Moringa oleifera L. CAKE AS A POTENTIAL SUBSTITUTE FOR ALUMINUM SULPHATE COAGULANT FOR THE TREATMENT OF SURFACE WATER

Jose Renato Robles Padilla

Federal University of Lavras (UFLA), Agricultural Engineering Department, Campus, P. O. Box 3037, ZIP Code 37,200-000, Lavras, MG, Brazil

Ednilton Tavares de Andrade

Federal University of Lavras (UFLA), Agricultural Engineering Department, Campus, P. O. Box 3037, ZIP Code 37,200-000, Lavras, MG, Brazil

Bárbara Lemes Outeiro Araújo

Federal University of Lavras (UFLA), Agricultural Engineering Department, Campus, P. O. Box 3037, ZIP Code 37,200-000, Lavras, MG, Brazil

Paula de Almeida Rios

Federal University of Lavras (UFLA), Agricultural Engineering Department, Campus, P. O. Box 3037, ZIP Code 37,200-000, Lavras, MG, Brazil

Abstract: Chemicals in water treatment are efficient, but they can bring problems to health and the environment, in addition to the possibility of corroding distribution pipes. *Moringa oleifera* L. grains contain a cationic protein that can be used in water treatment. The objective of the present work was to compare the efficiency of different dosages of the natural coagulant based on *Moringa oleifera* L. cake with the chemical coagulant based on aluminum sulphate. The preparation of the natural coagulant was done by saline extraction with NaCl, and the chemical coagulant was at a concentration of 5% m/v. The Jar Test was performed with different dosages of natural coagulant and with a single dosage of chemical coagulant to evaluate the efficiency in removing the turbidity and color parameters, in addition to verifying the pH variation. Treatments T1 (chemical coagulant), T4, T5 and T6 (natural coagulant) were statistically similar in terms of turbidity removal efficiency. For color, treatments T1, T5 and T6 were statistically similar. In conclusion, the natural coagulant prepared from *Moringa oleifera* L. cake can be a great alternative in water treatment, and can be improved.

Index terms: Water treatment, natural coagulant, chemical coagulant, by-products.

Received: April 28, 2023 - Accepted: May 3, 2023

INTRODUCTION

The growing demand for quality water to supply the population and the deterioration of water resources, worries and requires the search for new technologies to treat this resource (Neves & Baiardi, 2023). According to the UN (2022) in its report, by the year 2050 the consumption of drinking water will grow by around 20 to 30%. An amount of 80% of wastewater is returned to the environment without being treated (WWAP, 2015).

According to Sillanpää et al. (2018) the coagulants generally used in the coagulation process are inorganic such as aluminum or iron

salts, these in solution are dissociated in their trivalent ionic form (Al³⁺ and Fe³⁺), hydrolyzed and forming complexes with a positive charge that interact with negative charges of colloids. The coagulant allows the particles to associate through reducing the attractive forces of the colloidal suspension as a consequence of the lowering of the energy barrier (Metcalf and Eddy, 2012).

In water treatment station (WTS) aluminum sulfate is the most used chemical coagulant, due to its low cost, good efficiency and easy handling. In addition, this coagulant is used as a standard for comparing the efficiency of new coagulants (Seneda, Garcia and Reis, 2021).

A concern that arises with the use of inorganic coagulants based on aluminum and iron is the environmental impacts that the residual sludge can cause. These residues are rich in non-biodegradable metallic hydroxides, which must be treated for their correct environmental disposal (Skoronski et al., 2014). On the other hand, residual aluminum in treated water that is transported by the distribution network can cause damage to health, correlating this metal with neurodegenerative diseases such as Parkinson's and Alzheimer's (Driscoll and Letterman, 1988; Banks et al., 2006). Chemicals are a solution for water treatment, but there is a need to find biodegradable, economical products that can replace current solutions (Watanabe et al., 2016; Grenda et al., 2020).

Moringa oleifera is a plant native to India that is well adapted to conditions in Brazil (Azevedo et al., 2020). According to Gallão, Damasceno and Brito (2006) the seed is mainly composed of lipids and proteins. *Moringa oleifera* seeds have positively charged proteins which allow it to attract negatively charged particles such as clays, silts, among others, forming clots/ floccles that later settle (Amagloh and Benang, 2009). This protein allows the reduction of the amount of microorganisms and suspended particles, resulting in the removal of turbidity, color and total coliforms without changing the pH (Oliveira et al., 2018).

The objective of this work is to compare the efficiency of the natural coagulant of *Moringa oleifera* cake with the chemical coagulant Aluminum sulphate in waters from the catchment reservoir of the Federal University of Lavras.

MATERIAL AND METHODS

The grains of *Moringa oleifera* Lam. were collected in an area belonging to the Agricultural Engineering department at UFLA after they reached the point of physiological maturity according to (Agustini et al., 2015) in the second half of 2022. The oil extraction was a cold extraction through a Scott Tech expeller type mechanical press of the ERT 50 model, the grain was passed through the press and then the cake

was passed twice more to extract the largest amount of oil.

The water content (%) of the grains was determined (Brasil, 1992) by the oven method at 105 ± 3 °C for 24 hours. For the *Moringa oleifera* cake Lam. Bromatology was performed according to Detmann et al. (2021) and powder cake granulometric characterization according to Santana et al. (2010): 6 sieves with meshes of 1.00 mm, 0.500 mm, 0.425 mm, 0.210 mm, 0.150 mm and 0.074 mm were used.

To obtain the natural coagulant, the cake of Moringa oleifera Lam was used. As in Roveli et al. (2021) the extract solution was prepared at room temperature by mixing 10.816 g of sodium chloride (NaCl, Synth) in 200 ml of distilled water and 20.000 g of Moringa oleifera cake. They were mixed in a blender for 3 minutes and then placed on a magnetic stirrer for 30 minutes, finally filtered through a qualitative filter using vacuum filtration. The determination of total soluble protein in natural coagulant (mg mL-¹) was done using the Bradford method (1976). The aluminum sulfate coagulating solution was prepared at 5 % (m/v). Add 5g of aluminum sulfate to the powder in 100 mL of distilled water, to finally be mixed in a magnetic stirrer until complete dissolution.

The raw water sample was collected at the UFLA Water Treatment Station (ETA), the sample was taken to the Environmental Engineering and Sanitation Laboratory belonging to the same university where it was determined turbidity, being low, approximately 7.0 NTU. For 20 liters of raw water, add 305.78 g of soil from the edge of the reservoir and mixed with raw water, increase turbidity and take that prepared water as the initial plot.

It was set up in a completely randomized design (DCA) with two replications. In the first week all treatments were tested, for the second week the treatments with greater efficiency in removing the parameters of turbidity and color were tested.

The Jar Test apparatus was used with six reactors, in each reactor water was placed to treat in an amount of 1000 mL and then the coagulants were placed in predetermined doses in each of them. For the fast mix was used 120 RPM for 5 minutes, the slow mixing was used, 45 RPM was used for 20 minutes, with the aim of consolidating the coagulation/flocculation and to decantation time was used 30 minutes.

The treatments were, T0: control, T1 chemical coagulant aluminum sulfate in a dose of 5.0 mL, T2, T3, T4, T5 and T6 natural coagulant of saline extract of moringa pie in doses of 2.5, 5, 7.5, 10 and 12.5 mL, respectively. Response variables were determined as in Franco et al. (2017): Turbidity (measurement method: SMWW 2130 B); Color (measurement method: SMEWW 2120 B); pH (measurement method: SMWW 4500 H+B).

Removal efficiency was determined as done in Saltos et al. (2022), using the following equation:

$$E = \frac{S \text{ initial} - S \text{ final}}{S \text{ inicial}} x100$$

Where, E: Removal efficiency (%), $S_{initial}$: Load of contaminant before treatment, S_{final} : Load of contaminant after treatment.

The data was analyzed with the computer program SISVAR version 5.8, comparisons of means using the Tukey test at the significance level of 5%.

RESULTS AND DISCUSSION

Table 1 presents the results of the bromatology of Moringa cake after oil extraction by mechanical press. Results are on a dry matter (DM) basis.

Araújo et al. (2020), evaluating the drying effect at different temperatures of *Moringa oleifera* L. grains, determined the bromatology of the cake, obtained the following results; water content of 0.0703 b.s., ether extract of 8.03%, neutral detergent fibers of 38.60%, protein of 35.55% and mineral material of 4.82%.

Figure 1 shows the granulometry of the Moringa cake, characterized by the mass in grams retained in each sieve.

Santana et al. (2010), characterized the moringa grain physically and chemically, as well as the granulometry of the moringa cake, using

126.636 grams for sieving. In that work, sieves with dimensions of 1.180 mm, 0.590 mm, 0.420 mm, 0.250 mm, 0.149 mm, 0.074 mm were used, being retained in each 22.349 g, 32.224 g, 17.074 g, 15.801 g, 18.790 g and 17.492 g respectively.

Figure 2 shows the variation in total soluble protein concentration in mg/mL over 4 days since the preparation of the natural coagulant based on *Moringa oleifera* L.

Madrona et al. (2010), evaluating the coagulant of *Moringa oleifera* L. extracted with NaCl at different concentrations, determined the total soluble protein by the Lowry method, obtaining different protein contents in mg.L⁻¹. 0.01 M NaCl, 0.1 M NaCl and 1 M NaCl contents of 1290.86, 4388.01, 4499.19 respectively.

After preparing the water, the average turbidity reached for the first test was 247.00 NTU of turbidity, for the second test carried out in the second week, the same procedure was followed and the average turbidity reached the values of 243.00 NTU.

Figure 3 shows the treatments and their respective efficiencies in % in removing turbidity in the first week of the test, with treatment T0 without coagulant, T1 chemical coagulant, T2, T3, T4, T5 and T6 natural coagulant with dosages of 2.5, 5.0, 7.5, 10.0 and 12.5 mL, respectively.

It can be seen in Figure 3 that treatment T1 obtained excellent results, this treatment which achieved greater efficiency does not differ statistically from treatments T4, T5 and T6. Confirming that the natural coagulant based on the *Moringa oleifera* L. cake can be an interesting substitute for the chemical coagulant.

Meza-Leones et al. (2018) compared the efficiency of the aluminum sulfate coagulant with the *Moringa oleifera* seed coagulant in lowering the turbidity parameter in Malambo Ciénaga's waters. For this, they used the Jar Test and water samples with an initial turbidity of 56.5 NTU. They concluded that the chemical coagulant has an efficiency of 96% and the natural one 64%, the latter being an interesting alternative.

Table 1: Bromatology of Moringa oleifera L. cake - Results in Dry Matter

Dry matter	Water content	Crude Protein	Ethereal Extract	Fiber ND	Mineral matter
(%)	(%, wb)	(%)	(%)	(%)	(%)
92.339	7.661	29.890	18.588	47.641	4.492

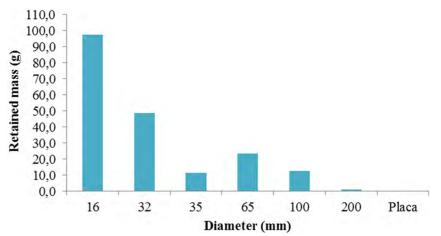


Figure 1: Granulometry of the Moringa oleifera L. cake.

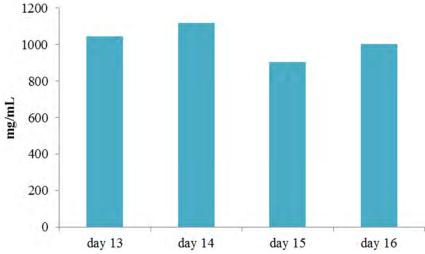


Figure 2: Variation in total soluble protein concentration in mg mL⁻¹ for September 13th, 14th, 15th and 16th.

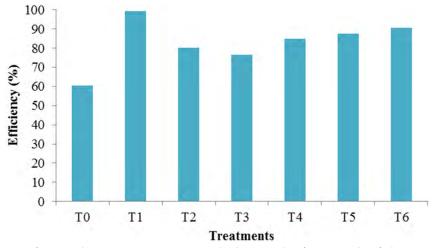


Figure 3: Efficiency of coagulants in removing turbidity in the first week of the test.

According to Okuda et al. (1999), the "salting in" phenomenon makes the protein more soluble, the protein becomes more soluble in the presence of salts because of an increase in the ionic strength caused by the charges at the root of the dissociation of the salt, which begin to interact with the protein molecules. According to Katalo et al. (2018), the greater efficiency of aluminum sulfate is due to the formation of larger flakes with higher density, which is called sweep coagulation.

Figure 4 presents the different treatments used and the efficiency in % achieved in removing the color parameter in the first week of the test.

It can be seen in Figure 4 that the efficiency in % for T1 (chemical coagulant) was superior to the others, but statistically it did not differ from treatments T4, T5 and T6 (natural coagulant). Prasad (2009), evaluating in a pilot station the natural coagulant of *Moringa oleifera* extracted in saline solution (1M) found a removal efficiency of the color parameter of 80%.

The color in the water is due to the content of metallic ions (iron and manganese), humus, industrial waste, among others. Color removal is required prior to discharge into water courses, in addition to suiting water for different industrial or general purposes (APHA, 2012).

The pH variations are shown in the Figure 5 for the different treatments used in the first week of the test.

It can be observed that when the natural coagulant based on *Moringa oleifera* L. cake is used (T2, T3, T4, T5, T6) there is no statistically significant difference when compared to the treatment without coagulant (T0). In the case of treatment with the chemical coagulant, there is a reduction in pH which differs statistically from the other treatments. Roveli et al. (2021) evaluating the coagulant function of the saline extract of *Moringa oleifera* with aluminum sulfate in textile effluents found a maintenance of the pH for the natural coagulant and a reduction for the latter from 9.00 to 5.73 when added to the dosage of 10 mL of prepared chemical.

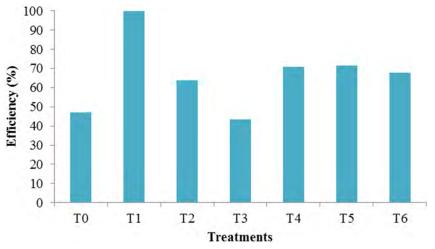


Figure 4: Efficiency in % in removing the color parameter for the different treatments in the first week of the test.

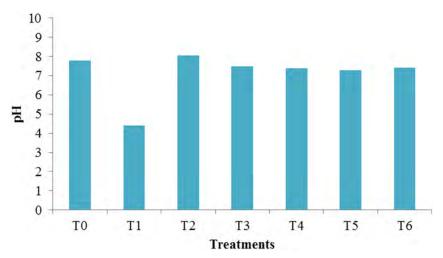


Figure 5: pH variation for different treatments in the first week of the test.

According to Mageshkumar and Karthikeyan (2015), the use of *Moringa oleifera* L. coagulant in the water treatment plant does not demand an additional cost for pH correction as compared to chemical coagulants.

The different treatments used and the efficiency in % achieved in removing the turbidity parameter in the second week of the test is shown in the Figure 6.

According to Pritchard et al. (2010), *Moringa oleifera* grains are highly efficient in removing turbidity for a storage period of 18 months, but for 24 months the coagulating power becomes inefficient. It is recommended to use freshly harvested beans.

The different treatments used and the efficiency in % achieved in removing the color

parameter in the second week of the test is shown in the Figure 7.

Valverde et al. (2014), evaluated the degradation time of *Moringa oleifera* L. coagulant powder, which was stored in a refrigerator at a temperature of 4 °C. The author applied the dosage of 50 mg.L⁻¹ of the powder. As a result, he obtained a decrease in the efficiency of removing the turbidity parameters over the course of one week after storage and for the color parameter over the course of two weeks. According to Katayon et al. (2006), the coagulant based on *Moringa oleifera* tends to lose efficiency over time and even tends to increase turbidity.

The different treatments used and the pH variation in the second week of the test is shown it the Figure 8.

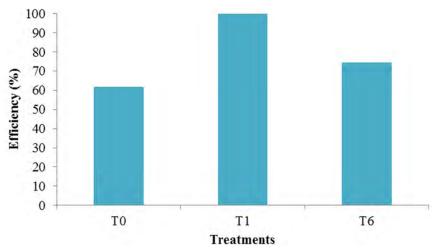


Figure 6: Efficiency in % in removing the turbidity parameter for the different treatments in the second week of the test.

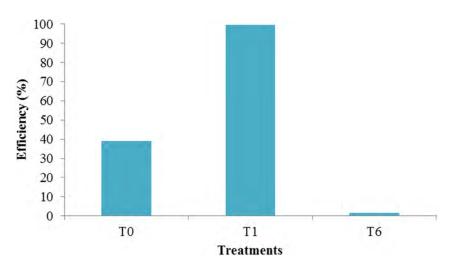


Figure 7: Efficiency in % in removing the color parameter for the different treatments in the second week of the test.

It can be seen in Figure 8 that in the pH parameter for evaluation of the second week and test, treatment T1 continues to decrease the pH being statistically different from treatments T0 and T6 which remained in the range of 7.780 to 9.165.

In the statistical analysis of the First week and test for the three studied parameters; turbidity, color and pH, there was a significant difference at 5%, comparisons of means were made using the Tukey test, which is shown in Table 2.

Thus treatment T1 (Chemical coagulant) T6, T5 and T4 (Natural coagulant; 12.5, 10.0, 7.5 mL respectively) are superior in removing turbidity as well as color for the first week and evaluation test. In the case of pH, the T1 treatment was different from the others, causing this parameter to decrease.

In the statistical analysis of the second week and test for the three studied parameters; turbidity, color and pH, there was a significant difference at 5%, comparisons of means were made using the Tukey test, which is shown in Table 3.

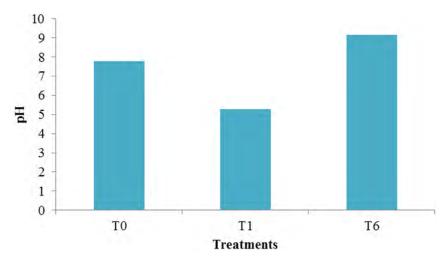


Figure 8: pH variation for different treatments in the second week of the test.

1	0 7		
Treatments	Turbidity Removal Efficiency (%)	Color Removal Efficiency (%)	pН
T1	99.1845 A	99.8650 A	4.3900 A
Т6	90.6075 A	67.9730 A	7.4100 B
Τ5	87.5505 A	71.3515 A	7.2700 B
T4	84.8585 A	71.0135 A	7.3900 B
Τ2	80.0610 B	63.7840 B	8.0650 B
Т3	76.7005 C	43.2430 B	7.4700 B
ТО	60.3645 C	47.0270 B	7.7800 B

Table 2: Comparisons of means using the Tukey 5% test.

Means followed by the same capital letter in the column do not differ according to Tukey's test at the 5% significance level.

Table 3: Comparisons of means using the Tukey 5% test.

<u>1</u>	0 /		
Treatments	Turbidity Removal Efficiency (%)	Color Removal Efficiency (%)	pН
T1	99.9590 A	99.7645 A	5.2650 A
Т6	74.4035 B	1.4900 B	9.1650 B
ТО	61.9750 B	39.0585 B	7.7800 B

Means followed by the same capital letter in the column do not differ according to Tukey's test at the 5% significance level.

Thus, treatment T1 (Chemical Coagulant) is superior both in removing turbidity and color for the second week and evaluation test. In the case of pH, the T1 treatment was also different, causing this parameter to decrease.

CONCLUSIONS

In conclusion, the use of a natural coagulant based on *Moringa oleifera* L. grain cake can be a promising substitute for chemical coagulant. This could be improved for greater ease of use.

The total soluble protein remained stable in quantity for three days after the preparation of the natural coagulant based on *Moringa oleifera* L.

The coagulant based on *Moringa oleifera* L. cake loses its efficiency over time, in this case, after a week, the cationic protein causing the coagulant effect lost its working efficiency.

The characterization of the grains in terms of water content and the cake in terms of granulometry and bromatology allows an idea of the conditions that can be used to improve future results.

REFERENCES

AGUSTINI, M. A. B. et al. Maturidade fisiológica de sementes de *Moringa oleifera* Lam. **Revista Inova Ciência & Tecnologia**, v.8, p. 267-278, 2015.

AMAGLOH, F. K.; BENANG, A. Effectiveness of Moringa oleifera seed as coagulant for water purification. **African Journal of Agricultural Research**, v. 4, n. 2, p. 119-123, 2009.

ARAÚJO, B. L. O. et al. Chemical prospection of moringa oil and bromatological quality of the pie from different types of grain processing. **Research**, **Society and Development**, v. 9, n. 11, p. e82291110599-e82291110599, 2020.

AZEVEDO, M. M. R. et al. Características de silagens de capim-elefante (Pennisetum purpureum Schum.) com níveis de inclusão de moringa (*Moringa oleífera* Lam.). **Brazilian Journal of Development**, v. 6, n. 9, p. 71418-71433, 2020.

BANKS, W. A. et al. Aluminum complexing enhances amyloid β protein penetration of blood-brain barrier. **Brain Research**, v. 1116, n. 1, p. 215-221, 2006.

BRADFORD, M.M. A Rapid and Sensitive Method for the Quantitation of Microgram Quantities of Protein Utilizing the Principle of Protein Dye Binding. **AnalyticalBiochemistry** 72, 248-254. 1976.

BRASIL. Ministério da Agricultura e Reforma Agrária. **Regras para análise de sementes.** Brasília: SNDA/DNDV/CLAV, 1992.

DETMANN, E. et al. **Métodos para Análise de Alimentos.** 2.ed. Visconde do Rio Branco, MG: Suprema. 350p. 2021.

DRISCOLL, C. T.; LETTERMAN, R. D. Chemistry and fate of Al (III) in treated drinking water. **Journal of Environmental Engineering**, v. 114, n. 1, p. 21-37, 1988.

FRANCO, C. S. et al. Coagulação com semente de moringa oleifera preparada por diferentes métodos em águas com turbidez de 20 a 100 UNT. **Engenharia Sanitária e Ambiental**, v. 22, p. 781-788, 2017.

GALLÃO, M. I.; DAMASCENO, L. F.; BRITO, E. S. (d). Avaliação química e estrutural da semente de moringa. **Revista Ciência Agronômica**, v. 37, n. 1, p. 106-109, 2006.

GRENDA, K. et al. Flocculation of silica nanoparticles by natural, wood-based polyelectrolytes. **Separation and Purification Technology**, v. 231, p. 115888, 2020.

KATALO, R. et al. Moringa oleifera coagulation as pretreatment prior to microfiltration for membrane fouling mitigation. **Environmental Science: Water Research & Technology**, v. 4, n. 10, p. 1604-1611, 2018.

KATAYON, S. et al. Effects of storage conditions of Moringa oleifera seeds on its performance in coagulation. **Bioresource technology**, v. 97, n. 13, p. 1455-1460, 2006.

MADRONA, G. S. et al. Study of the effect of saline solution on the extraction of the Moringa oleifera seed's active component for water treatment. **Water**, **Air**, **& Soil Pollution**, v. 211, n. 1, p. 409-415, 2010.

MAGESHKUMAR, M.; KARTHIKEYAN, R. Modelling the kinetics of coagulation process for tannery industry effluent treatment using Moringa oleifera seeds protein. **Desalination and Water Treatment**, v. 57, n. 32, p. 14954-14964, 2016. METCALF; EDDY. Inc. **Wastewater Engineering treatment Disposal Reuse**. 6. ed. New York, McGraw - Hill Book, 2012.

MEZA-LEONES, M. et al. Evaluación del poder coagulante del sulfato de aluminio y las semillas de Moringa oleífera en el proceso de clarificación del agua de la ciénaga de Malambo-Atlántico.**Revista UIS Ingenierías**, v. 17, n. 2, p. 95-104, 2018.

NEVES, V. S.; BAIARDI, A. Vicissitudes no acesso à água potável em municípios da Região Metropolitana deSalvador/BA.**Research,Society and Development**, v. 12, n. 4, p. e21012432922-e21012432922, 2023.

OKUDA, T. et al. Improvement of extraction method of coagulation active components from Moringa oleifera seed. **Water research**, v. 33, n. 15, p. 3373-3378, 1999.

OLIVEIRA, N. T. et al. Tratamento de água com Moringa oleifera como coagulante/floculante natural. **Revista Científica da Faculdade de Educação e Meio Ambiente**, v. 9, n. 1, p. 373-382, 2018.

UN (UNITED NATIONS). **WATER.** 2022. Disponível em: https://unric.org/pt/agua/. Acesso em 18 de jan. 2022.

PRASAD, R. K. Color removal from distillery spent wash through coagulation using Moringa oleifera seeds: Use of optimum response surface methodology. Journal of hazardous materials, v. 165, n. 1-3, p. 804-811, 2009.

PRITCHARD, M. et al. A study of the parameters affecting the effectiveness of Moringa oleifera in drinking water purification. **Physics and Chemistry of the Earth**, Parts A/B/C, v. 35, n. 13-14, p. 791-797, 2010.

ROVELI, A. R. et al. Utilização de extrato de semente de Moringa oleifera no tratamento de efluente têxtil. **Revista Geama**, v. 7, n. 1, p. 67-72, 2021.

SALTOS, H. T. V. et al. Remoción de contaminantes en aguas residuales mediante el polielectrolito catiónico extraído de las semillas de Moringa oleífera. **Ingeniería Hidráulica y Ambiental**, v. 43, n. 2, p. 84-96, 2022.

SANTANA, C. R. et al. Caracterização físico-química da moringa (*Moringa oleifera* Lam). **Revista Brasileira**

de Produtos Agroindustriais, Campina Grande, v. 12, n. 1, p. 55-60, 2010.

SENEDA, R. M.; GARCIA, G. F.; REIS, A. G. D. Flocculation kinetics: a comparative study on the use of polyaluminium chloride with high and low basicity and alum. **Engenharia Sanitaria e Ambiental**, p. 283-290, 2021.

SILLANPÄÄ, M. et al. Removal of natural organic matter in drinking water treatment by coagulation: A comprehensive review. **Chemosphere**, v. 190, p. 54-71, 2018.

SKORONSKI, E. et al. Estudo da aplicação de tanino no tratamento de água para abastecimento captada no rio Tubarão, na cidade de Tubarão, SC. **Revista Ambiente & Água**, v. 9, p. 679-687, 2014.

VALVERDE, K. C. et al. Avaliação do tempo de degradação do coagulante natural moringa oleifera lam em pó no tratamento de água superficial. **e-xacta**, v. 7, n. 1, p. 75-82, 2014.

WATANABE, R. et al. Efficient performance and the microbial community changes of submerged anaerobic membrane bioreactor in treatment of sewage containing cellulose suspended solid at 25 C. **Bioresource Technology**, v. 216, p. 128-134, 2016.

WWAP. United Nations World Water Assessment Programme. 2015. The United Nations World Water Development Report 2015: **Water for a Sustainable World.** Paris, Unesco. Disponível em: https:// unesdoc.unesco.org/ark:/48223/pf0000231823. Acesso em: 23 jul. 2022.