Análise de eficiência de diferentes sementes e cascas Oleaginosas para uso como coagulantes naturais

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Artigo recebido em 23/02/2023 e aceito em 13/03/2024

R E S U M O
O problema de pesquisa é reduzir o uso de coagulantes químicos buscando alternativa mais sustentáveis ao tratamento da água. O uso da Moringa oleifera (MO) apresenta-se como boa alternativa de agente coagulante, indicada no sistema de tratamento de água, no seu processo de clarificação, por ser biodegradável. No entanto, existe a necessidade de buscar outras sementes e frutas que também tenham potencial para ser utilizadas como coagulantes naturais. Neste contexto, justifica-se o estudo de outras sementes típicas de Portugal, onde foi realizado este estudo, para ser utilizada como alternativa de coagulante natural. A hipótese desse projeto é: encontrar-se-á, entre as sementes tipicamente europeias, selecionadas para avaliação, sementes com alto potencial de utilização como coagulante natural para uso em tratamento de água. Este projeto tem como objetivo a identificação de sementes, típicas de Portugal, com potencial semelhante ao da Moringa oleifera LAM para ser utilizada como coagulante natural. O projeto foi realizado em parceria da UFS, IFS e ISEP, onde foram realizados os experimentos com água sintética de turbidez que variaram de 40 a 350 NTU. As sementes utilizadas foram quatro tipos diferentes de castanhas europeias, nozes americanas e portuguesas, plátano e Moringa oleifera, além das cascas de castanha e de nozes. Dos resultados encontrados destaca-se a castanha espanhola, com eficiência de remoção de turbidez de até 69,8% em água de alta turbidez além da MO com eficiência de remoção de 89,8% em água de alta turbidez.
Palavras-chave: Castanha sativa; Coagulação; Moringa oleifera; Tecnologia social.

A B S T R A C T
The research problem is to reduce the use of chemical coagulants by seeking more sustainable alternatives to water treatment. The use of Moringa oleifera (MO) is a good alternative coagulant agent, indicated in the water treatment system, in its clarification process, in addition to being biodegradable, but there is a need to look for other seeds that also have potential for be used as natural coagulants, in this context, it is justified to study other typical seeds from Portugal, where this study was carried out, to be used as an alternative natural coagulant. The hypothesis of this project is: among the typically European seeds selected for evaluation, seeds with high potential for use as a natural coagulant for use in water treatment will be found. This project aims to identify seeds, typical of Portugal, with similar potential to Moringa oleifera Lam to be used as a natural coagulant. The project was carried out in partnership with UFS, IFS and ISEP, where experiments were carried out with synthetic water with turbidity ranging from 40 to 350 NTU. The seeds used were four different types of European chestnuts, American and Portuguese nuts, plantain and Moringa oleifera, in addition to...
chestnut and walnut shells. From the results found, the Spanish chestnuts stand out with a turbidity removal efficiency of up to 69.8% in high turbidity water, in addition to the MO with a removal efficiency of 89.8% in high turbidity water. Keywords: Castanea sativa; Coagulation; Moringa oleifera; Social Technology.

Introduction

The project is a theme social technologies and sustainable development, being the line of research alternative treatment of water for human consumption, with the motivation to reduce the use of chemical products in water treatment, in addition to allowing greater autonomy for riverside populations in water consumption of quality produced in a simple and safe way.

It is widely acknowledged the extensive use of chemical products in water treatment, primarily employing coagulating agents such as ferric chloride and aluminum sulfate. Hence, in efforts to diminish the reliance on these chemicals, an increasing number of studies are endeavoring to replace them with natural coagulants due to their sustainability and biodegradability. In this context, several well-known natural coagulants include chitosan, Leucaena leucocephala, Azadirachta indica, tannins, Moringa oleifera, among others. Consequently, this research, conducted in Portugal through a collaboration between ISEP, IFS, and UFS, aimed to explore seeds and husks indigenous to the northern region of Portugal for their potential efficacy as natural coagulants and flocculants in comparison to Moringa oleifera. (Alnawajha et al., 2022; Iber et al., 2021; Ahmad et al., 2021; Turunen, Karppinen & Ihme, 2019; Santos et al., 2023a).

For Dzuvor et al. (2021) the use of Moringa oleifera LAM (MO) presents a range of potential benefits for human health with anticancer, anti-diabetic, antioxidant, anti-inflammatory properties and, in addition, its seeds and cakes can be used in the treatment process of water in coagulation, flocculation and as adsorbents due to their ability to eliminate microbes and organic matter.

In this context, this cooperation project between UFS (Federal University of Sergipe), IFS (Federal Institute of Sergipe) and ISEP (Superior Institute of Engineering of Porto) aims to identify seeds, typical of Portugal, with similar potential to from Moringa oleifera LAM to be used as a natural coagulant.

The hypothesis of this project is that, similar to Moringa oleifera and other recently evaluated natural coagulants, predominantly of Indian origin, seeds typically of European, studied in this project will also exhibit high potential for utilization as natural coagulants in water treatment.

For this, we will use the MO, a tropical plant that belongs to the Moringaceae family, which, around fourteen species have already been identified and all have coagulant properties in different degrees of coagulation, as a reference for comparing the efficiency of removing turbidity from water (Oladoja & Pan, 2015).

Positive results in the water purification treatment were also presented by Owodunni et al. (2023), in the use of Moringa oleifera lam in a combined Coagulation/Flocculation process to reduce the amount of phosphate in water.

Other natural coagulants are already with well advanced studies that, according to Nimesha et al. (2022), are the most commonly used natural coagulants such as Chitosan, Moringa oleifera (MO), Nirmali, Cactus musilage and Tannins. In this case, for this research, we selected seeds that exist in abundance in the northern region of Portugal, easy to manipulate and with little research in the area, in addition to other oilseeds that were used as a comparison. The selected seeds were: European chestnut: wild, Portuguese longal and Jewish and Spanish, Moringa oleifera, Portuguese and American nuts and Plátano.

Theoretical Reference

In this topic we will present the theoretical framework that was the basis for our research, which is divided into the following topics: use of natural coagulants in water treatment, with special emphasis on Moringa oleifera LAM (MO) and Castanea sativa Mill (European chestnut) and chemical composition of selected oilseeds.

Use of natural coagulants in water treatment.

Coagulants are used in water treatment as chemical conditioning through the addition of chemicals in order to modify the properties of colloids thus increasing the efficiency of their removal. The most used coagulants are the inorganic ones based on aluminum and iron salts such as Aluminum Sulfate and Ferric Chloride. There is also the use of organic coagulant, the so-called cationic polymers, which can be synthetic or natural. (Pinto et al., 2020, apud., Faye; Zhang; Yang, 2017; Owodunni et al., 2023).

According to Nimesha et al. (2022) the most commonly used natural coagulants are Chitosan, Moringa oleifera (MO), Nirmali, Cactus musilage and Tannins, as we can see in Table 1.
Table 1. Summary of functional groups and mechanisms proposed for natural coagulants

<table>
<thead>
<tr>
<th>Natural coagulant</th>
<th>Source of extraction</th>
<th>Mechanism proposed</th>
<th>Functional groups</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nirmali (Strychnos pototorum)</td>
<td>seeds</td>
<td>Inter-particle bridging</td>
<td>Galactan and galactomannan</td>
<td>Vijayaraghavan <em>et al.</em>, 2011; Yin, 2010.</td>
</tr>
<tr>
<td>Chitosan</td>
<td>fungi, marine, invertebrates, yeasts</td>
<td>Charge neutralization and bridging</td>
<td>N-acetyl-D-glucosamine (acylated unit), B-(1-4)-linked D-glucosamine (deacetylated unit)</td>
<td>Saranya <em>et al.</em>, 2014.</td>
</tr>
<tr>
<td>Tannis</td>
<td>castanea, acacia, or Schinopsi</td>
<td>Adsorption and charge neutralization</td>
<td>Polyphenol compounds</td>
<td>Vijayaraghavan <em>et al.</em>, 2011; Yin, 2010.</td>
</tr>
</tbody>
</table>

Source: Nimesha *et al.* (2022, p. 104).

Besides these, Pinto *et al.* (2020), cites the use of Jatropha curcas, Prosopis juliflora, Guar gum, Calotropis procera, Cactus latifaria and Clidemia angustifolia as natural conditioners.

Second, Šćiban *et al.* (2009), some authors state that the active coagulating agents of MO are dimeric cationic proteins (*N*dainbigengesere *et al.*, 1995, Fink, 1984, apud. Šćiban *et al.* 2009), while other authors suggest that the active coagulant agent of MO they are not proteins, polysaccharides or lipids, but some other organic polyelectrolyte (Sanghi *et al.*, 2002; Okuda *et al.*, 2001a, *apud.* Šćiban *et al.*, 2009).

The coagulation activity, mainly in natural coagulants, is strongly related to three variables, namely initial turbidity, coagulant concentration and pH. Finding the ideal point between the coagulant concentration and the initial turbidity is fundamental for the best interaction between the molecules and, consequently, a greater efficiency in coagulation. This interaction is so important that the same coagulant agent can increase the final turbidity when the cation is added more than the anion, increasing the turbidity of the water because there are no charged particles to interact, or have an efficiency greater than 90% when used in a dosage correct, as can be seen in studies such as Hadadi *et al.*, (2022) and Desta & Bote (2021).

With emphasis on *Moringa oleifera* (MO), it has natural polymeric properties that have been notoriously gaining prominence in water treatment for its contribution to sustainable development by reducing the use of chemical products, acting as a clarifying agent due to the presence of a cationic protein that destabilizes particles contained in a liquid medium. (Madrona *et al.*, 2017).

It has active biocoagulants that can be used in water clarification treatment, filtration,
sedimentation and water flocculation, reducing the use of chemical-based coagulants (Camacho et al., 2017, Valverde et al., 2018).

Studies show that MO uses can reduce water turbidity by up to 85%, in addition to being able to remove 40% to 50% of the organic matter present in the water (Camacho et al., 2017). According to Nordmark et al. (2018), the results of their research conclude that Moringa oleifera proteins adsorb irreversibly with respect to rinsing in the water model used, suggesting that sand modified with moringa proteins is stable in repeated use of water filtration, thus making it a simple, effective and sustainable water filtration device.

Positive results in the water purification treatment were also found by Madrona et al. (2017), in the use of Moringa oleifera LAM in a combined coagulation/flocculation process with ultrafiltration, obtaining high quality water for human consumption after the process.

European chestnut tree (Castanea sativa Mill) has a significant economic contribution to Portugal, being among the first places as agricultural products most exported by the country, highlighting the North region where the largest number of chestnut groves are concentrated, with a representation of more than 83% both in terms of area and production volume. (Portugal – INRB, 2008).

According to the National Institute of Biological Resources (INRB, 2008) there is a wide variety of Portuguese chestnuts, from the European chestnut tree (Castanea sativa), among them we have: Aveleira, Matainha, Longal, Judia, Colarinha, Verdeal, Rebordã, Cota, Lada, Bária, Negral, Amaresal, Lamela, Zeive and Redonda.

Fruit varieties often differ significantly from each other in physical and nutritional characteristics. (Portugal – INRB, 2008).

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Fruit varieties often differ significantly from each other in physical and nutritional characteristics. (Portugal – INRB, 2008).

Chemical composition of selected oilseeds

Several oilseeds were selected to identify their effectiveness and efficiency as natural coagulants, including Moringa oleifera, four varieties of European chestnut, American and Portuguese walnuts and plantain (Platanus). The characteristics of these seeds reported in the literature are summarized in Table 2.

**Table 2.** Nutritional values of seeds selected for study.

<table>
<thead>
<tr>
<th></th>
<th>Proteins</th>
<th>Lipids</th>
<th>Carbohydrates</th>
<th>Ashes</th>
<th>Humidity</th>
<th>Tannins</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Europea</strong>n <strong>Chestnut</strong></td>
<td>6.26% to 7.55%</td>
<td>2.42% to 3.54%</td>
<td>41.5%</td>
<td>1.23% to 1.61%</td>
<td>48.87% to 50.64%</td>
<td>4.5% to 4.51%</td>
<td>Silva et al. (2021) e Souza et al. (2017)</td>
</tr>
<tr>
<td><strong>Nuts</strong></td>
<td>7.5% to 21.56%</td>
<td>42.88% to 66.71%</td>
<td>0.55% to 3.49% (total sugars)</td>
<td>1.16% to 3.28%</td>
<td>1.47% to 9.51%</td>
<td>0.01% to 0.88%</td>
<td>Venkatachalam et al. (2006)</td>
</tr>
<tr>
<td><strong>Moringa oleifera</strong></td>
<td>32.16% to 32.52%</td>
<td>25.0% to 33.68%</td>
<td>23.48% to 32.43%</td>
<td>4.5% to 4.51%</td>
<td>5.53% to 6.16%</td>
<td>1.20% (sheets)</td>
<td>Silva et al. (2020)</td>
</tr>
<tr>
<td><strong>Platanus</strong></td>
<td>13.7%</td>
<td>25%</td>
<td>1.20% (sheets)</td>
<td>Faye et al. (2023)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Materials and methods**

This project has a hypothetical-deductive method of approach, and can be classified as applied, quantitative, exploratory, experimental and case study research. The means used will be field research, laboratory, bibliographical, experimental and case study.

Santos, L.O., Sousa, I. F., Silva, G. F., Barbosa, S. A., Caetano, N. S.,
Synthetic water preparation

The synthetic water was prepared according to the methodology of Putra, Amri and Ayu (2020), using 1 gram of white clay for 1 liter of tap water. The methodology was changed because, instead of using white clay, gray clay was used to produce high turbidity synthetic water. The solution was stirred for 1 hour to obtain a uniform solution of clay particles. Soon after, the solution is used as a sample containing low organic content, seeking treatment for water turbidity.

![Image of water agitation with gray clay to prepare synthetic water.](image1)

Figure 1. Water agitation with gray clay to prepare synthetic water.

Seed Preparation

The seeds used were used in different ways of handling, ranging from: crushed; crushed and dried; crushed and sieved; crushed, dried and sieved, so that it could be verified how we would increase its coagulation potential. They were crushed in a blender and sieved through a domestic sieve with a mesh opening of 2.5 mm until obtaining a fine flour, as shown in figure 2. The natural coagulant based on MO was selected as a standard for comparing efficiency because it is widely known in academic and popular circles. In addition to this, wild chestnut seeds (Figure 4C), long Portuguese chestnuts (Figure 5A), Portuguese Jewish chestnuts (Figure 5B), Spanish chestnuts (Figure 3C), Portuguese walnuts and American walnuts (Figure 5B) were also used and prunus. Walnut shells (Figure 4B) and chestnut shells (Figure 4A) were also used, crushed in a blender and sieved through a domestic sieve with a mesh opening of 2.5 mm until obtaining a fine flour.

![Image of Moringa seed processing process to obtain powdered coagulant.](image2)

Figure 2. Moringa seed processing process to obtain powdered coagulant.
Figure 3. A: Ground and sieved MO seed; B: Crushed walnut seed; C: Crushed Spanish chestnut seed.

Figure 4. A: Crushed and sieved Spanish chestnut shell; B: Crushed and sieved walnut shell; C: Crushed and sieved wild chestnut seeds.

Figura 5. A: Portuguese Chestnut Longal Seed; B: Crushed Jewish Portuguese chestnut seed.
Jar Test and Turbidity Efficiency Calculation

To identify the ideal coagulant concentration, the jar test was performed, with concentrations of 0.1 g/L, 0.2 g/L and 0.3 g/L, fast stirring at 300 rpm for 2 minutes and slow stirring at 45 rpm for 10 minutes and a decantation period of 2 hours. For the same period of 2 h of decantation, it was awaited in the synthetic water without the addition of coagulant. After this procedure, the supernatant of each glass beaker was collected and the turbidity parameter was analyzed.

To calculate the turbidity efficiency, we used the equation below:

\[ \% \text{Turbidity removed} = \frac{T_f^{(as)} - T_f}{T_f^{(as)}} \times 100 \]  

Where; \( T_f^{(as)} \): Final turbidity of the synthetic water after decanting for 2 hours and \( T_f \): Final turbidity of the water with coagulant after decanting for 2 hours.

Physicochemical analysis

The physical-chemical parameters analyzed in this project were pH, color and turbidity, as described in Table 3. Note that all analyzes were performed in quintuplicate and the results treated statistically as mean and standard deviation.

Table 3. Physical-chemical and microbiological analysis methods

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Method</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>Potentiometric method</td>
<td>Dimensionless</td>
</tr>
<tr>
<td>Color</td>
<td>Colorimetric method</td>
<td>UC</td>
</tr>
<tr>
<td>Turbidity</td>
<td>Nephelometric method</td>
<td>NTU</td>
</tr>
</tbody>
</table>

Results and discussions

First, the physicochemical characterization of pH, color and turbidity of tap water and synthetic water will be presented, according to the methods listed in Table 4. Subsequently, the maximum efficiency of turbidity reduction obtained with Portuguese chestnut, Spanish chestnut, wild chestnut, Jewish chestnut, American walnuts, Portuguese walnuts, walnut shell, chestnut shell and MO seed will be presented, and the condition of coagulant concentration and initial turbidity of the synthetic water, which varies from 40 to 360 NTU in which that reduction efficiency was obtained.

As we saw in the previous topic, to calculate the turbidity removal efficiency, the final turbidity of the water using the natural coagulant is compared with the final turbidity of the synthetic water without the addition of coagulant, both after the decantation period. These results are shown in Table 4.

Table 4. Physical-chemical characterization of the samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Color</th>
<th>Turbidity</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tap water</td>
<td>&lt;2,0</td>
<td>4,64±0,04</td>
<td>7,50±0,08</td>
</tr>
<tr>
<td>Synthetic Water</td>
<td>&gt;550,00</td>
<td>130,95±28,53</td>
<td>7,80±0,04</td>
</tr>
</tbody>
</table>

The first phase of the research was based on analyzing all the selected seeds and peels, under different conditions of initial turbidity, coagulant concentration and method of preparation of the seeds, ranging from crushed seeds to crushed, dried and sieved, verifying those seeds with greater coagulation potential and in what condition that potential is obtained, so that further analyzes and comparisons could be carried out later, this time varying only the initial turbidity and fixing the other variables.
To facilitate the visualization of the tables and obtain a better understanding, the results will be presented in groups in the concentrations of 0.1 g/L, 0.2 g/L and 0.3 g/L for each element, as we will see in Table 5.

**Table 5.** Efficiency of the elements in reducing the turbidity of the synthetic water.

<table>
<thead>
<tr>
<th>% Efficiency in Reducing the Turbidity of Synthetic Water</th>
<th>Turbidity of synthetic water without decantation (NTU)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Portuguese nuts</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (S)</td>
<td>5.6%±0.4% 200±11.46</td>
</tr>
<tr>
<td>0.2 g/L (SD)</td>
<td>4.6%±0.4% 250±14.21</td>
</tr>
<tr>
<td>0.3 g/L (SD)</td>
<td>15.5%±0.5% 120.00±7.84</td>
</tr>
<tr>
<td><strong>American walnuts</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (S)</td>
<td>3.6%±0.4% 200±11.46</td>
</tr>
<tr>
<td>0.2 g/L (SD)</td>
<td>12.8%±0.4% 250±14.21</td>
</tr>
<tr>
<td>0.3 g/L (SD)</td>
<td>7.2%±0.0% 120±7.84</td>
</tr>
<tr>
<td><strong>Portuguese chestnut</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (S)</td>
<td>10.7%±0.4% 200±11.46</td>
</tr>
<tr>
<td>0.2 g/L (SD)</td>
<td>12.6%±0.4% 160±6.36</td>
</tr>
<tr>
<td>0.3 g/L (SD)</td>
<td>15.4%±0.4% 200±9.33</td>
</tr>
<tr>
<td><strong>Walnut shell</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (CDS)</td>
<td>8.9%±0.5% 160±6.36</td>
</tr>
<tr>
<td>0.2 g/L (S)</td>
<td>-5.1%±0.6% 120±7.84</td>
</tr>
<tr>
<td>0.3 g/L (SD)</td>
<td>-4.8%±0.6% 200±9.33</td>
</tr>
<tr>
<td><strong>Chestnut shell</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (S)</td>
<td>-28.0%±0.0% 120±7.84</td>
</tr>
<tr>
<td>0.2 g/L (CDS)</td>
<td>-27.7%±0.0% 160±6.36</td>
</tr>
<tr>
<td>0.3 g/L (SD)</td>
<td>-19.4%±1.3% 200±9.33</td>
</tr>
<tr>
<td><strong>Jewish Portuguese chestnut</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (SD)</td>
<td>27.3%±0.7% 160±6.36</td>
</tr>
<tr>
<td>0.2 g/L (SD)</td>
<td>47.2%±0.0% 350±17.72</td>
</tr>
<tr>
<td>0.3 g/L (SD)</td>
<td>37.7%±0.6% 140±5.51</td>
</tr>
<tr>
<td><strong>Wild chestnut</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (CDS)</td>
<td>16.3%±0.0% 200±9.33</td>
</tr>
<tr>
<td>0.2 g/L (S)</td>
<td>36.3%±0.3% 300±15.12</td>
</tr>
<tr>
<td>0.3 g/L (CDS)</td>
<td>9.6%±0.3% 200±9.33</td>
</tr>
<tr>
<td><strong>Platanus</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (CDS)</td>
<td>9.4%±0.4% 200±9.33</td>
</tr>
<tr>
<td>0.2 g/L (S)</td>
<td>24.5%±0.3% 300±15.12</td>
</tr>
<tr>
<td>0.3 g/L (S)</td>
<td>6.6%±0.3% 300±15.12</td>
</tr>
<tr>
<td><strong>Spanish chestnut</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (S)</td>
<td>21.3%±0.0% 120±7.84</td>
</tr>
<tr>
<td>0.2 g/L (S)</td>
<td>69.8%±0.0% 350±17.04</td>
</tr>
<tr>
<td>0.3 g/L (S)</td>
<td>24.9%±0.7% 120±7.84</td>
</tr>
<tr>
<td><strong>Moringa oleifera</strong></td>
<td></td>
</tr>
<tr>
<td>0.1 g/L (CDS)</td>
<td>50.7%±0.0% 120±7.84</td>
</tr>
<tr>
<td>0.2 g/L (CS)</td>
<td>89.8%±0.4% 300±15.12</td>
</tr>
<tr>
<td>0.3 g/L (CS)</td>
<td>88.2%±0.3% 300±15.12</td>
</tr>
</tbody>
</table>
Maple seeds, seeds and nut shells

In this topic, we will present and discuss the efficiency of plantain seeds, walnut seeds and walnut shells as natural coagulants.

For all statistical analyzes and graphs constructed in this article, Graphpad's prism 8 software was used with unidirectional ANOVA test and 5% significance for analysis of statistical differences. The same letters are used for samples that are not statistically significantly different and different letters for samples that are significantly different.

In this article, the positive values are those that obtained a reduction in the turbidity of the synthetic water and negative values come from the increase in the turbidity of the synthetic water.

**Table 5.** Efficiency of the elements in reducing the turbidity of the synthetic water.

<table>
<thead>
<tr>
<th>% Efficiency in Reducing the Turbidity of Synthetic Water</th>
<th>Turbidity of synthetic water without decantation (NTU)</th>
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Caption: S: Shredded; SD: Shredded and Dried; CDS: Crushed, Dry and Sieved; CS: Crushed and Sieved.

Figure 6. Graph of turbidity reduction efficiency of plantain seeds and walnuts, in addition to walnut shells.
As can be seen in figure 6, none of the walnut, plantain and walnut shell seeds were efficient in reducing turbidity, and the maximum reduction obtained was with plantain seed at a concentration of 0.2 g/L with 24.5% reduction efficiency. In research conducted by Ahmad et al. (2022), it was found that the turbidity removal results for natural coagulants were similar to those found in our study, with an average turbidity removal efficiency ranging between 10% and 30%.

The American and Portuguese walnuts had a low reduction efficiency in all analyzed situations, with the maximum efficiency of the Portuguese walnut in its crushed, dry and unsifted condition, in water with an initial turbidity of 100 to 120 NTU and at a concentration of 0.3 g/L, thus obtaining a reduction of 15.5%. American walnut also had its best efficiency in the crushed, dry and unsifted condition, in synthetic water of 100 to 120 NTU, but at a concentration of 0.2 g/L with a maximum reduction of 12.8%.

In most of the analyzed situations, the walnut shells increased the turbidity of the synthetic water, only in the case of crushed, dried and sieved walnut shells, in synthetic water with turbidity of 80 to 100 NTU and at a concentration of 0.1 g/L which achieved efficiency in reducing turbidity of 8.9%.

Chestnut Seeds and Peels
In this topic, we will present and discuss the efficiency of the seeds and husks of different European chestnuts as natural coagulants (figure 7).

As can be seen in figure 7, in most of the situations analyzed, the turbidity reduction efficiency was less than 50%. However, some situations stand out, they are: wild chestnut at a concentration of 0.2 g/L, Jewish chestnut at a concentration of 0.2 g/L and Spanish chestnut at a concentration of 0.2 g/L and for this motif will be analyzed later along with the MO seed.

It can be seen that the chestnut shell was not efficient in reducing the turbidity in any situation studied, contributing in all cases to the increase in the turbidity of the synthetic water.
As verified in the research by Šćiban et al. (2009), for synthetic water with high turbidity and a pH of 9, the efficiency of the coagulant activity of chestnut was below 40%, using the coagulant extract.

According to Šćiban et al. (2009) extracts from European chestnut trees had low efficiency in reducing turbidity in water with turbidity of 70 NTU. In this research, with some exceptions mentioned above in turbidity above 300 NTU, the chestnut seeds also obtained low efficiency with a pH of 7.80, indicating that these seeds also have low efficiency in removing turbidity under these conditions.

In Hatim, Almansoory & Al-Baldawi (2024) research’s, it is also noted that the natural coagulants investigated exhibit efficiency lower than 60% in most cases, even in highly turbid water.

Next we will see how the performance of Moringa oleifera seeds was for later comparison with European chestnuts.

Moringa oleifera LAM.

In this topic, we will present and discuss the efficiency of MO seed as a coagulant in synthetic water, as shown in figure 8.

These results are compatible with Desta & Bote (2021) who obtained a reduction of 99.5% of turbidity and 97.7% of color in basic waters and 98% of turbidity and 90.76% of color in acidic waters. They were also compatible with results found by Santos et al. (2023b) and Batista et al. (2023) who also obtained a reduction of approximately 97% in turbidity after treatment using MO and Valverde et al. (2018) who obtained a reduction of 70% of turbidity and 75% of apparent color with the use of MO seed as a coagulating agent. These results vary, as there are several factors that affect the coagulant activity of MO, mainly the pH and the initial turbidity of the raw water, which determine the level of interaction between the particles.

Comparing this graph with the previous ones in figure 6 and figure 7, we can see that the MO seed was the natural coagulant with the highest turbidity removal efficiency, among those studied in this article.

Analyzing the graphics in figures 6, 7 and 8, it can be seen that the seeds with the greatest efficiency in removing turbidity were: wild chestnut 0.2 g/L, Jewish chestnut 0.2 g/L, Spanish chestnut 0.2 g/L and MO 0.2 g/L. Thus, the next phase of the studies began, where we set the concentration of these natural coagulants at 0.2 g/L in their most efficient preparation and varied the initial turbidity of the synthetic water in concentrations in the range of: Range 1: 40 to 60 NTU; Range 2: 60 to 100 NTU; Range 3: 100 to 150 NTU; Range 4: 150 to 200 NTU; Range 5: 250 to 300 NTU and Range 6: 300 to 350 NTU.

Wild Chestnut, Jewish Portuguese Chestnut, Spanish Chestnut and Moringa oleifera.

In this topic, the coagulation efficiency of the seeds that showed a greater coagulant potential in previous experiments will be presented, now with a coagulant concentration fixed at 0.2 g/L and varying the initial turbidity in ranges ranging from 40 to 350 NTU, as shown in figure 9.

Figure 8. Graph of turbidity reduction efficiency of MO seed.

Figure 9. Graph of turbidity reduction efficiency of Castaha Brava, Judia, Española and Moringa oleifera seeds all at a concentration of 0.2 g/L.
In this graph, the relationship between initial turbidity and the coagulation efficiency of natural coagulants is clear. But, in general, it is also noticed that natural coagulants are more efficient when the initial turbidity is higher and less efficient when the initial turbidity is lower. Below we will present some information about each seed studied in this topic.

Wild chestnut: Wild chestnut did not show high potential as a natural coagulant in any situation, with an efficiency of less than 10%, whether without drying, after drying, crushed or crushed and sieved, with its best potential being freshly crushed, without drying or sieving, in synthetic water with turbidity between 250 and 300 NTU with a concentration of 0.2 g/L, obtaining a reduction of 36.3% in the turbidity of the synthetic water and soon after in water of 350 NTU it returned to an efficiency close to 5%

Jewish Portuguese Chestnut: The Portuguese nut of the Jewish type suffers great interference from drying in an oven at 60 °C for 1 hour, and in the same initial turbidity of 150 NTU and concentration of 0.2 g/L it increased its efficiency of removing turbidity from 1.1% to 43.1% after drying. With regard to the initial turbidity, it does not suffer major interference, with an initial turbidity of 40 NTU removal of 31.6%, initial turbidity of 100 NTU removal of 21.5%, 150 NTU removal of 43.1%, 240 NTU removal of 26.6%. 350 NTU 47.2% removal. Obtaining its greatest potential in synthetic water of greater turbidity, but showing that its removal power is not directly influenced by the initial turbidity.

Spanish Chestnut: The European chestnut harvested in Spain suffers a great negative influence from kiln drying and sifting, therefore, it is preferable only to crush it. With synthetic water of 200 NTU and concentration of 0.2 g/L of coagulant, the Spanish chestnut seed only ground reduced the initial turbidity by 62.5%, while the ground and dry seed reduced 42.5% and the ground seed, sieved and dried reduced the turbidity of the synthetic water by only 14.6%. The initial turbidity of the synthetic water also greatly interferes with the coagulation power of the Spanish chestnut, and in water with high turbidity it obtains the best results. In water with an initial turbidity of 40 NTU, the seed increased the turbidity of the water, obtaining an efficiency of -31.6% and thus, directly proportional to the increase in the initial turbidity of the water, as we can see in figure 10, the seed of Spanish chestnut also increases its turbidity removal potential reaching its apex at the highest studied turbidity of 350 NTU with 69.8% removal.

Moringa oleifera LAM: MO, among the studied seeds, was the one with the best efficiency for removing turbidity from high turbidity water, with efficiency of up to 90% in synthetic water with initial turbidity of 300 NTU. Like other natural coagulants, the coagulation potential of MO is highly influenced by the initial turbidity, concentration and manipulation of the coagulant, with ground, sieved and oven-free seeds having the greatest coagulation potential. As with the Spanish chestnut, the MO also increased the turbidity of the water with an initial turbidity of 40 NTU, obtaining an efficiency of -59.6%, increasing its removal potential directly proportional to the increase in the initial turbidity to the efficiency maximum of 89.8% at an initial turbidity of 300 NTU and soon after it had a reduction in this efficiency to 79.2% at an initial turbidity of 350 NTU.

Conclusions

In figures 6, 7, 8 and 9, it is clear how the concentration of the natural coagulant is a factor of high interference in the coagulation activity, depending on an optimal relation between the dose of coagulant and the divalent cations present in the water.

With the results presented, it can be seen that the chestnut shells and walnut shells did not have a good potential for removing turbidity and that, although all the nuts studied were European chestnuts of the genus Castanea sativa, the different types of nuts had very different results in terms of turbidity removal potential.

Among the seeds studied, the one with the best potential for removing turbidity was the MO with the best result of 89.8% removal, followed by Spanish chestnut with an efficiency of 69.8% and both obtained very similar behaviors with regard to the relationship between turbidity initial and coagulation activity, both with optimal concentration of 0.2 g/L. It is also worth noting that the Spanish nut obtained this removal efficiency by being crushed only, while the MO obtained its highest reduction efficiency by being crushed and sieved.

With this work, it was also noticeable the influence of the initial turbidity, concentration and method of preparation of the coagulant in the coagulation activity of the same, being that each coagulant has a different behavior and the best interaction between coagulant and the analyzed water must be studied so that maximum clotting potential can be extracted.

Another important point verified in this work was that, as presented in the theoretical
framework, some authors differ in relation to the agents that act in the coagulation of natural coagulants, whether they are dimeric cationic proteins or some other type of organic polyelectrolyte. As can also be seen in the theoretical references, Table 2, among the different varieties of Portuguese chestnuts, the amounts of proteins do not differ much from each other, being in values that vary from 3.9% to 7.8%, which in itself, does not justify a difference of more than 60% in the efficiency of turbidity reduction, thus suggesting that this difference is in the type of proteins found in each chestnut, or that the coagulant effect comes from other types of organic polyelectrolytes. In order for this to be verified, it is suggested to identify the types of proteins found in Jewish chestnut, wild chestnut and Spanish chestnut, which will remain as a suggestion for further research.

Acknowledgments

L. O. Santos thanks CAPES (Coordination for the Improvement of Higher Education Personnel) for the sandwich grant process No. 88881.690128/2022-01. Thanks to the IFS for the removal process Nº 23288.000359/2021-46 for the doctoral course.

References

http://dx.doi.org/10.1016/j.cej.2016.12.031


Kumar, V., Othman, N., & Asharuddin, S. (2017). Applications of natural coagulants to treat wastewater – A review. MATEC web of conferences, 103, 06016. https://doi.org/10.1051/matecconf/20171030616


Madrona, Grasiele; SCAPIM, Monica; Tonon, Lucineia Cestari; REIS, Miria Miranda; Paraíso, Carolina; Bergamasco, Rosangela. Use of Moringa oleifera in a combined coagulation-filtration process for water treatment. Chemical Engineering Transactions, [S.L.], v. 57, p. 1195-1200, maio 2017. AIDIC: Italian Association of Chemical Engineering. http://dx.doi.org/10.3303/CET1757200.


Santos, L.O., Sousa, I. F., Silva, G. F., Barbosa, S. A., Caetano, N. S.,