



Natural agents as auxiliaries in water clarification: literature review and experimental evaluation

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ABSTRACT. The increasing demand for water resources (primarily for human consumption and industrial and agricultural activity) is driven by socio-economic development, and population growth. Recent research has been focusing on alternative coagulants based on natural elements, as opposed to the commonly used iron and aluminum salts, for use in water and wastewater treatment processes. In this context, a thorough literature review on alternative coagulants and their application to water treatment processes has been conducted in this work. In addition, three experiments have been conducted with a widely used natural organic coagulant (*Moringa oleifera*). The alternative clarification system used is the helically coiled tube flocculator (HCTF), with high turbidity removal efficiency and low processing times. A comparative analysis of the turbidity reduction over time was performed with samples collected after 600, 900, 1200, 1500, 1800, 2100, 2400, and 2700 s from the hydraulic circuit. The process efficiency using the proposed alternative coagulant reached 95.3% (after 1800 s). The turbidity removal efficiency remained almost constant after 1800 s, with variations below 1%. These results prove that natural alternative agents can be powerful tools in the water treatment process, with efficiency values exceeding those obtained using chemicals (e.g., aluminum sulphate) as coagulant/ flocculant agents.

Keywords: *Moringa oleifera*; natural coagulants; natural flocculant; water treatment process.

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Introduction

Coagulation and flocculation are very important steps in water treatment processes. During coagulation, chemicals are commonly added to water in a rapid mixing stage, thereby enhancing particles destabilization and, consequently, flocs formation. The flocs formed are removed from water in the flotation/sedimentation step. The quality of the treated water should satisfy established governmental regulations, to avoid causing damages to the environment and human health (Santos, Pereira, Santana, & Silva, 2011). In this context, the development of new techniques and products to improve process efficiency becomes necessary.

In addition to this, the increasing demand for water resources and their newly proposed treatment and reuse processes, allow the development of scientific knowledge about alternative coagulants. These can be used in water treatment processes, for both industrial activity (Santos et al., 2011) and human consumption (Arantes, Paterniani, Rodrigues, Hatori, & Pires, 2015), where water resources are essential.

Oenning Junior and Pawlowsky (2007) pointed out that reuse of water resources in industrial activities is driven by the reduction in production costs, which positively affects profits. Reusing water resources also reduces maintenance and damage costs caused by impurities present in the water operated in machines and tools. Another driving factor is the reduction of environmental impact during production. Indeed, reusing water can reduce its contamination and interference with its chemical (such as pH), and physical characteristics (such as temperature) when it is finally returned to the natural environment.

According to Vargas-Camareno and Romero-Esquivel (2006), the use of iron and aluminum salts based coagulants may cause neurological diseases such as Alzheimer's disease (Bongiovani, Konradt-Moraes, Bergamasco, Lourenço, & Tavares, 2010). Llopis and Díez (2002) highlight the risks of exposure to aluminum. On the contrary, reduction in the incidence of neurological diseases within a population is observed to be correlated to alternative coagulants of vegetable origin (Li, Zhang, Hu, Gan, & Li, 2016). Therefore, natural coagulant alternatives constitute an important prevention strategy, leading to reduced impact on public health.

In the literature review, we highlight the research carried out in Latin American and Southeast Asian countries, where developing countries such as Brazil and China are located (Huang, Liu, Li, & Yang, 2017). This way, the connection between an adequate use of natural resources and sustainable development applied to the industry can be perceived, since sustainable production policies impact a company's image.

Thus, efficiency of the water treatment process and, consequently, characteristics of the treated water are important factors in defining public health and sanitation policies, as well as appropriate extraction rates of natural resources for production.

Therefore, this paper aims at performing a thorough literature review on alternative coagulants and their application to water treatment processes. In addition, three experiments have been conducted with a widely used alternative coagulant: *Moringa oleifera*. Results from the literature proved that *Moringa oleifera* can be a powerful tool in the water treatment process (Muniz, Duarte, & Oliveira, 2015; Camacho, Sousa, Bergamasco, & Teixeira, 2017; Landázuri et al., 2018).

Material and methods

The research work was undertaken between May 2017 and August 2018, using three search engines. Firstly, Google Scholar was used and the following keywords were searched for: 'tratamento com coagulantes alternativos', 'tratamento com floculantes alternativos', 'coagulante no tratamento de água', 'coagulantes naturais', and 'floculantes naturais'. Secondly, Portal de Periódicos Capes was searched for the following keywords: 'coagulantes alternativos', 'coagulantes naturais', 'coagulantes naturais em água', 'floculantes alternativos', 'floculantes naturais', and 'natural flocculants'. Lastly, Science Direct was searched for the following keywords: 'natural coagulants', and 'natural flocculants'.

Consequently, the retrieved literature was sorted by publication date (ascendant sorting), using Microsoft Excel. In the resulting table, articles were further sorted by the journal of publication, impact factor (2017), and Qualis score (considering Engenharias III area, when available; otherwise, Engenharias I area was considered). Based on the results encountered in the literature, *Moringa oleifera* was chosen as the alternative coagulant to be studied, due to its high potential for the removal of impurities (Muniz et al., 2015), and its availability in Brazil. The concentration of the *Moringa oleifera*-based coagulant was determined through experiments performed in the chemical laboratory of Federal Institute of Espírito Santo (campus Cariacica).

In the first experiment, 1×10^{-3} kg of *Moringa oleifera* (seeds in shell) and 8×10^{-5} m³ of distilled water were used to prepare the coagulant. They were blended at 157 rad s^{-1} (1500 rpm) for 60 s, as presented in Muniz et al. (2015). The obtained solution was then filtered. For the test, synthetic water was prepared mixing 5×10^{-4} m³ of natural water and 5×10^{-4} kg of clay (bentonite) with magnetic stirring. At this stage, a sample of the test water was collected (1×10^{-5} m³), for comparison purposes.

Then, 1×10^{-5} m³ of the coagulant was added to the test water and the resulting solution was mixed at 126 rad s^{-1} (1200 rpm) for 60 s. Subsequently, the rotation speed of the magnetic stirrer was reduced to 21 rad s^{-1} (200 rpm) and maintained for 600 s. Three samples were collected after 900, 1800, and 2700 s, to be compared with the test water. Before collecting samples, the used beakers were weighed. All collected samples were dried to remove the remaining water and to measure the solids present in each sample.

In the second experiment, 1×10^{-3} kg of *Moringa oleifera* (without shells) and 8×10^{-5} m³ of distilled water were used to prepare the coagulant, following the same procedure as the first experiment. Test water preparation and sample collection procedures remained unchanged, and four samples were thus prepared (one test water sample, and three proposed samples).

In the third experiment, similarly prepared coagulant solution and test water were evaluated in a hydraulic circuit. A schematic representation of the adopted hydraulic circuit is shown in Figure 1. The used flocculator was a helically coiled tube flocculator (HCTF), as presented by Oliveira and Teixeira (2017a), with a length of 10.53 m and a volume of 7.5×10^{-4} m³. This type of flocculator was chosen due to its low cost and high efficiency (Oliveira & Teixeira, 2017a; 2017b; 2018). The experiment was performed on a solution of 0.130 m^3 of test water and $1 \times 10^{-5} \text{ m}^3$ of coagulant solution.

For the third experiment, process efficiency was evaluated by observing the differences between turbidity values before and after coagulant application. To determine the initial turbidity, five samples were collected, and their average value was adopted in all comparisons. The coagulant solution was the same as that used in the first experiment (seeds with shell). After applying the coagulant, samples were collected at

the end of the hydraulic circuit. The final turbidity was measured for samples collected after 600, 900, 1200, 1500, 1800, 2100, 2400 and 2700 s.

Results and discussion

Table 1 presents the papers evaluated in this work to determine the alternative coagulant used in all experiments. From Table 1, the importance of *Moringa oleifera* can be easily highlighted, as it has been studied in more than 50% of presented papers.

Table 2 was then created to compile important information regarding the selected papers (Table 1), including journals of publication, Qualis score, impact factor (JCR), number of citations in the performed research, and percentage of citations in the performed research.

Due to the high percentage of citations in previous literature, *Moringa oleifera* was chosen to be evaluated in all experiments in this work.

Results of the first (seeds in shell) and second experiments (seeds without shells) are presented in Table 3 and 4, respectively. In these tables, a comparative analysis of bentonite mass reduction at each sample collection time is presented (after 900, 1800, and 2700 s); the comparison was done with the values for plain test water in every instance.

Data comparison shows that the highest efficiencies at all times were obtained using seeds in shell, where process efficiency reaches 78% at 2700 s. Additionally, the use of whole seeds of *Moringa oleifera* strongly contributes to two more aspects: first, it avoids the time-consuming process of removing seeds from their shells; secondly, it avoids disposal of removed shells in the natural environment.

Results of the third experiment (using the hydraulic circuit and seeds in shell) are presented in Table 5. The average value of the initial turbidity was 148 NTU, as shown for sample #1 (Table 5). Moreover, a comparative analysis of the turbidity reduction at each sample collection time is presented (after 600, 900, 1200, 1500, 1800, 2100, 2400, and 2700 s); the comparison was done with the initial turbidity value of test water in every instance.

As shown in Table 5, the efficiency obtained in the third experiment was higher than those in the first and second experiments. In the first experiment, the process efficiency peaked at 78.1%, whereas it only reached 43.8% in the second experiment. In the third experiment, however, the process efficiency reached a maximum of 95.3% (after 1800 s) and a minimum of 86.5%. Therefore, the lowest process efficiency obtained in the third experiment was higher than the best values obtained in the first two experiments. It is important to emphasize that the lowest process efficiency obtained in the third experiment (86.5%) was also higher than the highest value of process efficiency obtained by Oliveira and Teixeira (2017a) using aluminum sulphate (86.2%).

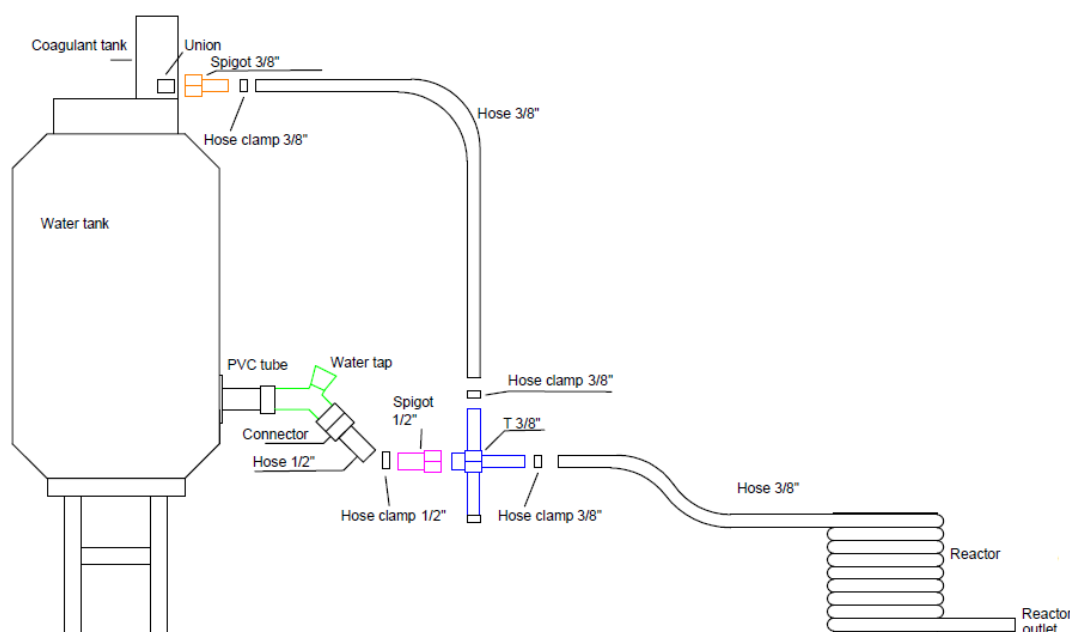


Figure 1. Schematic representation of the hydraulic circuit used in this work.

Table 1. Papers evaluated in this work.

Author (year)	Alternative coagulant applied
Díaz et al. (1999)	<i>Cactus latifaria</i> / <i>Prosopis juliflora</i>
Cruz, Oliveira, Araújo Filho, Hipólito, and Lima (2007)	<i>Moringa oleifera</i>
Cardoso, Bergamasco, Cossich, and Moraes (2008)	<i>Moringa oleifera</i>
Lédo, Lima, Paulo, and Duarte (2009)	<i>Moringa oleifera</i>
Šćiban, Klasnja, Antov, and Skrbic (2009)	Chestnuts / Acorns
Paterniani, Mantovani, and Sant'Anna (2009)	<i>Moringa oleifera</i>
Bongiovani et al. (2010)	Tanfloc SS (tannin)
Piazza, McAloon, and Garcia (2011)	Chicken blood
Santos et al. (2011)	<i>Moringa oleifera</i>
Madrona et al. (2012)	<i>Moringa oleifera</i>
Franco, Silva, and Paterniani (2012)	<i>Moringa oleifera</i>
Valverde, Moraes, Bongiovani, Camacho, and Bergamasco (2013)	<i>Moringa oleifera</i>
Lo Monaco et al. (2013)	<i>Moringa oleifera</i>
Tassinari, Doherty, and Marison (2013)	Colza (rapeseed)
Díaz, Roa, and Tordecilla (2014)	<i>Moringa oleifera</i>
Skoronski, Niero, Fernandes, Alves, and Trevisan (2014)	Tannin
Francisco, Silva, Roque, Nascentes, and Silva (2014)	<i>Moringa oleifera</i>
Martins, Oliveira, and Guarda (2014)	Chitosan, <i>Moringa oleifera</i> , and tannin
Muthuraman and Sasikala (2014)	<i>Moringa oleifera</i> / <i>Strychnos potatorum</i> / <i>Phaseolus vulgaris</i> (bean)
Arantes, Ribeiro, Paterniani, Tateoka, and Silva (2014)	<i>Moringa oleifera</i> and Tanfloc SG (tannin)
Gassara et al. (2015)	Chitin and chitosan
Arantes et al. (2015)	<i>Moringa oleifera</i>
Kukić, Šćiban, Prodanović, Tepić, and Vasić (2015)	Bean
Gutiérrez, Passos, Ferrer, Uggetti, and García (2015)	Ecotan / Tanfloc (tannin)
Lee, Chong, Robinson, and Binner (2015)	Abelmoschus esculentus (okra)
Freitas et al. (2015)	Abelmoschus esculentus (okra)
Muniz et al. (2015)	<i>Moringa oleifera</i>
Li et al. (2016)	Dextran
Kakoi, Kaluli, Ndiba, and Thiong'o (2016)	Banana
Ang, Mohammad, Benamor, and Hilal (2016)	Chitosan
Molina, Rodríguez, and Ariza (2016)	<i>Moringa oleifera</i> , <i>Cacto Opuntia</i> , seaweed, and starch
Wu, Liu, Yang, and Li (2016)	Starch
Choy, Prasad, Wu, Raghunandan, and Ramanan (2016)	Starch (Rice, wheat, corn, and potatoes)
Jia et al. (2016)	Modified Chitosan
Huang et al. (2017)	Starch
Liu, Wei, Li, and Yang (2017)	Starch
Camacho et al. (2017)	<i>Moringa oleifera</i>
Priya, Tarafdar, Gupta, and Mishra (2018)	<i>Moringa oleifera</i> and <i>Cyamopsis tetragonoloba</i>
Landázuri et al. (2018)	<i>Moringa oleifera</i>

Table 2. Journals evaluated in this work.

Journal (ISSN)	Qualis score	Impact Factor (JCR)	N. of citations	Percentage of citations
Acta Scientiarum Technology (1806-2563)	B2	0.231	4	10.31
Engenharia Agrícola (0100-6916)	B2	0.387	4	10.31
Water Research (0043-1354)	A1	7.051	3	7.69
Ecological Engineering (0925-8574)	A1	3.023	3	7.69
Revista Brasileira de Engenharia Agrícola e Ambiental (1415-4366)	B1	0.619	2	5.12
Journal of Environmental Management (0301-4797)	A2	4.005	2	5.12
Producción + Limpia (1909-0455)	**	**	2	5.12
Revista Ambiente & Água (1980-993X)	B3	**	2	5.12
Journal of Environmental Chemical Engineering (2213-3437)	B3	**	2	5.12
Journal of Environmental Sciences (1001-0742)	A2*	3.120	1	2.56
Información tecnológica (0718-0764)	B3	**	1	2.56
Bioresource Technology (0960-8524)	A1	5.807	1	2.56
Resources, Conservation and Recycling (0921-3449)	A2	5.120	1	2.56
Exacta (1678-5428)	B4	**	1	2.56
Journal of Industrial and Engineering Chemistry (1226-086X)	A1	4.841	1	2.56
Journal of Food Engineering (0260-8774)	A1	3.197	1	2.56
Algal Research (2211-9264)	A2*	3.745	1	2.56
Industrial Crops and Products (0926-6690)	A1	3.849	1	2.56
Chemosphere (0045-6535)	A1	4.427	1	2.56
Journal of Hazardous Materials (0304-3894)	A1	6.434	1	2.56
Chemical Engineering Journal (1385-8947)	A1	6.735	1	2.56
Process Biochemistry (1359-5113)	B1*	2.616	1	2.56
Revista Ciência Agronômica (1806-6690)	B2*	0.605	1	2.56
Fórum Ambiental da Alta Paulista (1980-0827)	**	**	1	2.56

*Qualis score of Engineering Group 1 (in portuguese, Engenharias I); **Impact factor and/or Qualis score not available.

Another interesting behavior noted in the third experiment was the increasing and then decreasing trend followed by the percentage reduction in turbidity over time. This is presented in Figure 2. Such behavior indicates that it is possible to obtain an optimal range of sedimentation time to maximize the process efficiency based on the specific reactor and coagulant characteristics.

Table 3. Reduction in bentonite mass - first experiment.

Sample number (-)	Time at which sample was collected (s)	Beaker mass ($\times 10^{-3}$ kg)	Beaker + bentonite mass after drying ($\times 10^{-3}$ kg)	Bentonite mass ($\times 10^{-3}$ kg)	Percentage reduction in bentonite mass (%)
#1	*	20.6161	20.6234	0.0073	-
#2	900	20.3588	20.3611	0.0023	68.5
#3	1800	21.2793	21.2814	0.0021	71.2
#4	2700	21.7720	21.7736	0.0016	78.1

*Refers to test water without coagulant addition.

Table 4. Reduction in bentonite mass - second experiment.

Sample number (-)	Time at which sample was collected (s)	Beaker mass ($\times 10^{-3}$ kg)	Beaker + bentonite mass after drying ($\times 10^{-3}$ kg)	Bentonite mass ($\times 10^{-3}$ kg)	Percentage reduction in bentonite mass (%)
#1	*	21.4222	21.4270	0.0048	-
#2	900	20.9636	20.9671	0.0035	27.1
#3	1800	21.3889	21.3916	0.0027	43.8
#4	2700	20.0707	20.0734	0.0027	43.8

*Refers to test water without coagulant addition.

Table 5. Turbidity reduction - third experiment.

Sample number (-)	Time at which sample was collected (s)	Turbidity (NTU)	Percentage reduction in turbidity (%)
#1	*	148	-
#2	600	19	87.2
#3	900	20	86.5
#4	1200	14	90.5
#5	1500	10	93.2
#6	1800	7	95.3
#7	2100	8	94.6
#8	2400	9	93.9
#9	2700	8	94.6

*Refers to test water without coagulant.

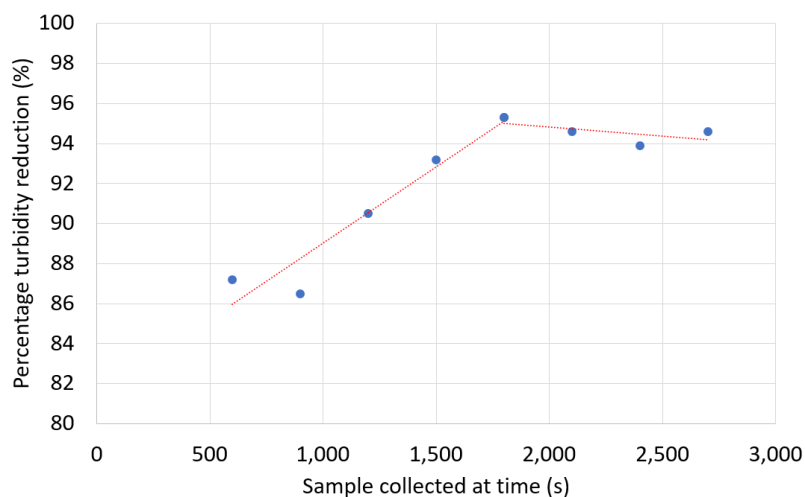


Figure 2. Behavior of the percentage reduction in turbidity over time - third experiment.

Conclusion

This paper analyzed alternative coagulants studied in the literature and their application to water treatment processes. Based on the literature analysis, *Moringa oleifera* was chosen as the natural agent to be used in experiments, reaching a process efficiency about 95.3%. Furthermore, the lowest process efficiency obtained in experiments was higher than the best results obtained in the literature when using aluminum

sulphate as coagulant. These results also prove that *Moringa oleifera* can be a powerful tool in water treatment processes, which can enhance high process efficiency while potentially lowering damages to the environment and human health.

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