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Xanthan gum as a novel flocculant aid employed in drinking water treatment

Laís Gimenes Vernasqui
laisvernasqui@gmail.com
Departamento de Engenharia Ambiental, Universidade Tecnológica Federal do Paraná, Campus Campo Mourão, Campo Mourão, PR, Brasil.

Patrícia Valderrama
patriciav@utfpr.edu.br
Departamento de Licenciatura em Química, Universidade Tecnológica Federal do Paraná, Campus Campo Mourão, Campo Mourão, PR, Brasil.

Flávia Vieira Silva-Medeiros
flaviamedeiros@utfpr.edu.br
Departamento de Engenharia Ambiental, Universidade Tecnológica Federal do Paraná, Campus Campo Mourão, Campo Mourão, PR, Brasil.

ABSTRACT

Aluminum salts used for drinking water treatment are strongly related to environmental implications and health hazards. In order to reduce aluminum concentration in water and enhance the quality of the flocs formed, flocculant additives are employed. This paper evaluated polyaluminum chloride as coagulant in combination with xanthan gum as a novel flocculant to improve drinking water quality. Optimum results for color removal of 97.4 %, were observed with 5.0 ppm of polyaluminum chloride combined with 0.6 ppm of xanthan gum, prepared at 65 ºC. Turbidity removal was complete with 5.0 ppm of polyaluminum chloride combined with xanthan gum solution, prepared at 65 ºC in all evaluated concentrations. Moreover, PACl combined with xanthan gum at 0.6 ppm, prepared at 65 ºC, rose to removal of other contaminants, which can enhance posterior stages of water treatment, as filtration and disinfection, resulting a greater control of organic load and DBPs formation.

KEYWORDS: coagulation/flocculation; natural flocculants; *Xanthomonas campestris*..

INTRODUCTION

Dissolved organic matter (DOM), particularly colored DOM, is currently increasing in surface water and, as a result, enhancing the color of the water. This non-expected color in surface water requires higher doses of chemical coagulants and disinfectants to maintain a high drinking water quality. (LAVONEN *et al*., 2015; EIKEBROKK *et al*., 2004).

However, this addition of conventional chemicals is prohibitively expensive, especially in developing countries, and also generates disinfection by-products (DBPs), known for their negative health effects (RICHARDSON, 2011; PRITCHARD *et al*., 2009; RICHARDSON, 2007). Likewise, low concentrations of DOM in drinking water are particularly required because it can also avoid bacterial growth. In addition, surface water generally presents high turbidity levels, which are related to high-suspended solid concentrations, which require a coagulation-flocculation process followed by a sedimentation unit. (PACKMAN *et al*., 1999; NDABIGENGESERE and NARASIAH, 1998).

Coagulation-flocculation is still the most common process for clarification and particle removal of surface water to make it acceptable for consumption (BROSTOW *et al*., 2009; WANG *et al*., 2008). Aluminum salts are the most widely used in water treatment; however, they are associated with environmental implications and health hazards (KAWAHARA AND KATO-NEGISHI, 2011; RENAULT *et al*., 2009; ZHAO AND BACH, 2002; AWWA, 1990; KOO AND KAPLAN, 1988). Moreover, aluminum-based coagulants are not able to control the enhancement in DOM and turbidity caused by dumping of domestic sewage and industrial effluents on watersheds, even in higher dosages (RONG *et al.,* 2013; WEI *et al*., 2009; SHI *et al.*, 2007).

It has been previously demonstrated that flocculent additives have the role of strengthen the flocs formed in coagulation, which are otherwise weak when chemical coagulants are employed alone (GUPTA AND AKO, 2005; BRATBY, 2006; BOLTO AND GREGORY, 2007; OWEN *et al*., 2007). In practice, synthetic flocculants are employed, such as Poly (diallyldimethyl ammonium chloride (PDADMAC), epichlorohydrin, polyacrylamide (PAM), and its methacrylate analogue is also commercially available, nonetheless they have been associated with health risks due to monomer residues in water (GARCÍA, 2011; GUPTA & AKO, 2005). Nonetheless, previous studies highlighted that natural flocculants could be used, with the advantages of low sensitivity to pH and larger and stronger flocs (KONO & KUSUMOTO, 2015; ZHANG *et al*., 2013; GARCÍA, 2011; BROSTOW *et al*., 2009; WEI *et al*., 2009; WANG *et al.*, 2008; BOLTO & GREGORY, 2007; GUPTA & AKO, 2005).

Among them, natural polysaccharides are becoming more and more relevant due to their low toxicity and high effectiveness in low concentrations. The most studied natural polysaccharide used as flocculant is certainly guar gum, which can be obtained from the seed of *Cyamopsis tetragonalobus* (Gavar, Guwar or Guvar bean), and has been previously reported as an effective flocculant used in water and wastewater treatment, applied in its natural composition or grafted with synthetic polymers (LEE *et al*., 2014; VERMA *et al*., 2012; BOLTO & GREGORY, 2007; SANGHI *et al*., 2006; GUPTA & AKO, 2005; SINGH *et al.,* 2000).

Xanthan gum, produced from the fermentation of glucose, sucrose or lactose by the bacterium *Xanthomonas campestris* (GARCÍA-OCHOA *et al.*, 2000) is chemically similar to guar gum, therefore, it presents the potential to be employed as a novel flocculant aid, once there have been few specific studies evaluating the application of this natural polysaccharide in drinking water treatment. Xanthan gum has been widely explored for various uses in pharmaceutical, cosmetics and food industries as tablet excipient, viscosity modifier, stabilizing agent, hardening agent, and suspending and emulsifying agent (PRAKASH *et al*., 2011; SHARMA *et al*., 2006; SANTOS *et al*., 2005).

This paper aims to evaluate polyaluminum chloride as coagulant in combination with xanthan gum solution as flocculant in different concentrations and temperatures in order to improve drinking water quality.

MATERIAL AND METHODS

SAMPLING AND CHARACTERIZATION OF WATER

Surface water used in the assays was retrieved from Campo River watershed managed by Companhia de Saneamento do Paraná (SANEPAR), in Campo Mourão, Paraná, Brazil. The raw water samples collected were characterized and stored under refrigeration so their original characteristics could be maintained till analysis.

Raw and treated water were characterized in relation to color (spectrophotometry DR 5000 Hach) and turbidity (turbidimeter Policontrol AP2000 IR), in accordance with Standard Methods (APHA, 2005). These parameters were measured at the collection of raw water and during coagulation-flocculation assays.

The parameters suspended solids (APHA 2005), dissolved solids (APHA 2005), total coliforms and *E. coli* (Petrifilm® count plates) were also evaluated at the collection of raw water. In the case of treated water, these analyses were restricted to those that achieved the optimum results for remaining color and turbidity.

The selection of these parameters is due to their relation to recent degradation in the quality of superficial water. Concerns about suspended solids are increasing because they are correlated with anthropogenic perturbations, such as industrial and wastewater discharges (MULLIGAN *et al*., 2009; BILOTTA & BRAZIER, 2008). In addition, dissolved solids showed a relevant concentration, which could demand higher amounts of coagulants and disinfectants with economic and health implications, such as esthetic rejection, disinfection by-products formation, and potential microbial growth (LAVONEN *et al*., 2015; RITSON *et al*., 2014). Moreover, it is increasingly recognized that even water from improved sources could present fecal contamination, so monitoring total coliforms and *E. coli* is still important (RODRIGUEZ-ALVAREZ *et al*., 2015; FATEMEH *et al*., 2014).

PREPARATION OF THE SOLUTIONS

Saturated solution of polyaluminum chloride (PACl) was kindly donated by SANEPAR and was diluted to 5.0 ppm and used alone and in combination with xanthan gum in the coagulation-flocculation assays.

The xanthan gum used was food grade and the flocculant solution was prepared in a concentration of 0.01% (m/v), at room temperature and at 65 ºC, named, respectively, Gum Solution at Room Temperature (GRT) and Gum Solution at Hot Temperature (GHT). The solutions were used fresh to avoid polysaccharides degradation (GUPTA AND AKO, 2005).

The temperatures used in this work was based in previous studies that show the high water solubility of xanthan gum at low and high temperatures (NIKNEZHAD et al., 2015) and also the stability and low variation of viscosity of xanthan gum solutions in a large range of temperatures, from 10 to 90 °C (TIAN et al., 2015; LUVIELMO & SCAMPARINI, 2009).

COAGULATION-FLOCCULATION ASSAYS

The combined employment of PACl as coagulant and xanthan gum as flocculant was evaluated in relation to the color and turbidity removal efficiency. Experimental tests were conducted in Jar Test equipment Nova Ética model 218/LDB with six samples of raw water inserted simultaneously into beakers of 1 L capacity. The coagulation/flocculation process was performed with the pH of surface water, without any correction or adjustment.

Jar test operational conditions comprised a 1-min rapid mixture with a rapid mixture gradient at 120 rpm and a 15 min. slow mixture with a mixture gradient at 60 rpm, followed by 30 min. of sedimentation at the end of the process.

PACl concentration was fixed in 5.0 ppm. Thenceforth, xanthan gum solution was evaluated as flocculant for concentrations varying from 0.2 to 1.2 ppm, added 4 minutes after the slow mixture started. The characterization of treated water was restricted to the assays that presented better results for color and turbidity removal.

STATISTICAL ANALYSIS

After that, the data obtained from this characterization was submitted to a statistical test to assess significant differences between results at 95% confidence level. The test considers, simultaneously, the confidence interval between slope and intercept. The interval is a region in the plane of two variables (slope and intercept), showing an elliptical shape. Then, a statistical test investigates if the point (1.0) for slope and intercept, respectively, is contained within the elliptical joint confidence region (RIU & RIUS, 1996; MASSART *et al*., 1997), described by the following equation (GONZÁLEZ *et al*., 1999):

$$q(\beta - B)^2 + 2(\alpha - A)(\beta - B)\sum_{i=1}^{q} x_i + (\alpha - A)^2 \sum_{i=1}^{q} x_i^2 = 2s_{y/x}^2 F_{2,q-2} \quad (1)$$

Where, α and β are the variables that correspond to the dimensions of the plane in which the elliptical region is represented; $F_{2,q-2}$ is the F statistic value with 2 and q-2 degrees of freedom for 95% confidence level.

RESULTS AND DISCUSSION

RAW WATER CHARACTERIZATION

Average characteristics of surface water used in coagulation-flocculation assays are shown in Table 1.

Table 1 – Characterization of raw water and treated water.

	Raw water
Color (mg PtCo/L)	390.0
Turbidity (uT)	69.0
pH	7.7
Suspended Solids mg.L-1	87.7
Dissolved Solids mg.L-1	35.5
Total Coliforms (UFC.mL-1)	83.0
E. coli (UFC.mL-1)	3.0

As shown in Figure 1, raw water presented considerable values of color and turbidity, which indicates that an appropriate treatment should be employed to improve water quality before distribution to consumers. Missed the discussion of the treated water, if it has? (This discussion is presented in item 3.3)

Suspended and dissolved solids also presented high levels, which may demand higher concentrations of coagulants (LAVONEN *et al.*, 2015; RITSON *et al.*, 2014). In this case, the combination with xanthan gum as flocculant could improve the coagulation-flocculation process, maintaining the lower coagulant concentration.

Total coliforms and *E. coli* colonies were also found, which indicate that the collected raw water is pathogenically contaminated. The role of flocculant agent in this case is to improve transport of bacteria in sedimentation stage, due to flocs size growth.

COAGULATION, FLOCCULATION AND SEDIMENTATION ASSAYS

The results for remaining color obtained in coagulation-flocculation assays employing PACl as coagulant, alone and combined with xanthan gum as flocculant, are presented in Figure 1.

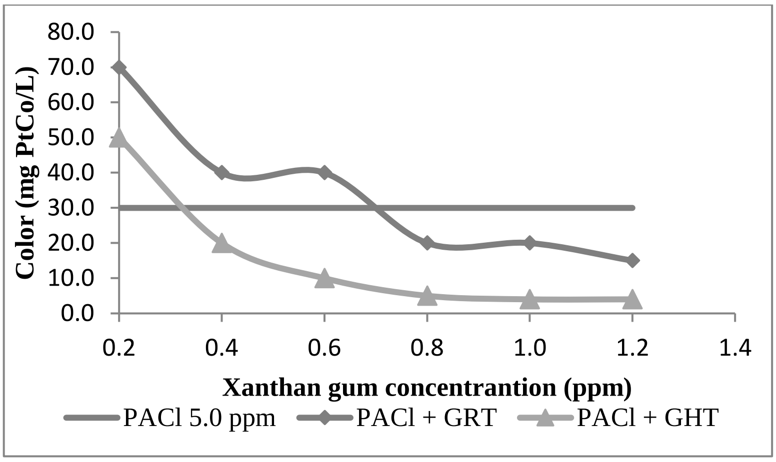

Figure 1 – Remaining color results for assays with 5.0 ppm of PACl as coagulant and xanthan gum as flocculant at concentrations of 0.2, 0.4, 0.6, 0.8, 1.0, 1.2 and 2.0 ppm.

Figure 1 illustrates a major color removal when GHT flocculant is applied, especially in dosages higher than 0.4 ppm, reaching values of 4 mg PtCo/L when using 1.2 ppm of GHT, while values of 15 PtCo/L where obtained employing GRT at the same concentration.

Besides, it is evident that the temperature of the flocculant solution affected color removal. As illustration, 5.0 ppm of PACl combined with 0.6 ppm of GHT is sufficient to achieve color removal of 97.4 %, while this same combination with GRT provides 92.3% of color removal.

As the application of xanthan gum as flocculant has not been explored yet, it was chosen to draw a parallel with reported results obtained with guar gum due to the chemical similarities, as well as with cellulose ampholytes (CAms), derivatives of cellulose, considering the results for color removal.

Sanghi et al. (2006) observed that guar gum solution in very low concentrations is very efficient as flocculant, when combined with PACl, for dyes removal. The authors found 73 to 87%, considerably greater when compared to the values reached with PACl when used alone, near 25%. Values obtained in the current study were slightly superior, up to 90%, due to minor differences between xanthan gum and guar gum, and differences in systems behaviors: dyes removal against drinking water treatment.

Kono and Kusumoto (2015) investigated the flocculation ability of cellulose ampholytes and observed high color removal at lower concentrations and lower pH values. In this current work, pH was not controlled, but authors also observed that the remaining color (Figure 1) has a tendency to be around the same value, despite the increase in xanthan gum concentration.

Figure 2 presents the results for remaining turbidity obtained in coagulation-flocculation assays employing PACl as coagulant, combined with xanthan gum as flocculant.

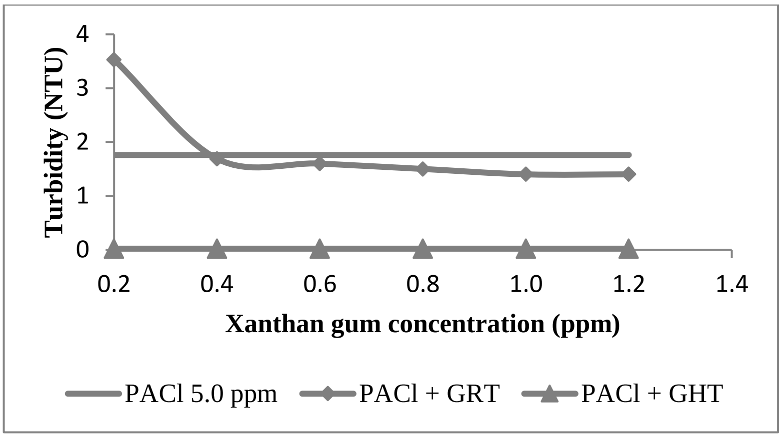

Figure 2 - Remaining turbidity results for assays with 5.0 ppm of PACl as coagulant and xanthan gum as flocculant, at concentrations of 0.2, 0.4, 0.6, 0.8, 1.0, 1.2 and 2.0 ppm.

It is remarkable in Figure 2 that the application of GHT combined with PACl reached values of 0.02 NTU.

Moreover, it should be underscored that the application of GRT presented turbidity removal greater than 97.6% with concentrations higher than 0.4 ppm, which indicates that this combination is even better than PACl used alone.

As the application of xanthan gum as flocculant has not been explored yet, it was chosen to draw a parallel with reported results obtained with guar gum due to chemical similarities.

Gupta and Ako (2005) collected water with average turbidity of 26.5 NTU and reached 96.2 % of removal using optimum values of 44.97 and 49.39 ppm of $Al_2(SO_4)_3$ and 1.80 and 1.88 ppm of guar gum, properly combined. In this current paper, greater results were achieved using lower concentrations of aluminum-based coagulant and gum flocculant. As examples, it is possible to indicate 5.0 ppm of PACl combined with 1.0 ppm of GRT and 5.0 ppm of PACl combined with 0.2 of GHT, which achieved 97.9 and 99.9 % of turbidity removal, respectively.

In addition, Pritchard et al (2009) employed guar gum solution alone and observed that higher concentrations are needed to achieve considerable turbidity removal. In this case, 95% of turbidity removal was observed when 50 ppm of guar gum solution was used in surface water with 49 NTU. This result indicates that aluminum-based coagulants should be applied in small quantities to avoid high concentrations of the polymeric flocculant, which may increase the organic load in drinking water and affect DBPs formation at the disinfection stage.

According to the results shown and discussed herein, the combination of PACl and xanthan gum presented in this paper allows the possibility to reduce chemical coagulants, consequently minimizing costs and health hazards, although the contribution in organic matter.

CHARACTERIZATION OF THE TREATED WATER

Considering the results presented in Figures 1 and 2, the optimum results were obtained with 5.0 ppm of PACl combined with 0.6 ppm of GHT, since this combination has produced water with remaining color of 10 mg PtCo/L, and remaining turbidity of 0.02 NTU, which is in accordance with Guidelines for drinking-water quality (WHO 2011).

The experimental results from the physical-chemical and bacteriologic characterization of the treated water at optimum treatment conditions, in comparison to the raw water characteristics, are shown in Table 2.

Table 2 - Characterization of raw water and treated water with 5.0 ppm PACl and 0.6 ppm GHT

	Raw water	**5.0 ppm PACl**	**5.0 ppm PACl + 0.6 ppm GHT**
Color (mg PtCo/L)	390.0	30.0	10.0
Turbidity (NTU)	69.0	1.76	0.02
pH	7.7	6.0	6.2
Total Solids mg.L-1	123.2	90.5	68.6
Suspended Solids mg.L-1	87.7	56.8	38.4
Dissolved Solids mg.L-1	35.5	33.7	30.2
Total Coliforms (CFU.mL-1)	83	14	1
E. coli (CFU.mL-1)	3	2	0

It is noticeable in Table 2 that the proposed treatment with 5.0 ppm of PACl combined with 0.6 ppm of GHT, when compared to 5.0 ppm of PACl used alone, further reduced the content of total, suspended and dissolved solids, besides the contents of total coliforms and *E. coli* presented in raw water, which indicates that the GHT employed as flocculant definitely improves the coagulation-flocculation process, and provides a safer and competitive treatment, since it does not require higher concentrations of aluminum-based coagulants. Furthermore, these results contribute to posterior stages in the treatment, such as avoiding an overload in filters and savings in disinfectants products.

STATISTICAL TEST RESULTS

In order to statically prove the efficiencies of GHT as flocculant, the elliptical joint confidence regions, at 95%, for the slope and intercept of the regression of variables' results was considered for Table 2, using ordinary least squares. These results are presented in Figure 3 and show that the ellipse does not contain the ideal point (1.0) for slope and intercept, respectively, indicating a significant difference between raw and treated water at the 95% confidence level.

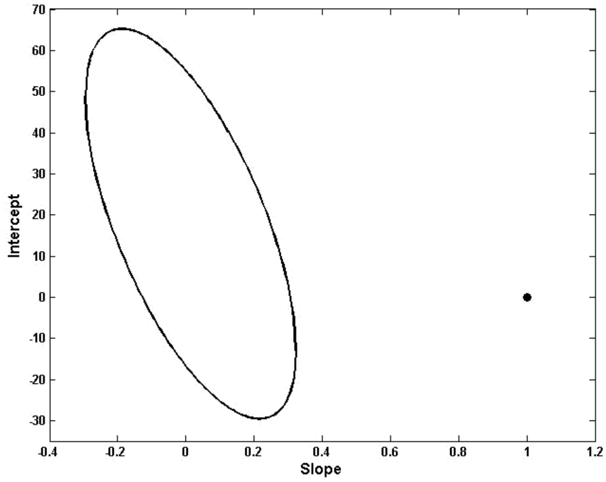

Figure 3 - Elliptical joint confidence regions at 95% of confidence for the slope and intercept of the regression of variables' results for raw water and water sample with GHT.

CONCLUSIONS

Xanthan gum combined with chemical coagulants may be successfully employed as flocculant, enhancing coagulation-flocculating performance, although few specific studies have been found in literature until now. This study demonstrates that temperature influences the flocculant efficiency of the xanthan solution. As demonstrated by color removal results, xanthan gum solution prepared at 65 ºC at 0.6 ppm, combined with PACl at 5.0 ppm, achieves color removal of 97.4 %, which is higher than the value achieved by xanthan gum solution prepared at room temperature, combined with PACl at 5.0 ppm (89.7 %).

Furthermore, PACl combined with xanthan gum prepared at 65 ºC enhanced the removal of other contaminants, such as suspended and dissolved solids, total coliforms and *E. coli*. This result indicates that the evaluated combination could improve the efficiency of posterior stages of water treatment, extending filter careers and saving disinfectants and, as a consequence, there is greater control of organic load and DBPs formation in the disinfection stage.

This study highlighted the possibility to reduce chemical coagulants in drinking water treatment with the use of xanthan gum as a novel flocculant aid, which has improved turbidity and color removal and, as a consequence, may lead to savings in conventional chemicals that could be prohibitively expensive and could generate hazardous by-products in contact to organic matter.

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Correspondência:
Flávia Vieira Silva-Medeiros
Departamento de Engenharia Ambiental, Universidade Tecnológica Federal do Paraná, Campus Campo Mourão, Campo Mourão, PR, Brasil.