

## Original Article

<https://doi.org/10.22463/0122820X.2960>

### Clarification of superficial waters of the Magdalena river using seeds of *Tamarindus indica* as bio-coagulants

Clarificación de las aguas superficiales del río Magdalena utilizando semillas de *Tamarindus indica* como biocoagulantes

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**How to cite:** S.D. Buelvas-Caro, Y.del Rosario Aguas-Mendoza, R.E. Olivero-Verbel, "Clarification of superficial waters of the Magdalena river using seeds of *Tamarindus indica* as bio-coagulants". *Respuestas*, vol. 25, no. 2, pp. 170-176, 2020.

Received on February 07, 2020; Approved on April 5, 2020

## ABSTRACT

### Keywords:

Coagulant,  
Clarification,  
Tamarind seeds,  
Turbidity.

Biocoagulants have emerged as an environmentally friendly and efficient alternative to surface water clarification, allowing the use of agro-industrial waste as a bioavailable source for the treatment of high turbidity water. The objective of this research was to compare the efficiency in the surface water coagulation process of the Magdalena river using aluminum sulfate and tamarind seeds (*Tamarindus indica*) dissolved in aqueous solution and saline solution. The clarification process was carried out by testing jar, for this was performed an extraction of the hot and cold tamarind coagulant which were evaluated in conjunction with an inorganic coagulant in different mother dilutions (distilled water and saline) at concentrations of 35 and 40 mg/L and at a stirring speed of 100 and 200 rpm. The parameters of pH, electrical conductivity (S/cm), turbidity (NTU) and color (UPC) were measured during the clarification process. The results show a 97.2% reduction in inorganic treatment (SAL) compared to initial turbidity and a decrease in natural cold extracted (STF) and hot (STC) treatments of 58.2% and 39.1% respectively, when diluted in aqueous solution. While diluting in saline solution there was a greater removal of turbidity when applying aluminum sulfate, followed by STF and STC treatment, with removal values of 99%, 75% and 53% respectively. The highest coagulating activity occurred when applying a dose of 40mg/L and when diluting treatments in NaCl with maximum reported values for SAL, STF and STC treatment of 93%, 85% and 59%. Tamarind seeds proved to be an efficient coagulant for water clarification.

## RESUMEN

### Palabras clave:

Coagulante,  
Clarificación,  
Semillas de tamarindo,  
Turbidez.

Los biocoagulantes han surgido como una alternativa eficiente y amigable con el medio ambiente para la clarificación de aguas superficiales, permitiendo el uso de residuos agroindustriales como fuente biodisponible para el tratamiento de aguas de alta turbidez. El objetivo de esta investigación fue comparar la eficiencia en el proceso de coagulación del agua superficial del río Magdalena utilizando sulfato de aluminio y semillas de tamarindo (*Tamarindus indica*) disueltas en solución acuosa y solución salina. El proceso de clarificación se llevó a cabo por medio de una jarra de ensayo, para ello se realizó una extracción del coagulante de tamarindo en frío y en caliente los cuales fueron evaluados en conjunto con un coagulante inorgánico en diferentes diluciones madre (agua destilada y solución salina) a concentraciones de 35 y 40 mg/L y a una velocidad de agitación de 100 y 200 rpm. Durante el proceso de clarificación se midieron los parámetros de pH, conductividad eléctrica (S/cm), turbidez (NTU) y color (UPC). Los resultados muestran una reducción del 97,2% en el tratamiento inorgánico (SAL) con respecto a la turbidez inicial y una disminución en los tratamientos naturales de extracción en frío (STF) y en caliente (STC) del 58,2% y 39,1% respectivamente, cuando se diluye en solución acuosa. Al diluir en solución salina hubo una mayor eliminación de la turbidez al aplicar sulfato de aluminio, seguido del tratamiento STF y STC, con valores de eliminación del 99%, 75% y 53% respectivamente. La mayor actividad coagulante se produjo al aplicar una dosis de 40mg/L y al diluir los tratamientos en NaCl con valores máximos reportados para el tratamiento SAL, STF y STC de 93%, 85% y 59%. Las semillas de tamarindo demostraron ser un coagulante eficiente para la clarificación del agua.

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## Introduction

Access to drinking water sanitation systems for many communities around the world is limited [1], due to the constant discharges of effluents caused by anthropogenic activities, which has led to contamination of bioavailable water sources with impurities, gases, dust and pathogenic microorganisms [2]. Chemical coagulants such as inorganic salts and synthetic polymers have been implemented for the primary treatment of this type of water resources which have been associated with the generation of diseases such as cancer, Alzheimer's and bone diseases [3] - [5]. In the face of the cluster of problems generated for the primary treatment of water, green alternatives have emerged such as the use of biologically sourced coagulants/flocculants which have aroused great interest due to the advantages they provide, such as biodegradability, bioavailability, low cost and mainly the elimination of toxic risk to humans [6].

Among the many options for the coagulation process we find tamarind seeds, which are mostly composed of carbohydrates, protein and water, the protein fraction is mostly made up of glutamic, aspartic acids, glycine and leucine. Recent research carried out by [7] and [8] has shown the effectiveness of tamarind seed as a natural coagulant with removal values between 85% and 97.6%. Positioning tamarind seed as a potential source for the coagulation/ flocculation process. The objective of this research was to compare the efficiency in the surface water coagulation process of the Magdalena river using aluminium sulfate and tamarind seeds (*Tamarindus indica*) dissolved in aqueous solution and saline solution.

## Materials and methods.

### *Preparation of natural coagulant (Tamarindus indica)*

The natural coagulant was obtained by cold and hot extraction. The cold extraction was carried out by a tamarind peeling process followed by manual depulping, washing, drying at room temperature and a sizing process, to finally subject the obtained flour to a sieving process in order to have a homogeneous particle size. Hot extraction followed the same procedure described above, unlike tamarind was first subjected to a cooking process for 10 minutes.

### *Jar test*

To perform the coagulation-flocculation process an E&Q model FP4 flocculator was used, where cold extracted organic coagulant (STF) and hot coagulant (STC) was added, aluminum sulfate (SAL) previously diluted in distilled water and saline to three jugs containing the samples collected in the Magdalena River. Applying a quick mixture (200 rpm) for 1 minute, followed by a slow mix (40 rpm) for 30 minutes finally let the floc settle for 1 hour according to ASTM No. D2035-80. Physicochemical properties (pH, electrical conductivity (S/cm), turbidity (NTU) and color (UPC)) were measured at the supernatant of each sedimented jug using the methodologies referred to in the APHA-AWWA-WEF method manual.

### *Coagulating activity and removal efficiency.*

The determination of the coagulating activity of the different treatments used were measured according to (1). Where coagulating activity is defined by reducing turbidity from the initial,

$$\% \text{ Coagulant Activity} = \frac{RT_{\text{Control}} - RT_{\text{Sample}}}{RT_{\text{Control}}} * 100 \quad (1)$$

### *Experimental design*

A categorical multifactorial design was carried out with 2x3x2 arrangement in which 2 concentrations, 3 treatments and 2 stirring speeds were evaluated. The Statgraphics centurion version XV statistical program was used for data processing and analysis.

## Results and Discussions.

### *Physicochemical properties of coagulants diluted in aqueous solution.*

The initial characteristics of the water samples collected in the Magdalena river are shown in Table I. Where highly murky water is evident according to decree 15705 of the Ministry of Social Protection.

**Table I.** Initial water characteristics

Turbidity (UNT)	Color (UPC)	pH	Conductivity ( $\mu\text{S}/\text{cm}$ )
$170 \pm 0,31$	$28 \pm 0,24$	$6,96 \pm 0,06$	$117 \pm 0,5$

### *Physicochemical properties of coagulants diluted in water and saline solution*

The physicochemical parameters for the different treatments diluted in distilled water and saline solution are shown in Table II, where water-diluted treatments show significant differences in turbidity between aluminium sulfate (SAL) with respect to cold and hot tamarind seed coagulants (STF), showing a 97.2% reduction in initial turbidity, STF and STC treatments achieved a reduction of 58.2% and 39.1% respectively. When assessing the same property, in saline solution, greater elimination of turbidity could be demonstrated when applying aluminium sulfate, followed by treatment with STF and STC, with turbidity elimination values of 99%, 75% and 53% respectively. It can be observed that when diluting the different saline treatments, the elimination values are higher than those obtained in distilled water. The color, a variable closely related to turbidity, showed the same trend in terms of decrease in both mother dilutions, placing the SAL treatment followed by the STF as the best for color reduction in the waters of the Magdalena River. As for the pH variable it was found that diluted treatments in water and saline solution did not suffer great variations showing pH values close to the initial value. However, for both cases inorganic treatment had significant differences from STF and STC and a slight decrease of 2.1% from the initial value. In terms of conductivity, there was a growth between 20% and 30% for the treatment of SAL in the different dilutions. The results of physical properties for water-diluted and saline-diluted treatments show that the best conditions occur when treatments undergo 100 rpm and a concentration of 40 mg/L.

**Table II.** Effect of factors assessed on the physicochemical properties of water.

	Factor	Level	Turbidity (UNT)	Color (CU)	pH	Conductivity ( $\mu\text{S}/\text{cm}$ )
dilution in water	Treatments	SAL	$5,025 \pm 2,3^a$	$8,95 \pm 3,9^a$	$6,81 \pm 0,6^a$	$141,01 \pm 1,5^a$
		STF	$71,03 \pm 2,8^b$	$18,16 \pm 2,8^b$	$7,01 \pm 0,3^b$	$99,65 \pm 2,3^b$
		STC	$103,40 \pm 2,1^c$	$22,32 \pm 5,7^b$	$7,04 \pm 0,5^b$	$106,24 \pm 2,5^b$
	stirring speed (rpm)	100	$53,94 \pm 1,2^a$	$17,72 \pm 0,3^a$	$6,95 \pm 0,5^a$	$116,66 \pm 1,3^a$
		200	$72,36 \pm 0,6^b$	$15,85 \pm 0,7^a$	$6,93 \pm 0,4^a$	$109,29 \pm 0,9^a$
	Dosage (mg/L)	35	$64,56 \pm 0,8^a$	$17,72 \pm 1,4^a$	$6,95 \pm 0,2^a$	$119,66 \pm 1,8^a$
40		$61,75 \pm 1,0^a$	$15,83 \pm 2,1^a$	$6,93 \pm 0,7^a$	$106,29 \pm 1,4^b$	
salt dilution	Treatments	SAL	$2,18 \pm 2,3^a$	$8,08 \pm 3,9^a$	$5,89 \pm 0,6^a$	$153,25 \pm 1,5^a$
		STF	$47,12 \pm 2,8^b$	$13,61 \pm 2,8^b$	$7,07 \pm 0,3^b$	$107,55 \pm 2,3^b$
		STC	$71,40 \pm 2,1^c$	$17,75 \pm 5,7^b$	$7,01 \pm 0,5^b$	$104,42 \pm 2,5^b$
	stirring speed (rpm)	100	$45,34 \pm 1,2^a$	$12,36 \pm 0,3^a$	$6,95 \pm 0,5^a$	$121,76 \pm 1,3^a$
		200	$63,27 \pm 0,6^b$	$14,78 \pm 0,7^a$	$6,95 \pm 0,4^a$	$119,90 \pm 0,9^a$
	Dosage (mg/L)	35	$61,86 \pm 0,8^a$	$15,14 \pm 1,4^a$	$6,95 \pm 0,2^a$	$121,68 \pm 1,8^a$
40		$58,95 \pm 1,0^a$	$13,83 \pm 2,1^a$	$6,97 \pm 0,7^a$	$110,21 \pm 1,4^b$	

**Note:** the lowercase letters correspond to the Tukey test and those that share an equal letter indicate that there is no statistically significant difference.

Within the physicochemical parameters evaluated, the removal of turbidity is explained by the behavior of the coagulation mechanism associated with natural coagulants, which are mainly adsorption and neutralization of loads, as well as adsorption and bonding between particles [9]. These types of mechanism are related to the attraction of two ion particles with opposite charge, while a bridge occurs between particles when the coagulant provides a polymer chain of suction particles [10]. Tamarind seed is polymerically loaded with carbohydrates and proteins. In the latter we can find mainly aliphatic amino acids such as glycine and leucine which are not soluble in water and water-soluble amino acids such as glutamic and aspartic acid [11]. In research carried out by [12] the presence of heterogeneous polysaccharides in tamarind with a high content in galactose, mannose and glucose, which, when in contact with water, swell and form highly viscous solutions that can allow particle agglomeration. The majority components of tamarind seed share similarity to what was reported by [13] in oily Moringa, where the author discloses that the aforementioned compounds are the possible components responsible for the removal of turbidity.

Samples showed an improvement in color by applying the different treatments as described in the results, this improvement could be due to increased removal of suspended organic and inorganic matter and microscopic organisms that are also responsible for the turbidity of water [14]. pH is one of the most important factors affecting the performance of the coagulation and flocculation system, directly influences the removal mechanism [15]. The pH values as described in the research results make it known that the biocoagulants obtained values around pH 7, while the pH of aluminium sulfate treatment (SAL) had values below the natural STF and STC treatments. The difference of this factor influences clotting activity because the load of the polyelectrolyte molecule depends on it, as well as the coagulation process [16]. The pH effect also correlates with the protein solubility of the active compounds present that is affected by the hydrophilic and hydrophobic balance of the system [17]. Additionally, research conducted by [18] discloses that when the pH is less than 6 the surface charge of tamarind powder is positive and that above this tamarind seed value would have a negative net load.

Conductivity was not significantly altered for STF and STC treatments, whereas for inorganic treatment (SAL) there was a significant increase due to the metallic nature of aluminum sulfate. The physicochemical properties in general terms for biocoagulant were more optimal for cold-obtained treatment than hot, this could be because the temperature destabilized the protein structure of the seed thus decreasing the active sites that could have been used for the coagulation process. Similarly the results were more satisfactory when diluting the samples in saline than in water. These results are consistent with those reported by [19]. Where it states that dilution of samples in NaCl accelerates the separation of protein interactions and the solubility of proteins due to increased ion strength.

### *Coagulating activity*

Figure 1 shows the treatment-concentration interactions in which it is possible to identify that aluminium sulfate (SAL) exhibited increased clotting activity compared to organic treatments when applying different doses, obtaining 86% with a dose of 40mg/L and 77% with 35 mg/L. In terms of organic treatments, STF had a higher coagulating activity ranging from 61% to 52%, while STC showed the lowest coagulating activity values between 36% and 27%. The variation in the percentage of coagulating activity is strongly related to the applied dose, with 40 mg/L being the best concentration. Saline solution treatments had a significant increase in coagulation activity (figure 2), identifying that aluminium sulfate (SAL) has a higher clotting activity compared to organic treatments when applying the different doses, getting 93% with a dose of 40mg/L and 85% with 35 mg/L. As for organic treatments, STF had a higher coagulating activity ranging from 76% to 85%, while STC yielded the lowest coagulating activity values between 51% and 59%. The variation in the percentage of coagulating activity is strongly related to the applied dose, with 40 mg/L being the optimal dose.

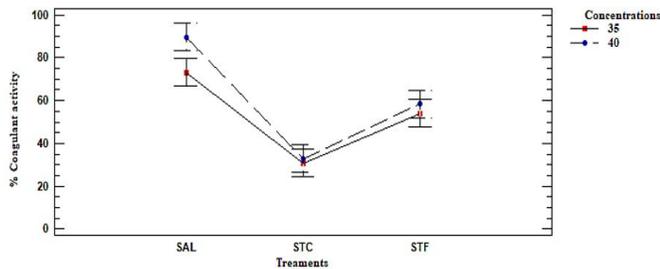


Figure 1. Graph of treatment-dose relationships for the evaluation of coagulating activity.

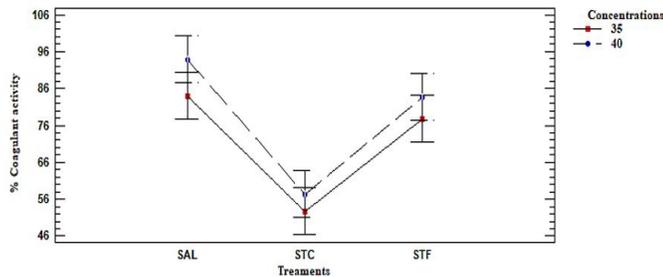


Figure 2. Graph of treatment-dose ratios for the evaluation of coagulant activity in saline solution.

The coagulating activities of polymeric treatments are associated with the structural nature of the long chain, which confers high molecular weights, leading to a large increase in the number of adsorption sites available [20]. research carried out by [21] publicize sizing through FTIR spectra of the existence of hydroxyl, amino and carboxylic acid powder groups of tamarind seeds. The presences of these groups can destabilize colloidal particles by increasing ion force and absorbing opposite-loading ions into assessed water samples [22]. It has been shown that tamarind seed has anionic behavior that depends on pH variation, as shown [23] where it discloses the z-potential value of -35 mV at pH of 7 in indica tamarind seeds. Similarly [7] reports that the potential Z in aqueous solution is -25.2 mV. Coagulation activity is due to the mechanism of polymer bridge formation during the coagulation process, anionic polymers provides the number of active sites available for surface bonding of particles found in water samples, due to the particle collision promoted by the stirring process [24].

## Conclusion

Within the coagulation process evaluated the best values of turbidity removal and coagulating activity were reported by inorganic treatment. However, bio coagulants from tamarind seeds proved to be a high-potential alternative for primary surface water treatment. Specifically the cold

extracted tamarind seed coagulant, which showed better efficiency than hot extracted with turbidity removal values between a range of 56.6% in aqueous solution and 75% in saline, with a coagulating activity of 61% and 85% respectively when applying the optimal dose corresponding to 40 mg/L. While hot treatment obtained a maximum coagulating activity of 59% in saline. In addition, it was demonstrated that the application of such natural coagulants does not significantly alter variables such as color, pH and electrical conductivity while inorganic treatment yes.

## References

- [1] Rodiño Arguello, J. P., Feria Diaz, J. J., Paternina Uribe, R., & Marrugo Negrete, J. L. Sinú River raw water treatment by natural coagulants, *Revista Facultad de Ingeniería*, vol. 76, pp. 90-98. 2015.
- [2] Olivero Verbel, R. E., Mercado Martínez, I. D., & Montes Gazabón, L. E. Removing turbidity from Magdalena river by the use of opuntia ficus-indica cactus mucilage. *Producción + Limpia*, vol. 8, no. 1, pp. 19-27. 2013.
- [3] Bondy, S. C. The neurotoxicity of environmental aluminum is still an issue. *NeuroToxicology*, vol. 31, no. 5, pp. 575-581. 2010.
- [4] Poddar, S., Talukder, G., & Sharma, A. Chromosome Damage Induced by Ferric Chloride in Human Peripheral Lymphocytes. *International Journal of Human Genetics*, vol. 4, no. 4, pp. 261-264. 2004.
- [5] Flaten, T. p. Aluminium as a risk factor in Alzheimer's disease, with emphasis on drinking water. *Brain Research Bulletin*, vol. 55, no. 2, pp. 187-196. 2001.
- [6] Saritha, V., Srinivas, N., Srikanth V. Analysis and optimization of coagulation and flocculation process. *Applied Water Science*, vol. 7, no. 1, pp. 451-460. 2017.

- [7] Ramírez, L. I. Evaluación de las semillas de tamarindo (*tamarindus indica*) como coagulante para disminuir la carga contaminante en el tratamiento de aguas, en relación a un coagulante comercial. Universidad Politécnica Salesiana. Cuenca: Trabajo de pregrado para obtener el título de Ingeniería en biotecnología de los recursos naturales. 2019.
- [8] Carrasquero, S., Martínez, M. F., Castro, M., López, Y., Díaz, A., & Colina, G. remoción de turbidez usando semillas de *tamarindus indica* como coagulante en la potabilización de aguas. *Revista Bases de la Ciencia*, vol. 4, no. 1, pp. 19-44. 2019.
- [9] Choy, S. Y., Prasad, K. M. N., Wu, T. Y., Raghunandan, M. E., & Ramanan, R. N. Utilization of plant-based natural coagulants as future alternatives towards sustainable water clarification. *Journal of Environmental Sciences*, vol. 26, no. 11, pp. 2178–2189. 2014.
- [10] Feria Díaz, J. J., Rodiño Arguello, J. P., & Gutiérrez Ribon, G. E. Behavior of turbidity, pH, alkalinity and color in Sinú River raw water treated by natural coagulants. *Revista Facultad de Ingeniería*, no. 78, pp. 119-128. 2016.
- [11] Hernández, B. M. Semillas de tamarindo (*Tamarindus indica*) como coagulante en aguas con alta turbiedad. *REDIELUZ*, vol. 3, no. 1, pp. 91-96. 2013.
- [12] Gurdían López, R., & Coto Campos, J. M. Estudio preliminar del uso de la semilla de tamarindo (*Tamarindus indica*) en la coagulación-floculación de aguas residuales. *Tecnología en Marcha*, vol. 24, no. 2, pp. 18-26. 2010.
- [13] Campos, J., Colina, G., Fernández, N., Torres, B., Sulbarán, & Ojeda, G. Caracterización de agentes coagulante activo de semillas de moringa oleifera mediante HPLC. Maracaibo, Venezuela: Universidad de Zulia, *Boletín del Centro de Investigaciones Biológicas*, vol. 37, no. 1. 2003.
- [14] Shilpa, A., & Kavita, G. Evaluation of Cactus and Hyacinth Bean Peels as Natural Coagulants. *International Journal of Chemical and Environmental Engineering*, vol. 3, no. 3, pp. 187-191. 2012.
- [15] Lek, B. L., Peter, A. P., Chong, K. H., Ragu, P., Vasanthi, S., Anurita, S., & Senthil K. A. 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. *Journal of Environmental Chemical Engineering*, vol. 6, no. 5, pp. 6243-6255, 2018.
- [16] Antov, M. G., Marina B. Šćiban, J. M., Prodanović, D. V., Kukić, V. M., Vasić, T. R., & Đorđević, M. M. Common oak (*Quercus robur*) acorn as a source of natural coagulants for water turbidity removal. *Industrial Crops & Products*, vol. 117, pp. 340-346. 2018.
- [17] Yang Teh, CY, Wu, TY & Juan, JC. Optimization of agro-industrial wastewater treatment using unmodified rice starch as a natural coagulant. *Industrial Crops and Products*, vol. 56, pp. 17–26. 2014.
- [18] Agarwal, G. S., Bhuptawat, H. K., & Chaudhari, S. Biosorption of aqueous chromium(VI) by *Tamarindus indica* seeds. *Bioresource Technology*, vol. 97, no. 7, pp. 949–956. 2006.
- [19] Okuda, T., Baes, A. U., Nishijima, W., & Okada, M. Isolation and characterization of coagulant extracted from moringa oleifera seed by salt solution. *Water Research*, vol. 35, no. 2, pp. 405–410. 2001.
- [20] Chun-Yang, Y. Emerging usage of plant-based coagulants for water and wastewater treatment. *Process Biochemistry*, vol. 45, no. 9, pp. 1437-

1444. 2010.

- [21] Buenaño, B., Vera, E., & Aldás, M. B. Study of coagulating/flocculating characteristics of organic polymers extracted from biowaste for water treatment. *Ingeniería e Investigación*, vol. 39, no. 1, pp. 24-35. 2019.
- [22] Özacar, M., & Şengil, İ. Evaluation of tannin biopolymer as a coagulant aid for coagulation of colloidal particles. *Colloids and Surfaces. Physicochemical and Engineering Aspects*, vol. 229, pp. 85-96. 2003
- [23] Aziz, H., Yii, Y., Syed Zainal, S., Ramli, S., & Akinbile, C. Effects of using *Tamarindus indica* Seeds as a natural coagulant aid in landfill leachate treatment. *Global NEST Journal*, vol. 20, no. 2, pp. 373-380. 2018.
- [24] Sethu, V., Selvarajoo, A., Wei, L. C., Ganesan, P., Lim, G. S., & Yuan, M. X. *Opuntia cactus* as a novel bio-coagulant for the treatment of Palm Oil Mill Effluent (POME). *Progress in Energy and Environment*, vol. 9, pp. 11-26.