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# Dissolved air flotation optimization for treatment of dairy effluents with organic coagulants



Magno do Santos Pereira, Alisson Carraro Borge[s\\*,](#page-0-0) Gustavo Lopes Muniz, Fernanda Fernandes Heleno, Leda Rita D'Antonino Faroni

*Federal University of Viçosa, Brazil*



# **1. Introduction**

Dairy industries consume a high volume of water for a variety of purposes, including milk processing units, cleaning structures, and sanitizing and disinfecting structures. On average, 2.5 L of water are spent for each liter of processed milk [\[1,](#page-4-0)[2](#page-4-1)]. Dairy wastewater is characterized by high biochemical oxygen demand (BOD), chemical oxygen demand (COD), high dissolved solids, suspended solids, oils & greases, and nutrients, such as ammonia or minerals and phosphates, which require adequate attention before discharge to the environment [[3](#page-5-0)[,4\]](#page-5-1).

Among the methods used to treat dairy wastewater, most are based on physicochemical or biological principles. Biological methods require large area, long treatment time, and low efficiency when applied as a single treatment system. On the other hand, physicochemical methods, such as dissolved air flotation (DAF), have been a promising technology that has gained prominence and has been applied as a Chemically Enhanced Primary Treatment (CEPT) in countless wastewater treatment industries.

CEPT is a technology that uses coagulants to enhance the removal of pollutants in the primary phase of treatment. The addition of coagulants in the process aims to assist the flocculation of colloidal particles

present in the medium so that they are removed by sedimentation or flotation. However, the use of chemical coagulants in the process generates secondary pollution due to sludge produced in large volumes that contains toxic substances. This becomes an environmental liability [[5](#page-5-2)] and causes companies to invest even more in technologies for the treatment and disposal of this sludge, increasing operating costs [[6](#page-5-3),[7](#page-5-4)].

Several organic and plant-based coagulants have been tested in the treatment of water and wastewater in place of chemical coagulants [[8](#page-5-5),[9](#page-5-6)[,10](#page-5-7),[11\]](#page-5-8).

Polyacrylamide (PAM) is an acrylamide-derived flocculant polymer widely used as a coagulant due to its high molecular weight, water solubility, and low cost [[12](#page-5-9)[,13](#page-5-10)]. Ahmad et al. [\[14](#page-5-11)] proved that the addition of PAM improved coagulation performance using aluminum polychloride (APC) in wastewater treatment. Aguilar et al. [\[15](#page-5-12)] observed that coagulation efficiency and flocs settling rate when using ferric sulfate, aluminum sulfate, and APC coagulants can be improved by the addition of anionic polyacrylamide (APAM), and the required amount of coagulants can also be reduced by the addition of APAM.

Plant tannin-based coagulants have also been widely used in water and wastewater treatment systems. Tanfloc SG (Tanac) is a tanninbased product, modified by physicochemical process, and has great

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<span id="page-0-0"></span><sup>⁎</sup> Corresponding author.

*E-mail address:* [borges@ufv.br](mailto:borges@ufv.br) (A.C. Borges).

potential as a flocculant [\[16](#page-5-13)]. Its use has become attractive due to the advantages presented by natural coagulants, such as little sludge generated, lower concentration of heavy metals in sludge, and water alkalinity not being consumed during treatment, in addition to being organic and biodegradable.

However, little is known about coagulation/flocculation efficiency when using Tanfloc associated with PAM in the treatment of dairy wastewater. Considering this and the available technologies, there is a need to achieve greater efficiency in the wastewater treatment process at a lower cost and in a sustainable manner. Thus, the present work aims to evaluate the performance of Tanfloc associated with PAM in the treatment of dairy wastewater by dissolved air flotation through the planning of experiments in a central composite rotatable design (CCRD).

#### **2. Material and methods**

The experiment was conducted both at the Agricultural Product Pre-Processing and Storage Sector and at the Environmental Quality Laboratory of the Department of Agricultural Engineering, Federal University of Viçosa (UFV), located in Viçosa, Brazil. All analyses performed followed methods available in American Public Health Association et al. [\[17](#page-5-14)].

# *2.1. Wastewater materials*

The experiment used synthetic dairy wastewater (SDW), as suggested by Silva et al. [\[18](#page-5-15)]. According to the authors, this formulation satisfactorily characterizes a synthetic effluent, which does not contain whey from cheese making.

[Table 1](#page-1-0) presents the average composition of the synthetic effluent used in the experiment.

A sample of real wastewater was obtained from the Monte Celeste dairy factory (Viçosa, MG, Brazil), whose characteristics are presented in [Table 2,](#page-1-1) and it was also used for the final evaluation of the treatment efficiency of the proposed system.

# *2.2. Coagulation/flocculation*

In each assay, a volume of 2 L of SDW sample was used and placed in a 2 L beaker. The samples were agitated in a jar test apparatus at 320 rpm (G = 220 s<sup>-1</sup>) for solubilization of the products and coagulation (destabilization) of the particles.

In each assay, the cationic PAM and Tanfloc pop coagulants were added. The pH of the samples was adjusted after dosing and complete dissolution of the coagulants using a pH meter and with the addition of 1:1 v/v either NaOH or HCl solutions.

After adjusting the pH, a reduced flocculation was performed by stirring the SDW for 5 min at 120 rpm (G =  $80 s^{-1}$ ) for the formation of microflocs which, according to Richter [\[19](#page-5-16)] and Edzwald [\[20](#page-5-17)], are suitable for high particle removal efficiency in the process.

After flocculation, the samples were transferred to the flotation

# <span id="page-1-0"></span>**Table 1**





COD: chemical oxygen demand; O&G: oils and greases; TS: total solids; TSS: total suspended solids; SD: standard deviation.

<span id="page-1-1"></span>





TSS: total suspended solids; COD: chemical oxygen demand; BOD: biochemical oxygen demand; O&G: oils and greases; TN: total nitrogen; TP: total phosphorus; SD: standard deviation.

# <span id="page-1-2"></span>**Table 3**

Flotation system operating conditions.



column slowly along the flotation wall, so that the flocs formed did not break. Then, the flotation tests were performed.

# *2.3. Flotation process and system used*

The operating conditions of the flotation system are presented in [Table 3](#page-1-2). The fixed parameters were adopted according to the recommendation of Edzwald [\[20](#page-5-17)] and based on preliminary tests.

The flotation process was performed in the same way for all tests. Air was injected through the lower inlet and dissolved in potable water under pressure in the saturation chamber until it reached the pressure of 10 bar, being adjusted by the pressure regulating valve.

After reaching the pressure inside the saturation chamber, the air injection was maintained for 2 min for water saturation. Afterwards, the piping valve connecting the saturation chamber to the flotation column was opened, injecting supersaturated air water into the flotation column.

To maintain the pressure set in the saturation chamber during valve opening, air was injected through the upper opening of the saturation chamber to compensate for pressure drop because of water exiting the chamber. A maximum pressure drop of 2 bar was achieved during the entire flotation column water release process.

The system inlet flow rate was adjusted to 5 L min−1, and the flotation rate in the flotation column was set to 10.0  $\pm$  1 cm min<sup>-1</sup> using both needle valves.

After injection of the water volume corresponding to the pre-established recirculation ratio (20 %), the chamber water outlet valve was closed, sealing the water and gas injection in the flotation column. Then 5 min after the start of the flotation, 500 mL of samples were collected from the lower portion of the flotation column in each run.

# *2.4. Experimental planning*

The central composite rotatable design (CCRD) was used to determine the best pH value and the best coagulant doses, thus determining the best treatment by the surface response method. Thus, an experimental design was organized in 3 blocks with  $2<sup>3</sup>$  factorial points, 2  $\times$  3 axial points, 6 repetitions in the central point, and an  $\alpha$  = 1.633, totaling 20 trials.

<span id="page-2-0"></span>**Table 4**

CCRD values for the three factors tested.

| Variable  | Symbol               | Level          |                        |                        |                        |                        |  |
|---|----------------------|----------------|------------------------|------------------------|------------------------|------------------------|--|
|   |                      | -α             | $-1$                   | $\Omega$               | $+1$                   | $+\alpha$              |  |
| pH (dimensionless)<br>Tanfloc (mg $L^{-1}$ )<br>PAM $(mg L^{-1})$ | X1<br>$X_2$<br>$X_3$ | 4.37<br>0<br>0 | 5.00<br>100.0<br>100.0 | 6.00<br>350.0<br>350.0 | 7.00<br>600.0<br>600.0 | 7.63<br>758.3<br>758.3 |  |

PAM: polyacrylamide.

[Table 4](#page-2-0) presents the factors and levels used in the planning.

In this stage, residual turbidity was the response variable chosen to obtain the optimal treatment. This is because it can be quickly analyzed and mainly because it presents good representation of the flotation process behavior. Residual turbidity was measured using the Thermo Scientific Orion AO3010 portable turbidity meter. At the end of each flotation test, the residual turbidity value was corrected by a correction factor of 1.2 (corresponding to the recirculation ratio  $= 20$  %), due to the dilution of the sample by water injection into the flotation column.

The statistical planning of the CCRD, the results analysis, and the generation of the prediction model and graphs were made using tools from Minitab 17 software. The optimal response was determined using the *Response Optimizer* tool, which determines the combination of values of each model prediction factor to generate the best response, adopting a residual turbidity value of 0 NTU.

In the next step, the obtained model was validated. This was also done using the synthetic effluents through 3 tests at the determined optimum point. The measured and estimated residual turbidity values of the model were compared with the parameters generated by the statistical program after response optimization (confidence interval and model prediction interval at a significance level of 5 %).

At the end of the study, the best condition obtained was applied to the treatment of real wastewater collected from the Monte Celeste dairy factory to evaluate the system efficiency, applying the optimal treatment previously determined. The following parameters were analyzed: residual turbidity, COD, BOD, color, O&G, TSS, TN, and TP.

For statistical analysis of the results, the Minitab 17 (Minitab) [\[33](#page-5-18)] computer application was used for generation of the predictive model, response optimization, and graph construction.

#### **3. Results and discussion**

#### *3.1. Results obtained in experimental runs*

The residual turbidity results, as well as the removal efficiencies, are presented in [Table 5.](#page-2-1)

Based on the results, turbidity removal efficiency was above 90 % for most of the applied treatments. Lower efficiency was observed when 600 mg L−1 of PAM and 100 mg L−1 TanFloc at pH 5.00 was used, where the residual turbidity observed was 157.2 NTU (removal efficiency 74.8 %). The decrease in turbidity removal efficiency can be explained by the increased dosage of both coagulants in the process, which contributed to the increase in concentration of suspended solids in the medium.

Similar results were observed by Hameed et al. [[11\]](#page-5-8), using Tanfloc to treat municipal wastewater. According to the authors, the increase in coagulant dosage from a given dose (35 mg  $L^{-1}$ ) has no significant effect on the removal of water turbidity. On the contrary, it shows a decrease in turbidity removal efficiency, especially in waters with low initial turbidity value. Similarly, Ma et al. [\[21](#page-5-19)] observed that overdosing of PAM prevents floc growth, reducing sedimentation efficiency.

However, it can be observed that at pH 7.00, the dosage of 600 mg L<sup>-1</sup> of PAM and 100 mg L<sup>-1</sup> TanFloc removed 90.5 % of turbidity, and the observed residual turbidity was only 59.2 NTU. This result justifies the influence of pH, which was significant at 5 % significance.

<span id="page-2-1"></span>**Table 5** Results of central composite design used to optimize residual turbidity.

| $\mathbf{1}$<br>1<br>350<br>350<br>46.3<br>92.6<br>6.0<br>$\overline{2}$<br>$\mathbf{1}$<br>6.0<br>350<br>350<br>53.9<br>91.4<br>3<br>$\mathbf{1}$<br>100<br>600<br>70.4<br>7.0<br>88.7<br>4<br>$\mathbf{1}$<br>7.0<br>600<br>100<br>59.2<br>90.5<br>5<br>$\mathbf{1}$<br>5.0<br>600<br>600<br>130.8<br>79.1<br>6<br>1<br>100<br>5.0<br>100<br>28.1<br>95.5<br>7<br>$\overline{2}$<br>6.0<br>350<br>350<br>34.1<br>94.5<br>8<br>2<br>6.0<br>350<br>350<br>29.2<br>95.3 | Efficiency (%) |
|--|----------------|
|  |                |
|  |                |
|  |                |
|  |                |
|  |                |
|  |                |
|  |                |
|  |                |
| 9<br>2<br>100<br>600<br>24.8<br>96.0<br>5.0  |                |
| 10<br>$\overline{2}$<br>100<br>100<br>58.0<br>7.0<br>90.7  |                |
| $\overline{2}$<br>600<br>11<br>7.0<br>600<br>19.4<br>96.9  |                |
| $\overline{2}$<br>12<br>600<br>100<br>5.0<br>157.2<br>74.8   |                |
| 3<br>13<br>57.1<br>6.0<br>758<br>350<br>90.9   |                |
| 3<br>14<br>6.0<br>350<br>758<br>68.9<br>89.0   |                |
| 3<br>15<br>350<br>350<br>88.8<br>85.8<br>4.4   |                |
| 3<br>16<br>6.0<br>$\mathbf{0}$<br>350<br>15.2<br>97.6  |                |
| 3<br>17<br>350<br>350<br>44.9<br>92.8<br>7.6   |                |
| 3<br>18<br>22.7<br>6.0<br>350<br>350<br>96.4   |                |
| 3<br>19<br>6.0<br>350<br>350<br>34.4<br>94.5   |                |
| 3<br>20<br>6.0<br>350<br>0<br>86.9<br>81.6   |                |

PAM: polyacrylamide; T: turbidity.

<span id="page-2-2"></span>**Table 6**





\*: significant; ns: non-significant; PAM: polyacrylamide; DoF: degree of freedom; SS: sum of squares; MS: mean square.

### *3.2. Analysis of variance*

The total variation of the turbidity dependent variable to the regression and residue model (ANOVA) is presented in [Table 6](#page-2-2).

The quadratic regression shows that the model was significant (p  $\leq$ 0.05), since the calculated F value, equal to 47.91 (MS/SS), was greater than the value of F critical ( $F_{0.05; 9,10} = 3.02$ ). In addition, Box et al. [[22\]](#page-5-20) explains that for a regression to be not only significant but also useful for predictive purposes, the F<sub>value</sub>/F<sub>critical</sub> ratio must be greater than three, a condition that was met by the model.

It can also be observed that the model lack of fit was not significant (p  $> 0.05$ ), since the F value for the misadjustment (MS Lack of fit / MS Pure error) was less than the F critical ( $F_{0.05; 7,3} = 8.89$ ), which is desirable in obtaining a regression model.

The p-value  $(p < 0.001)$  of the model shows that the second-order polynomial model fit well with the experimental data. This is because when this value is lower, the model fit is better. The values of the coefficients of determination, which measure the fit of the model to the observed data, are presented in [Table 7.](#page-3-0)

The standard deviation (SD) of the distance between the data values and the adjusted values was 7.45 NTU ([Table 7](#page-3-0)). Such a measurement

# <span id="page-3-0"></span>**Table 7**





SD: standard deviation;  $R^2$ : coefficient of determination.

represents how far the turbidity data values fall from the values adjusted by the model. A lower value of S suggests better description of the response by the model. Therefore, a good result was obtained regarding this parameter.

The value of the coefficient of determination  $(R^2 = 0.9784)$  indicates that only 2.16 % of the total variation cannot be explained by the empirical model. This is good because more variation that is explained by the model results in data points that are closer to the fitted regression line. According to Olmez [\[23](#page-5-21)], a larger  $R^2$  suggests a better model and lesser error, and models with  $R<sup>2</sup>$  values greater than 0.80 are more reliable for predictive purposes.

Once the model has been adjusted, the value of the adjusted coefficient of determination (adjusted  $R<sup>2</sup>$ ) should be considered. This value it is the percentage of variation in response that is explained by the model and is adjusted for the number of model predictors relative to the number of observations. Thus, 95.89 % variation in response can be explained by the adjusted model.

On the other hand, the predicted  $R^2$  should be used to predict answers for new observations. In this study, the value was high (90.92 %), and thus, the model has a high predictive capacity. Predicted  $R^2$  may be more useful than adjusted  $R^2$  for model comparison because it is calculated from observations that are not included in model calculation (Minitab 17). The difference between  $R^2$  and predicted  $R^2$  must be less than 0.2 for the model to be reliable. This relationship was met by the proposed model, indicating the absence of over-adjustment, and therefore, it is useful for predictive purposes.

#### *3.3. Mathematical model, contour plots, and response surface*

The mathematical model obtained to represent the residual turbidity as a function of the pH values and coagulant doses with their respective coefficients is presented in Eq. [\(1\).](#page-3-1)

<span id="page-3-1"></span>**Tresidual** = 272.7−113.1 pH + 0.9865 PAM - 0.179 TanFloc + 12.3  $pH^{2}$  + 0.000288 TanFloc<sup>2</sup> - 0.1424 pH  $\times$  PAM - 0.000151 PAM  $\times$  $TanFloc$  (1)

From this mathematical model, contour and response surface graphs were generated for the turbidity response variable, shown in [Fig. 1](#page-3-2). These analyses kept one variable at the central point and the other two varying within the range studied. Through the graphs obtained, it is possible to confirm the significant interaction of the factors studied.

[Fig. 1](#page-3-2) shows that the maximum and minimum points are within the experimental region. Thus, the contour plot shows hyperbolic characteristic [\(Fig. 1](#page-3-2)a). In contrast, when the PAM dosage was fixed at the midpoint and Tanfloc doses varied, the contour plot exhibited a circle ([Fig. 1b](#page-3-2)). According to Nair et al. [\[24\]](#page-5-22), the hyperbolic characteristic presents a saddle point that is an inflection point between a relative maximum and a relative minimum and is neither a minimum point nor a maximum point.

The contour plot of Tanfloc dosage in relation to pH shows that the ideal conditions for turbidity removal were located in the region where the Tanfloc dose ranged from 250 to 550 mg L<sup>-1</sup> and pH from 5.8 to 7.2, maintaining PAM dosage at 350 mg L<sup>-1</sup> ([Fig. 1](#page-3-2)a).

The process of coagulation of colloidal particles occurred through charge neutralization, adsorption, and bridging, since the coagulants employed are organic polymers. Thus, chemical bridges are formed when the colloidal particles are adsorbed on the surface of the various polymer chains [[25\]](#page-5-23).

According to Hameed et al. [\[11](#page-5-8)], Tanfloc is a tannin-based coagulant, which are polyphenolic compounds with high water solubility and molecular weight ranging from 500 to a few thousand Daltons, presenting cationic character. Thus, the modified compound, Tanfloc, has the same characteristics as pure tannin and other added features. These new features give it numerous applications in the coagulation process through charge neutralization.

In turn, PAM exhibits flocculating action similar to conventional polymer flocculants by destabilizing and neutralizing negatively charged colloidal particle charges, making them unstable. It also presents coagulant action in various pH ranges [[26,](#page-5-24)[10](#page-5-7)], a fact that justifies the results found in this work.

Rozeno et al. [[27\]](#page-5-25) studied biodiesel industry effluent treatment with tannin as a coagulant associated with PAM. They observed that with a dosage of 860 mg L<sup>-1</sup> of coagulant at pH 8.00, there was 92 % removal of turbidity. The removal mechanism, being the same observed in this work, was the formation of bridges, adsorption, and neutralization of charges.

Ribeiro et al. [\[28](#page-5-26)] used tannin (250 mg L<sup>-1</sup>) associated with PAM (8 mg  $L^{-1}$ ) in the treatment of industrial laundry effluent, and the authors observed that the application of PAM-associated tannin provided values close to 100 % turbidity removal in acidic pHs and near the neutral pH

<span id="page-3-2"></span>

**Fig. 1.** Contour plots relating residual turbidity as a function of pH for PAM (a) and TanFloc (b) doses.

<span id="page-4-2"></span>

Model validation test data obtained.

| рH  | PAM $(mg L^{-1})$ | Tanfloc (mg $L^{-1}$ ) | <sup>1</sup> residual measured | <sup>1</sup> residual estimated | CI(95%)      | PI (95 %)    |
|-----|-------------------|------------------------|--------------------------------|---------------------------------|--------------|--------------|
| 7.6 | 758.3             | 212.3                  | 62.0                           | 0.0                             | $-22.2:22.2$ | $-27.7:27.7$ |

<span id="page-4-3"></span>**Table 9**

Results of CEPT with dissolved air flotation using real wastewater.



COD: chemical oxygen demand; BOD: biochemical oxygen demand; O&G: oils and greases; TSS: total suspended solids; TN: total nitrogen; TP: total phosphorus; CV: coefficient of variation.

region, as well as approximately 80 % reduction in the basic range. According to the same authors, the tannin coagulation process in this dosing range is explained by the sweep mechanism. For this, interactions occur in greater quantity and involve all particles in the effluent suspension and participate in the system, minimizing turbidity in the medium, as also observed in this work.

#### *3.4. Optimization and validation of the mathematical model*

Response surfaces can be analyzed for maximum or minimum responses and corresponding optimum conditions. With multiple responses, optimal conditions can be met when all parameters meet the desirable criteria [[24\]](#page-5-22). When there are more than three independent variables, Montgomery [[29\]](#page-5-27) mentions that it becomes more difficult to find the conditions that satisfy all the answers simultaneously, thus multicriteria methodologies can be followed.

In this work, the optimized condition was defined using the *Response Optimizer* from the Minitab software in order to minimize the residual turbidity. This resulted in obtaining the following point: PAM dose of 758.3 mg L<sup>-1</sup>, with Tanfloc dose of 205.4 mg L<sup>-1</sup> at pH 7.6. At this point, the model was validated.

To confirm the validity of the mathematical model, additional tests with three repetitions were performed at the optimal point for confirmation. The results are presented in [Table 8.](#page-4-2)

The average residual turbidity obtained in the experimental tests was 62.0 NTU. In contrast, the model estimated that under this condition, the effluent after treatment would be free of turbidity, expressed by the value of predicted residual turbidity of 0.0 NTU. On other hand, the results are according to CI and PI predicted ranges.

Using the mathematical model (Eq.  $(1)$ ) to foresee the residual turbidity at the optimized pH value of 7.6, PAM value of 758.3 mg L<sup>-1</sup> and Tanfloc concentration of zero, a Tresidual of 50.4 NTU and an efficiency of 91.9 % are predicted. On other hand, testing PAM concentration equal to zero, at pH 7.6 and Tanfloc concentration of 212.3 mg L<sup>-1</sup>, a T<sub>residual</sub> of 98.0 NTU and an efficiency of 84.3 % are predicted.

Comparing this results with the values in [Table 5](#page-2-1), it can be seen that at pH of 7.6 the TanFloc added isolated and with an lower dose compared to the PAM can reach treatment efficiency above 90 %, indicating that depending on the desirable efficiency required for the treatment, the TanFloc is more indicated due to less sludge generation and potential reduction on treatment costs of the dairy wastewater, transport and sludge disposal.

#### *3.5. System application in real wastewater treatment*

After obtaining the best conditions and validating the model obtained, real raw wastewater was collected from the Monte Celeste dairy factory, whose initial characteristics are shown in [Table 2.](#page-1-1) The results obtained from the treatment are shown in [Table 9](#page-4-3).

Excellent results were observed when the treatment was applied to the real raw dairy effluent, presenting expressive reductions in solids, organic matter, and nutrients.

Ayeche [[30\]](#page-5-28) treated dairy effluent with an alternative coagulant obtained from the residue from acetylene production, and the authors reported reductions in COD, BOD, turbidity, TSS, and TP by 49 %, 54 %, 92 %, 93 %, and 83 % with a coagulant dose equal to 4000 mg  $L^{-1}$  at alkaline pH (7–11). Only TP removal, found by this author, was above the removal efficiency observed in this study, but it is noteworthy that the dosage used in this study was well below that used by Ayeche [\[30](#page-5-28)].

Mansoorian et al. [[31\]](#page-5-29) reported maximum removal efficiency of COD, BOD, NH<sub>3</sub>, NH<sub>4</sub><sup>+</sup>, dissolved and suspended phosphorus,  $SO_4^2^-$ , and TSS at 90.46 %, 81.72 %, 73.22 %, 69.43 %, 31, 18 %, 72.45 %, 39.43 %, and 70.17 %, respectively, in the treatment of dairy effluents with microbial fuel cells. According to Taufer et al. [[32\]](#page-5-30), the treatment of dairy effluent as an industrial effluent is difficult to maintain consistent and achieve a homogeneous result of contaminant removal because of the large variation in pollutant loads.

The use of coagulants PAM and Tanfloc in the dissolved air flotation for the treatment dairy wastewater showed promising results due the high efficiencies obtained when used combined or isolated. Other advantage is that the sludge generated during the treatment process can be used as an fertilizer in agriculture after proper treatment.

#### **4. Conclusion**

The use of Tanfloc associated with polyacrylamide has shown great potential to be used in the treatment of dairy wastewater by CEPT with dissolved air flotation. The central composite rotatable design was efficient in modeling the responses of the wastewater treatment process, demonstrating the significant effect of the independent variables studied on the turbidity removal response variable. The optimal treatment point was obtained with PAM dosage of 758.3 mg L−1 and Tanfloc dosage of 212.3 mg L<sup>-1</sup> at pH 7.6.

# **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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