Evaluation of the effect of *Moringa oleifera* and *Caesalpinia spinosa* mixtures on surface water turbidity

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Evaluación del efecto de mezclas de Moringa oleifera y Caesalpinia spinosa en la turbidez de las aguas superficiales

Avaluació del efecte de mescles de Moringa oleifera i Caesalpinia spinosa en la superfície

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SUMMARY

The chemical coagulants used in the purification of surface waters generate a negative impact on the environment, with long-term effects on human health. The present study was carried out with the purpose of evaluating the coagulation-flocculation from *Moringa oleifera* and *Caesalpinia spinosa* seeds. The factors were evaluated: drying temperature of the seeds (45, 55 and 65 ° C), the concentration of NaCl in the solutions (0 and 5 g / L), as well as the agitation regime. Also, it was determined that the mixture of the solutions of *M. oleifera* and *C. spinosa* favor the removal of turbidity from the mixture dose of 15 mL / L of each solution. The water to be treated had a turbidity and initial color of 206 NTU and 2510 UPC respectively, the results for the mixtures showed values of turbidity and color removal of 98.03% and 95.56% respectively.

Keywords: *Moringa oleifera*; *Caesalpinia Spinosa*; natural coagulant; mixture; superficial water.

RESUMEN

Los coagulantes químicos utilizados en la purificación de aguas de la superficie generan un impacto negativo en la purificación del ambiente, con efectos a largo plazo en la salud del ser humano. El presente estudio fue llevado a cabo con el objetivo de evaluar la coagulación-floculación de semillas de *Moringa oleifera y Caesalpinia spinosa*. Se evaluaron los factores siguientes: temperatura de secado de las semillas (45, 55 y 65 ° C), la concentración de NaCl en las soluciones (0 y 5 g / L), así como el régimen de agitación. Además se determinó que la mezcla de las soluciones de M. oleifera y C. spinosa favorece la eliminación de la turbidez de la dosis de mezcla de 15 ml / l de cada solución. El agua que debía ser tratada tenía una turbidez y un color inicial de 206 NTU y 2510 UPC respectivamente, y los resultados para las mezclas mostraban valores de turbidez y eliminación del color del 98.03% y 95.56% respectivamente.

Palabras clave: *Moringa oleifera; Caesalpinia* Spinosa; coagulante natural; mezcla; agua superficial.

RESUM

Els coagulants químics utilitzats en la purificació d'aigües de la superfície generen un impacte negatiu en la purificació del ambient, amb efectes a llarg termini en la salut del ser humà. El present estudio va ser portat a terme amb el objectiu d'avaluar la coagulació-floculació de llavors de *Moringa oleifera i Caesalpinia spinosa*. Es van avaluar els factors següents: temperatura de assecament de llavors (45, 55 y 65 ° C), la concentració de NaCl en les solucions (0 y 5 g / L), així com el règim de agitació. També es va determinar que la mescla de les solucions de M. oleifera i C. spinosa afavoreix la eliminació de la terbolesa de la dosis de mesura de 15 ml / l de cada solució. L'aigua que devia ser tractada tenia una terbolesa i un color

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inicial de 206 NTU i 2510 UPC respectivament, i els resultats per les mescles mostraven valores de terbolesa i eliminació del color del 98.03% i 95.56% respectivament.

Paraules clau: *Moringa oleifera; Caesalpinia Spinosa*; coagulant natural; mescla; aigua superficial.

INTRODUCTION

Nowadays, due to industrialization, the humans consume a greater volume of water than at other times, so there is less availability of the resource[1]. The sources of drinking water are under the threat of contamination, with far-reaching consequences for human health, as well as for the economic and social development of communities and even nations[2]. According to the reports from the World Health Organization (WHO) and the United Nations Children's Fund (UNICEF), more than 20% of the population does not have access to safe water (2.6 billion people) and nearby 40% suffer from shortages[3]. It is reported that 3 out of 10 people lack access to safe drinking water in the home and 6 out of 10 lack safe sanitation. Latin America holds 26% of the planet's water resources, which are available to 6% of the population. In contrast, Asia concentrates 30% of the availability of the resource and 60% of the inhabitants of the planet [4].

It is estimated that by 2030 the world will face a global deficit of 40% of water [5]. One of the great challenges of the 21st century will be to improve water management, to ensure that the world population of nine billion or more by 2050 has access [4].

Natural coagulants have high efficiency in breaking thermodynamically stable contaminating systems. In contrast to chemical coagulants, the so-called natural coagulants have not reported a danger to the flora or fauna, nor negative effects on human health [6]. The present work proposes to evaluate the action of the mixture of seeds of *Moringa oleifera* and *Caesalpinia spinosa* in surface water.

MATERIALS AND METHODS

Water sampling

To achieve the proposed objective, 300 L of surface water were used, which were taken from the Poza Honda irrigation canal in the city of Portoviejo, Manabí, Ecuador, located at the geographic coordinates of 1 ° 04'32.1 "S and 80 ° 25'56.0 "W. Turbidity and color were measured as water interest characteristics by means of a portable spectrophotometer (Hach DR900) in nephelometric turbidity units (NTU) and platinum-cobalt units (UPC), respectively.

Coagulant solutions

Aqueous seed solutions of two plant species were used: *Moringa oleifera* and *Caesalpinia spinosa*, which acted as coagulating agents. The seeds of *Moringa oleifera* and *Caesalpinia spinosa* were obtained in Manabí and Tungurahua province, respectively. For the preparation of the solutions, the *M. oleifera* and *C. spinosa* seeds were first processed to obtain flours. The moisture content was reduced in a solids dehydrator, three drying temperature (DT) were tested (45, 55 and 65 ° C). Then the particle size was reduced by means of a mechanical hammer mill.

The solutions were prepared with distilled water. For each liter of solution, 20 g of the obtained flours were added. A total of 12 solutions were prepared as shown in Table 1. To 6 of the solutions, 5 g of sodium chloride were added for each liter of water, this parameter was identified as SC. The prepared solutions are presented below:

Table 1. *Prepared coagulant solutions.*

Species	*Moringa oleifera*						*Caesalpinia spinosa*					
SC (g/L)	5			0			5			0		
DT (°C)	45	55	65	45	55	65	45	55	65	45	55	65

Jars Test

Prior to the evaluation of the seeds mixtures, the effect of the agitation on the turbidity decrease was analyzed. For this, jars tests were carried out in 2 agitation speed regimes: R1 (300, 150 and 15 rpm in turns of 10 minutes each) and R2 (200 rpm and 60 rpm in turns of 1 and 15 minutes respectively). Both regimens considered a sedimentation time of 20 minutes. It should be noted that jar tests used 1 L of sample water, the dosage of coagulant solutions was 15 mL per liter of water to be treated.

After defining the conditions of agitation, drying temperature and concentration of NaCl, a design of experiments of the central type composed with axial points was carried out to evaluate the dosage of the solutions in mixtures. Table 2 shows the combinations that were obtained.

Table 2. *Composite central design for the evaluation of mix dosages.*

Moringa oleifera *(mL/L)*	***Caesalpinia spinosa*** *(mL/L)*
15	15
15	25
20	20
25	15
25	25

For the evaluation of the mixtures, the final turbidity (NTU_f) and final color (C_f) data were processed by means of the expressions:

$$\%R_{Turbidity} = \frac{(NTU_o - NTU_f)}{NTU_o} 100 \quad eq.1$$

$$\%R_{Color} = \frac{(C_o - C_f)}{C_o} 100 \quad eq.2$$

For the development of the research, each experimental run was carried out in triplicate. Statgraphics

Table 3. *Percentages of turbidity removal after treatment with natural coagulants*

Species	DT (°C)	CS (g/L)	R1 Filtered Turbidity (Removal %)	R2 Filtered Turbidity (Removal %)	R2 Unfiltered Turbidity (Removal %)
Moringa oleifera	45	0	96.05±0.25	98.67±0.58	75.00±1.73
		5	91.55±8.14	96.67±1.53	45.10±2.65
	55	0	94.89±0.85	98.33±0.58	90.01±2.00
		5	94.22±1.26	92.33±2.08	57.67±3.06
	65	0	91.73±5.46	96.67±2.08	77.67±1.15
		5	90.36±7.59	95.67±4.16	57.67±8.50
Caesalpinia spinosa	45	0	92.14±3.01	94.67±2.31	64.67±3.21
		5	85.86±9.48	81.67±2.31	67.33±4.51
	55	0	93.98±1.49	90±3.46	69.67±1.15
		5	92.61±1.39	93±2.65	70.33±2.31
	65	0	91.28±0.69	94.33±2.52	80.33±3.51
		5	91.6±1.45	92.67±2.08	88.67±2.52

Centurion XV software was used to process the data. The confidence level proposed for the analysis was 5%.

RESULTS

Influence of agitation conditions, NaCl concentration and drying temperature for each species.

In the evaluation of the agitation regime, surface water with turbidity of 100 NTU was used. The action of each species was evaluated individually. Table 3 shows the results of the turbidity removal by means of the dosage of the coagulant species.

In the case of *M. oleifera*, the turbidity removal values showed significant differences in relation to the factors agitation regime and concentration of NaCl, whose P-value were 0.0081 and 0.0279, respectively. No differences were detected between the reported turbidity values in relation to the drying temperature (P-value = 0.3746). In the case of *C. spinosa* also, significant differences are observed in relation to the concentration factor of NaCl, whose P-value is 0.0326. However, the drying temperature and the agitation rate (P-value of 0.8941 and 0.0507, respectively) did not influence the turbidity removal values.

The agitation regime R2 was selected for the next experimental phase, due to the fact that it presented the best results for *M. oleifera*, whereas for *C. spinosa* this factor had no influence. With regard to the temperature of drying, it was established that the temperatures of 45 and 65 ° C were those indicated for the processing of *M. oleifera* and *C. spinosa*, respectively. Only NaCl (5 g / L) was added in the solution of *C. spinosa*, because in this condition the lowest removals were obtained for the treated and unfiltered waters in the R2 regime.

Optimization of the dosage of the solutions.

The surface water sample used to determine the optimum dose of *M. oleifera* and *C. spinosa* showed turbidity values of 206 NTU, as well as 2510 UPC of color. Table 4 shows the results of the treatments with mixtures of *M. oleifera* and *C. spinosa*.

When comparing tables 3 and 4 it is observed that the mixtures led to greater uniformity in the results (minor standard deviations) and that turbidity removal values were reached above 90% without filtering the treated water. Regarding the removal of color, the mixture of coagulating agents in the treated and unfiltered samples does not reach 91%, but filtered it achieves 95% of color removal.

From these results of the experimental design with the mixtures, 4 polynomials were adjusted (one for each response variable). These include factors related to the dosages of *M. oleifera* and *C. spinosa*, as well as an interaction factor between both species.

The analysis of the turbidity removal models, indicates that the concentrations of *M. oleifera* and *C. spinosa*, do not present interactions (P-value > 0,05) or effects related to their concentrations (P-value of 0.6831 and 0.7462, respectively), when the samples are filtered. In the unfiltered samples it was observed that the interaction between coagulant agents is significant (P-value less than 0.0001). The maximization of

Table 4. *Percentage of turbidity and color removal after treatment with Moringa oleifera and Caesalpinia spinosa mixture.*

Dosage (mL/L)		Turbidity (Removal %)		Color (Removal %)	
Moringa oleifera	*Caesalpinia spinosa*	Filtered	Unfiltered	Filtered	Unfiltered
15	15	98.30±0.34	91.26±4.81	95.08±1.55	90.96±0.85
15	25	98.06±0.10	95.39±0.34	92.05±0.37	89.94±1.16
20	20	98.09±0.7	93.66±3.32	93.43±0.86	90.41±1.86
25	15	98.20±0.69	91.02±1.72	93.76±0.87	90.18±0.48
25	25	97.57±0.69	92.96±1.72	91.25±2.28	87.91±3.30

Table 5. *Results of physicochemical analysis of surface water before and after treatment with M. oleifera and C. spinosa.*

Parameter	Water sample		
	Untreated sample	*Treated and filtered water*	*Treated and unfiltered water*
Temperature *(°C)*	28.7	28.5	28.5
Turbidity *(NTU)*	557.2	1.77	3.37
pH	7.14	7.10	7.13
Alkalinity *(ppm)*	204.525	199.98	195.435
Suspended solids *(ppm)*	1240	<25	<25
Total dissolved solids *(ppm)*	619	690.1	666.4
Chlorides *(ppm)*	41.85	75.04	71.19
Hardness *(ppm)*	322.46	302.00	308.86
Hydrogen sulfide *(ppm)*	0.74	<0.1	<0.1
Iron *(ppm)*	2.37	0.15	0.25
Aluminium *(ppm)*	0.36	<0.1	<0.1
Silica *(ppm)*	36.1	2	2
Chemical Oxygen Demand *(mg/L)*	100.39	64.45	254.22

response variables estimated that doses of 13.33 mL / L of *Moringa oleifera* and 18.56 mL / L of *Caesalpinia spinosa*, with a reduction of 98.18%.

The analysis of the polynomials for the reduction of color indicated that both for the filtered samples, and without filtering the interactions between *M. oleifera* and *C. spinosa* are significant. The P-values reported in both cases were less than 0.0001. From the optimization of the model it was determined that the dosage that maximizes the removal of color is 15.4934 mL / L of *Moringa oleifera* and 12.9289 mL / L of *Caesalpinia spinosa*, with an estimated color removal of 95.56 %.

To achieve a compromise response in which turbidity and color removals were maximized, a maximization of multiple responses was performed. The result of this analysis reveals that the doses of 14.8061 mL / L and 12.9289 mL / L of *Moringa oleifera* and *Caesalpinia spinosa* respectively, achieve a 98.03% removal of turbidity and 95.56% of color removal.

To verify the influence of the mixture of *Moringa oleifera* and *Caesalpinia spinosa* on physicochemical parameters of the surface water, a water sample with higher initial turbidity was used, unlike the previous experiments. In addition to turbidity, physicochemical parameters were measured such as pH, alkalinity, suspended solids, total solids, hydrogen sulfide, among others. Table 5 reports the results of the physicochemical analyzes performed on the mother sample and surface water after treatment with the seed mixture.

DISCUSSION

Drying temperature

According to the statistical analysis of the experimental results, it was established that the drying temperature of the seeds of *M. oleifera* and *C. spinosa*, has a significant effect on the removal of turbidity. It was reported that the coagulating action of *Moringa oleifera* is due to the presence of a cationic protein [7], which is preserved at drying temperatures close to 40 °C, so other organic components do not lose their chemical properties, this statement agrees with the result obtained in the experimentation [8]. Therefore, in the investigated interval, the greatest removal (98.67%) of turbidity was obtained at a temperature of 45°C.

The seeds of *Caesalpinia spinosa* have a high content of gums with contents of galactomannans, which are neutral polysaccharides, with a linear structure of mannan with side chains of a single unit of galactose [9]. Galactomannans are used as stabilizers in the chemical industry, due to their ability to increase the particle size from the formation of conglomerates [10]. It was estimated that at temperatures close to 60 °C, the concentration of carbohydrates and other components is favored [8]. These affirmations agree with the obtained result, at a temperature of 65 °C the *Caesalpinia spinosa* presented the greater removal of turbidity (92,67%).

Agitation

The increase in the speed of agitation had a negative effect on the removal of turbidity in the treatment with *M. oleifera*. The treatment with *C. spinosa* did not present significant differences, therefore, the speed of agitation does not influence this species. The agitation speed and the mixing time, influence the formation of flocs, affecting the size and the force that the particles generate to stay together [11]. Excessive mixing can cause the breaking of the formed floc particles, whose breaking is related to the strength of the flocs, giving rise to a colloidal suspension, thus reducing the effectiveness in the removal of solids[12]. The colloidal stability increases according to the molecular weight of the polymer, for which the efficiency of flocculation increases [13]. The coagulant components of the *M. oleifera* have molecular weights between 3 and 30 kDa [9a]. In the other side, *C. spinosa* seeds, contain gums that have a high molecular weight between 50 and 8000 kDa [14]. This information agrees with the results obtained in the experimentation, however, the greatest removal is evident in the species *M. oleifera* with 98.67%, compared to *C. spinosa* with a turbidity removal of 92.67%.

Sodium chloride concentration

For the *Moringa oleifera* species, the obtained experimental results refer that the solutions without the presence of NaCl remove greater amount of turbidity in relation to the solutions with NaCl. According to what is presented in the specialized literature, the addition of NaCl allows the extraction of the cationic protein that makes up the coagulant of *M. Oleifera*. This causes greater contact with the particles present in the water, which potentiates the coagulant [15]. The coagulation efficiency of the crude extract of *M. Oleifera* is considerably improved by using saline solutions as a solvent due to the increase in the solubility of the proteins by the addition of salts. This mechanism suggests the breakdown of protein-protein or protein-polysaccharide or other associations existing in the seed powder of *M. oleifera*, which increases the soluble proteins in the saline solutions resulting in an increase in their coagulant activity, the solutions of sodium chloride and seawater act, therefore, better than distilled water [7, 16]. This evidence contradicts the results obtained experimentally, however [17] states that tannins diminish their capacity to form aggregates in solutions with the presence of NaCl. This suggests that both protein and tannins are the active components for coagulation-flocculation in surface water.

In the scientific literature, there are no reports that refer to the study of the addition of NaCl in *Caesalpinia spinosa* solutions as a coagulating agent for water treatment in general. Reason for which, the corresponding experimentation was carried out, the experimental results obtained refer that the presence of NaCl in *Caesalpinia spinosa* solutions potentiates the effect of turbidity reduction in superficial turbid water.

Optimal dose in the removal of turbidity and color

According to [2, 7a] the cationic proteins present in the solution of *M. oleifera* act as an active coagulant agent, in the form of soluble polypeptide chains, which allow contact with the colloidal particles present in the water to be treated. According to [13a, 18] *C. spinosa* allows the formation of larger particles, due to high weight components molecular like the gums and galactomannans (50 and 8000 kDa) contained in the seed. [19] stated that the dosage of the polymer chains should be sufficient to form suitable connecting bonds between the particles, so that they can spread from one particle to another and most of the particles join together, this causes the destabilization of the suspensions.

According to the experimental data recorded in Table 5, natural coagulants based on mixtures of *Moringa oleifera* seeds and *Caesalpinia spinosa*, remove turbidity, as well as important physicochemical parameters for the water purification process. According to [20], *Moringa oleifera* flour manages to reduce other physicochemical parameters of water such as acidity (85.71%), alkalinity (11.53%), chlorides (66.47%) and hardness (17.02%). According to [21] the biochemical oxygen demand (BOD_5) decreases up to 52% and the fecal coliforms reduce up to 99.3 %. As mentioned by [22] refers that the flour of *Caesalpinia spinosa* has a potential to decrease hardness and BOD_5 to 16.26% and 69.29% respectively. [23] stated that the COD in treated samples reduced 51.4% after treatment.

However, the dose of mixture of natural coagulants does not decrease parameters such as total dissolved solids and chlorides, on the contrary, they increase by 11% and 79% in the filtered treated samples and by 7% and 70% in the unfiltered treated sample respectively. This result is contradictory with what was stated by [20], who observed that the treatment with *M. oleifera* seed reduced the total dissolved solids in the samples of water to be treated. However, according to [21, 24] the TDS and chlorides increase due to the presence of mineral elements, charged macromolecules, salts and other ionic compounds that dissolve or dissociate in the treated water.

According to [25] the *Moringa oleifera*-based coagulant also increases COD and conductivity in waters of low initial turbidity (15 NTU). [22] mention that the *Caesalpinia spinosa* seed decreases the conductivity to 18% in after the coagulation-flocculation process in the water treatment.

CONCLUSIONS

The preparation conditions of the solutions of *M. oleifera* and *C. spinosa* were determined. It was evidenced that the drying temperature of the seeds has a effect on coagulation process, as well as the addition of NaCl in the solutions. The agitation of the jar test influenced the removal of turbidity for the *M. oleifera*, in which the coagulant effect was decreased with the increase in the velocity of the impeller.

The dosage of 15 mL / L of both solutions manages to remove 98.30% of turbidity and 95.08% of color of an initial sample of surface water of 206 NTU of turbidity and 2510 UPC of color. In addition, it was found that the mixture of coagulants has an effect in the reduction of iron, silica, hydrogen sulfide and chemical oxygen demand.

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