

Removal of Suspended Particles from Water Sources Using Natural Coagulants: *Leucaena Leucocephala*

Angel Villabona-Ortiz^a, Candelaria Tejada-Tovar^a, Rodrigo Ortega-Toro^b, Betsy Torres-Castillo^a, Gabriela Cuten-Trespalacio^a

^aProcess Design, and Biomass Utilization Research Group (IDAB), Chemical Engineering Department Universidad de Cartagena, Avenida del Consulado St. 30. Cartagena de Indias, Colombia, 130015.

^bFood Engineering Department, Food Packaging and Shelf-Life Research Group (FP&SL), Universidad de Cartagena, Avenida del Consulado St. 30. Cartagena de Indias, Colombia, 130015.
ctejadat@unicartagena.edu.co

Coagulation/flocculation is the process of destabilization of colloidal particles, forming flocs that precipitate. Although chemical coagulants such as aluminum sulfate are effective, they have long-term negative effects on health and the environment. For this reason, natural alternatives are sought for the removal of turbidity in waters for human consumption that are safe and low cost. This study evaluates the capacity of the *Leucaena leucocephala* (LL) plant from the Colombian Caribbean region as a natural coagulant compared to aluminum sulfate. The active coagulant compound was extracted from the LL seeds by drying, grinding and salt extraction. The optimum dose was 4.5 mL/L, prepared with 10 g of seeds at 4% NaCl, achieving an efficiency of 86% and 97.54% with oil extraction. A 1% solution of aluminium sulphate showed an efficiency of 99.05% with a lower dose (2.4 mL/L) and took longer to settle (5 min) compared to LL (3-4 min). Furthermore, the pH remained stable with LL, while aluminum sulfate lowered it to 5.04. It is concluded that LL is feasible for water clarification and is presented as a promising alternative.

1. Introduction

Water is a resource of immense value globally, widely utilised in industries and essential for all living organisms. However, a significant proportion is unsuitable for human consumption due to the presence of sediments and colloids that contribute to contamination. Therefore, it is necessary to implement strategies to improve water quality, including enhancing taste, odour, and the control of suspended particles (Marrugo et al., 2022).

Previous studies have explored sustainable methods for removing suspended particles by utilising biomasses as coagulants in water treatment. This approach aims to reduce dependence on inorganic coagulants, which have health and environmental concerns. It has been demonstrated that biomasses, such as chickpea (*Cicer arietinum*), *Strychnos potatorum* (Nirmali), apricot seeds (*Prunus armeniaca*), moringa seeds (*Moringa oleifera*), okra mucilage (*Abelmoschus esculenta*), prickly pear seeds (*Opuntia ficus indica*) are promising plants to be used as coagulants in water treatment and to replace the use of chemical coagulants due the presence of molecules such as carbohydrates, proteins and polysaccharides that are associated with coagulant properties (Karnena & Saritha, 2022).

Because of this, the use of this coagulant is considered an opportunity to replace chemical coagulants such as aluminium sulfate in the industrial field, whose use is associated with severe health conditions due to its accumulative characteristic (Bryliński et al., 2023), so that the exploration of new plants represents an alternative of great potential for the treatment of raw water and wastewater with a low production cost (Vargas et al., 2022). This study contributes to the existing literature on the turbidity removal capacity of *Leucaena leucocephala* (LL) seeds, a plant widely available in Colombia (Perez Almaro et al., 2024). This leguminous species (*Fabaceae*) is commonly utilised in silvopastoral systems due to its high protein content (Hernández-Melchor et al., 2023). The effectiveness of plant coagulants from the *Fabaceae* family is verified, supporting previous studies that report turbidity and other pollutant removal rates above 70% (Karnena & Saritha, 2022). This research also

serves as a guide for the evaluation of active coagulants capable of removing turbidity in raw water. For this purpose, the raw material is subjected to extraction procedures for the coagulant actives, both without oil extraction and using the Soxhlet method (Arenas Diaz, 2019). The data obtained will serve as a reference for optimal preparation and will enable a comparison of the efficiency of these natural coagulants with well-known chemical coagulants, such as aluminium sulfate.

2. Methodology

2.1 Material

The seed powder was prepared using a Black+Decker convection oven, a Corona manual mill grinder, and a 7-sieve Orto-alresa sieve shaker. The coagulant extract was obtained from the seed powder using a sodium chloride solution and filtered with coffee filter paper. Similarly, hydrated aluminium sulfate, $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$, was used as a chemical coagulant, with which synthetic turbid water, prepared with bentonite and distilled water, was treated in a 4-seater F4-300-EQ flocculator. The characteristics of the turbid water before and after treatment were measured with an HM Digital Aquapro conductivity meter, an Orion Star A321 pH meter, and a HANNA HI98703 turbidity meter.

2.2 Process for Obtaining Seed Powder

A total of 811 LL pods were collected in Cartagena, from which 10 were selected to determine the weight and quantity of seeds and to establish the correlation between the number of pods and the quantity of raw material obtained. The pods had an average length of 15 cm, an average width of 2 cm, and an average weight of 1.46 g with seeds and 0.74 g without seeds. Each pod contained on average 20 seeds, with an average total weight of 0.74 g. The extracted seeds were washed with distilled water to remove impurities and dried in an oven at 80 °C for 5 hours. Subsequently, the seeds were crushed using a grinder and the particulate material was sieved to obtain a particle size of < 500 µm (Chee *et al.*, 2020).

For the extraction of oil present in the seed powder, 10 g of *Leucaena leucocephala* (LL) seeds were placed in filter paper and inserted into a laboratory Soxhlet apparatus. The seeds were then subjected to extraction using 300 mL of 96% ethanol as the organic solvent at a temperature of 200 °C. The process involved three cycles with a total duration of approximately 3 hours (Arenas Diaz, 2019).

2.3 Coagulant extract and synthetic water preparation

For the extraction of the coagulant solution, 10 g of powder were mixed in 100 mL of a 3% NaCl solution for 30 minutes at 200 rpm. After the mixing time, the solution was filtered under vacuum using a coffee filter (Kristanda *et al.*, 2021). The second and third coagulating solutions, prepared for the NaCl percentage variation, were made by mixing 10 g of powder in 100 mL of 2% and 4% NaCl solutions, respectively. Then, a fourth and fifth coagulating solution were prepared, in which 9 and 11 g of powder were mixed in 100 mL of a 4% NaCl solution, corresponding to the variation in the amount of seed.

Furthermore, the synthetic turbid water was prepared using clay (bentonite) to achieve a turbidity of 100 ± 2 NTU, for which base water was made by mixing 2.5 g of clay in 1 litre of water at 200 rpm for 1 hour, and then allowing it to settle for 24 hours to homogenise the solution. The solution was finally diluted until the required turbidity was reached (Villabona-Ortíz *et al.*, 2023).

2.4 Jar test

Four 1-litre beakers were filled with the synthetic turbid water, whose initial turbidity, conductivity, and pH were previously measured, and placed in a four-place flocculator. Different doses of coagulant were then added, and rapid agitation at 200 rpm was initiated for 3 minutes, followed by slow stirring at 60 rpm for 30 minutes. Finally, the water was left to settle for 1 hour, and the conductivity, pH, and final turbidity were determined. The turbidity removal efficiency and sedimentation time of the natural coagulant were then compared with those of aluminium sulfate. A stopwatch was used to record the time in minutes from the cessation of slow mixing until the sludge displacement stopped (Alnawajha *et al.*, 2023).

3. Results and Discussion

3.1 Obtaining Seed Powder

Initially, the seeds were washed with distilled water, which increased their moisture content and, consequently, their weight from 600 g to 762 g. Drying the 762 g of wet seeds resulted in a final weight of 568.2 g of dry seeds, representing an overall loss of 5.30% compared to the initial weight. From the milling process, 561.7 g of seed powder were obtained, representing an overall loss of 6.38% compared to the initial weight. The sieving process resulted in a loss of 11.02% relative to the initial weight, yielding a total of 533.9 g of seed powder.

During oil extraction, it was observed that, upon contact of the solvent with the seed powder, the solvent's colour changed from colourless to dark green, indicating that the oil dissolved in the solvent, as reported by Arenas Diaz (2019). Of the 10 g of seeds subjected to extraction, 9.12 g of seed powder were obtained, resulting in losses of 8.8%. This indicates that 0.56 g of oil was extracted, a value higher than those reported by De Angelis et al. (2021).

3.2 Jar test

3.2.1 NaCl concentration effects on coagulant capacity

Figure 1a shows that increasing the concentration of NaCl improves the percentage of turbidity removed, with the best results at 4% NaCl. This indicates that higher NaCl concentrations enhance the extraction of proteins responsible for coagulation (Correia et al., 2022), which are present in LL seeds and make up about 30% of their weight (De Angelis et al., 2021). The results allow us to identify the 4% NaCl concentration as the best for the preparation of the coagulant from LL, given that with this concentration the highest turbidity removal of 81.3% was achieved using a dose of 4.5 mL/L.

Figure 1b shows that the concentration of NaCl does not have a significant effect on the pH of the water, as occurs in experiments carried out with natural coagulants by Carrizales Condori & Enríquez Nateros (2019). The initial pH values range between 8.54 and 7.71, and the final values between 7.5 and 7.01, representing a slight decrease once the natural coagulant is added. These results can be considered within normal and neutral values, as established in the Colombian Resolution (Ministerio de la Protección Social, Ministerio de Ambiente, 2007b).

Analyzing Figure 1c, the concentration of sodium chloride affects the conductivity of the evaluated water, which has initial values of 16 $\mu\text{S}/\text{cm}$, thanks to the Na^+ and Cl^- ions contained in the coagulant solution, causing an increase in this parameter, a behaviour that coincides with that described by Navarro & Quintero (2021). A maximum final conductivity value of 355 $\mu\text{S}/\text{cm}$ is observed. This indicates that it is within the acceptable values according to Colombian Resolution 2115 of 2007, Article 3°, which establishes a maximum acceptable value for conductivity of up to 1000 $\mu\text{S}/\text{cm}$ (Ministerio de la Protección Social, Ministerio de Ambiente, 2007a). Because of this, we did not work with higher NaCl concentrations.

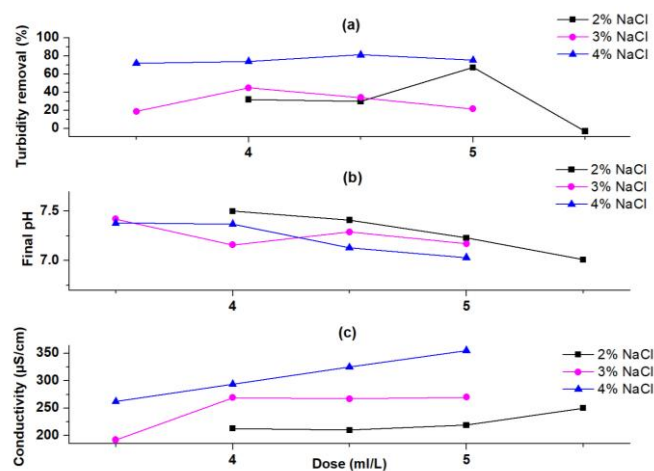


Figure 1. Effects of NaCl concentration on turbidity removal percentage (a), final pH (b), and final conductivity (c) in relation to coagulant dosage.

3.2.3 Effects of seed quantity on coagulant capacity

Figure 2a illustrates that the quantity of seeds significantly impacts the coagulant capacity. Turbidity removal was comparable when using 8 g and 10 g of seeds in coagulant preparation, both yielding satisfactory results. However, employing 10 g resulted in higher turbidity removal (86%), establishing it as the best amount for the preparation of the natural coagulant. It can also be noted in Figure 2a that when using 8 g of powder, its effectiveness decreases to 85.7%, and as reported by Martínez & González (2012), when a solution does not have enough destabilising charges, the removal efficiency is reduced, while with 12 g, there is a greater amount of charges, thus the solution is saturated, generating residual turbidity due to the coagulant solution, and the removal effectiveness decreases to 79.7%. From Figure 2b and 2c, it can be seen that the pH and the

conductivity are minimally affected by the variation in the amount of seeds. In the end, using 10 g achieved greater stabilisation in turbidity removal with the doses evaluated and was selected as the best alternative.

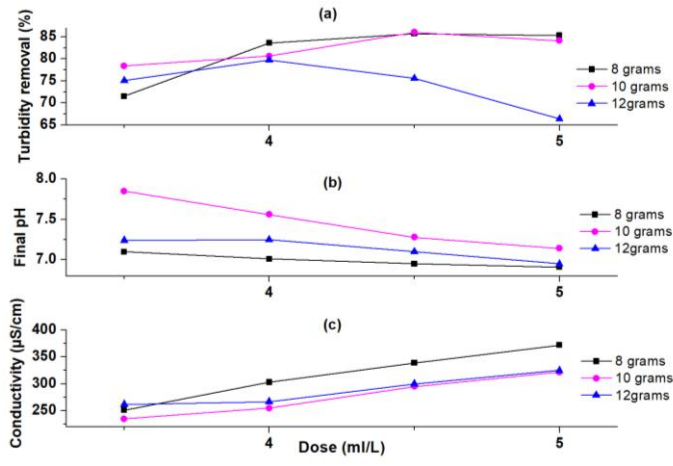


Figure 2. Seeds concentration effects on turbidity removal percentage (a), final pH (b), and final conductivity (c) in relation to coagulant dose

3.3 Efficiency comparison of the natural coagulant with aluminum sulfate

Based on the data in Figure 3, the natural coagulant evaluated had acceptable results when compared with the chemical coagulant aluminium sulfate and with other studies that have been done with this same plant and other plants as coagulant. First, the coagulating capacity for LL was 86% and 94.7% with and without oil respectively, however, higher doses need to be implemented with oil extraction, while the chemical coagulant aluminium sulfate was 99.05%. In addition, the concentration and doses applied for the evaluation of aluminum sulfate were 1% (w/v) and 2.4 mL/L, lower than those used for LL extracts, which means that its yield is higher since it requires minimum doses. This is consistent with what has been described in several investigations, where chemical coagulants tend to have higher efficiency compared to natural coagulants (Barreto Pardo et al., 2019).

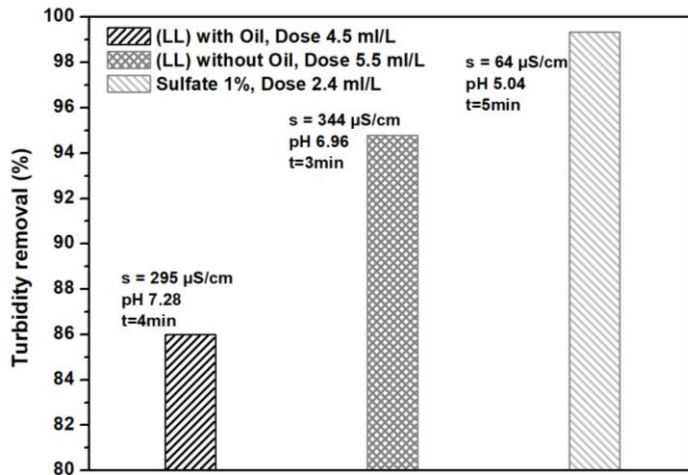


Figure 3. Comparison of natural coagulants and aluminum sulfate

Regarding the final pH of the evaluated water, its values ranged between 6.95 and 7.28 for the natural coagulant, which means a neutral pH. This is in contrast to aluminium sulfate, where the pH was 5.02, which in turn leads to a decrease in its quality. It can also be observed that the sedimentation time in the case of LL was shorter than the sedimentation time of aluminium sulfate by 1 minute without oil extraction and by 2 minutes with oil extraction, which indicates a slight advantage in relation to the chemical coagulant.

Comparing these results with other studies reported in the literature (Table 1), it can be observed that the prepared coagulant achieves an acceptable and competitive removal percentage of 86%, outperforming other seeds tested under similar conditions with an initial turbidity concentration close to 100 NTU. This positions the prepared coagulant as a promising low-cost alternative for water treatment. Other studies demonstrate higher efficiency at greater turbidity levels, indicating that the preparation of the coagulant will depend on the specific properties of the water to be treated.

Table 1. Comparison of seeds used as a source of natural coagulants

Plant	Water to be treated	Initial concentration	Coagulant solution	Particle size (mm)	Dose	Removal %	Reference
Cassia fistula	Domestic wastewater	90.28 NTU	25% of seed; Distilled water	0.4	160 mg/L	62.18	(Dunoyer <i>et al.</i> , 2022)
Trigonella foenum	POME	12667 NTU	2% of seed; Distilled water	0.2-0.5	4.09 mg/L	94.97	(Lanan <i>et al.</i> , 2021)
Carica papaya	Synthetic Water (Kaolinite); River Water	100 NTU 200 NTU	1% of seed; 1% NaCl	0.4	50 mg/L	90.7 74.6	(Marrugo <i>et al.</i> , 2022)
Leucaena leucocephala	Synthetic Water (Kaolinite)	326 NTU	1% of seed; 1M NaCl	0.062	5 mL/L	99.7	(Putra <i>et al.</i> , 2022)
	Synthetic Water (Bentonite)	100 NTU	10% of seed; 4% NaCl	0.5 – 0.355	4.5 mL/L	86	This study

Abbreviations: POME - Palm Oil Mill Effluent.

4. Conclusions

The coagulating effectiveness of the LL plant can be attributed to its protein content, as evidenced by its high efficiency in turbidity removal. An 86% removal was achieved using a dosage of 4.5 mL/L prepared with 10 g of seeds and 4% NaCl. It was observed that the efficiency of the coagulating extract is determined by the NaCl concentration. A very low salt concentration reduces the presence of proteins in the extract, thereby lowering its coagulating capacity. Conversely, increasing the salt concentration saturates the opposite charges on the proteins, resulting in reduced turbidity removal and increased conductivity of the treated water. It was noted that for LL seed oil extraction, this resulted in an increase in removal efficiency from 86% to 94.7%.

It was determined that aluminium sulfate is more effective in turbidity removal compared to the extracted natural coagulants. However, an important consideration is the pH impact: natural coagulants had no significant effect on water pH, maintaining an ideal neutral range. In contrast, aluminium sulfate acidified the water. This highlights natural coagulants as a promising and safe alternative without adverse effects on human health, offering potential for partial replacement of chemical coagulants in water treatment processes.

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