

**Tannin and chemical-based agents for coagulation and flocculation of landfill leachate**

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## SUMMARY

Effluents generated in landfills are difficult to treat and require different forms of treatment to reach the necessary polishing for disposal. Leachate is usually treated in stabilization ponds where pollutants are significantly removed, but it might not be enough for disposal. Thus, we aimed at evaluating the applicability of the natural coagulant Tanfloc SG and the chemical coagulant PAC, with the addition of the anionic flocculation auxiliary polymer (PA), in the coagulation / flocculation processes as a post-treatment of landfill effluents. The tests were carried out by means of a jar test, with coagulant dosage ranging from 0 - 1,250 mg.L<sup>-1</sup> and pH values ranging from 5 - 9, in three decantation times. The results showed that both Tanfloc SG and PAC are efficient in clarifying the effluents, even though they act well in different dosage values of coagulants and pH. The polymer dosage did not generate significant improvements in the variations of the variables studied, however, in some experiments it was shown to expand removal range in regards to some variables. The PAC coagulant showed to be efficient in the removal of suspended solids, dissolved materials, colloids and humic substances, and Tanfloc SG acted mainly in the removal of solids.

**KEYWORDS:** Waste. Effluents. Coagulants. Flocculation. Pollutant removal.

## 1 INTRODUCTION

The disposal of solid waste can be a serious environmental problem depending on how it happens. Disposal in inappropriate places, such as in dumps, causes degradation and direct pollution of the environment that receives the waste (RABONI et al., 2013). The destination in landfills reduces the risks of direct contamination, however, the release of gases and leachate must be controlled (KORF et al., 2008).

The formation of leachate (chorume) is complex, and it is related to several factors, such as: the origin of the waste and its composition; local climate; the shape of the landfill and its operation and age. Leachate demands, as well as other effluents, for treatment before discharge into water resources (MORAVIA et al., 2011).

Effluent treatment is an important operation, since the removal of contaminants will depend on it. As the contaminants have different physical, chemical and biological characteristics, different treatments must be used for the effluents so that they reach quality for disposal in water bodies (FELICI et al., 2013; LIMA et al., 2015; MOODY; TOWNSEND, 2016).

In general, the treatment of effluents that contain organic matter occurs by biological processes, such as stabilization ponds. These can achieve high removals of biodegradable organic matter, however, they do not remove recalcitrant organic materials, which for the most part are humic substances, responsible for the color of the effluent (SPERLING, 2006; VIVAN et al., 2009).

Thus, depending on the concentrations and on the presence of other pollutants, there is a need for alternative complementary treatments, such as coagulation / flocculation (YUSOFF et al., 2018; YONG et al., 2018; KAWAHIGASHI, 2014; VELIZ et al., 2015). The main function of the physical-chemical process based on coagulation / flocculation and subsequent decantation is to remove suspended and dissolved solids, which are difficult to remove in effluents such as the landfill leachate (PEDROSO et al., 2012).

Several parameters can affect the coagulation / flocculation process. In addition to the nature of the effluent, the type of coagulant and their dosage, fast and slow mixing speeds, the pH value of the effluent and the settling time are important factors in the process (IBRAHIM; YASER, 2019; QUEIROZ et al., 2011; KUMAR; VERMA, 2016).

Coagulants can be chemical or natural products. Chemical coagulants such as aluminum sulphate, ferrous sulphate, ferric sulphate, ferric chloride, chlorinated ferrous

sulphate, sodium aluminate and aluminum polychloride (PAC) are the most used ones due to their proven efficiency and low cost. Natural coagulants such as *Moringa oleifera* Lam, Tanfloc and Chitosan have been the object of many papers in order to check the feasibility of its use and to reduce the consumption of chemicals (CHOWDHURY et al., 2013).

The use of the chemical coagulant PAC is widespread in Brazil, including in the treatment of water supplies and effluents. Natural coagulants can also be used in water and wastewater treatment. The Tanfloc SG coagulant is a cationic polymer of low molecular weight, acting in the processes of coagulation and flocculation. Its main advantage in relation to chemical coagulants is the production of biodegradable sludge, which due to its organic composition, can be biologically degraded (PEDROSO et al., 2012).

In addition to the coagulant, the presence of flocculation aids can change the conditions and effectiveness of the coagulation / flocculation process, since the flocculation auxiliary polymer allows for an increase in the size of the flakes and, which thus leads to an increase in their decantation speed (CASTRO et al., 2012).

In this sense, the objective of this paper was to evaluate the process of coagulation / flocculation of leachate previously treated in stabilization lakes, depending on the type of coagulant, dosage of coagulants, initial effluent pH, and decantation times.

## **2 MATERIALS AND METHODS**

### **2.1 Effluent**

The effluent (leachate) used in the coagulation / flocculation tests was sampled at a landfill from the city of Sorriso, Mato Grosso State - Brazil. The leachate was collected at the exit of the facultative pond, after undergoing treatment in an anaerobic and facultative pond.

We carried out the leachate characterization in the laboratory, by determining parameters such as pH, UV absorbance at 254 nm (UV abs), color, turbidity, chemical oxygen demand (COD), electrical conductivity, total suspended solids (TSS), total Kjeldhal nitrogen and total phosphorus. All analytical procedures followed those established by the Standard Methods for the Examination of Water and Wastewater (APHA, 2012).

### **2.2 Coagulation/Flocculation tests**

The coagulation / flocculation / decantation tests were performed at different pH values (5, 7 and 9 respectively), and their adjustment was performed by using either concentrated sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) or 1 N sodium hydroxide (NaOH).

The coagulants we used in the tests were aluminum polychloride (PAC), a chemical coagulant, and Tanfloc SG, a natural coagulant. The anionic polymer, PA 0823, simply referred to as PA, was used as the flocculant. The Sanitation Company of Paraná (SANEPAR) supplied us with the PAC coagulant, and the company TANAC (Montenegro, Rio Grande do Sul), provided the anionic polymer. These coagulants and coagulants plus the PA were defined in the tests as four types of treatments: PAC, Tanfloc SG, PAC + PA and Tanfloc SG + PA.

The PAC coagulant was prepared in a 1% v/v solution and the Tanfloc SG coagulant was prepared in a 0.5% w/v solution. The coagulant dosages used in the tests were 0, 50, 150, 300, 500, 750, 1,000 and 1,250 mg L<sup>-1</sup>.

The anionic polymer, used as a flocculant, was prepared in 0.1% w / v solution and at a dosage of 1.0 mg L<sup>-1</sup>.

The coagulation / flocculation / decanting tests were performed on a bench scale, by jar testing with 0.25 L samples.

The experimental tests were carried out with the following operating conditions: rapid mixing time (RMT) of 2 minutes, rapid mixing speed (RMS) of 100 rpm, slow mixing time (SMT) of 20 minutes and slow speed of mixing (SMS) of 20 rpm. Subsequently, the samples were poured into Imhoff cones for decantation. The settling times evaluated were 20, 40 and 60 minutes. After each decantation time, the volume of the sludge decanted in the Imhoff cone was measured and samples of the treated leachate were collected for analysis of pH, turbidity, color and abs UV.

The determinations of turbidity and pH were performed in a turbidimeter and in a pH meter, respectively. The color determinations were performed in a colorimeter and the absorbance measurements in 254 nm were performed in a UV spectrophotometer.

### **2.3 Experimental design and statistical analysis**

The experimental design used was completely randomized, in an 8x3 factorial scheme, with eight different dosages of coagulant and 3 different pH levels, with a portion subdivided in the decantation time (3 times), applied in 4 combinations of coagulants, with 3 repetitions per treatment.

For parameters such as pH, UV abs, color and turbidity, the statistical analysis was performed according to the variation of these parameters. For the sludge, the statistical analysis was performed according to the volume produced.

An analysis of variance (ANOVA) was performed for comparison of results, and when differences were found, means comparison tests and regression analyses were performed, at a 5% significance level. Statistical analyzes were performed using the computer program Sisvar (FERREIRA, 2010).

### **2.4 Effluent Characterization – Leachate**

Table 1 below shows the characteristics of the effluent in terms of pH, turbidity, apparent color, UV abs, total suspended solids (SST), electrical conductivity (CE), chemical oxygen demand (COD), total nitrogen Kjeldahl (NTK) and total phosphorus.

Although turbidity is relatively low, color was quite high, indicating the presence of dissolved and colloidal solids in high concentrations. In addition, there is the presence of humic substances (measured indirectly by the amount of UV absorbed), and high concentrations of total phosphorus.

It is worth emphasizing that the leachate used in the tests was a leachate previously treated by means of anaerobic, facultative and maturation lagoons, located in the landfill area. When crossing the lagoons, part of the suspended solids were removed, and going through the facultative and maturing lagoons promoted the development of algae.

**Table 1: Characteristics of landfill leachate after treatment in lagoons and limits established by legislation for launching**

Leachate Parameters	Release limits*	Max concentration after mixing**				
		Class 1	Class 2	Class 3	Class 4	
Turbidity (NTU)	54,2	--	40,0	100	100	---
DQO (mg L <sup>-1</sup> )	712	--	----	----	----	----
Color (mg Pt L <sup>-1</sup> )	600 <sup>1</sup>	--	natural	75	75	----
PH	9,19	5-9	6-9	6-9	6-9	6-9
UV absorption (cm <sup>-1</sup> )	2,165	--	----	----	----	----
CE (S m <sup>-1</sup> )	7,19	--	----	----	----	----
NTK (mg L <sup>-1</sup> )	28,84	--	---	---	---	----
Total Phosphorus (mg L <sup>-1</sup> )	1,25	--	0,1	0,050	0,15	----
SST (mg L <sup>-1</sup> )	5040	--	----	----	----	----

\* Limit of discharge of effluents into surface water bodies in accordance with CONAMA Resolution No. 430, of 2011 (BRAZIL, 2011); \*\* Limits established according to the class of freshwater bodies defined in CONAMA Resolution No. 357, 2005 (BRAZIL, 2005). 1 Apparent color.

### 3 RESULTS AND DISCUSSION

The analysis of variance showed that there was a significant interaction ( $p < 0.5$ ) between the sources of initial pH variation and the coagulant dosage for the variation of pH, turbidity, color and UV abs. There was no variation in their values between the different settling times (20, 40 and 60 minutes).

For each of the tested treatments, PAC, PAC + PA, Tanfoc SG and Tanfloc SG + PA, polynomial regression models were adjusted to each of the evaluated parameters (pH, turbidity, color, UV abs and sludge volume) and they showed to be significant and satisfactory, posing high coefficients of determination. The coefficients of the adjusted models (regression models) and the coefficients of determination can be seen in the supplementary material.

#### 3.1 PH variation in regards to initial pH and dosage

The results of the pH variation showed the dynamics of this variation in relation to the initial pH values and the dosages used. Regarding the initial pH, we found that the variations were positive (reduction of the effluent pH from the beginning to the end of each experiment) for the initial acidic and basic pH values (close to 5 and 9), and neutral and negative variations (little variation or slight increase in pH) for the initial pH values close to the neutral value (7).

As for dosage, the higher it was the bigger the increase in pH variation for all the initial pH values, with variations of up to 20%. Regarding the PAC, PAC + PA, Tanfloc SG and Tanfloc SG + PA treatments, there were few differences between them, even with the use of the polymer.

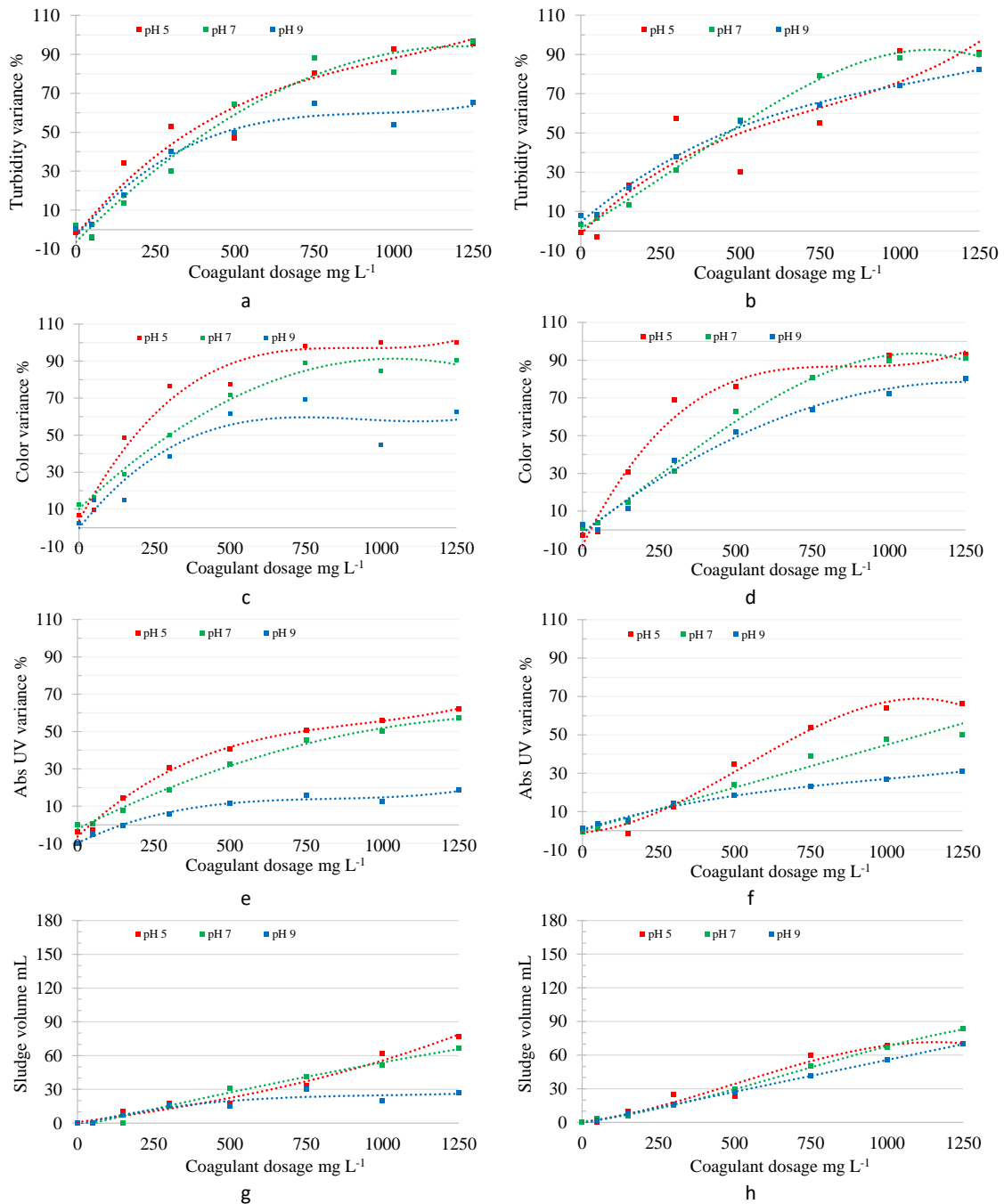
Small pH variations are reported for PAC (ZHANG et al., 2008) and for tannin (LEITE; HOFFMANN; DANIEL, 2019), as they are coagulants whose alkalinity consumption is low and null, respectively, which does not lead to significant changes in values of the leachate pH.

#### 3.2 Variation of turbidity, color, UV abs and sludge volume in regards to initial pH and dosage

In general, particle removal was obtained using coagulants and flocculants (PAC, PAC + PA, Tanfloc SG and Tanfloc SG + PA), because turbidity, color and UV abs were removed. The maximum removal values varied according to the initial pH and dosage, and were different for each of the coagulants and coagulants + PA.

The results from the PAC coagulant show that the largest removals of turbidity and color (> 90%) (Figures 1a and 1c) and UV abs (> 50%) (Figure 1e) took place when the effluent was acidic (pH = 5) and at higher dosage rates. The highest volumes of generated sludge were detected under these same conditions (Figure 1g).

**Figure 1: Variation of turbidity vs initial pH and dosage of the coagulant PAC (a), PAC + PA (b). Color variation vs initial pH and dosage of PAC (c), PAC + PA (d) coagulant. Variation of UV absorption vs initial pH and dosage of the coagulant PAC (e), PAC + PA (f). Sludge volume vs initial pH and dosage of the coagulant PAC (g), PAC + PA (h)**



Not only does the pH determine the surface loads of natural organic matter (NOM) molecules, it also significantly affects the hydrolysis of the PAC coagulant, that is, the interaction of the aluminum in the PAC coagulant with the NOM present in the effluent (Al-MON) (YANG et al., 2011), reflecting on the removal of turbidity, color and UV abs.

Yong et al. (2018) found the best removal rates for COD, total solids and color at a basic pH using the aluminum sulfate coagulant. According to the authors, this removal happened due to the "sweep floc-coagulation" phenomenon, where the dominant species of amorphous aluminum hydroxide ( $\text{Al}(\text{OH})_3$ ) are normally formed during the hydrolysis of aluminum in water. And then  $\text{Al}(\text{OH})_3$  traps or adsorbs the colloidal material and color in the metal hydroxides formed by hydrolysis.

The highest volumes of sludge produced by the PAC coagulant are in the highest dosage rates of the coagulants and in the pH values from neutral (pH = 7) to acid (pH = 9), which is where UV abs, color and turbidity vary the most (Figure 1g).

The addition of an anionic polymer to the PAC (PAC + PA) (Figures 1 b-d-f-h) promoted an increase in the removal of turbidity, color and UV abs in relation to pH, that is, the removal occurred in acidic and basic values. The sludge volume increased when the polymer was added to the PAC coagulant at a higher pH (7 and 9), corroborating the increased removal of turbidity, color and UV abs.

Polymers are elements that contribute to the increase of size and strength of particle aggregation, improving the quality of the effluent and accelerating the decantation of particles (LIMA JUNIOR; ABREU, 2018). Due to their high molecular weight and, therefore, appreciable length, polymers are able to form bridges or interconnect particles (LETTERMAN; YIACOUMI, 2011).

The presence of PA did not cause a coagulant dosage reduction effect, nor did it reduce the sludge volume, due to the formation of a denser sludge. In general, the PA was of little importance in the process of treating the landfill leachate. This result may be due to the low dosage used ( $1 \text{ mg L}^{-1}$ ). On the other hand, Lee et al. (2014) reinforce that polymeric flocculants must have a high flocculation capacity even at low dosage rates, since they are non-biodegradable products.

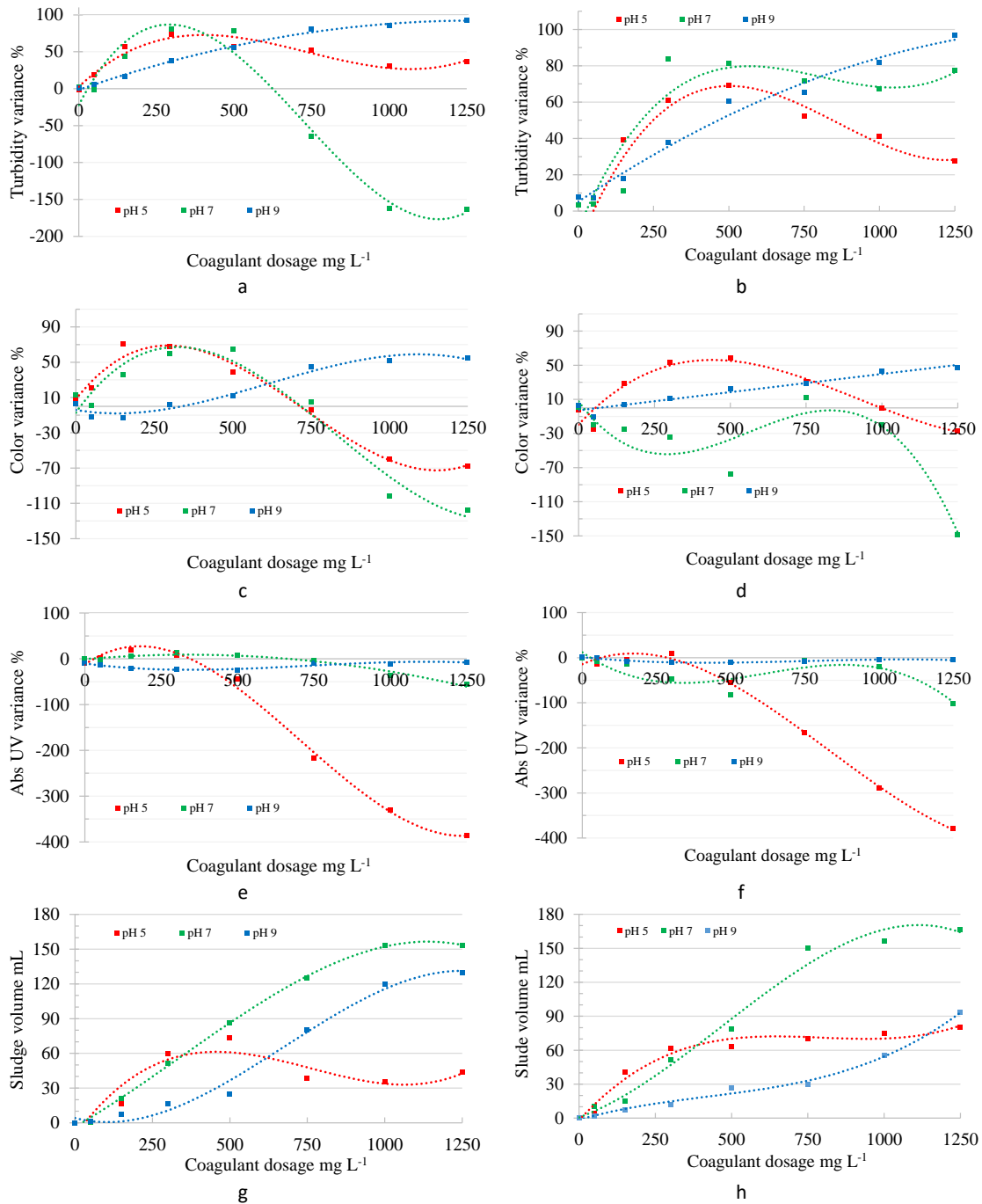
The experiments with the Tanfloc SG coagulant showed complex results when compared to the PAC coagulant. The removal of organic pollutants, such as humic substances, is due to the ability of polyphenols present in tannins of adsorbing organic ions and the electrical effect of the double layer formed by phenolic, carboxylic and amino groups (BRANCH et al., 2019).

The greatest turbidity removals (Figure 2a) were obtained in acid, neutral and basic values, in the coagulant dosage ranges close to 400, 300 and  $1,000 \text{ mg L}^{-1}$ , respectively. For color, the greatest removal rates (Figure 2c) were also obtained at all pH values, but in a value range of  $300 \text{ mg L}^{-1}$  for acidic and neutral pH values, while for basic pH values the best dosage rates were in the range close to  $1,000 \text{ mg L}^{-1}$ . The removal of UV abs (<30%) was verified only at an acidic pH, in dosage rates close to  $150 \text{ mg L}^{-1}$ .

The tannin-based coagulant is expected to have a positive charge, which is decreased as the pH increases, due to the deprotonation of amine and hydroxyl groups (LOPES et al., 2019). This behavior was reported by Ibrahim and Yaser (2019). The authors claimed that as the pH of

the sample increased, there was a decrease in the color removal of the leachate. Possibly because of the decrease in positive coagulant charges, the interaction by neutralizing charges diminished.

**Figure 2: Variation of turbidity as a function of initial pH and dosage of the coagulant Tanfloc SG (a), Tanfloc SG + PA (b). Color variation depending on the initial pH and dosage of the coagulant Tanfloc SG (c), Tanfloc SG + PA (d). Variation of UV absorption in regards to initial pH and dosage of the coagulant Tanfloc SG (e), Tanfloc SG + PA (f). Sludge volume in regards to the initial pH and dosage of the coagulant Tanfloc SG (g) and Tanfloc SG + PA (h).**





On the other hand, Banch et al. (2019) found an increase in the removal of color and chemical oxygen demand at pH values from 3 to 9. According to the authors, with pH between 7 and 9, particle absorption will be high because of the neutral electrical charge. Leachate cations can improve the coagulation process, neutralizing and destabilizing the negative charges of the residue and the functional groups of the coagulants by binding with tannin particles. Still, Barrado-Moreno; Beltrán-Herédia; Martín-Gallardo (2016) found that the pH value was not an important factor in the efficiency of algae removal by tannin at pH values minor than 8.

In the paper, we detected that the pH effect decreased as the coagulant dosage increased, with good color and turbidity removal at basic pH values, in high coagulant dosage rates.

Regarding the coagulant dosages, we found that, unlike what was observed for the PAC, increases in dosages did not necessarily lead to an increase in removals.

The study of coagulant dosages showed the behavior of particle destabilization. Upon reaching the ideal dosage it was found that the removal decreased. The reason for this may be the re-stabilization of organic particles when the optimum value has been reached.

Trends of increasing and decreasing the removal of elements related to organic material after an optimum point are characteristics of a coagulation-flocculation process controlled by charge neutralization (BOLTO, 1995). According to Ibahim and Yaser (2019), the excess coagulant is adsorbed by the colloidal particles present in the leachate. This leads to reversal of the colloid charges from negative to positive, and these colloids are re-stabilized, and have started to repel each other, causing less crowding.

The low removal of UV abs by Tanfloc SG shows that it was not efficient in removing humic substances. In addition, it appears that high doses of coagulant contributed to the increase in turbidity and color. Haamed et al. (2016) report that there was a serious deterioration in COD removal when the dose of Tanfloc SG was above 40 mg L<sup>-1</sup> (point of maximum COD removal). This may be due to the residue of Tanfloc SG in the water which acts as an organic pollutant. Furthermore, the authors claim that the coagulant remains as dissolved material if it is not incorporated to solids by the flocculation process.

Leite; Hoffmann; Daniel (2019), when studying the removal (harvesting) of algae by organic coagulants, found that the neutralization of loads and the formation of bridges acted together in the coagulation of the material. The concentration of algae cells is significant in effluents treated by lagoons, and the removal of these cells mainly promotes the removal of turbidity from the effluent (Kaya; Dilek; Gökçay, 2007).

The addition of the auxiliary flocculation polymer to the treatment with Tanfloc SG did not increase the removal of turbidity, but expanded the removal range in relation to the pH value. For color, the introduction of the polymer reduced the removals at a neutral pH.

For Tanfloc SG, the generated volumes of sludge are not related to the removal of color and UV abs, and are related to the removal of turbidity. In addition, the values obtained in the production of sludge with the Tanfloc SG coagulant are twice the volume compared to the PAC coagulant, indicating that possibly the sludge from Tanfloc SG does not settle as well as the PAC sludge.

Color removal, turbidity, UV abs have been studied by other authors. Ibrahim and Yaser (2019) observed an increase in color removal according to the increase in the dosage of

tannin-based coagulant for pre-treated effluent in a biological system. Pedroso et al. (2011), in tests with Tanfloc SG as a primary treatment for leachate, obtained better results when removing the apparent color (96%) at a basic pH (pH equal to 9) and concentration of  $100 \text{ mg L}^{-1}$ . These authors claim that the removal of apparent color points to an important removal of dissolved compounds.

Pedroso et al. (2012) studied the application of different doses of Tanfloc SG in different leachate samples, collected over a period of three months. The authors chose to evaluate different leachate samples over time because their composition is extremely variable. Each sample represented the possible values (high, medium and low) found for the leachate in relation to the parameters studied such as color, turbidity and humic substances present in the landfill in Maringá, PR. The leachate collected was corrected to pH equal to 9 and a dosage of  $1,500 \text{ mg L}^{-1}$  was used, resulting in a color removal of 59.97% and turbidity of approximately 50%, a result lower than that found in this paper.

Felici et al. (2013) used ferric chloride in the treatment of biologically pretreated leachate and found high color removals (98%) at acidic pH values (pH 3). Despite the good removal, the authors reported low sedimentation speed, with volume stabilization after 20 hours. In this work, we verified volume stability after 20 min of sedimentation.

Wolf et al. (2015), aiming at the treatment of effluent from agrobusiness (dairy), under slightly acid pH conditions (pH equal to 6) and dosage of  $20 \text{ mg L}^{-1}$  of the coagulant Tanfloc SG with initial turbidity of 109 NTU, reported a turbidity removal of 71.2%.

Maximo (2007), evaluating the treatment of crude leachate, used dosages of Tanfloc SG that varied from 600 to  $2,400 \text{ mg L}^{-1}$ , and in the dosage of  $1,800 \text{ mg L}^{-1}$ , he verified the best conditions for removing turbidity, around 66%, and color removal reached a value around 56%. The author also stated that the addition of anionic polymer in dosage in the range of 1 to  $2.25 \text{ mg.L}^{-1}$ , did not promote significant variations in the removal of turbidity and color.

According to Pedroso et al. (2012), the efficiency in the treatment by coagulation / flocculation for the removal of UV abs with the application of the Tanfloc coagulant in samples of raw leachate studied was obtained when the pH of the leachate was corrected to a basic value (pH equal to 9) and used dosages of  $1,500 \text{ mg L}^{-1}$ , with variable removals ranging from 19 to 32%.

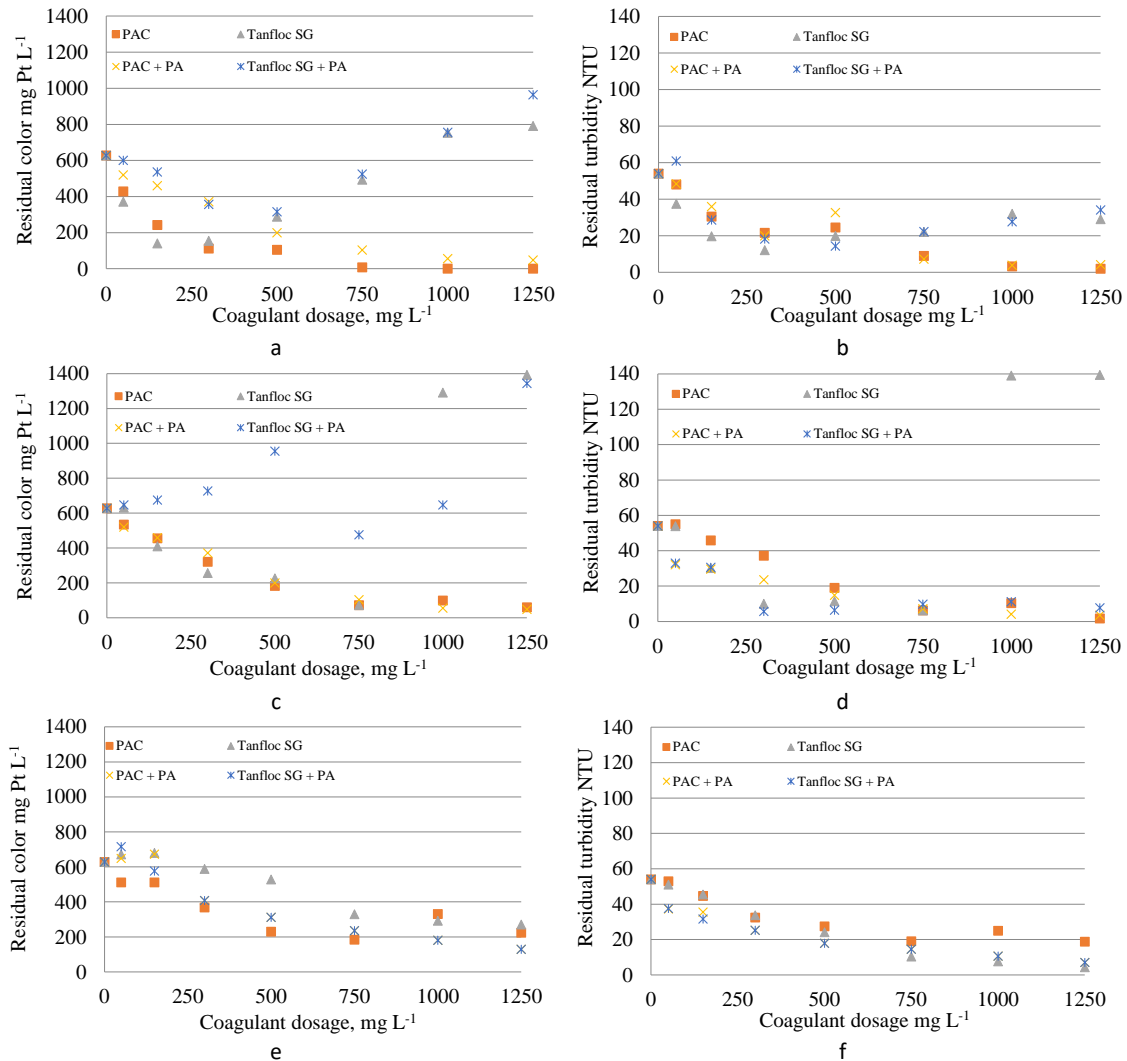
### 3.3 Color and residual turbidity

The residual values of color and turbidity of the effluent treated by the coagulation / flocculation process demonstrated that, in all the initial pH values analyzed, the treatments with PAC and PAC + PA, in dosages above  $300 \text{ mg L}^{-1}$ , reduced at least 50% color and turbidity. This behavior was not verified for Tanfloc SG or Tanfloc SG + PA (Figure 3).

The residual color varied a lot depending on the pH and the use of the PA. However, the lowest residual color values considering the lowest dosages were found at the initial pH values of five and seven. Residual turbidity got progressively lower at pH nine, when the treatment dosage was increased, a fact that did not happen at other pH levels, which had their lowest residual turbidity levels at the dosage of  $300 \text{ mg L}^{-1}$ .

At pH values of five and seven, both Tanfloc SG and PAC coagulants showed similar removals at low dosage values, without the need for a polymer. At pH 9, PAC, PAC + PA and Tanfloc SG + PA brought the residual color to very similar values.

**Figure 3: Color and residual turbidity as a function of initial pH, dosage of coagulants and treatment (types of coagulants). Color pH 5.0 (a), color pH 7.0 (c), color pH 9.0 (e), turbidity pH 5.0 (b), turbidity pH 7.0 (d), turbidity pH 9.0 (f).**



## 4 CONCLUSIONS

The coagulation / flocculation process is efficient for removing suspended, dissolved, colloidal organic matter and humic substances from the landfill leachate, except humic substances by Tanfloc SG.

The dosage of the PA used did not demonstrate important results regarding the improvement in the removal of the investigated parameters.

Under conditions of natural pH and basic pH, the Tanfloc SG coagulant shows good results, but the PAC coagulant is superior, achieving high percentages of turbidity, color and UV

abs removal in all analyzed pH values, in addition to resulting in low pH changes and lower volumes of sludge produced.

## 5. ACKNOWLEDGMENT

The authors would like to thank the Institute of Agricultural and Environmental Sciences (ICAA) and the Graduate Program in Environmental Sciences (PPGCAM), Sinop-MT; they also thank the Federal University of Mato Grosso (UFMT), the National Council for Scientific and Technological Development (CNPq) and the Coordination for the Improvement of Higher Education Personnel (CAPES).

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