treatment units are being created at an increasing rate, and treatment technologies are being developed and improved [2]. Efforts have been undertaken to increase the efficacy of the wastewater treatment systems now in circulation through the use of chemical reagents for their purification and potabilization, which have resulted in adverse health consequences [3–5].

Current wastewater treatment methods include adsorption [6], advanced oxidation processes (AOPs) [7], biological treatment [8], as well as membrane technologies [9]. Despite this, the traditional coagulation-flocculation technique is still commonly used owing to its simplicity and effectiveness.

The term “turbidity” refers to the cloudiness of water that is generated by suspended particles such as clay and silt, chemical precipitates such as manganese and iron, and organic particles such as human and animal waste, plant debris, and organisms. Turbidity is used in the operational monitoring of control measures included in water safety as a recommended approach to managing drinking water quality [10].

Turbidity reduction is, therefore, an essential step in water treatment processes either for large-scale treatment plants, small communities, or households. In developing countries and most especially in rural areas without water treatment systems and that rely heavily on polluted water sources, such as rivers, small community water supply, small dams, ponds, and traditional wells, there may be a need to render the water safe for drinking at household level through turbidity reduction followed by sand filtration and then disinfection [11].

Coagulants and flocculants are positively charged chemical substances that are often utilized in the treatment of drinking water and wastewater. Negatively charged polymers are widely employed in water treatment as well, particularly as high molecular weight flocculants, iron salts (FeCl_3 or $\text{Fe}_2(\text{SO}_4)_3$), aluminum salts ($\text{Al}_2(\text{SO}_4)_3$), hydrated lime, magnesium carbonate, and polymers (aluminum chlorohydrate, polyaluminum chloride (PAC), polyaluminum sulfate chloride, and polyferric sulfate) are among the chemical compounds typically employed as coagulants/flocculants, their high efficiency, ease of usage, and low cost are well recognized. Some of the above-mentioned chemicals have been found to be very effective at reducing suspended solid concentrations in water; they have also been used extensively in textile/dye wastewater treatment with remarkable results; In addition, their application has been reported for the treatment of wastewater in a variety of sectors, including agriculture, food processing, pulp and paper, tanning, and landfill leachate, among others [12,13]. The use of these substances is not always without consequences. Several environmental issues are presently being considered, particularly for environmental observers globally, as a result of the long-term toxicity of coagulants/flocculants. An in-depth investigation has been undertaken into the influence that the usage of chemical compounds as coagulants and flocculants can have. The environmental effects include increased corrosion of metallic utilities, pH alteration, root elongation limitation, and seed germination inhibition. Water and wastewater treatment with traditional coagulants/flocculants creates excessive chemical sludge in addition to the suspended particles to be removed; hence, chemical sludge management becomes another difficulty in handling. Aside from the environmental consequences, there are worries about human health. Metallic-based coagulants/flocculants are nonbiodegradable or nondegradable, and their residuals in drinking water can have a direct influence on human health when ingested and accumulate in body cells. Chemical coagulants/flocculants residuals in wastewater treatment effluent released to the environment may be trapped in food chains. Some indicators of the effects of chemical coagulants/flocculants on human health have been recorded, including central nervous system dysfunction, dementia, Alzheimer’s disease, and severe shaking [5].

For these reasons, attempts have been carried out to explore natural sources for the development of ecologically friendly flocculants and coagulants, with higher accessibility for rising economies and minimum or no toxicity, without disrupting the food chain. In addition, the use of natural and renewable materials for the reduction in pollution has gained scientific attention [14].

Organic coagulants induce a coagulation process distinct from that of metal-based coagulants. Coagulation aims to destabilize a colloidal solution by either (1) adsorption and neutralization of charges or (2) adsorption and bridging of colloids [15,16]. Regarding the adsorption and neutralization phenomena, ions with opposing charges are absorbed on the surface of colloids, leading to a reduction in the thickness of the electrical water double layer surrounding colloids [16,17]

Several natural substances, such as cellulose, proteins, starch, and polysaccharides, have a high molecular weight and numerous electrical charges across their atom chains in relation to the adsorption and bridging phenomena. These chemicals have extensive

chain lengths and can destabilize negatively charged colloidal particles [17]. Several scientists have proposed a hypothesis on chemical bridges that may be employed to explain the empirical behavior of these molecules [16]; coagulant molecules are responsible for the construction of bridges between colloidal solids: the free sites of multiple coagulant molecules attach to the colloidal solids, and coagulants bind colloids by hydrogen bonds, coulombic attraction force, van der Waals forces, or ion exchange processes [17,18].

Moringa oleifera, *Stryconus potatorum*, Cactus species, *Phaseolus vulgaris*, surjana seed, maize seed, tannin, gum arabic, *Prosopis juliflora*, and *Ipomoea dasycarpa* seed gum are prominent bio-coagulants that have been examined in the past. Among them, the Cactaceae family has exceptional adaptability to severe and contrasting situations, including drought, high and low temperatures, and nutrient- and organic-poor soils. It has been known for its therapeutic benefits, and the pulp material is typically constituted of a variety of polysaccharides and proteins [19,20]. The plant-based coagulants are simple to use since minimal processing is required, and they may provide a sustainable treatment option [21,22].

The present paper focuses on comparing the efficiency of four plant-based coagulants with traditional ones such as Alum and Ferric chloride; hence, in consideration of the lack of knowledge on natural coagulants, it is important to investigate the potential of various bio-coagulants without purification/extraction processes. For the first time, PHS have been described as a bio-coagulant for the treatment of synthetic turbid water after a first successful attempt in treating synthetic dye wastewater containing Congo red dye [23]; another bio-coagulant (AV) with few studies was selected for this paper owing to its widespread in Algeria, these two natural coagulants were discussed along with two known bio-coagulants, OFI which provided promising results in previous works [24–28] and MO which is considered as a widely established vegetal resource in the field of natural coagulants for the treatment of drinking water and wastewater, their seeds are rich in a water-soluble cationic coagulant protein that can lower turbidity in treated water. Multiple parameters were taken into consideration, such as the type of solvent, pH, coagulant dosage, initial turbidity, aging effect, and produced sludge amount.

2. Materials and Methods

2.1. Chemicals and Materials

The plant pads of bio-coagulants OFI and AV were harvested in Bouira's countryside (northeast of Algeria) during the month of March 2022, PHS and MO seeds were purchased in a local herbal market.

The following chemicals were used in this study: HCl, NaOH, NaCl, KCl, KNO₃, NaNO₃, and bentonite (all from Sigma Aldrich Chemical Company, USA). Metal salts used in this work were supplied either by Sigma-Aldrich or Biochem-Chemopharma, aluminum sulfate octadecahydrate (Al(SO₄)₃·18 H₂O), molecular weight: 666.42 g/mol, purity: 54–59% assay, iron (III) chloride hexahydrate (FeCl₃·6 H₂O), molecular weight: 270.33 g/mol, purity: 98–102%.

2.2. Apparatus and Measurements

Using a turbidity meter (HANNA, Cluj-Napoca, Romania), the formazine turbidity unit was used to measure turbidity (FTU). The jar test was conducted using a jar tester fitted with an illuminator paddle (VELP Scientifica Srl, Usmate Velate, Italy) to evaluate the performance of natural coagulants isolated from the vegetal materials.

2.3. Preparation of the Bentonite Synthetic Wastewater

For the coagulation experiments, 4 g of bentonite was added to 1 L of distilled water (DW) to obtain synthetically turbid water. The resulting bentonite suspension was then agitated at 200 rpm for one hour to achieve uniform dispersion of its particles. The suspension was then allowed to stand for 24 h to allow for complete hydration of the bentonite [29]. This suspension was used as a stock solution to prepare water samples of varying turbidities for subsequent coagulation tests. A total of 0.5 M NaOH or 0.5 M HCl were used to change the stock solution's initial pH [30].

2.4. Preparation, Extraction, and Characterization of the Bio-Coagulants

Preparation of OFI- and AV-based bio-coagulants: The pads of OFI and AV were repeatedly washed with tap water to eliminate dust and other impurities and then rinsed with deionized water. They had their thorns and external envelope removed. The mucilage was sliced into little pieces and oven-dried at 40 °C (Memmert GmbH + Co. KG, Bavaria, Germany) for 24 h. It was then reduced to a fine powder using a mortar and sieved through a 500 µm stainless steel sieve to achieve solubilization of active ingredients in the coagulant [31]. The powder was kept in an airtight polythene bag until it was used, without any chemical pretreatment.

Preparation of PHS and MO seeds-based bio-coagulants: MO seeds were deshelled and air-dried before grinding. Whole kernels were crushed and sieved through a small mesh. PHS was thoroughly washed with distilled water to eliminate dust and impurities, oven-dried at 40 °C for 24 h, crushed and powdered, then sieved through a 500 µm stainless steel sieve. The obtained precursors were both stored in sterile bottles for further use.

The precursors were subjected to the extraction of coagulation active agents using different solvents (distilled water, NaCl, KCl, KNO₃, NaNO₃) to increase repeatability and minimize any aging effects, such as changes in pH, viscosity, and coagulation activity related to the microbial degradation of organic components during storage, a new solution of the coagulant was prepared for each series of tests. The bio-coagulant precursors were also characterized by Fourier Transform Infrared Spectroscopy using an FTIR spectrophotometer-Thermo Scientific-Nicolet iS10 (Thermo Fisher Scientific, Waltham, MA, USA).

2.5. Coagulation-Flocculation Experiments

A batch test of coagulation-flocculation was conducted using jar test equipment that supports six beakers with a maximum capacity of 1000 mL each (Figure 1). All studies were conducted at 25 °C (room temperature), and each beaker contained 200 mL of the synthetic wastewater at specific turbidities. The bio-coagulant-turbid water mixture was stirred vigorously at 200 rpm for 3 min, followed by a slow speed at 30 rpm for 20 min; flocs were allowed to settle for 30 min. Samples (20 mL) were subsequently withdrawn using a volumetric pipette at 3 cm below the mixture surface, the final turbidity was determined using a turbidimeter, and the effect of several parameters, including pH, coagulant dosage, initial turbidity, extraction solvent nature, and aging effect was investigated. The experiments were repeated three times; all calculations were made using the average of the three data. The % removal of turbidity was calculated by the straightforward equation.

$$\text{Turbidity removal (\%)} = \frac{T_0 - T}{T} \times 100 \quad (1)$$

where T_0 and T represent the initial and final turbidity, respectively.

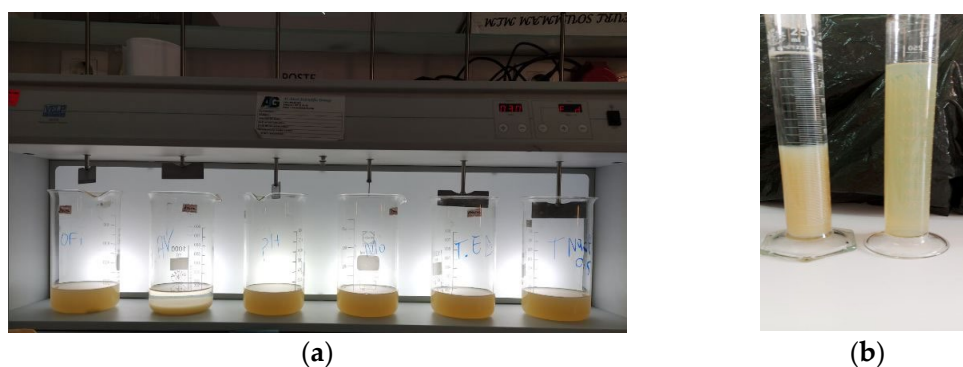


Figure 1. (a): Jar test apparatus used in this study; (b): treated water.

2.6. Comparison of the Sludge Production

Using the Imhoff cones and the volumetric approach, the volume of decanted sludge was calculated. After performing the experiment in the jar test (optimal conditions), the contents of the beakers were transferred to appropriate graduated conical containers (Imhoff cones). After one hour of settling, the sludge production was directly measured in milliliters of sludge per liter of treated wastewater [32].

3. Results and Discussion

3.1. FTIR Bio-Coagulants Characterization

The infrared spectra of the four bio-coagulants were recorded with a Fourier Transform Infrared Spectrometer (Thermo Fisher Scientific, Waltham, MA, USA); from Figure 2 and Table 1, the peaks at 1320 cm^{-1} and 1050 cm^{-1} indicate the presence of $-\text{COOH}$ groups, phosphoric groups, polysaccharides and aromatic proteins [28,33]. During the coagulation phase, the carboxyl group supplies adsorption sites for suspended particles enhancing the coagulation-flocculation process [12], the proteinaceous profile of the bio-coagulants (amino groups) suggests their involvement in the coagulation mechanism by charge neutralization.

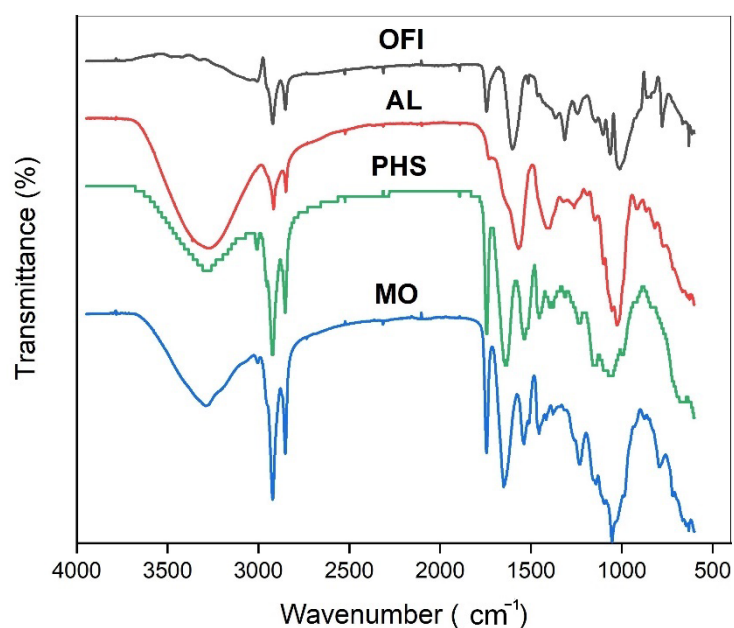


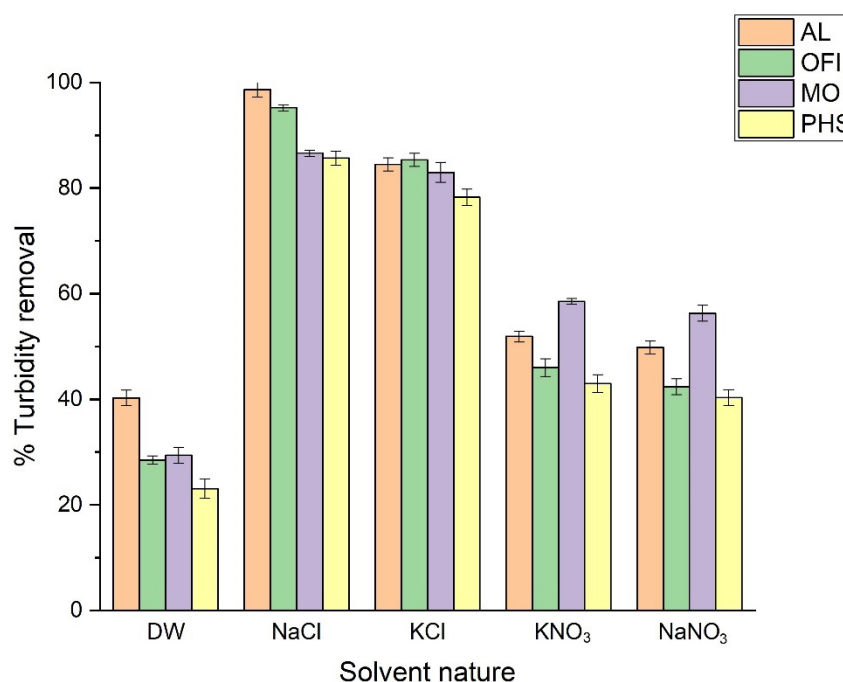
Figure 2. FTIR analysis of the studied bio-coagulants.

Table 1. Active functional groups in the four bio-coagulants.

Wave Number (cm ⁻¹)	Functional Group	References
3278.85	Hydroxyl group OH/N-H bonds	
2916.66	C-H asymmetric stretching in CH ₂	
2848.6	C-H symmetric stretching in CH ₂	
1567.72	Carbonyl function C=O (primary amides)	[34–37]
1398.82	CH ₃ primary aromatic amines	
1537	NH ₂ amine group	
1025.83	CO group/C-N stretching vibration of amine groups	

3.2. Effect of Solvent Nature on Turbidity Removal

The four bio-coagulant precursors were subjected to extraction using distilled water, NaCl, KCl, NaNO₃, and KNO₃, 0.5 M each, at a ratio of 5% (5/100 *w/v*). Figure 3 depicts the turbidity removal rate of each solvent (Dose 1,5 mL/200 mL, Natural pH (7,5), 500 FTU). It is more than obvious that each investigated solvent provided a different turbidity removal percentage, with a maximum attained with NaCl salt followed by KCl for the four bio-coagulants. The minimum turbidity removal was observed with distilled water. Similar results were obtained in previous studies, de Souza et al. [38] found that NaCl salt was more efficient in extracting OFI active component. They assumed that this tendency may be attributed to the increase in the solubility of the protein responsible for the coagulation activity; they considered that due to enhanced solvation, the addition of a neutral salt to a species may improve its protein solubility (salting-in effect). Okuda et al. [39], in a study of the enhancement of coagulation active component extraction from *Moringa oleifera* seeds, reported that MO-enhanced coagulation activity is a result of the proposed mechanism of promoting the breakdown of protein connections, resulting in greater solubilities upon salt addition. NaCl solution performed as a more effective solvent than pure water in dissolving protein-protein, protein-polysaccharide, and other associations in the seeds powder, resulting in an increase in protein solubility in the salt solution.

**Figure 3.** Effect of solvent nature on turbidity removal.

According to Madrona et al. [40], *Moringa oleifera* seed cationic protein does not dissolve effectively in water without the addition of salt. Baquerizo-Crespo et al. [41] and Dalvand et al. [42] also found that NaCl was the best salt for the extraction of coagulants from *Moringa stenopetala* seeds. Due to the proteinaceous profile of PHS, similar interpretations could also explain such results. Benalia et al. [35] used *Aloe vera* as a bio-coagulant for the treatment of drinking water and found that a concentration of 0.5 M NaCl was the most effective one to extract active agents, they attributed the improvement of coagulation activity to salting, as explained by the Debye-Hückel theory (increasing solvating power).

3.3. Effect of pH on Turbidity Removal

The surface charge of the coagulants is affected by the pH of the water, which in turn affects the degree to which the suspension is stabilized [43]. In this context, the effect of the initial pH (in the range of 2 to 12) of the turbid water was investigated. Figure 4 indicates that the coagulants showed significant turbidity removal almost over the entire pH range (2 to 8) with a percentage removal of 97.12%, 95.19%, 87.28%, 84.2%, 99.07%, and 99.37% for AV, OFI, MO, PHS, FeCl₃, and Alum, respectively. Beyond this pH value, a sharp decrease in turbidity removal% was observed, especially for seeds-based bio-coagulant (PHS, MO) and for Alum. A lower decrease was observed concerning AV, OFI, and FeCl₃ since most of the plant-based organic coagulants predominantly work on polymer bridging and charge neutralization [44].

The results obtained for OFI and AV support the hypothesis of adsorption facilitated by an inter-particle bridging mechanism where particle destabilization takes place in the adsorption of colloids onto the main chain and side chains of polysaccharides and forming bridge in terms of complexes of particle-polysaccharide-particle [45]. The higher turbidity removal in acidic media may be attributed to the protonation of hydroxyl and carboxyl groups of protein and carbohydrates present in the bio-coagulants [46–48]. Miller et al. [49] confirmed the involvement of –C–O–C, –OH, –C=O, N–H, and –CH groups of the carboxylic acids, amino acids, and polysaccharides present in OFI in the removal of Pb (II) ions by coagulation-flocculation. Because of the greater negative charge on the surface of the polysaccharide in alkaline media, turbidity removal efficiency decreases.

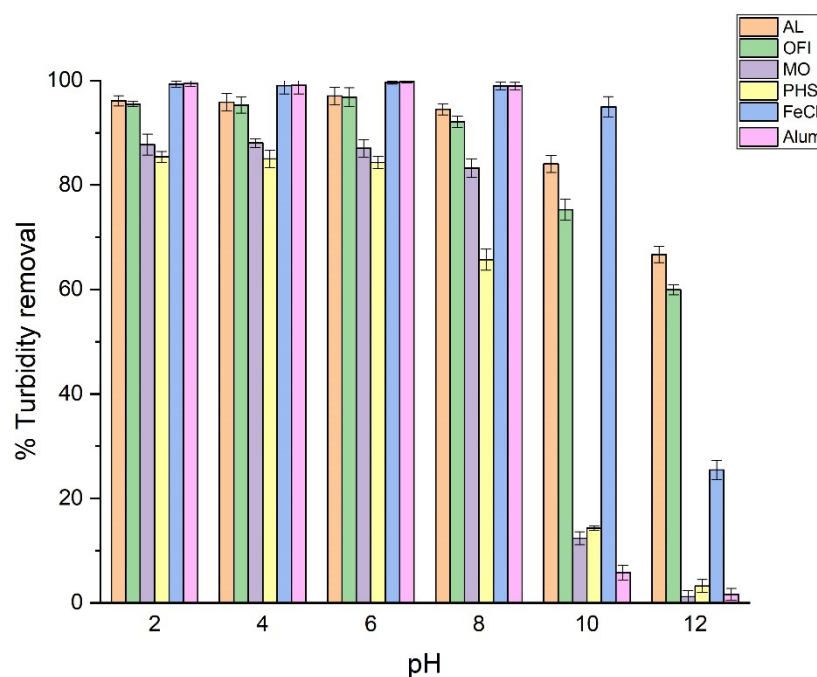


Figure 4. Effect of initial pH on turbidity removal.

The turbidity removal activity of the bio-coagulants may also be affected by changes in the molecular size or rearrangement of the monosaccharides of the polysaccharide chains in alkaline media [46,50]. The higher turbidity removal rates at acidic media by MO- and PHS-based bio-coagulant may be explained by the protonation of their protein amino groups providing better interaction between them and suspension's colloids [51,52]. These findings indicate that the use of the four bio-coagulants does not require pH adjustment. This non-dependence on pH is an advantage in the coagulation-flocculation process [53,54].

3.4. Effect of Bio-Coagulant Dosage on Turbidity Removal

In the coagulation and flocculation process, the dose of the coagulant is one of the most essential criteria to consider. Insufficient dose or overdosing leads to poor flocculation performance of the coagulant. Therefore, the optimal coagulant dose must be identified to reduce dosing costs and obtain optimal removal rates [55]. Due to the filtration stage, which retains approximately 70% of the insoluble powder precursors during bio-coagulant preparation, we chose to quantify the coagulant dose in units of (mL/200 mL) rather than (mg/L).

The dosage ranges were determined based on preliminary tests performed for this investigation. The results of these tests have also been supported by data from prior research [56–58]. Figure 5 illustrates the percentage of turbidity removed by each bio-coagulant with various dosages (in the range of 0.2 to 3.5 mL/200 mL). As it can be observed, increasing the bio-coagulant dose enhances the percentage elimination of turbidity until a plateau is reached, after which an additional increase in dosage does not improve the percentage removal of turbidity. As mentioned earlier, only charge neutralization and polymer bridging are the possible coagulation mechanisms for natural coagulants. Polymer bridging is preceded by polymer adsorption, a process by which long-chain polymers stick to the surface of colloidal particles due to the affinity between them. Only a portion of the polymers is bound to the particle, with the rest forming loops and tails [59]. These loops and tails constitute the primary structure of polymer bridging because they facilitate attachments to additional colloidal particles, resulting in bigger flocs. As a result, there must be enough vacant particle surface to provide efficient polymer bridging, and the bridging must spread long enough to overcome inter-particle repulsion. These criteria are addressed by providing a sufficient dosage of the bio-coagulant or, to be more specific, a required amount of polymer, which offers adequate bridging connections and a bare particle surface for strong polymer bridging; thus, a better turbidity removal performance [60].

3.5. Effect of Initial Turbidity

The influence of initial turbidity on turbidity removal effectiveness was also a significant aspect that needed thorough examination since wastewater streams produced by various sectors or sources might have varying concentrations. To assess the effect of initial turbidity on the efficiency of the four bio-coagulants, five bentonite suspensions with varying initial turbidity (50, 200, 500, 800, and 1000 FTU) were treated under optimal conditions (pH 7.5, coagulant dose 1.5, 3, 3, 3.5, 1.5, and 1.5 mL/200 mL for AV, OFI, MO, PHS, Alum, and FeCl₃, respectively). Using a 0.5 M NaCl salt solution, the coagulants were extracted from the four precursors. The results in Figure 6 show that the percentage of turbidity reduction increased as initial turbidity in bentonite suspensions increased from low to very high levels. This is likely the result of a higher particle concentration in water with a high turbidity level, which enhanced the frequency of collisions between suspended particles and coagulants, hence facilitating a more efficient coagulation-flocculation process. With increased contact and interaction between coagulating and suspending particles, larger and denser flocs would develop, leading to a greater sedimentation rate in the treatment process according to Stokes law [46,61]; these findings are comparable to those of prior studies [62–64].

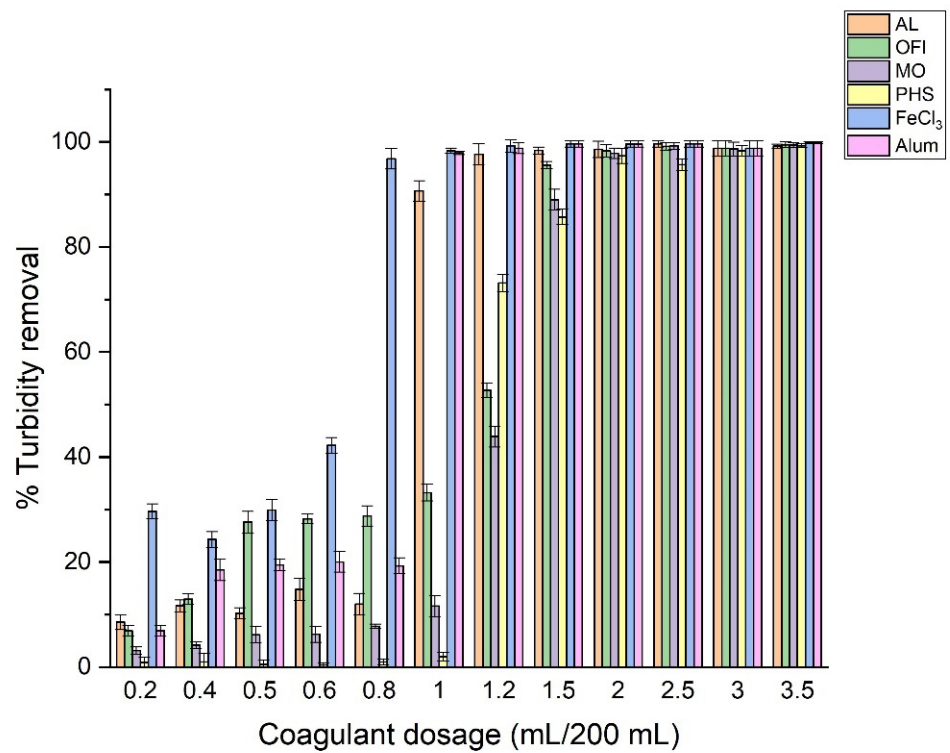


Figure 5. Effect of coagulant dosage on turbidity removal.

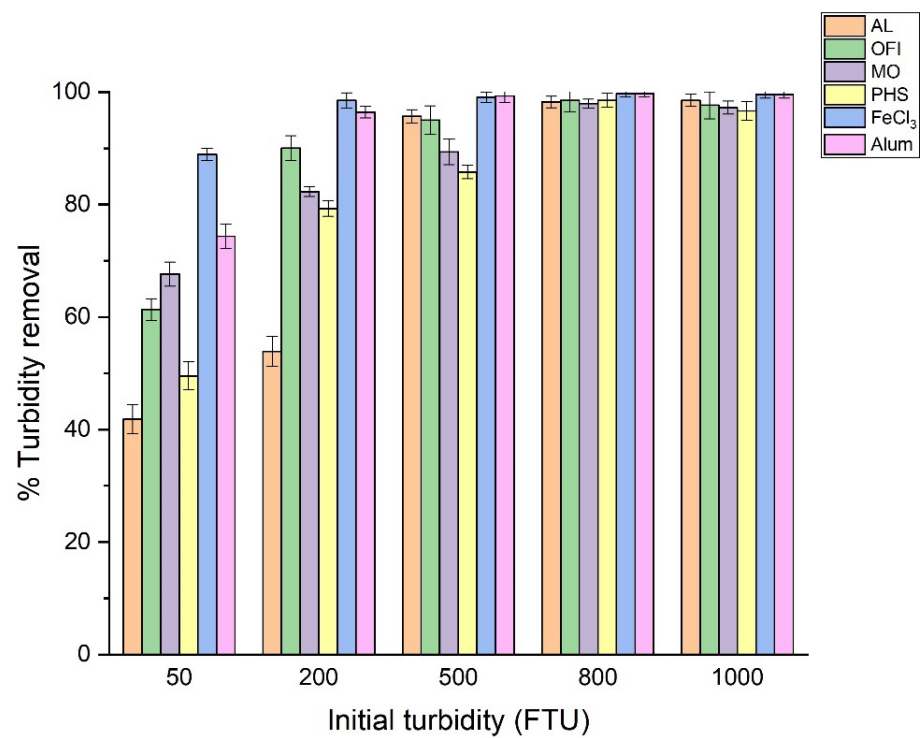


Figure 6. Effect of initial turbidity on the effectiveness of the coagulants.

Kurniawan et al. [65] attributed this phenomenon to the mechanism of floc formation by the vast majority of bio-coagulants/bio-flocculants, in which particle bridging becomes more efficient when more dense suspended solids are present; in contrast, when initial turbidity is low, there are fewer collisions between coagulant and pollutant due to an insufficient number of pollutants. The creation of smaller flocs should lower the sedimentation rate, extending the sedimentation time [66]. For high turbidity levels, the six coagulants have approximately the same efficiency removal, with a percentage of 90.76%, 99.76%, 98.5%, 98.04%, 99.92%, and 100% for AV, OFI, MO, PHS, FeCl₃ and Alum, respectively). These findings demonstrate the versatility of the four bio-coagulants to be used at a wide range of water conditions [29] and the potential of AV and PHS as green alternatives to traditional coagulants when compared to previously studied natural coagulants (Table 2).

Table 2. Comparison of the studied bio-coagulants with prior research.

Bio-Coagulant	Preparation	Effluent Type	Optimum pH	Optimum Dosage	Turbidity Removal	References
AV	NaCl extraction 0.5 M	Bentonite synthetic water	2–8	2 mL/200 mL	100%	This study
OFI	NaCl extraction 0.5 M	Bentonite synthetic water	2–8	2.5 mL/200 mL	100%	This study
MO	NaCl extraction 0.5 M	Bentonite synthetic water	2–8	3 mL/200 mL	99.71%	This study
PHS	NaCl extraction 0.5 M	Bentonite synthetic water	2–8	3.5 mL/200 mL	99.43%	This study
<i>Malva sylvestris</i>	Mucilage	Synthetic	7	12 mg/L	96.3–97.4%	[67]
<i>Luffa cylindrica</i>	Powder	Surface water	9.4	8000 mg/L	85%	[68]
<i>Cicer arietinum</i>	Water extract	Clay	/	100 mg/L	95.9%	[67]
<i>Cocos nucifera</i>	Water extract	/	8	250 mg/L	25%	[69]
<i>Cassia obtusifolia</i>	Powder	Palm oil	acidic	2470 mg/L + 1115 mg/L aluminum sulfate	82%	[70]
<i>Gossypium spp</i>	Water extract	Phosphate	6	500 mg/L	>70%	[71]
<i>Ocimum basilicum</i>	Water extract at 50 °C + 0.9% NaCl/1 h	Textile (cationic dye)	6.5	9.6 mg/L + 20 mg of aluminum sulfate	68.5%	[72]
<i>Moringa oleifra</i>	0.5 M (NH ₄) ₂ SO ₄ extract	Synthetic	5	5 mL	94%	[73]
<i>Jatropha curcas</i>	5% water extract	Synthetic	3	80 mg/L	90%	[74]
<i>Vicia faba</i>	Textile wastewater	Textile wastewater	7	6.75 mg/L	60–70%	[75]
<i>Phaseolus vulgaris</i>	NaCl extraction	Kaolinturbid water	7	1 mL/L	95%	[76]

3.6. Effect of Bio-Coagulants Aging on Turbidity Removal

As bio-coagulants are constituted of a variety of polysaccharides and organic chemicals, they are susceptible to enzymatic degradation over time. To investigate the influence of this gradual deterioration on their coagulation effectiveness, the four bio coagulants were open-air stored in opaque bottles. The coagulation/flocculation tests were performed for up to 20 days in optimal conditions, and the findings are shown in Figure 7. Due to the enzymatic degradation of polysaccharides, the coagulants' removal performance diminishes with time. The enzymes attack the different chemical bonds present in polysaccharides and depolymerize them, which might affect the polysaccharide's physicochemical characteristics and ultimately reduce its coagulation efficiency [77,78].

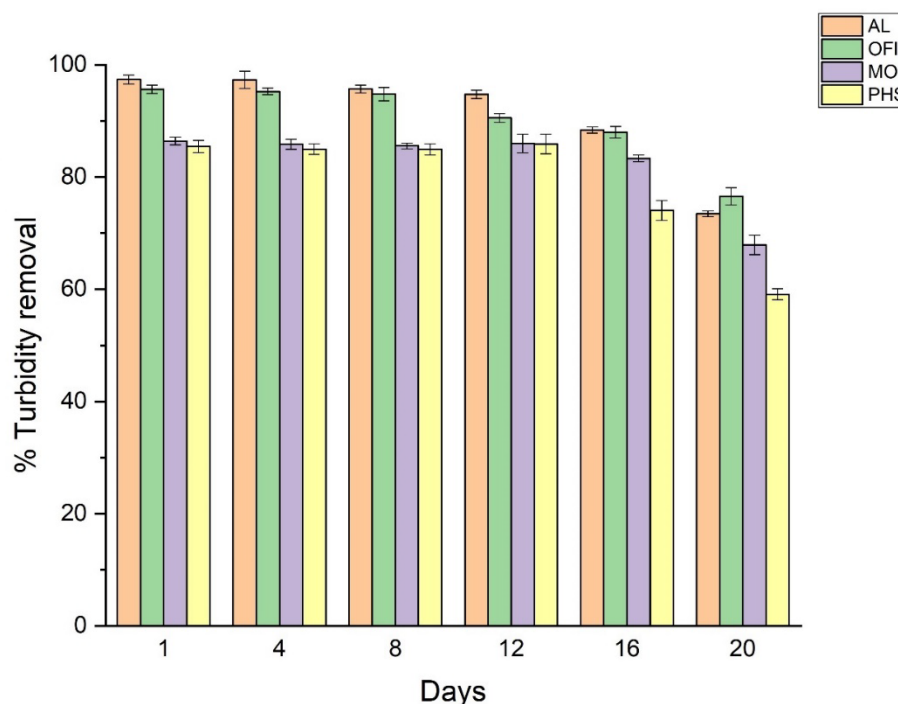


Figure 7. Effect of bio-coagulants aging on turbidity removal.

3.7. Comparison of Sludge Production

According to Table 3, there was a significant difference in the sludge volume produced when bio-coagulants were used compared to inorganic ones, The formation of aluminum hydroxide precipitates by Alum, as stated by Kristiano et al. [79] is a factor that contributes to the overall amount of sludge that is formed during the coagulation process. On the other hand, natural coagulants cause agglomeration formation, which leads to greater settleable floc with no further creation of precipitates. This is because natural coagulants do not include any additives. Organic polymers often generate less sludge than inorganic salts because they do not add weight to the water or react with other ions to form precipitates. Thus, the bio-coagulant produced a compact and volume-reduced sludge. This may also be ascribed to the disparity in floc formation between FeCl_3 and bio-coagulant [42,80].

In addition, since these studied coagulants are derived from plant materials, their byproducts are organic and biodegradable, making them a more environmentally friendly alternative to the other treatment technique using chemical coagulants. These findings provide a useful indication of the appropriateness of the four tested bio-coagulants as a potential replacement for chemical ones, such as FeCl_3 and Alum.

Table 3. Comparison of bio-coagulant sludge production with FeCl_3 and Alum. (Mean \pm SD, $n = 3$).

Coagulant	Coagulant Dosage (mL/200 mL)	% Turbidity Removal	Sludge Volume (mL/L)
MO	1.5	87 \pm 2.5	25 \pm 1
PHS	1.5	84.2 \pm 3.7	30 \pm 1
OFI	1.5	94.81 \pm 2.5	26 \pm 1
AV	1.5	87.1 \pm 3.0	22 \pm 1
FeCl_3	1.5	99.12 \pm 3.7	52 \pm 1
Alum	1.5	98.84 \pm 3.7	62 \pm 1

3.8. Variation of pH

The performances of the bio-coagulants were compared to Alum and FeCl₃ in terms of the alteration of the final pH. Table 4 shows that a slight change occurred in the initial pH in the treated water when the bio-coagulants were applied, while a significant decrease in pH value was observed when Alum and FeCl₃ were used. These findings are consistent with the results of prior research; Adnan et al. [81] reported that mushroom chitosan-based bio-coagulant slightly alters the pH value, whereas a considerable drop was noticed when the inorganic coagulant (Alum) was applied. According to the findings of Abidin et al. [74], the use of the extract of *Jatropha curcas* resulted in a minor shift in the initial pH of the water samples that were treated. Amagloh et al. [82] suggested that when there are basic amino acids present in protein that can accept protons from water, it releases hydroxyl groups into water. This could explain the slight increase in pH of the water treated with MO, PHS, AV, and OFI. These results imply that a treatment without acidifying or alkalinizing water is possible, adding a major advantage compared to commercial inorganic coagulants, which require post-pH modification of the treated water.

Table 4. Values of pH of water samples treated with the coagulants (mean \pm SD, $n = 3$).

Coagulant	Initial pH of Water (before Addition of Coagulant)	Final pH
AV		7.28 \pm 0.42
OFI		7.25 \pm 0.15
MO	7.23 \pm 0.32	7.80 \pm 0.28
PHS		7.68 \pm 0.31
FeCl ₃		3.28 \pm 0.46
Alum		4.12 \pm 0.22

4. Conclusions

The present study analyzed the potential of two novel bio-coagulants in the treatment of turbid water by comparing their performances to two well-established bio-coagulants (MO and OFI) and two chemical ones (Alum and Ferric chloride). The coagulation-flocculation research demonstrates that AV and PHS seeds have a high potential to perform as suitable as well-established bio-coagulants and chemical coagulants, removing turbidity from bentonite suspension by more than 90% across the entire pH range of 2 to 8, with a maximum turbidity removal of approximately 100% obtained at natural pH of 7.5 using optimal doses of 1.5, 3, 3, 3.5, 1.5, and 1.5 mL/200 mL for AV, OFI, MO, PHS, Alum, and FeCl₃, respectively. The active molecules involved in the coagulation-flocculation process were assumed to be a combo of polysaccharides and proteins responsible for inter-particle bridging and charge neutralization. The four bio-coagulants had the advantage of not altering the pH of the treated water and producing less sludge than Alum and Ferric chloride, which caused a significant drop in pH values.

Despite the fact that Alum and Ferric chloride performed better in terms of turbidity removal, the results of this study are sufficient to encourage the use of AV and PHS seeds for the treatment of turbid water, although there is a clear need for further research into the development of feasible technologies using these bio-coagulants for the elimination of turbidity in water and wastewater.

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Abbreviations

AV	<i>Aloe vera</i>
OFI	<i>Opuntia ficus indica</i>
MO	<i>Moringa oleifera</i>
PHS	<i>Pinus halepensis</i>
FeCl ₃	Ferric chloride
Alum	Aluminum sulfate
HCl	Hydrochloric acid
NaOH	Sodium hydroxide
NaCl	Sodium chloride
KCl	Potassium chloride
KNO ₃	Potassium nitrate
NaNO ₃	Sodium nitrate

References

- Larsen, T.A.; Hoffmann, S.; Lüthi, C.; Truffer, B.; Maurer, M. Emerging solutions to the water challenges of an urbanizing world. *Science* **2016**, *352*, 928–933. [[CrossRef](#)] [[PubMed](#)]
- Wang, X.-H.; Wang, X.; Huppel, G.; Heijungs, R.; Ren, N.-Q. Environmental implications of increasingly stringent sewage discharge standards in municipal wastewater treatment plants: Case study of a cool area of China. *J. Clean. Prod.* **2015**, *94*, 278–283. [[CrossRef](#)]
- Chen, C.-L.; Chang, K.-Y.; Pan, T.-M. *Monascus purpureus* NTU 568 fermented product improves memory and learning ability in rats with aluminium-induced alzheimer's disease. *J. Funct. Foods* **2016**, *21*, 167–177. [[CrossRef](#)]
- Lugo-Arias, J.; Lugo-Arias, E.; Ovallos-Gazabon, D.; Arango, J.; de la Puente, M.; Silva, J. Effectiveness of the mixture of nopal and cassava starch as clarifying substances in water purification: A case study in Colombia. *Heliyon* **2020**, *6*, e04296. [[CrossRef](#)]
- Kurniawan, S.; Abdullah, S.; Imron, M.; Said, N.; Ismail, N.; Hasan, H.; Othman, A.; Purwanti, I. Challenges and opportunities of Biocoagulant/Biofloculant application for drinking water and wastewater treatment and its potential for sludge recovery. *Int. J. Environ. Res. Public Health* **2020**, *17*, 9312. [[CrossRef](#)]
- Adebisi, G.A.; Chowdhury, Z.Z.; Alaba, P.A. Equilibrium, kinetic, and thermodynamic studies of lead ion and zinc ion adsorption from aqueous solution onto activated carbon prepared from palm oil mill effluent. *J. Clean. Prod.* **2017**, *148*, 958–968. [[CrossRef](#)]
- Miklos, D.B.; Remy, C.; Jekel, M.; Linden, K.G.; Drewes, J.E.; Hübner, U. Evaluation of advanced oxidation processes for water and wastewater treatment—A critical review. *Water Res.* **2018**, *139*, 118–131. [[CrossRef](#)]
- Imran, M.; Crowley, D.E.; Khalid, A.; Hussain, S.; Mumtaz, M.W.; Arshad, M. Microbial Biotechnology for decolorization of Textile Wastewaters. *Rev. Environ. Sci. Bio/Technol.* **2014**, *14*, 73–92. [[CrossRef](#)]
- Dasgupta, J.; Sikder, J.; Chakraborty, S.; Curcio, S.; Drioli, E. Remediation of textile effluents by membrane based treatment techniques: A state of the art review. *J. Environ. Manag.* **2015**, *147*, 55–72. [[CrossRef](#)]
- World Health Organization (WHO). *Guidelines for Drinking-Water Quality*; WHO: Geneva, Switzerland, 2004; Volume 1.
- Soros, A.; Amburgey, J.E.; Stauber, C.E.; Sobsey, M.D.; Casanova, L.M. Turbidity reduction in drinking water by coagulation-flocculation with chitosan polymers. *J. Water Health* **2019**, *17*, 204–218. [[CrossRef](#)]
- Vijayaraghavan, G.; Sivakumar, T.; Kumar, A.V. Application of plant based coagulants for waste water treatment. *Int. J. Adv. Eng. Res. Stud.* **2011**, *1*, 88–92.
- Teh, C.Y.; Budiman, P.M.; Shak, K.P.; Wu, T.Y. Recent advancement of coagulation–flocculation and its application in wastewater treatment. *Ind. Eng. Chem. Res.* **2016**, *55*, 4363–4389. [[CrossRef](#)]
- Rivera Cañón, A.F.; Hernández Carrillo, C.G.; Gómez Cuaspuud, J.A. Evaluation of physicochemical properties of Nopal (*Opuntia Ficus-Indica*) as bio coagulant-flocculant for water treatment. *J. Phys. Conf. Ser.* **2021**, *2046*, 012057. [[CrossRef](#)]
- Ang, W.L.; Mohammad, A.W. State of the art and sustainability of natural coagulants in water and wastewater treatment. *J. Clean. Prod.* **2020**, *262*, 121267. [[CrossRef](#)]
- Saleem, M.; Bachmann, R.T. A contemporary review on plant-based coagulants for applications in water treatment. *J. Ind. Eng. Chem.* **2019**, *72*, 281–297. [[CrossRef](#)]
- Bratby, J. *Coagulation and Flocculation*; Uplands: Croydon, UK, 1980.
- Williams, R.B.; Culp, G.L. *Handbook of Public Water Systems*; Van Nostrand Reinhold Co: New York, NY, USA, 1986; p. 379.

19. Dick, M.; Dal Magro, L.; Rodrigues, R.C.; de Oliveira Rios, A.; Flôres, S.H. Valorization of *Opuntia monacantha* (Willd.) haw. Cladodes to obtain a mucilage with hydrocolloid features: Physicochemical and functional performance. *Int. J. Biol. Macromol.* **2019**, *123*, 900–909. [[CrossRef](#)]
20. Albano, C.; Negro, C.; Tommasi, N.; Gerardi, C.; Mita, G.; Miceli, A.; De Bellis, L.; Blando, F. Betalains, phenols and antioxidant capacity in cactus pear [*Opuntia ficus-indica* (L.) mill.] fruits from Apulia (South Italy) genotypes. *Antioxidants* **2015**, *4*, 269–280. [[CrossRef](#)]
21. Gassenschmidt, U.; Jany, K.D.; Bernhard, T.; Niebergall, H. Isolation and characterization of a flocculating protein from *Moringa oleifera* Lam. *Biochim. Biophys. Acta (BBA)-Gen. Subj.* **1995**, *1243*, 477–481. [[CrossRef](#)]
22. Mandloi, M.; Chaudhari, S.; Folkard, G.K. Evaluation of natural coagulants for direct filtration. *Environ. Technol.* **2004**, *25*, 481–489. [[CrossRef](#)]
23. Hadadi, A.; Imessaoudene, A.; Bollinger, J.-C.; Bouzaza, A.; Assadi, A.A.; Mouni, L. An eco-friendly approach to dye wastewater treatment using a novel biocoagulant: Aleppo Pine Seeds (*Pinus halepensis* Mill.) Parametrical Study and Response Surface Methodology. 2022, *Under examination for publication*.
24. Ihaddaden, S.; Aberkane, D.; Boukerroui, A.; Robert, D. Removal of methylene blue (basic dye) by coagulation-flocculation with biomaterials (bentonite and *Opuntia Ficus indica*). *J. Water Process Eng.* **2022**, *49*, 102952. [[CrossRef](#)]
25. Vargas-Solano, S.V.; Rodríguez-González, F.; Martínez-Velarde, R.; Morales-García, S.S.; Jonathan, M.P. Removal of heavy metals present in water from the Yautepéc River Morelos México, using *Opuntia ficus-indica* mucilage. *Environ. Adv.* **2021**, *7*, 100160. [[CrossRef](#)]
26. Putra, R.S.; Putri, C.I.; Tyagustin, N.S. The combination of electroflotation-biocoagulation process using aloe vera for river water treatment. *IOP Conf. Ser. Mater. Sci. Eng.* **2021**, *1087*, 012047. [[CrossRef](#)]
27. Benalia, A.; Derbal, K.; Khalfaoui, A.; Bouchareb, R.; Panico, A.; Gisonni, C.; Crispino, G.; Pirozzi, F.; Pizzi, A. Use of Aloe Vera as an organic coagulant for improving drinking water quality. *Water* **2021**, *13*, 2024. [[CrossRef](#)]
28. Bouaouine, O.; Bourven, I.; Khalil, F.; Bressollier, P.; Baudu, M. Identification and role of *Opuntia Ficus indica* constituents in the flocculation mechanism of Colloidal Solutions. *Sep. Purif. Technol.* **2019**, *209*, 892–899. [[CrossRef](#)]
29. Zurina, A.Z.; Mohd Fadzli, M.; Abdul Ghani, L.A. Preliminary study of rambutan (*Nephelium lappaceum*) seed as potential biocoagulant for turbidity removal. *Adv. Mater. Res.* **2014**, *917*, 96–105. [[CrossRef](#)]
30. ElSayed, E.M.; El-Den, A.N.; Elkady, M.F.; Zaatout, A.A. Comparison of coagulation performance using natural coagulants against traditional ones. *Sep. Sci. Technol.* **2020**, *56*, 1779–1787. [[CrossRef](#)]
31. Ghebremichael, K.A.; Gunaratna, K.R.; Henriksson, H.; Brumer, H.; Dalhammar, G. A simple purification and activity assay of the coagulant protein from *Moringa oleifera* seed. *Water Res.* **2005**, *39*, 2338–2344. [[CrossRef](#)]
32. Aboulhassan, M.A.; Souabi, S.; Yaacoubi, A.; Baudu, M. Improvement of paint effluents coagulation using natural and synthetic coagulant aids. *J. Hazard. Mater.* **2006**, *138*, 40–45. [[CrossRef](#)]
33. Yang, W.; Shang, J.; Sharma, P.; Li, B.; Liu, K.; Flury, M. Colloidal stability and aggregation kinetics of biochar colloids: Effects of pyrolysis temperature, cation type, and humic acid concentrations. *Sci. Total Environ.* **2019**, *658*, 1306–1315. [[CrossRef](#)]
34. Marques, T.L.; Alves, V.N.; Coelho, L.M.; Coelho, N.M. Removal of ni(ii) from aqueous solution using *Moringa oleifera* seeds as a bioadsorbent. *Water Sci. Technol.* **2012**, *65*, 1435–1440. [[CrossRef](#)]
35. Benalia, A.; Derbal, K.; Khalfaoui, A.; Pizzi, A.; Medjahdi, G. The use of as natural coagulant in algerian drinking water treatment plant. *J. Renew. Mater.* **2022**, *10*, 625–637. [[CrossRef](#)]
36. Kandasamy, K.; Venkatesh, M.; Khadar, Y.S.; Rajasingh, P. One-pot green synthesis of cds quantum dots using *Opuntia ficus-indica* fruit sap. *Mater. Today Proc.* **2020**, *26*, 3503–3506. [[CrossRef](#)]
37. León-Martínez, F.M.; Cano-Barrita, P.F.; Castellanos, F.; Luna-Vicente, K.B.; Ramírez-Arellanes, S.; Gómez-Yáñez, C. Carbonation of high-calcium lime mortars containing cactus mucilage as additive: A spectroscopic approach. *J. Mater. Sci.* **2020**, *56*, 3778–3789. [[CrossRef](#)]
38. De Souza, M.T.; Ambrosio, E.; de Almeida, C.A.; de Souza Freitas, T.K.; Santos, L.B.; de Cinque Almeida, V.; Garcia, J.C. The use of a natural coagulant (*Opuntia ficus-indica*) in the removal for organic materials of textile effluents. *Environ. Monit. Assess.* **2014**, *186*, 5261–5271. [[CrossRef](#)]
39. Okuda, T.; Baes, A.U.; Nishijima, W.; Okada, M. Improvement of extraction method of coagulation active components from *Moringa oleifera* seed. *Water Res.* **1999**, *33*, 3373–3378. [[CrossRef](#)]
40. Madrona, G.S.; Serpelloni, G.B.; Vieira, A.M.S.; Nishi, L.; Cardoso, K.C.; Bergamasco, R. Study of the effect of saline solution on the extraction of the *Moringa oleifera* seed's active component for water treatment. *Water Air Soil Pollut.* **2010**, *211*, 409–415. [[CrossRef](#)]
41. Baquerizo-Crespo, R.J.; Nuñez, Y.; Albite, J.; Macías-Alcívar, J.A.; Cedeno-Zambrano, N.; Dueñas-Rivadeneira, A.A.; Gomez-Salcedo, Y.; Rodríguez-Díaz, J.M. Biocoagulants as an Alternative for Water Treatment. In *Advances in the Domain of Environmental Biotechnology*; Springer: Singapore, 2021; pp. 313–334. [[CrossRef](#)]
42. Dalvand, A.; Gholibegloo, E.; Ganjali, M.R.; Golchinpoor, N.; Khazaei, M.; Kamani, H.; Hosseini, S.S.; Mahvi, A.H. Comparison of moringa stenopetala seed extract as a clean coagulant with alum and *Moringa stenopetala*-Alum hybrid coagulant to remove direct dye from textile wastewater. *Environ. Sci. Pollut. Res.* **2016**, *23*, 16396–16405. [[CrossRef](#)]
43. Oladoja, N.A. Headway on natural polymeric coagulants in water and wastewater treatment operations. *J. Water Process Eng.* **2015**, *6*, 174–192. [[CrossRef](#)]

44. Kristianto, H. The potency of Indonesia native plants as natural coagulant: A Mini Review. *Water Conserv. Sci. Eng.* **2017**, *2*, 51–60. [[CrossRef](#)]
45. Beyene, H.D.; Hailegebrial, T.D.; Dirersa, W.B. Investigation of coagulation activity of Cactus Powder in water treatment. *J. Appl. Chem.* **2016**, *2016*, 7815903. [[CrossRef](#)]
46. Choudhary, M.; Ray, M.B.; Neogi, S. Evaluation of the potential application of cactus (*Opuntia ficus-indica*) as a bio-coagulant for pre-treatment of oil sands process-affected water. *Sep. Purif. Technol.* **2019**, *209*, 714–724. [[CrossRef](#)]
47. Adjeroud-Abdellatif, N.; Hammoui, Y.; Boudria, A.; Agab, S.; Choulak, F.; Leclerc, J.-P.; Merzouk, B.; Madani, K. Effect of a natural coagulant extract from *Opuntia ficus-indica* cladode on electrocoagulation-electroflotation water treatment process. *Int. J. Environ. Anal. Chem.* **2020**, *100*, 1–25. [[CrossRef](#)]
48. Othmani, B.; Gamelas, J.A.; Rasteiro, M.G.; Khadhraoui, M. Characterization of two cactus formulation-based flocculants and investigation on their flocculating ability for cationic and anionic dyes removal. *Polymers* **2020**, *12*, 1964. [[CrossRef](#)] [[PubMed](#)]
49. Miller, S.M.; Fugate, E.J.; Craver, V.O.; Smith, J.A.; Zimmerman, J.B. Toward understanding the efficacy and mechanism of *Opuntia* spp. as a natural coagulant for potential application in water treatment. *Environ. Sci. Technol.* **2008**, *42*, 4274–4279. [[CrossRef](#)] [[PubMed](#)]
50. Nasim, T.; Pal, A.; Giri, A.; Goswami, L.; Bandyopadhyay, A. Exploring polyelectrolytic features of the exudate from native acacia nilotica for flocculating aqueous kaolin suspension. *Sep. Purif. Technol.* **2014**, *131*, 50–59. [[CrossRef](#)]
51. Tie, J.; Li, P.; Xu, Z.; Zhou, Y.; Li, C.; Zhang, X. Removal of congo red from aqueous solution using *Moringa oleifera* seed cake as natural coagulant. *Desalination Water Treat.* **2014**, *54*, 2817–2824. [[CrossRef](#)]
52. Chethana, M.; Sorokhaibam, L.G.; Bhandari, V.M.; Raja, S.; Ranade, V.V. Green approach to dye wastewater treatment using biocoagulants. *ACS Sustain. Chem. Eng.* **2016**, *4*, 2495–2507. [[CrossRef](#)]
53. Aboulhassan, M.A.; Harif, S.; Souabi, S.; Yaacoubi, A. Efficient and sustainable treatment of industrial wastewater using a tannin-based polymer. *Int. J. Sustain. Eng.* **2021**, *14*, 1943–1949. [[CrossRef](#)]
54. Okaiyeto, K.; Nwodo, U.U.; Okoli, S.A.; Mabinya, L.V.; Okoh, A.I. Implications for public health demands alternatives to inorganic and synthetic flocculants: Bioflocculants as important candidates. *MicrobiologyOpen* **2016**, *5*, 177–211. [[CrossRef](#)]
55. Ramli, S.F.; Aziz, H.A. Use of ferric chloride and chitosan as coagulant to remove turbidity and color from landfill leachate. *Appl. Mech. Mater.* **2015**, *773–774*, 1163–1167. [[CrossRef](#)]
56. Barbot, E.; Dussouillez, P.; Bottero, J.Y.; Moulin, P. Coagulation of bentonite suspension by polyelectrolytes or ferric chloride: Floc Breakage and Reformation. *Chem. Eng. J.* **2010**, *156*, 83–91. [[CrossRef](#)]
57. Al-Husseini, T.R.; Ghawi, A.H.; Ali, A.H. Performance of hydraulic jump rapid mixing for enhancement of turbidity removal from synthetic wastewater: A comparative study. *J. Water Process Eng.* **2019**, *30*, 100590. [[CrossRef](#)]
58. Baghvand, A.; Zand, A.D.; Mehrdadi, N.; Karbassi, A. Optimizing coagulation process for low to high turbidity waters using aluminum and iron salts. *Am. J. Environ. Sci.* **2010**, *6*, 442–448. [[CrossRef](#)]
59. Bolto, B.; Gregory, J. Organic polyelectrolytes in water treatment. *Water Res.* **2007**, *41*, 2301–2324. [[CrossRef](#)]
60. Tripathy, T.; De, B.R. Flocculation: A new way to treat the waste water. *J. Phys. Sci.* **2006**, *10*, 93–127.
61. Zhou, Z.; Yang, Y.; Li, X.; Gao, W.; Liang, H.; Li, G. Coagulation efficiency and flocs characteristics of recycling sludge during treatment of low temperature and micro-polluted water. *J. Environ. Sci.* **2012**, *24*, 1014–1020. [[CrossRef](#)]
62. Ramavandi, B. Treatment of water turbidity and bacteria by using a coagulant extracted from plantago ovata. *Water Resour. Ind.* **2014**, *6*, 36–50. [[CrossRef](#)]
63. Gaikwad, V.T.; Munavalli, G.R. Turbidity removal by conventional and ballasted coagulation with natural coagulants. *Appl. Water Sci.* **2019**, *9*, 130. [[CrossRef](#)]
64. Asrafuzzaman, M.; Fakhruddin, A.N.; Hossain, M.A. Reduction of turbidity of water using locally available natural coagulants. *ISRN Microbiol.* **2011**, *2011*, 632189. [[CrossRef](#)]
65. Kurniawan, S.B.; Abdullah, S.R.; Othman, A.R.; Purwanti, I.F.; Imron, M.F.; Ismail, N.I.; Ahmad, A.; Hasan, H.A. Isolation and characterisation of bioflocculant-producing bacteria from aquaculture effluent and its performance in treating high turbid water. *J. Water Process Eng.* **2021**, *42*, 102194. [[CrossRef](#)]
66. Bahrodin, M.B.; Zaidi, N.S.; Hussein, N.; Sillanpää, M.; Prasetyo, D.D.; Syafiuddin, A. Recent advances on coagulation-based treatment of wastewater: Transition from chemical to natural coagulant. *Curr. Pollut. Rep.* **2021**, *7*, 379–391. [[CrossRef](#)]
67. Anastasakis, K.; Kalderis, D.; Diamadopoulos, E. Flocculation behavior of Mallow and okra mucilage in treating wastewater. *Desalination* **2009**, *249*, 786–791. [[CrossRef](#)]
68. Sowmeyan, R.; Santhosh, J.; Latha, R. Effectiveness of herbs in community water treatment. *Int. Res. J. Biochem. Bioinform.* **2011**, *1*, 297–303.
69. Fatombi, J.K.; Lartiges, B.; Aminou, T.; Barres, O.; Caillet, C. A natural coagulant protein from copra (*Cocos nucifera*): Isolation, characterization, and potential for water purification. *Sep. Purif. Technol.* **2013**, *116*, 35–40. [[CrossRef](#)]
70. Shak, K.P.; Wu, T.Y. 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. Crop. Prod.* **2015**, *76*, 1169–1178. [[CrossRef](#)]
71. Babayemi, K.A.; Onukwuli, O.D. Phosphate removal from phosphorus containing waste water by coagulation/flocculation process using gossypium spp. (GS) as coagulant. *Curr. Adv. Environ. Sci.* **2015**, *3*, 1–5. [[CrossRef](#)]
72. Shamsnejati, S.; Chaibakhsh, N.; Pendashteh, A.R.; Hayeripour, S. Mucilaginous seed of ocimum basilicum as a natural coagulant for textile wastewater treatment. *Ind. Crop. Prod.* **2015**, *69*, 40–47. [[CrossRef](#)]

73. Tie, J.; Jiang, M.; Li, H.; Zhang, S.; Zhang, X. A comparison between *Moringa oleifera* seed presscake extract and polyaluminum chloride in the removal of direct black 19 from synthetic wastewater. *Ind. Crop. Prod.* **2015**, *74*, 530–534. [[CrossRef](#)]
74. Abidin, Z.Z.; Ismail, N.; Yunus, R.; Ahamad, I.S.; Idris, A. A preliminary study on *Jatropha curcas* as coagulant in wastewater treatment. *Environ. Technol.* **2011**, *32*, 971–977. [[CrossRef](#)]
75. Kukić, D.V.; Šćiban, M.B.; Prodanović, J.M.; Tepić, A.N.; Vasić, M.A. Extracts of fava bean (*Vicia faba* L.) seeds as natural coagulants. *Ecol. Eng.* **2015**, *84*, 229–232. [[CrossRef](#)]
76. Muthuraman, G.; Sasikala, S. Removal of turbidity from drinking water using natural coagulants. *J. Ind. Eng. Chem.* **2014**, *20*, 1727–1731. [[CrossRef](#)]
77. Ward, O.P.; Moo-young, M.; Venkat, K. Enzymatic degradation of cell wall and related plant polysaccharides. *Crit. Rev. Biotechnol.* **1989**, *8*, 237–274. [[CrossRef](#)] [[PubMed](#)]
78. Byong, H.L. *Fundamentals of Food Biotechnology*; Wiley-VCH Publishers Inc.: New York, NY, USA, 1996.
79. Kristianto, H.; Rahman, H.; Prasetyo, S.; Sugih, A.K. Removal of congo red aqueous solution using *Leucaena leucocephala* seed's extract as natural coagulant. *Appl. Water Sci.* **2019**, *9*, 88. [[CrossRef](#)]
80. Ndabigengesere, A.; Narasiah, K.S. Quality of water treated by coagulation using *Moringa oleifera* seeds. *Water Res.* **1998**, *32*, 781–791. [[CrossRef](#)]
81. Adnan, O.; Abidin, Z.Z.; Idris, A.; Kamarudin, S.; Al-Qubaisi, M.S. A novel biocoagulant agent from Mushroom Chitosan as water and wastewater therapy. *Environ. Sci. Pollut. Res.* **2017**, *24*, 20104–20112. [[CrossRef](#)]
82. Amagloh, F.K.; Benang, A. Effectiveness of *Moringa oleifera* seed as coagulant for water purification. *Afr. J. Agric. Res.* **2009**, *4*, 119–123.