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Use of *Aloe vera* as an Organic Coagulant for Improving Drinking Water Quality

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Abstract: The coagulation–flocculation–sedimentation process is widely used for removal of suspended solids and water turbidity reduction. The most common coagulants used to conduct this process are aluminum sulfate and ferric sulfate. In this paper, the use of *Aloe vera* as a natural-based coagulant for drinking water treatment was tested. The bio-coagulant was used in two different forms: powder as well as liquid; the latter was extracted with distilled water used as a solvent. The obtained results showed that the use of the natural coagulant (*Aloe vera*) in both powder (AV-Powder) and liquid (AV-H_2O) forms reduced the water turbidity at natural pH by 28.23% and 87.84%, respectively. Moreover, it was found that the use of the two previous forms of bio-coagulant for drinking water treatment had no significant influence on the following three parameters: pH, alkalinity, and hardness. The study of the effect of pH on the process performance using *Aloe vera* as a bio-coagulant demonstrated that the maximum turbidity removal efficiency accounted for 53.53% and 88.23% using AV-Powder and AV-H_2O, respectively, at optimal pH 6.

Keywords: *Aloe vera*; extract; turbidity; powder; drinking water; jar test.

1. Introduction

Water treatment plants are facilities designed to correct the characteristics of influent water to the plant and make it suitable for its final use (e.g., drinking water, productive process water, discharged water, etc.). Different treatments are therefore needed as both influent water and final uses vary widely [1]. In general waters to be treated are grouped as follows: (i) municipal wastewaters that are characterized by a high organic content as well as nutrients, and their treatment is mainly conducted with biological processes [2–5] and other alternative methods [6]; (ii) industrial wastewaters that are mainly loaded with potential toxic elements (PTEs), and their treatment is performed with several processes, usually chemical, but also biological such as bioremediation [7], and (iii) surface waters and groundwater, which generally represent the main source of drinking water. However, these sources are in most cases loaded with suspended solids and colloids. The presence of these solids increases the water turbidity. For the removal of suspended solids and the reduction of turbidity, several physicochemical processes can be applied [8,9]. Among them, the coagulation–flocculation–sedimentation process is the most widely used [10,11].

Currently, in Algeria as well as in many other countries in the world, aluminum- or iron-based coagulants are the most used in drinking water treatment [12–14]. They are used for their effectiveness in removing suspended and colloidal solids, reducing water turbidity, removing color and, in some cases, also microorganisms. The addition of coagulants with trivalent metal ions as $Al_2(SO_4)_3$ or $FeCl_3$ in raw water, allows flocs composed of $Al(OH)_3$ or $Fe(OH)_3$ to form and precipitate at neutral or acid pH [1]. The formed flocs entrap the negatively charged colloids and promote their separation from water during the sedimentation phase [1,15].

Despite the performance and cost-effectiveness of these metal-based coagulants, they require, in most cases, pH and alkalinity adjustments to properly treat the water [11,16]. Moreover, when $Al_2(SO_4)_3$ is used as a coagulant, an excess of dosage, an inefficient coagulant use, and unexpected changes in water chemical composition can result in a high concentration of dissolved Al^{3+} in treated water [1,13]. Such residual metal concentration can be responsible for several health diseases such as Alzheimer's disease, neurotoxic disturbances, and cancer [17–19].

Moreover, the use of metal-based coagulants affects the quality of the sludge resulting from the coagulation–flocculation–sedimentation process. Such sludge is loaded with metals such as Al^{3+} and Fe^{3+} and its final disposal can require high costs and be a source of environmental concerns [20,21].

On the basis of such critical aspects, recent studies have focused on evaluating sustainable and eco-friendly natural coagulants as an alternative to metal-based coagulants for the coagulation–flocculation–sedimentation process [14].

A significant number of studies have proved that replacing metal salts with natural coagulants may reduce the environmental impact associated with the coagulation–flocculation–sedimentation process. In detail, efficient organic coagulants have been obtained from several vegetal materials such as *Moringa oleifera* [22–25], Cactus [26], Acorn [12,27], Alyssum seeds [28], Citrus fruit peel [29], Fava bean [30], *Cassia obtusifolia* seed gum [31], Dolichos lablab [32], and *Lens culinaris* [33]

The mechanism of coagulation induced by organic coagulants differs from that induced by metal-based coagulants. Coagulation is aimed at destabilizing the colloidal suspension by the following two phenomena: (1) adsorption and neutralization of charges, or (2) adsorption and bridging of colloids [34,35].

As regards the adsorption and neutralization phenomenon, ions of opposite charges are absorbed on the surface of colloids resulting in the reduction of the thickness of the electrical water double layer around colloids [35,36].

As regards the adsorption and bridging phenomenon, several natural compounds such as cellulose, proteins, starch, and polysaccharides, are characterized by their high molecular weight and, in addition, have multiple electrical charges throughout their atom chains. These compounds show a long chain length and are capable of destabilizing the negatively charged colloidal solids [36]. Several researchers have developed a theory on chemical bridges, useful to explain the empirical behavior of these compounds [35]. Such a phenomenon is due to the formation of bridges between colloidal solids thanks to coagulant molecules: the free sites of several coagulant molecules adhere to the colloidal solids, and coagulants bind colloids by means of hydrogen bonds, coulumbic attraction force, Van der Waals forces or ion exchange processes [10,36].

To evaluate the efficiency of natural coagulants for destabilizing colloidal particles, two water parameters were considered: turbidity and suspended solids concentration. The aim of this paper is to investigate the potential of *Aloe vera* as a coagulant for drinking water treatment. This natural element was selected according to its effectiveness of treatment and its worldwide availability.

2. Materials and Methods

This study was conducted according to the sequence reported in Figure 1.

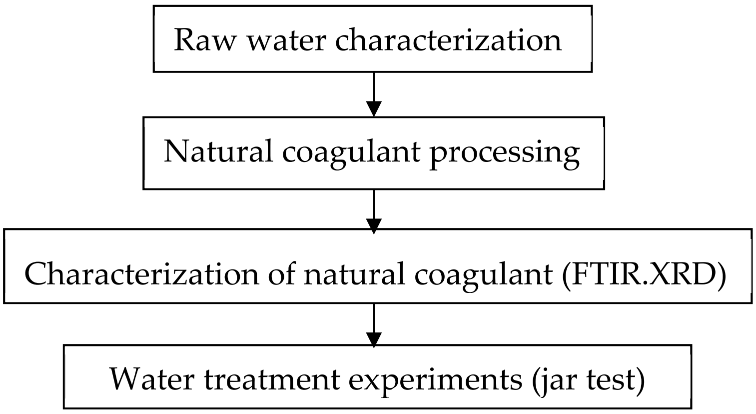

Figure 1. Flow chart of the experimental activities.

2.1. Analytical Methods

Raw water was collected from the drinking water treatment plant of Oued El Athmania (36°14′35.40″ N; 6°17′6.00″ E), Algeria. Table 1 reports the following elements: (i) some chemical parameters measured in raw water; (ii) the used measurement instrument or method, and (iii) the corresponding quality thresholds set by the Algerian water quality law [37].

Measurements of water turbidity were performed using a Turbidimeter (HANNA Code: HI 98713, Hanna instruments, Cluj-Napoca, Romania). Water pH and salinity were determined with a multi-parameter instrument (Jenway model 3540, Camlab, Cambridge, United Kingdom). The total alkalinity, total hardness, and organic matter content in water were detected according to the standard methods for water and wastewater characterization [38].

Table 1. Raw water characteristics; measurement instruments, and methods; Algerian water quality standards.

Parameters	Measurement Instrument or Method	Raw Water	Algerian Standards [37]
Turbidity (NTU)	Turbidimeter (HANNA Code: HI 98713, Hanna instruments, Cluj-Napoca, Romania)	13	5
pH	Multi-parameters instrument (Jenway model 3540, Camlab, Cambridge, United Kingdom)	7.94	6.5–9
Salinity (g/L)		0.7	/
Alkalinity (F°), Hardness (F°) and Organics content (mgO_2/L)	Standard titrimetric methods [38]	16, 34.6 and 2.1	20, 50 and 5

2.2. Preparation of the Bio-Coagulant

The *Aloe vera* was collected from fields located in the city of Mila (36°27′1.01″N; 6° 15′51.98″ E), which is located in the North East of Algeria. Prior to use as a coagulant, *Aloe vera* was treated as follows: (1) washed by tap water; (2) dried at a relatively low temperature of 50°C to prevent active component content decomposition; (3) ground; (4) sieved through a 0.35 mm pore size sieve as shown in Figure 2.

After this treatment, 25 g of *Aloe vera* powder was added to 1000 mL of distilled water and mixed for 20 min by a magnetic stirrer to extract active coagulating agents. After 30 min of settling, the supernatant was filtered through a standard filter (porosity < 8 μm), and the filtrate was used as a liquid coagulant [10,12,13].

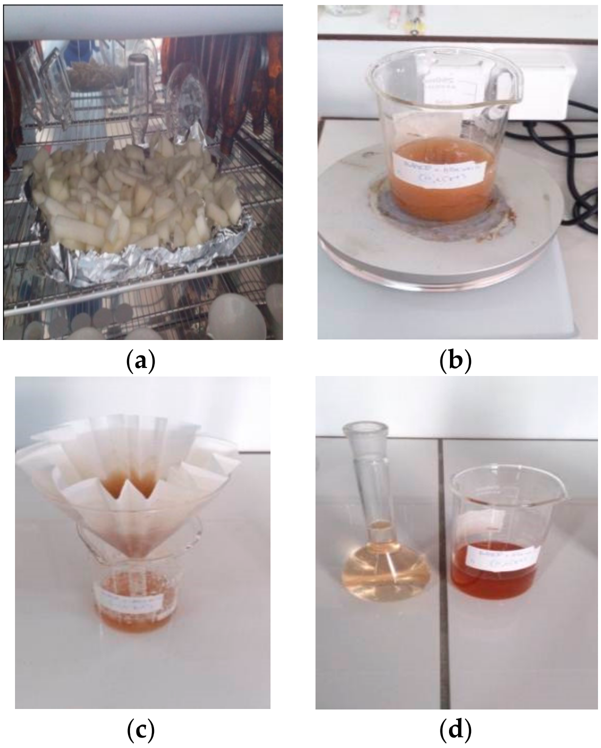

Figure 2. Preparation of the bio-coagulant: (**a**) drying at T < 50°C; (**b**) mixing for 20 min; (**c**) filtering; (**d**) extracting.

2.3. Characterization of *Aloe vera*

Aloe vera was characterized using both FTIR and XRD analysis.

2.3.1. Fourier-Transform Infrared Spectrophotometry (FTIR)

The infrared spectrum of *Aloe vera* was obtained in the wavelength range between 4000–500 cm^{-1} with a Fourier-Transform Infrared Spectrometer (Shimadzu Code: HI 98713, Shimadzu, Cluj Napoca, Romania).

Figure 3 shows the infrared spectra of *Aloe vera*. The strong stretch at 3385.1 cm^{-1} was due to the bound OH stretching vibration [39,40]. The CH group was observed at 2927.7 cm^{-1} and 2858.3 cm^{-1} [41,42].The bands at 2360.7 cm^{-1} and 2148.6 cm^{-1} could be representative of C≡N and C≡C, respectively [12].

The carbonyl function (C=O) and CH_3 primary aromatic amines were detected at 1635.5 cm^{-1} and 1423.4 cm^{-1}, respectively [31,43]. These peaks confirm the presence of NH groups in amides [44] and the carboxyl group (COOH) [31,45], which are responsible for water turbidity removal.

The presence of CN stretching (aromatic primary amine stretch) was confirmed by the peak at 1326.9 cm^{-1} [46].

The bands at 1257.6 cm^{-1} and 1018.3 cm^{-1} indicate the presence of COO stretching Carboxylic acid salt [46]and CO of ester [31], respectively.

The peaks at 830.0 cm^{-1}, 793 cm^{-1}, 607 cm^{-1}, and 535 cm^{-1} indicate the presence of aromatic CH out-of-plane deformation [46].

During the coagulation–flocculation–sedimentation process conducted with *Aloe vera,* its functional groups (the carboxyl group) act as adsorption sites for the suspended and colloidal solids [45]. In addition, the amide groups in *Aloe vera* form intermolecular bonds between the suspended solids and the coagulant, thus increasing the efficiency of the coagulation process [44].

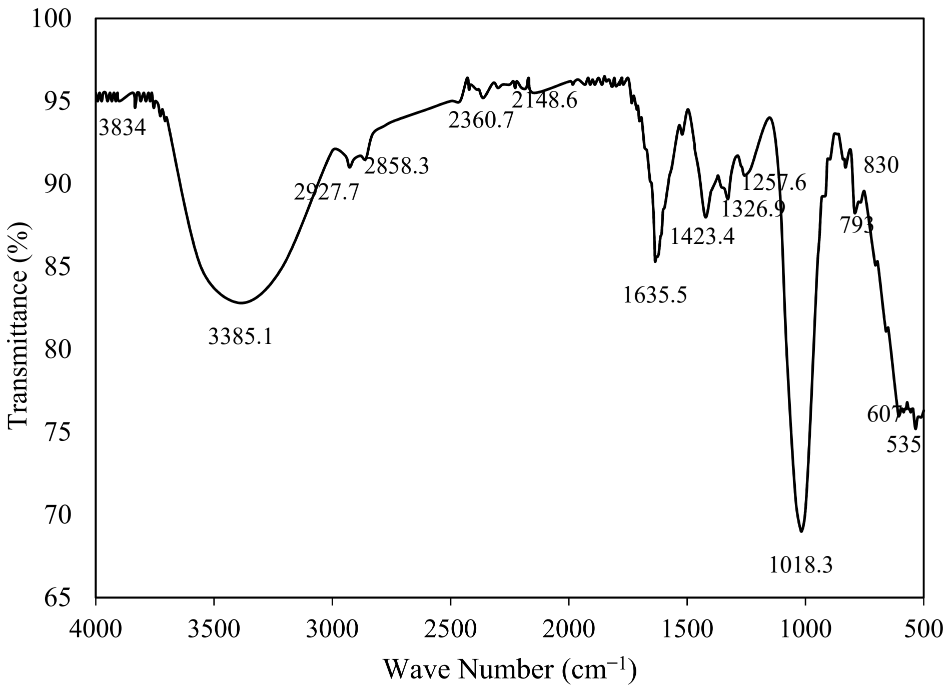

Figure 3. Infrared spectra of *Aloe vera*.

2.3.2. XRD Analysis

The crystalline structure of the powdered coagulant was evaluated by an X-ray diffractometer using Cu with wavelength K-Alpha (1.54) at 40 kV and 30 mA with a scan analysis. The spectra of *Aloe vera* were obtained in the range of 10–90 degrees.

X-ray diffraction analysis (XRD) is used for the determination of crystalline phases in natural material. In general, these materials can be classified in three phases: amorphous, crystalline or semi-crystalline [47]

Figure 4 shows the XRD pattern of *Aloe vera* powder. The percentage of crystallinity was 38%.

The peaks between 15 and 40 degrees could be representative of various substances such as lipids, proteins, carbohydrates, and ash [48,49].

The specific nature of this coagulant allows contaminants to enter the coagulant surface [50]. According to the obtained results, it was noticed that the different coagulation agents present in *Aloe vera* facilitate the removal of suspended solids and negatively charged colloidal particles, thus promoting the turbidity reduction and, as consequence, water quality improvement [47,51].

Similar results were obtained by Hirendrasinh Padhiyar et al. (2020) by using *Moringa Oleifera* as a natural coagulant [51].

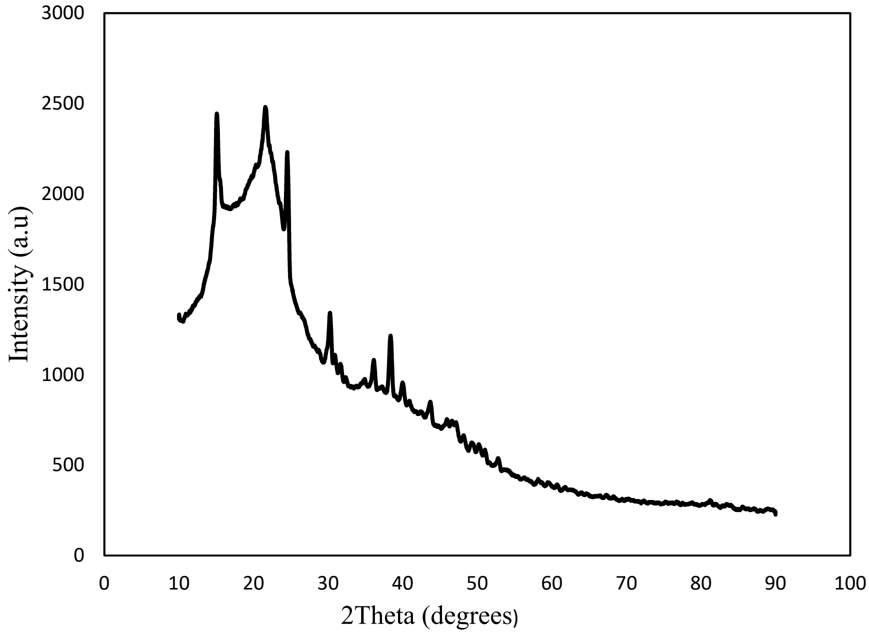

Figure 4. X–ray diffractogram for *Aloe vera*.

2.4. Experimental Procedure

The coagulation–flocculation–sedimentation process was carried out using a Jar test equipment (LI-JTA-125, LABARD, labard instruments, Bengale, India) to evaluate the performance of the natural coagulant: powder (AV-Powder) and liquid extracted with distilled water (AV-H_2O) *Aloe vera* were both tested at several pH values and coagulant dosages since pH is one of the main parameters to be optimized in a Jar test experiment [22].

A Jar test was conducted according to the following sequence of operations: (i) six 500 mL beakers were filled with raw water; (ii) a defined coagulant dose was added; (iii) beakers were mixed rapidly at a mixing speed of 160 rpm for 3 min (coagulation); (iv) beakers were slowly mixed at mixing speed of 30 rpm for 20 min (flocculation); (v) the formed flocs were allowed to settle for 30 min (sedimentation). At the end of the coagulation–flocculation–sedimentation process, the supernatant from each beaker was characterized in terms of turbidity, pH, total alkalinity, total hardness, salinity, and organic matter content. Turbidity removal efficiency (TRE) was calculated using Equation (1):

$$\text{TRE (\%)} = [(\text{Initial Turbidity} - \text{Residual Turbidity})/(\text{Initial Turbidity})] \times 100 \quad (1)$$

3. Results and Discussion

In the present experimental study, the effect of bio-coagulant concentration on turbidity removal efficiency, water pH, total alkalinity, total hardness, salinity, and organic matter content was investigated. In addition, the effect of the pH adjustment was taken into account in further experiments as discussed in the following sections.

3.1. Effect of Bio-Coagulant Dosage on Water Turbidity Removal

Figure 5 shows the effect of the coagulant dosage on the water turbidity removal efficiency using two coagulants: AV-Powder and AV-H_2O. Different coagulant dosages were tested in the ranges 0.1–50 mg/L and 0.1–2 mL/L for AV-Powder and AV-H_2O, respectively.

The obtained results show that the AV-Powder was responsible for a water turbidity reduction of 28.23% as shown in Figure 5a. This can be explained by the low solubility of

AV-Powder in water. Conversely, the water turbidity reduction was more pronounced when AV-H_2O was added to water, achieving a reduction rate of 87.84%.

Several papers from the international literature have confirmed the positive effect of bio-coagulants on water turbidity reduction when they are added in powder or liquid form; the latter was extracted by distilled water. For instance, the study published by Benalia et al. (2019), proves a turbidity reduction in drinking water by 71.6% and 84.77% from an initial value of 13 NTU when acorn was used as a coagulant in powder and liquid form, respectively [12].

A further study, conducted by Gandiwa et al. (2020), showed that water turbidity reduction was enhanced when liquids extracted from *Moringa Oleifera* and Cactus Opuntia with distilled water were used as bio-coagulants. The authors demonstrated that water turbidity was reduced to 3.2 and 3 NTU from the initial value of 29 NTU [52].

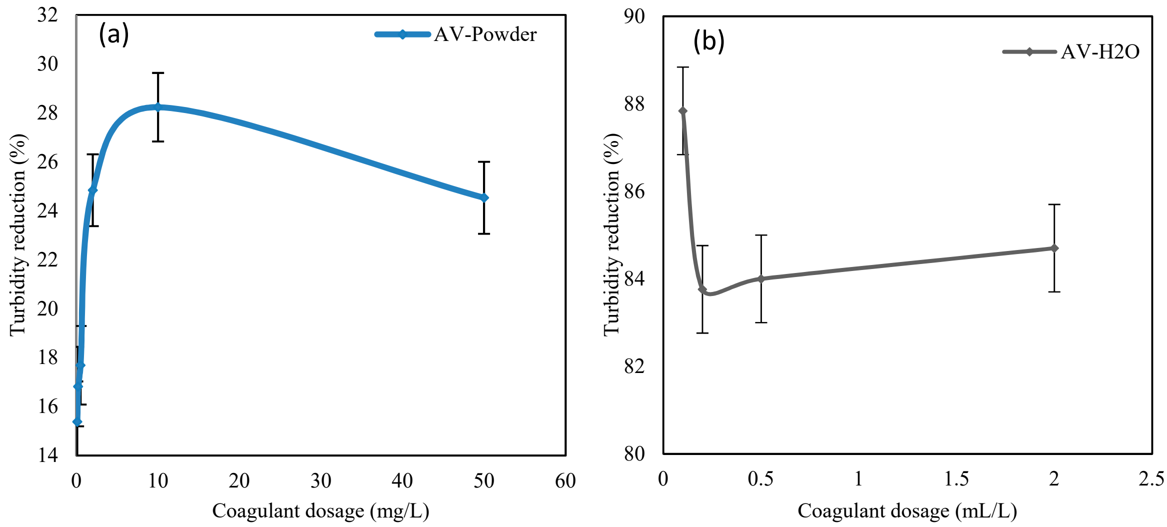

Figure 5. Effect of bio-coagulant dosage on water turbidity reduction by using: (a) AV Powder; (b) AV-H_2O.

3.2. Effect of Bio-Coagulant Dosage on Water pH, Total Alkalinity, total Hardness, and Salinity

Figure 6 shows the effect of the coagulant dosage on water pH and alkalinity. There was a slight variation in pH and total alkalinity values when the two coagulants, i.e., AV-Powder and AV-H_2O, were added to water. This result can be related to the organic nature of the bio-coagulant [12]. As mentioned in the literature, the activity of natural coagulants is due to the long chains of proteins contained in their molecules, which are responsible for pollutant aggregation and floc formation [48]. On the contrary, metal-based coagulants hydrolyze in water and release H^+ ions, resulting in a pH decrease. For such reason, coagulation using chemical-based coagulants requires pH adjustment after coagulation, thus increasing the costs of chemical reagents used in water treatment [53].

Similar results were obtained by different researchers such as Narasiah et al. (1998); Arnoldsson et al. (2008); Chu et al. (2017) and Villareal et al. (2018), when *Moringa oleifera* was used as a bio-coagulant [20,54–56].

Figure 7a,b show the effect of coagulant dosage on total hardness variations in treated water when AV-Powder and AV-H_2O were used. The sum of the magnesium (Mg^{2+}) and calcium (Ca^{2+}) ion equivalent concentration represents the total hardness. Such a value was almost independent of the coagulant dosage. This result is due to absence of any interaction between ions (Ca^{2+} and Mg^{2+}) responsible for the total hardness and the bio-coagulant used.

Concerning the water salinity content, its value depends on the concentration of the main dissolved ions, calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), chlorides (Cl^-), sulfates (SO_4^{2-}), and bicarbonates (HCO_3^-). According to Figure 7c,d, the water salinity content generally remained stable when the bio-coagulant dosage was changed.

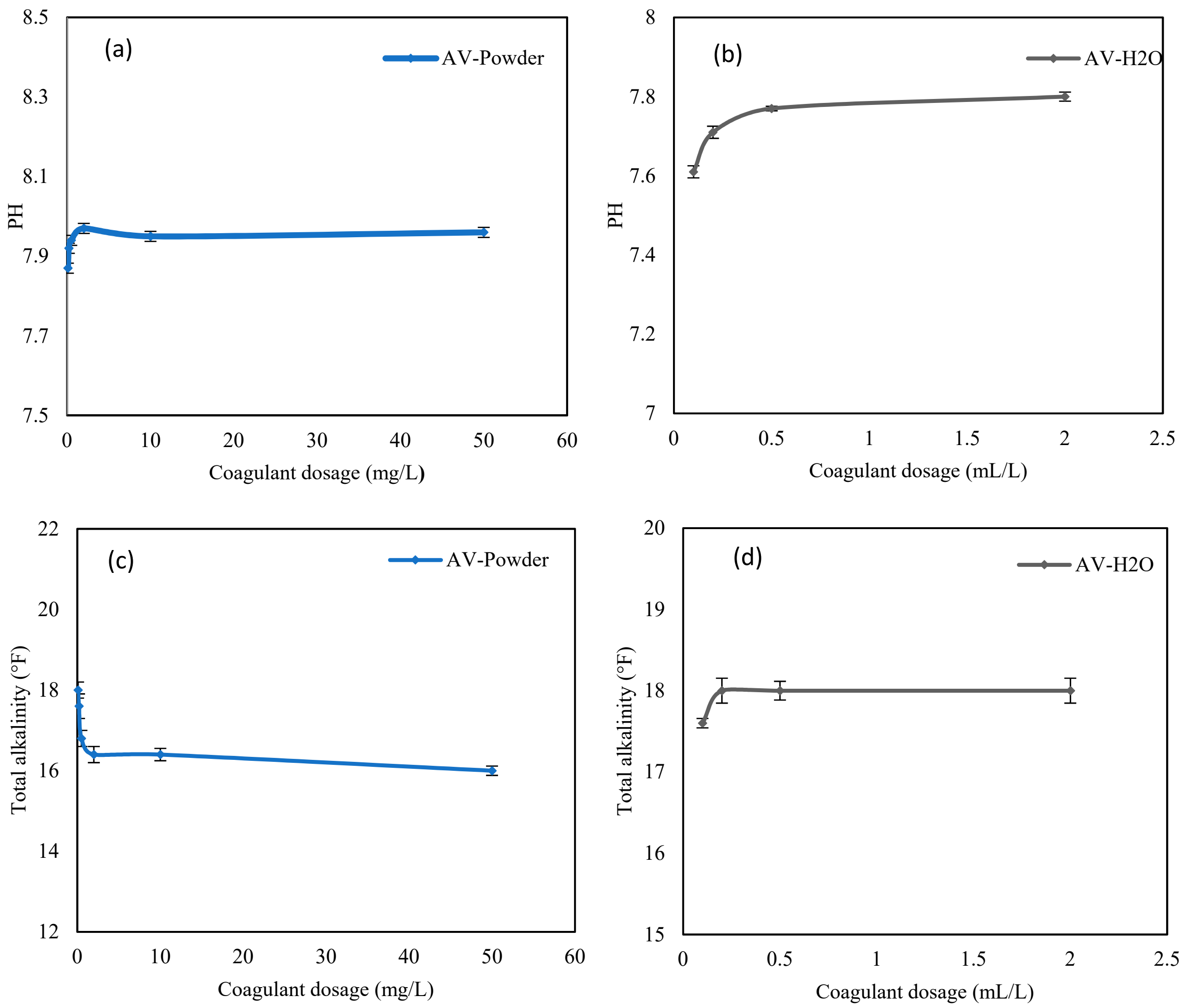

Figure 6. Effect of bio-coagulant dosage on: (**a**,**b**) pH; (**c**,**d**) total alkalinity by using AV-Powder and AV–H_2O. (1°F = 10 mg $CaCO_3$/L).

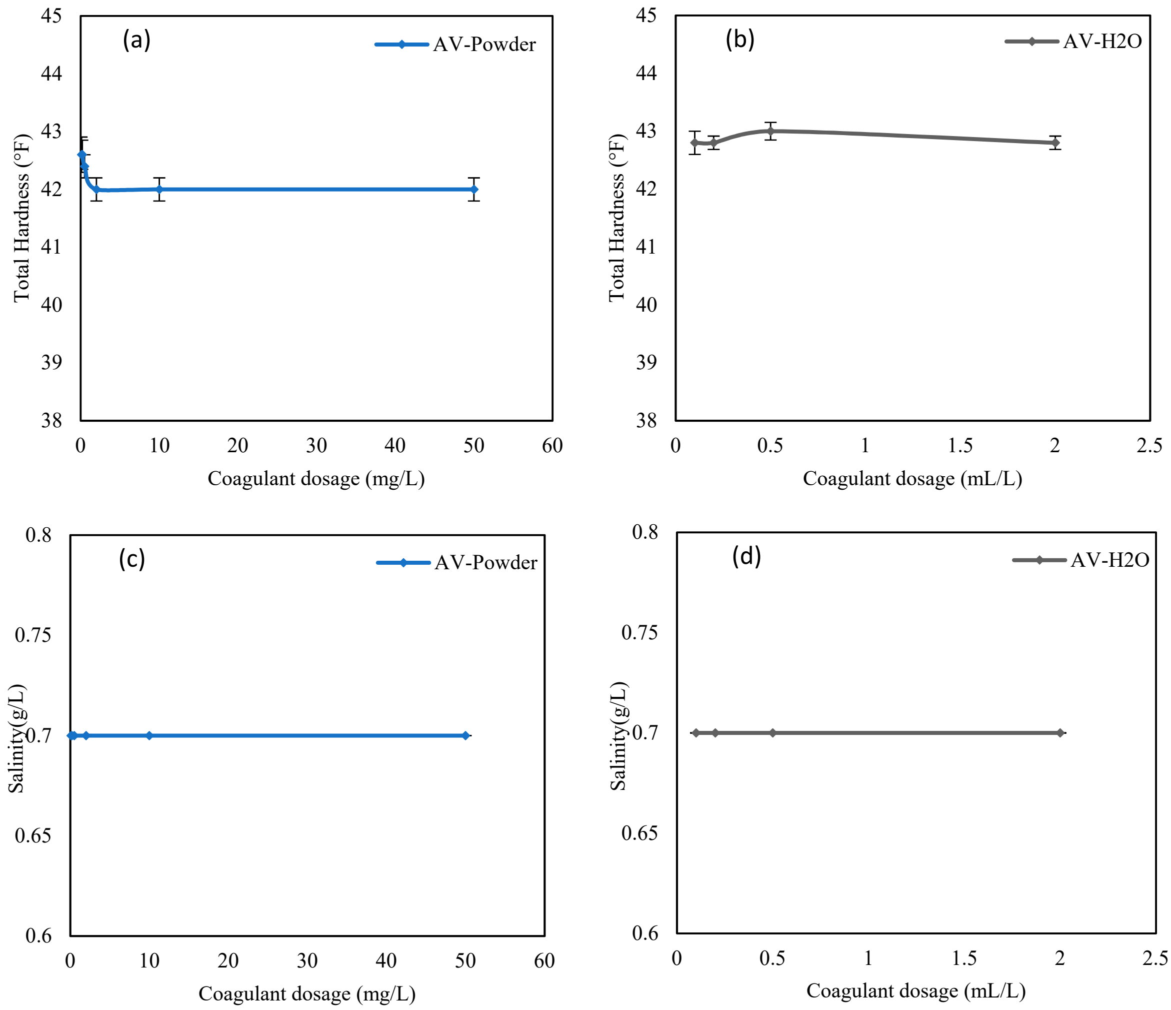

Figure 7. Effect of bio-coagulant dosage on: (**a**,**b**) total hardness; (**c**,**d**) salinity by using AV-Powder and AV-H_2O.

3.3. Effect of Bio-Coagulant Dosage on Organic Matter Concentration

Figure 8 shows the effect of bio-coagulant dosage on organic matter concentration, and it can be noticed that the concentration of organic matter increased in treated water. This can be due to the natural organic origin of the bio-coagulant that partially dissolved in the aqueous solution during raw water treatment. Actually, the organic matter content after coagulation was higher than the initial value.

Similar trends in organic matter variations were found by other researchers, such as Nacoulma et al. (2000) and Jacques et al. (2009), when *Moringa oleifera* was used as an organic coagulant [57,58].

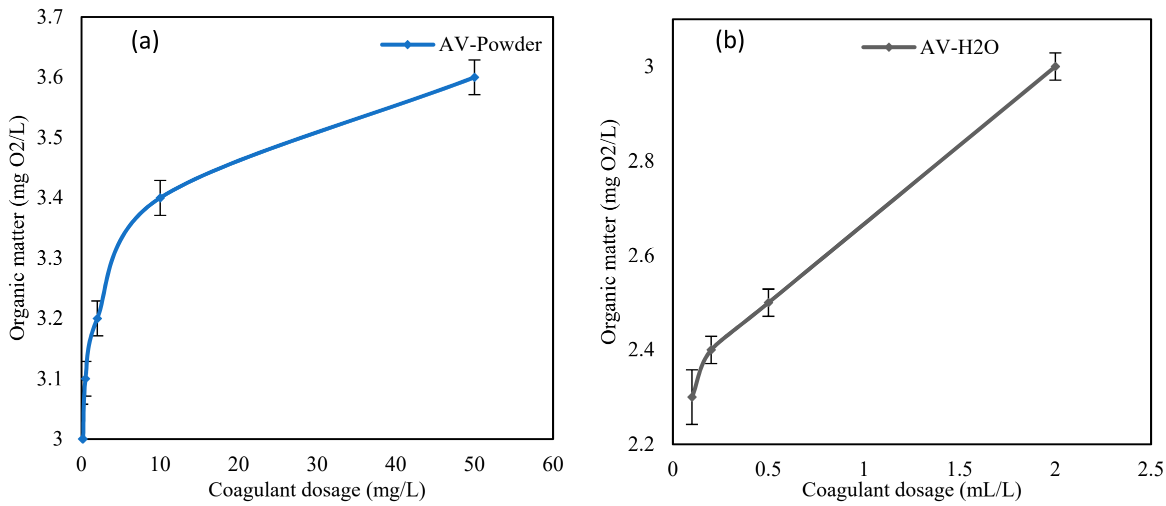

Figure 8. Effect of bio-coagulant dosage on organic matter by using (**a**) AV Powder; (**b**) AV H_2O.

3.4. Effect of pH on the Water Turbidity Reduction

The coagulation–flocculation–sedimentation process performance is affected by many parameters, such as coagulant type and dosage, initial water turbidity, pH, temperature, water chemical composition, etc. Among them, pH plays a crucial role as coagulants often are associated with electric charges [59].

Therefore, the coagulation process performance was investigated at different pH values (from 5.5 to 8) and at an initial turbidity of 13 NTU, with the optimal dosages (10 mg/L of AV-Powder and 0.1 mL/L of AV-H_2O). pH was adjusted by adding NaOH (1.0 M) or HCl (1.0 M). Results from this study are shown in Figure 9: the most promising process performance in water turbidity reduction was obtained at a pH value of 6 by using AV-H_2O and AV-Powder. According to the obtained results, at the optimal pH value, the water turbidity was reduced by 53.53% and 88.23% with AV-Powder and AV-H_2O, respectively.

Similar results were obtained by Marina et al. (2015), Tie et al. (2015), and Saritha et al. (2019), thus confirming the effect of pH on turbidity reduction efficiency by using organic coagulants [30,60,61].

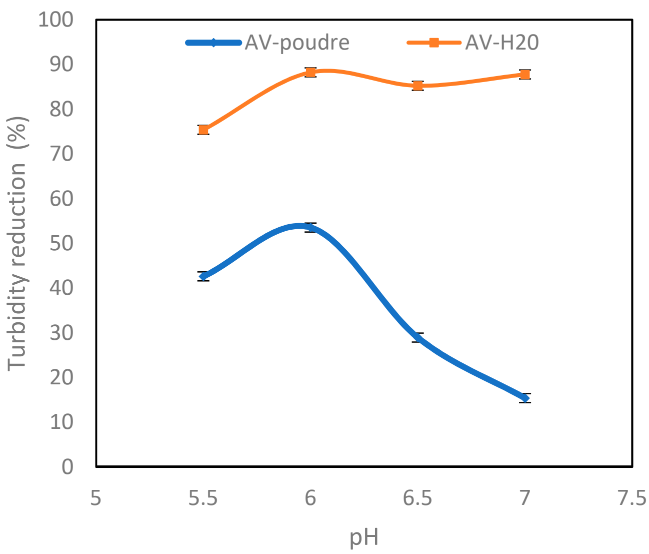

Figure 9. Effect of pH on water turbidity reduction by using AV-Powder and AV-H_2O.

Table 2 shows the effectiveness of some organic coagulants, in powder as well as liquid form, to remove turbidity from water. Such values are compared to those obtained from the present study.

Table 2. Turbidity removal efficiency of different organic coagulants.

Coagulant	Optimal Dosage	Initial Turbidity (NTU)	Removal Turbidity (%)	Year	References
Aloe vera Powder	10 mg/L	13	28.23	-	Current study
Aloe vera Liquid	0.1 mL/L	13	87.47	-	Current study
Pine cones	0.1 mL/L	13	84.77	2019	[12]
	0.5 mL/L	67	55	2019	[59]
	0.5 mL/L	75	62	2019	[59]
Cactus	1500 mg/L	500	97	2018	[62]
	33 mg/L	214	96	2017	[63]
	10 mg/L	160	84.43	2015	[13]
	6 mg/L	9.5	49.78	2015	[13]
	1 mL/L	9.5	53.05	2015	[13]
	55 mg/L	29	89.4	2020	[52]
Dolichos lablab	100 mg/L	100	88.9	2011	[64]
	100 mg/L	35	60.85	2011	[64]
	0.6 mL/L	62	98.84	2018	[32]
Fava bean	0.25 mL/L	20	38	2015	[30]
	0.25 mL/L	45	54	2015	[30]
Jatropha curcas seeds	120 mg/L	500	99	2013	[65]
Moringa Oleifera	100 mg/L	100	94.1	2011	[64]
	100 mg/L	25	60	2011	[64]
	50 mg/L	146	83.7	2010	[66]
	50 mg/L	131	84.9	2010	[66]
	50 mg/L	29	88.7	2020	[52]
Banana pith	0.1 kg/m^3	279	98.56	2016	[67]
	0.6 mL/L	62	98.14	2018	[32]
pearl millet (*Pennisetum glaucum*)	80 mg/L	200	99.2	2019	[68]
black-eyed pea (*Vigna unguiculata*)	20 mg/L	200	97.6	2019	[68]
Citrus Microcarpa	30 mg/L	29.8	75.6	2019	[29]
Citrus Aurantiifolia	60 mg/L	20.1	74	2019	[29]
Cicer arietinum	100 mg/L	95	95.89	2011	[64]
	100 mg/L	31	71.29	2011	[64]
Peanut seeds	20 mg/L	200	31.5	2013	[69]

4. Conclusions

All the experiments conducted in this study were performed on natural raw water with an initial turbidity of 13 NTU obtained from a drinking water treatment plant. In this work, two forms of bio-coagulants obtained from *Aloe vera* were used. The obtained results show that the turbidity was reduced to 6.0 NTU at pH 6 and to 1.42 NTU at pH 7.5 when AV-Powder and AV-H_2O were used, respectively. It can be highlighted that the residual turbidity from AV-H_2O addition was less than 5 NTU and fulfills the quality standards of the Algerian drinking water law [37]. Moreover, the use of natural coagulants in water treatment had no significant effect on several parameters such as pH, total hardness, total alkalinity, and salinity, whereas the concentrations of organic matter increased.

This experimental study has proved that *Aloe vera* can be successfully used as a natural coagulant for drinking water treatment. Both AV-Powder and AV-H_2O can improve the final drinking water quality. Therefore metal-based coagulants, such as Aluminum sulfate, Ferrous sulfate, Ferric Chloride, etc., can be replaced with natural coagulants without reducing the process performance.

This study opens new perspectives of research on the development of natural coagulants to treat surface water, groundwater, wastewater, and industrial water, as well as on extraction processes to obtain liquid coagulants from *Aloe vera*, and finally on the solvent recovery procedure after extracting process. However, further investigations in pilot and full-scale plants are necessary in addition to a cost–benefit analysis to comprehensively understand the potential of bio-coagulants in water treatment.

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Abbreviations

$Al(OH)_3$	Aluminum hydroxide
$Al_2(SO_4)_3$	Aluminium sulphate
Al^{3+}	Aluminum Ion
AV	*Aloe vera*
COD	Chemical Oxygen Demand
$Fe(OH)_3$	Ferric hydroxide
Fe^{3+}	Ferric ion
$FeCl_3$	Ferric chloride
FTIR	Fourier-Transform Infrared Spectrophotometry
HCl	Hydrochloric acid
NaOH	Sodium hydroxide
NTU	Nephelometric Turbidity Unit
PTEs	Potential Toxic Elements
Rpm	Revolutions per minute

References

1. Desjardins, R. *Traitement Des Eaux*, 2nd ed.; Presses Polytechnique de de Montréal: Montréal, QC, Canada, 1997.
2. Zekker, I.; Raudkivi, M.; Artemchuk, O.; Rikmann, E.; Priks, H.; Jaagura, M.; Tenno, T. Mainstream-sidestream wastewater switching promotes anammox nitrogen removal rate in organic-rich, low-temperature streams. *Environ. Technol.* **2020**, 1–23, doi:10.1080/09593330.2020.1721566.
3. Mandel, A.; Zekker, I.; Jaagura, M.; Tenno, T. Enhancement of anoxic phosphorus uptake of denitrifying phosphorus removal process by biomass adaption. *Int. J. Environ. Sci. Technol.* **2019**, *16*, 5965–5978, doi:10.1007/s13762-018-02194-2.
4. Bouchareb, E.M.; Kerroum, D.; Bezirhan Arikan, E.; Isik, Z.; Dizge, N. Production of bio-hydrogen from bulgur processing industry wastewater. *Energy Sources Part A Recovery Util. Environ. Eff.* **2021**, 1–14, doi:10.1080/15567036.2021.1877853.
5. Rokaya, B.; Kerroum, D.; Hayat, Z.; Panico, A.; Ouafa, A.; Pirozzi, F. Biogas production by an anaerobic digestion process from orange peel waste and its improvement by limonene leaching: Investigation of H2O2 pre-treatment effect. *Energy Sources Part A Recovery Util. Environ. Eff.* **2019**, 1–9, doi:10.1080/15567036.2019.1692975.

6. Kamyab, H.; Ravi, A.V.; Chelliapan, S.; Tin, C. Fabrication of nanocomposites mediated from aluminium nanoparticles/Moringa oleifera gum activated carbon for effective photocatalytic removal of nitrate and phosphate in aqueous solution. *J. Clean. Prod.* **2020**, 124553, doi:10.1016/j.jclepro.2020.124553.
7. Suresh, G.; Balasubramanian, B.; Ravichandran, N.; Ramesh, B.; Kamyab, H.; Velmurugan, P.; Siva, G.V.; Ravi, A.V. Bioremediation of hexavalent chromium-contaminated wastewater by Bacillus thuringiensis and Staphylococcus capitis isolated from tannery sediment. *Biomass Convers. Biorefin.* **2021**, *11*, 383–391, doi:10.1007/s13399-020-01259-y.
8. Alam, S.; Khan, M.S.; Bibi, W.; Zekker, I.; Burlakovs, J.; Ghangrekar, M.M.; Bhowmick, G.D.; Kallistova, A.; Pimenov, N.; Zahoor, M. Preparation of activated carbon from the wood of paulownia tomentosa as an efficient adsorbent for the removal of acid red 4 and methylene blue present in wastewater. *Water* **2021**, *13*, 1453, doi:10.3390/w13111453.
9. Valiron, F. *Gestion Des Eaux; Alimentation En Eau—Assainissement*; ENPC Ed.; Paris, France, 1989; ISBN 2859781315.
10. HDR Engineering Inc. *Handbook of Public Water Systems*, 2nd ed.; John Wiley & Sons, Inc: New York, NY, USA, 2001.
11. Degrémont, S.A. *Mémento Technique De L'eau*, 10th ed.; Degrémont Ed.; Paris, France, 2005; ISBN 9782743007171.
12. Benalia, A.; Derbal, K.; Panico, A.; Pirozzi, F. Use of Acorn Leaves as a Natural Coagulant in a Drinking Water Treatment Plant. *Water Res.* **2019**, *11*, 57, doi:10.3390/w11010057.
13. Benalia, A.; Derbal, K. *Etude Expérimentale et Modélisation du Processus de la Coagulation Floculation: Application Aux Eaux Destinée a la Consommation*; Université Constantine 3: El Khroub, Algeria, 2015.
14. Matilainen, A.; Lindqvist, N.; Tuhkanen, T. Comparison of the Effiency of Aluminium and Ferric Sulphate in the Removal of Natural Organic Matter During Drinking Water Treatment Process. *Environ. Technol.* **2005**, *26*, 867–875, doi:10.1080/09593332608618502.
15. Desjardins, C. *Simulation et Étude en Laboratoire de la Floculation Lestée (Actiflo) a L'aide D'une Procédure Modifiée de Jar Test*; Université de Montréal: Montréal, QC, Canada, 1999.
16. Benalia, A.; Derbal, K. Comparative Study between Aluminum Sulfate and Ferric Chloride in Water Treatment: Turbidity Removal. 2015.Available online: file:///C:/Users/MDPI/AppData/Local/Temp/numero-01-JDOC-vf-4-9.pdf(accessed on 22 July 2021)
17. Exley, C. La toxicité de l'aluminium chez l'homme. *Morphologie* **2016**, *100*, 51–55, doi:10.1016/j.morpho.2015.12.003.
18. Rondeau, V.; Commenges, D.; Jacqmin-Gadda, H.; Dartigues, J. Relation between Aluminum Concentrations in Drinking Water and Alzheimer's Disease: An 8-year Follow-up Study. *Am. J. Epidemiol.* **2000**, *152*, 59–66, doi:10.1093/aje/152.1.59.
19. Zengjin, W.; Xiaomin, W.; Junlin, Y.; Jinning, S.; Jingyi, C.; Xianchen, L.; Zhao, X. Chronic exposure to aluminum and risk of Alzheimer's disease: A metal-analysis. *Neurosci. Lett.* **2016**, *610*, 200–206, doi:10.1016/j.neulet.2015.11.014.
20. Narasiah, K.S. Quality Of Water Treated By Coagulation Using Moringa Oleifera Seeds. *Water Res.* **1998**, *32*, 781–791, doi:10.1016/S0043-1354(97)00295-9.
21. Megersa, M.; Beyene, A.; Ambelu, A.; Triest, L. Comparison of purified and crude extracted coagulants from plant species for turbidity removal. *Int. J. Environ. Sci. Technol.* **2019**, *16*, 2333–2342, doi:10.1007/s13762-018-1844-2.
22. Bouchareb, R.; Derbal, K.; Özay, Y.; Bilici, Z.; Dizge, N. Combined natural/chemical coagulation and membrane filtration for wood processing wastewater treatment. *J. Water Process Eng.* **2020**, *37*, 101521, doi:10.1016/j.jwpe.2020.101521.
23. Pandey, P.; Khan, F.; Mishra, R.; Singh, S.K. Elucidation of the potential of Moringa oleifera leaves extract as a novel alternate to the chemical coagulant in water treatment process. *Water Environ. Res.* **2020**, *92*, 1051–1056, doi:10.1002/wer.1300.
24. Singh, G.; Patidar, S.K. Water quality restoration by harvesting mixed culture microalgae using Moringa oleifera. *Water Environ. Res.* **2020**, *92*, 1268–1282, doi:10.1002/wer.1322.
25. Adelodun, B.; Ogunshina, M.S.; Ajibade, F.O.; Abdulkadir, T.S.; Bakare, H.O.; Choi, K.S. Kinetic and prediction modeling studies of organic pollutants removal from municipal wastewater using Moringa oleifera biomass as a coagulant. *Water* **2020**, *12*, 2052, doi:10.3390/w12072052.
26. Maran, J.P.; Manikandan, S.; Mekala, V. Modeling and optimization of betalain extraction from Opuntia ficus-indica using Box—Behnken design with desirability function. *Ind. Crops Prod.* **2013**, *49*, 304–311, doi:10.1016/j.indcrop.2013.05.012.
27. Antov, M.G.; Šć, M.B.; Prodanovi, J.M.; Kuki, D.V.; Vasi, V.M.; Tatjana, R.Đ.; Milo, M.M. Industrial Crops & Products Common oak (Quercus robur) acorn as a source of natural coagulants for water turbidity removal. *Ind. Crops Prod.* **2018**, *117*, 340–346, doi:10.1016/j.indcrop.2018.03.022.
28. Fard, M.B.; Hamidi, D.; Yetilmezsoy, K.; Alavi, J.; Hosseinpour, F. Utilization of Alyssum mucilage as a natural coagulant in oily-saline wastewater treatment. *J. Water Process Eng.* **2021**, *40*, 101763, doi:10.1016/j.jwpe.2020.101763.
29. Dollah, Z.; Abdullah, A.R.C.; Hashim, N.M.; Albar, A.; Badrealam, S.; Zaki, M. Citrus fruit peel waste as a source of natural coagulant for water turbidity removal. *J. Phys. Conf. Ser.* **2019**, *1349*, 012011, doi:10.1088/1742-6596/1349/1/012011.
30. Marina, B.S.; Prodanovi, J.M.; Dragana, V.K.; Aleksandra, N.T.; Mirjana, A.V. Extracts of fava bean (Vicia faba L.) seeds as natural coagulants. *Ecol. Eng.* **2015**, *84*, 229–232, doi:10.1016/j.ecoleng.2015.09.008.
31. Subramonian, W.; Wu, T.Y.; Chai, S. A comprehensive study on coagulant performance and floc characterization of natural Cassia obtusifolia seed gum in treatment of raw pulp and paper mill effluent. *Ind. Crops Prod.* **2014**, *61*, 317–324, doi:10.1016/j.indcrop.2014.06.055.
32. Daverey, A.; Tiwari, N.; Dutta, K. Utilization of extracts of Musa paradisica (banana) peels and Dolichos lablab (Indian bean) seeds as low-cost natural coagulants for turbidity removal from water. *Environ. Sci. Pollut. Res.* **2019**, *26*, 34177–34183, doi:10.1007/s11356-018-3850-9.

33. Chua, S.C.; Malek, M.A.; Chong, F.K.; Sujarwo, W.; Ho, Y.C. Red lentil (Lens culinaris) extract as a novel natural coagulant for turbidity reduction: An evaluation, characterization and performance optimization study. *Water* **2019**, *11*, 1686, doi:10.3390/w11081686.
34. Ang, W.L.; Mohammad, A.W. State of the art and sustainability of natural coagulants in water and wastewater treatment. *J. Clean. Prod.* **2020**, *20*, 121267, doi:10.1016/j.jclepro.2020.121267.
35. Saleem, M.; Bachmann, R.T. A Contemporary Review on Plant-Based Coagulants for Applications in Water Treatment. *J. Ind. Eng. Chem.* **2018**, *72*, 281–297, doi:10.1016/j.jiec.2018.12.029.
36. Bratby, J. *Coagulation and Flocculation with an Emphasis on Water and Wastewater Treatment*; Uplands Press Ltd.: Craydon, U.K., 1980.
37. République Algérienne Démocratique et Populaire .Conventiont et accords internationaux-Lois et decrets arretes, decisions, avis, communications et annonces. *J. Off. Repub. Alger.* **2006**, *26, 1-27*.
38. Rice, E.W.; Baird, R.B.; Eaton, A.D. (Eds.) *Standard Methods for the Examination of Water and Wastewater*, 22nd ed.; American Public Health Association (APHA), Ed.; American Water Works Association (AWWA) and Water Environment Federation (WEF): Washington, DC, USA, 2012; ISBN 9780875530130.
39. Freitas, T.; Oliveira, V.M.; de Souza, M.T.F.; Geraldino, H.C.L.; Almeida, V.C.; Fávaro, S.L.; Garcia, J.C. Optimization of coagulation-flocculation process for treatment of industrial textile wastewater using okra (A. esculentus) mucilage as natural coagulant. *Ind. Crops Prod.* **2015**, *76*, 538–544, doi:10.1016/j.indcrop.2015.06.027.
40. Luz, D.; Pablo, J.; Baracaldo, D.; Andr, J.; Muñoz, E. Using Co ff ee Pulp as Bioadsorbent for the Removal of Manganese (Mn (II)) from Synthetic Wastewater. *Water* **2020**, *12*, 2500, doi:10.3390/w12092500.
41. Bougeard, D. *Infrared and Raman Spectroscopy: Methods and Applicaitons*; Schrader, B., Ed.; VCH Verlagsgesellschaft mbH, Weinheini (Federal Republic of Germany), 1995; ISBN 3527264469.
42. Brian, A.; Choong, L.; Peter, A.P. Treatment of palm oil mill effluent (POME) using chickpea (Cicer arietinum) as a natural coagulant and flocculant: Evaluation, process optimization and characterization of chickpea powder. *J. Environ. Chem. Eng.* **2018**, *6*, 6243–6255, doi:10.1016/j.jece.2018.09.038.
43. Abonele, M.; Chidi, A.; Ifedi, O.; Okoye, P. Equilibrium Dynamics of m-Xylene Removal from Aqueous Solution by Organoclay. *Iran. J. Sci. Technol. Trans. A Sci.* **2019**, *43*, 119–125, doi:10.1007/s40995-017-0422-7.
44. 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, doi:10.1016/j.seppur.2013.05.015.
45. Yin, C. Emerging usage of plant-based coagulants for water and wastewater treatment. *Process Biochem.* **2010**, *45*, 1437–1444, doi:10.1016/j.procbio.2010.05.030.
46. Nharingo, T.; Moyo, M. Application of *Opuntia ficus-indica* in bioremediation of wastewaters. A critical review. *J. Environ. Manag.* **2016**, *166*, 55–72, doi:10.1016/j.jenvman.2015.10.005.
47. Araújo, C.S.T.; Carvalho, D.C.; Rezende, H.C.; Almeida, I.L.S.; Coelho, L.M.; Coelho, N.M.M.; Marques, T.L.; Alves, V.N. Bioremediation of Waters Contaminated with Heavy Metals Using Moringa oleifera Seeds as Biosorbent. *Appl. Bioremediat. Act. Passiv. Approaches* **2013**, *23*, 227–255.
48. Bouchareb, R.; Derbal, K.; Benalia, A. Optimization of active coagulant agent extraction method from Moringa Oleifera seeds for municipal wastewater treatment. *Water Sci. Technol.* **2021**, doi:10.2166/wst.2021.234.
49. Kebede, T.G.; Dube, S.; Mhuka, V.; Nindi, M.M. Toxic/Hazardous Substances and Environmental Engineering Bioremediation of Cd (II), Pb (II) and Cu (II) from industrial effluents by Moringa stenopetala seed husk. *J. Environ. Sci. Health Part A* **2019**, *54*, 337–351, doi:10.1080/10934529.2018.1551648.
50. Nath, A.; Mishra, A.; Prakash, P. Materials Today: Proceedings A review natural polymeric coagulants in wastewater treatment. *Mater. Today Proc.* **2020**, doi:10.1016/j.matpr.2020.03.551.
51. Padhiyar, H.; Thanki, A.; Kumar, N.; Pandey, S.; Yadav, M. Parametric and kinetic investigations on segregated and mixed textile e ffl uent streams using Moringa oleifera seed powders of different sizes. *J. Water Process Eng.* **2020**, *34*, 101159, doi:10.1016/j.jwpe.2020.101159.
52. Gandiwa, B.I.; Moyo, L.B.; Ncube, S.; Mamvura, T.A.; Hlabangana, N. Optimisation of using a blend of plant based natural and synthetic coagulants for water treatment: (Moringa Oleifera-Cactus Opuntia-Alum Blend). *S. Afr. J. Chem. Eng.* **2020**, *34*, 158–164, doi:10.1016/j.sajce.2020.07.005.
53. Camacho, F.P.; Sousa, V.S.; Bergamasco, R.; Teixeira, M.R. The use of Moringa oleifera as a natural coagulant in surface water treatment. *Chem. Eng. J.* **2017**, *313*, 226–237, doi:10.1016/j.cej.2016.12.031.
54. Arnoldsson, E.; Bergman, M.; Matsinhe, N.; Persson, K.M. Assessment of drinking water treatment using Moringa Oleifera natural coagulant. *Vatten* **2008**, *64*, 137–150.
55. Chu, T.; Manaf, S.; Matar, A.; Makky, E.A.; Ali, E.N. The use of Moringa oleifera seed as a natural coagulant for wastewater treatment and heavy metals removal. *Appl. Water Sci.* **2017**, *7*, 1369–1376, doi:10.1007/s13201-016-0499-8.
56. Villarreal, J.S.; Pico, M.M.; Esteban, N. Experimental evaluation of crushed Moringa oleifera Lam. seeds and powder waste during coagulation-flocculation processes. *J. Environ. Chem. Eng.* **2018**, *6*, 5443–5451, doi:10.1016/j.jece.2018.08.021.
57. Nacoulma.G.; Piro, J.; Ali, B. Etude de l'activité floculante d'un complexe protéine-mucilage végétal dans la clarification des eaux brutes. *J. Société Ouest Afr. Chim.* **2000**, *9*, 43–57.
58. Jacques, F.K.; Josse, R.G.; Mama, D.; Aminou, T. Étude de l'activité floculante de la caséine acide extraite de la crème de Cocos nucifera dans la clarification des eaux de surface. *Rev. Sci. L'eau* **2009**, *22*, 93–101, doi:10.7202/019826ar.

59. Hussain, S.; Sattar, A.; Ahmad, A. Pine cone extract as natural coagulant for purification of turbid water. *Heliyon* **2019**, *5*, e01420, doi:10.1016/j.heliyon.2019.e01420.
60. 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. Crops Prod.* **2015**, *74*, 530–534, doi:10.1016/j.indcrop.2015.04.004.
61. Saritha, V.; Karnena, M.K.; Dwarapureddi, B.K. "Exploring natural coagulants as impending alternatives towards sustainable water clarification"—A comparative studies of natural coagulants with alum. *J. Water Process Eng.* **2019**, *32*, 100982, doi:10.1016/j.jwpe.2019.100982.
62. 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, doi:10.1016/j.seppur.2018.09.033.
63. Bouaouine, O.; Baudu, M.; Khalil, F.; Chtioui, H.; Zaitan, H. Comparative study between Moroccan cactus and chemicals coagulants for textile effluent treatment. *J. Mater. Environ. Sci.* **2017**, *2*, 2687–2693.
64. Asrafuzzaman, M.; Fakhruddin, A.N.M.; Hossain, A. Reduction of Turbidity of Water Using Locally Available Natural Coagulants. *Int. Sch. Res. Netw. ISRN Microbiol.* **2011**, *19*, 7–12, doi:10.5402/2011/632189.
65. Abidin, Z.Z.; Mohd, N.S.; Madehi, N.; Sobri, S. Optimisation of a method to extract the active coagulant agent from Jatropha curcas seeds for use in turbidity removal. *Ind. Crops Prod.* **2013**, *41*, 319–323, doi:10.1016/j.indcrop.2012.05.003.
66. Pritchard, M.; Craven, T.; Mkandawire, T.; Edmondson, A.S.; Neill, J.G.O. A comparison between Moringa oleifera and chemical coagulants in the purification of drinking water—An alternative sustainable solution for developing countries. *Phys. Chem. Earth* **2010**, *35*, 798–805, doi:10.1016/j.pce.2010.07.014.
67. Kakoi, B.; Wambua, J.; Ndiba, P.; Thiong, G. Banana pith as a natural coagulant for polluted river water. *Ecol. Eng.* **2016**, *95*, 699–705, doi:10.1016/j.ecoleng.2016.07.001.
68. Hussain, G.; Haydar, S. Exploring potential of pearl millet (Pennisetum glaucum) and black-eyed pea (Vigna unguiculata subsp. unguiculata) as bio-coagulants for water treatment. *Desalination Water Treat.* **2019**, *143*, 184–191, doi:10.5004/dwt.2019.23255.
69. Birima, A.H.; Hammad, H.A.; Desa, M.N.M.; Mudam, Z.C. Extraction of natural coagulant from peanut seeds for treatment of turbid water. *Earth Environ. Sci. Open* **2013**, *16*, 012065, doi:10.1088/1755-1315/16/1/012065.