Guazuma ulmifolia: an environmentally friendly coagulant aid for water treatment

Leticia Cardoso Madureira Tavares, Alisson Carraro Borges, Teresa Cristina Fonseca da Silva and André Pereira Rosa

ABSTRACT

In the present work the use of a promising novel coagulant aid, Guazuma ulmifolia, was optimized to treat synthetic water using central composite being highly efficient at rotatable design (CCRD). The factors evaluated for the coagulation-flocculation process were coagulants dosages and pH. A model to describe the coagulation-flocculation process was successfully obtained. The model was validated using 5 mg L\(^{-1}\) aluminum sulfate, 2.5 mg L\(^{-1}\) G. ulmifolia and pH 9, achieving excellent agreement with observed values.

Key words | CCRD, mutamba, natural polymer, water treatment

HIGHLIGHTS

- Novel coagulant aid.
- CCRD used to optimize turbidity removal by dual coagulants.
- Potential alternative to synthetic polymers.

INTRODUCTION

Water treatment has faced many challenges, some of which relate to operational/environmental concerns such as energy consumption, environmental footprint and sludge production. The scenario is even more challenging for the coming years. The growing population and the need to treat water, the requirements to control issues arising from the treatment process, and the increasing regulatory pressure to ensure societal well-being are important reasons to seek alternatives to minimize these impacts.

The coagulation-flocculation process is widely used for water treatment for removing suspended and dissolved solids (Katrivesis et al. 2019). Aluminum salts, traditionally, are most commonly used in this process due to being easy to obtain and efficient. However, their use has been questioned due to adverse health effects and the difficulty of disposing of or using the sludge that is generated in large quantities (Antov et al. 2018).

An alternative to reduce residual aluminum in treated water is to use natural polymers as coagulant aids, at the same time reducing the amount of sludge generated and increasing the sedimentation speed, thus reducing the process costs (Jones & Bridgeman 2016; Lim et al. 2018). In recent years, natural polymers have been widely studied by the scientific community as alternatives to metallic coagulants and synthetic polymers (Yang et al. 2009; Mehdinejad & Bina 2018). Among the natural polymers most mentioned in the literature, Moringa oleifera and chitosan stand out, being highly efficient at removing water turbidity (>80%) (Muyibi & Alfugara 2003; Mehdinejad & Bina 2018). In addition, natural polymers stand out for being biodegradable, generating less sludge and being widely available in the environment. Furthermore, when these are used together with metallic coagulants, they provide combined coagulation mechanisms, which improves coagulant activity (Bo et al. 2012; Priya et al. 2017).

Guazuma ulmifolia, commonly known as ‘West Indian elm’, ‘bay cedar’ or ‘mutamba’, is native to Latin America. Its mucilaginous extract, obtained by cooking pieces of the stem, is widely used in sugarcane regions in Brazil for the artisanal manufacture of rapadura, used as a clarifying agent for sugarcane juice during boiling (Carvalho 2007).
Only two studies were found in the scientific literature that evaluated the use of *G. ulmifolia* as a natural coagulant, reporting efficiencies in removing turbidity in the range of 50–70% (Rodríguez-Argüello *et al.* 2015; Feria-Díaz *et al.* 2016). These results suggest that *G. ulmifolia* has the potential to be studied as a coagulation aid, since reasonable efficiencies have been reported, although not yet high enough to be used as the main coagulant (>80%).

Thus, this study hypothesized that the use of mutamba as a coagulant aid to aluminum sulfate makes it possible to decrease the dose of the main coagulant. This hypothesis was tested using central composite rotatable design (CCRD) in order to optimize the dose (main and auxiliary coagulant) and pH factors.

**MATERIALS AND METHODS**

**Synthetic turbid water**

The synthetic water used in this study was prepared by mixing 10 g of powdered kaolin in 1 L of distilled water. The suspension was mixed under magnetic stirring for 1 hour, and then left to stand for 24 hours for complete hydration (Muthuraman & Sasikala 2014). Subsequently, the solution was placed in an Imhoff cone for 30 min. Finally, the 300 mL at the bottom of the cone was discarded and the remaining 700 mL was placed in a flask. This solution was used as the stock solution for the preparation of raw synthetic water with an initial turbidity of 80 NTU, and all tests used the jar test (Zhao *et al.* 2012).

**Primary coagulant**

The standard solution of alum was prepared by adding 10 g of aluminum sulfate (Al₂(SO₄)₃·18H₂O – Synth) to 1 L of distilled water (aluminum sulfate 1% w/v). This solution was subjected to magnetic stirring for complete homogenization (Mehdinejad *et al.* 2009).

**Obtaining coagulant aid**

Mutamba husks were collected in the northern region of Minas Gerais State, Brazil. The material was left in an oven with forced air circulation (model MA 035, Marconi) for 18 hours at 50 °C. Then, the dry husks were crushed in a domestic blender. The material obtained was homogenized with a 45 mesh sieve. The *G. ulmifolia* extract was prepared by adding 10 g of plant bark powder to 1 L of 1% NaCl (w/v) solution. The solution was mixed for 1 hour using a magnetic stirrer, and was prepared before the tests were performed in the jar test in order to avoid interference due to storage (Feria-Díaz *et al.* 2016).

**Coagulation-flocculation tests**

The jar tests were carried out on a 218 – LDB/06 six-paddle stirrer flocculator (Nova Ética). The pH adjustments were performed with the addition of 1 mol·L⁻¹ HCl or 1 mol·L⁻¹ NaOH in order to obtain the desired pH values (5–9). This parameter was read with a pH meter (model MP-6, Hach). The primary coagulant, aluminum sulfate, was added as soon as the rapid mixing speed was reached and the *G. ulmifolia* extract was added after 1 min. The doses of aluminum sulfate and extract of *G. ulmifolia* tested were within the ranges of 0 to 10 mg·L⁻¹ and 0 to 5 mg·L⁻¹, respectively. The fast and slow mixing rates and their respective mixing times were 100 rpm, 2 min and 40 rpm, 30 min. The sedimentation time was 30 min and, in the sequence, the samples were collected to determine the final turbidity using an Orion turbidimeter, model AQ4500, ThermoFisher Scientific. The removed turbidity was used as the response (dependent) variable and it was calculated as the average initial turbidity subtracted from the final turbidity for each test. All tests were conducted in random order to minimize the effect of any unexplained variability in the responses observed due to external errors.

**Experiment design**

CCRD was used in this experiment. The independent variables were the dosages of aluminum sulfate and *G. ulmifolia* extract and pH.

The high and low levels (+1, −1) of the experimental design were determined from preliminary tests and a literature review (Muthuraman & Sasikala 2014). Axial points (+α, −α) were calculated based on the number of independent variables (k = 3), using Equation (1). The value of ‘α’ represents the distance between an axial point and the central point (= 0).

\[
α = (2^k)^{1/4} = 1.68
\]  
(1)

The number of tests was obtained by substituting values in Equation (2).

\[
N = 2^k + 2k + nc
\]  
(2)
where \( k \) is the number of factors \((k = 3)\) and \( nc \) is an arbitrary number of replicates at the central points.

**Statistical analysis**

In order to estimate the statistical parameters and evaluate the predictive capacity of the mathematical models, the data variance analysis (ANOVA) was performed. Individual effects, quadratic effects and interactions were assessed. The model was chosen based on the following quality parameters: significance of the regression models, non-significance of lack of fit, coefficient of determination \((R^2)\), adequate precision \((AP)\) coefficient and standard deviation of the residuals \((S)\).

The optimization step was performed considering the best responses determined by selecting the target option and matching it to 77 NTU. Finally, the model was validated using two independent tests. In one of the tests, the conditions chosen as promising, considering the aim of the work, were reproduced in three replicates. In the second trial, six combinations of random levels of the dependent variables were obtained.

---

### RESULTS AND DISCUSSION

Table 1 shows the turbidity removal and treatment efficiency obtained. The World Health Organization (WHO) recommends a residual turbidity of 0.5 NTU before disinfection for drinking water (WHO 2011). Table 1 shows very satisfactory turbidity removals prior to filtration for some experiments. For instance, the efficiency of runs 4, 6, 13, and 14 was above 85%. Therefore, we conclude that satisfactory turbidity removals can be obtained with a combination of a dose of \( G. \ ulmifolia \) in the range of 1.01–3.99 mg·L\(^{-1}\), a dose of aluminum sulfate in the range of 5.00–7.97 mg·L\(^{-1}\) and a pH in the range of 5–9.

A comparison between runs 6 (pH 8.19) and 2 (pH 5.81) suggests that the basic medium was more effective, while keeping the aluminum sulfate and \( G. \ ulmifolia \) doses fixed. In contrast, the first run presented the worst overall performance, probably because the dose used was not enough to neutralize the electrical charges, consequently showing an increase in the remaining turbidity (Table 1).

The mathematical model obtained by regression in non-coded units to represent the turbidity removal in NTU \((T_{\text{removed}})\) as a function of the aluminum sulfate dose in

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Alum dose</th>
<th>( G. \ ulmifolia ) dose</th>
<th>pH</th>
<th>Alum dose (mg·L(^{-1}))</th>
<th>( G. \ ulmifolia ) dose (mg·L(^{-1}))</th>
<th>pH</th>
<th>Turbidity removal (NTU)</th>
<th>Efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>−1</td>
<td>−1</td>
<td>−1</td>
<td>2.03</td>
<td>1.01</td>
<td>5.81</td>
<td>−20.2</td>
<td>−22.9</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>−1</td>
<td>−1</td>
<td>7.97</td>
<td>1.01</td>
<td>5.81</td>
<td>35.7</td>
<td>43.5</td>
</tr>
<tr>
<td>3</td>
<td>−1</td>
<td>1</td>
<td>−1</td>
<td>2.03</td>
<td>3.99</td>
<td>5.81</td>
<td>39.7</td>
<td>48.9</td>
</tr>
<tr>
<td>4</td>
<td>1</td>
<td>1</td>
<td>−1</td>
<td>7.97</td>
<td>3.99</td>
<td>5.81</td>
<td>73.9</td>
<td>90.1</td>
</tr>
<tr>
<td>5</td>
<td>−1</td>
<td>−1</td>
<td>1</td>
<td>2.03</td>
<td>1.01</td>
<td>8.19</td>
<td>15.3</td>
<td>16.0</td>
</tr>
<tr>
<td>6</td>
<td>1</td>
<td>−1</td>
<td>1</td>
<td>7.97</td>
<td>1.01</td>
<td>8.19</td>
<td>80.8</td>
<td>98.7</td>
</tr>
<tr>
<td>7</td>
<td>−1</td>
<td>1</td>
<td>1</td>
<td>2.03</td>
<td>3.99</td>
<td>8.19</td>
<td>51.0</td>
<td>61.4</td>
</tr>
<tr>
<td>8</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>7.97</td>
<td>3.99</td>
<td>8.19</td>
<td>14.1</td>
<td>18.8</td>
</tr>
<tr>
<td>9</td>
<td>−1.68</td>
<td>0</td>
<td>0</td>
<td>0.00</td>
<td>2.50</td>
<td>7.00</td>
<td>9.2</td>
<td>5.1</td>
</tr>
<tr>
<td>10</td>
<td>1.68</td>
<td>0</td>
<td>0</td>
<td>10.00</td>
<td>2.50</td>
<td>7.00</td>
<td>4.1</td>
<td>7.2</td>
</tr>
<tr>
<td>11</td>
<td>0</td>
<td>−1.68</td>
<td>0</td>
<td>5.00</td>
<td>0.00</td>
<td>7.00</td>
<td>−1.2</td>
<td>−6.1</td>
</tr>
<tr>
<td>12</td>
<td>0</td>
<td>1.68</td>
<td>0</td>
<td>5.00</td>
<td>5.00</td>
<td>7.00</td>
<td>13.0</td>
<td>16.8</td>
</tr>
<tr>
<td>13</td>
<td>0</td>
<td>0</td>
<td>−1.68</td>
<td>5.00</td>
<td>2.50</td>
<td>5.00</td>
<td>71.1</td>
<td>87.4</td>
</tr>
<tr>
<td>14</td>
<td>0</td>
<td>0</td>
<td>1.68</td>
<td>5.00</td>
<td>2.50</td>
<td>9.00</td>
<td>65.9</td>
<td>80.3</td>
</tr>
<tr>
<td>15</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>5.00</td>
<td>2.50</td>
<td>7.00</td>
<td>30.8</td>
<td>38.5</td>
</tr>
<tr>
<td>16</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>5.00</td>
<td>2.50</td>
<td>7.00</td>
<td>5.0</td>
<td>3.6</td>
</tr>
<tr>
<td>17</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>5.00</td>
<td>2.50</td>
<td>7.00</td>
<td>−1.8</td>
<td>−2.1</td>
</tr>
<tr>
<td>18</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>5.00</td>
<td>2.50</td>
<td>7.00</td>
<td>0.2</td>
<td>3.6</td>
</tr>
</tbody>
</table>
mg·L$^{-1}$ (A), the *G. ulmifolia* dose in mg·L$^{-1}$ (B) and pH (C), is presented in Equation (3). All effects and interactions considered non-significant (at the 5% significance level) were removed from the model. The adjustment parameters for the selected model are displayed in Table 2.

$$T_{\text{rem}} = 590.3 + 5.5A + 79.6B - 213.9C - 2.5AB + 2.6AC + 0.5BC - 12.2B^2 + 16.1C^2 - 1.9ABC + 2.5AB^2$$

(3)

From the results shown in Table 2, it was concluded that the model obtained is effective in predicting the removal of turbidity in synthetic water, since its coefficient of determination is greater than 75% (Naik & Setty 2014), adequate precision is greater than 4 and the lack of fit was not significant ($p = 0.7872$).
Optimization

The selected optimum values, when applying the model presented in Equation (3) and selecting the target removed turbidity of 77 NTU, were 5 mg·L$^{-1}$ of aluminum sulfate, 2.5 mg·L$^{-1}$ of *G. ulmifolia* extract and pH 9. These values were chosen given the aims of this work (to study the feasibility of mutamba as a coagulant aid). However, this is not the only optimum condition for the coagulation-flocculation process. The response surfaces indicated other combinations of levels with excellent turbidity removals for both the basic and acidic media.

The pH value calculated to achieve the optimum (pH = 9) was also reported by Bazrafshan et al. (2015). The authors evaluated the use of pistachio seeds (0.4 mg·L$^{-1}$) as a coagulant aid to iron chloride (10 mg·L$^{-1}$). They concluded that the basic medium is efficient for high turbidity removal (97%) for some types of natural polymers. It is noteworthy that the authors also used synthetic water with initial turbidity values similar to this work (100 NTU).

Bo et al. (2011), studying the use of a bioflocculant compound (CBF) as an aid to aluminum sulfate to treat synthetic water (15 NTU) in the presence of humic acid, found results similar to this research: dosage of aluminum sulfate (7 mg·L$^{-1}$), coagulant aid dosage (2 mg·L$^{-1}$) and the tests were conducted in a basic medium (8.3–8.7).

In a complementary study, Bo et al. (2012) sought to understand the effect of pH on the formation and structure of the flocs formed by the same coagulants mentioned above. The researchers observed that the most basic medium provided larger, more compact flocs that formed faster than in an acid medium.

The contour plot generated from the model obtained shows the relationship between an adjusted response and two continuous variables in a two-dimensional view, in which points of the same response value are connected to produce the contour lines. Figure 1(a)–1(c) present the turbidity removal for the chosen conditions found by the model (A = 5 mg·L$^{-1}$, B = 2.5 mg·L$^{-1}$, C = 9). Furthermore, Figure 1(d) shows the turbidity removal for pH 7.

An unexpected observation was made from Figure 1(c): high turbidity removal when the dose of *G. ulmifolia* was higher than 2 mg·L$^{-1}$ and the dose of aluminum sulfate was adjusted to 0 mg·L$^{-1}$ for pH 9. However, conclusions from this finding should be reached with care as it can be observed from the contour plot that there is only one experimental point around these conditions. Moreover, CCRD can present less power of prediction around these conditions (axial points).

From Figure 1(d), it can be noted that an increase in the dose of *G. ulmifolia* for doses of aluminum sulfate smaller than 5 mg·L$^{-1}$ results in greater removal of turbidity. This observation corroborates the hypothesis that this novel coagulant aid has the potential to reduce the dose of aluminum sulfate employed and still achieve satisfactory turbidity removal.

Several conditions were observed for which satisfactory turbidity removal occurred (>77 NTU). This is interesting and should be evaluated along with the cost of production and operation. In this way, the optimum conditions can be selected based both on technical and economic aspects. Therefore, it is necessary to evaluate the cost of production of *G. ulmifolia* as a coagulation aid.

Considering the consequences of the proposed equation, the performance of the *G. ulmifolia* as a coagulant aid...
showed the potential to reduce the dose of aluminum sulfate, the main coagulant.

**Experimental validation**

In order to verify the model obtained, validation tests were carried out for the selected point (Figure 2) and for another six random points drawn considering the interval studied for each variable (Figure 3).

In Figure 3, the values predicted by the model are related to the observed values of removed turbidity resulting from the validation tests. The continuous line illustrates the ideal correspondence \( y = x \) where the observed values correspond to the values predicted by the model.

It is assumed that the model is deficient in predicting the behavior of extreme points, since these conditions are far from the central point and are not tested in this type of experimental design.

The selected model is considered satisfactory, as in the validation stage, six out of nine points yielded good results. Moreover, the three validation tests performed at the chosen optimal point (highlighted in yellow in the graph), with observed responses of 75.8 ± 0.3 NTU, versus a response predicted by the model of 77 NTU.

**CONCLUSIONS**

The results showed individual, quadratic and effects found as significant in coagulation-flocculation process studied \((p \leq 0.05)\). The model obtained presented good values for predicting turbidity removal, with an \( R^2 \) equal to 93.2%. The promising conditions obtained were pH 9, a dose of aluminum sulfate of 5 mg·L\(^{-1}\) and a dose of \( G. \) ulmifolia extract of 2.5 mg·L\(^{-1}\) with a prediction of 77 NTU of removed turbidity. Under these conditions, with the addition of the same influent solution (80 NTU), there was an excellent performance in the validation tests. Therefore, \( G. \) ulmifolia as a coagulant aid proved to be promising for reducing the dosage of the primary coagulant.

**DATA AVAILABILITY STATEMENT**

All relevant data are included in this paper.

**REFERENCES**


First received 5 April 2020; accepted in revised form 28 August 2020. Available online 8 September 2020.