Use of Natural Coagulants/Flocculants in the Treatment of Hospital Laundry Effluents

Maykon Johnny De Souza Abreu[a], Adão Lincon Bezerra Montel[b], Elisandra Scapin[c*]

[a] Postgraduate Program in Environmental Sciences, Federal University of Tocantins, Palmas, Tocantins 77001-090, Brazil. maykon.abreu@uft.edu.br

[b] Civil Engineering Course, Chemistry Laboratory, Room 02, Block II, Federal University of Tocantins, Palmas, Tocantins 77001-090, Brazil. montel@uft.edu.br

[c*] Environmental Engineering Course, Graduate Program in Environmental Sciences, Graduate Program in Biodiversity and Biotechnology, Chemistry Laboratory, Room 02, Block II, Federal University of Tocantins, Palmas, Tocantins 77001-090, Brazil. scapin@uft.edu.br

ABSTRACT: Water is a fundamental substance for the existence of life on earth. However, globally there is a freshwater crisis. Hospitals generate exorbitant volumes of effluents (5 to 15 times more toxic than urban ones). Hospital laundry is known for demanding the highest volumes of water, generating a proportional amount of complex effluents with high toxicity and recalcitrance. Adequate treatment for hospital wastewater is always an essential solution. Among all treatment methods, coagulation/flocculation emerges as one of the best alternatives. However, the use of traditional compounds such as aluminium sulfate has caused secondary pollution; its residues are harmful to public and environmental health. In this sense, the present study used natural compounds that do not cause adverse effects, such as chitosan/hydroxyapatite, to clarify the laundry effluents of the largest hospital from the Tocantins. The results showed that the hydroxyapatite associated with chitosan, at pH 6 and dosage of 50 mg/L, reduced the turbidity and apparent colour of these wastewaters by up to 67 and 55%, respectively. With lower performance and higher dosage (60 mg/L), the chitosan gel used (pH 6) promoted a maximum reduction of 35% of the apparent colour and 40% of turbidity.

KEYWORDS: hospital wastewater; hospital laundry effluent; wastewater treatment; chitosan; hydroxyapatite; coagulation and flocculation

1. INTRODUCTION

It is difficult to conceive of any other element that is more central to human existence than water.[1] It plays a decisive role in all aspects of life and is the defining characteristic of our planet.[2,3] However, more and more easily accessible water sources have already been drained, reserves are approaching their physical limits and new supplies for populations, with increasing consumption levels, are only available at higher costs than before.[3,4] On a global scale, there is a freshwater crisis.[5,6]

In a context in which water scarcity combined with surface water pollution represents one of the major problems today, the multiple activities that take place in health facilities, both medical and auxiliary, generate an exorbitant volume of wastewater,[7–11] with varying compositions, different types, and concentrations of different pollutants may be being released into the environment[11–19] through disposal without treatment in public sewage.[7,18,20–27]

Wastewater generated from health facilities poses a potential threat to the environment and public health due to the discharge of toxic chemicals that affect various aquatic species.[10,28–34] In this sense, hospital effluents are 5 to 15 times more toxic than urban effluents.[9,28] Lutterbeck et al.[35] listed the primary sources of hospital effluents with the potential to generate some refractory and persistent products and by-products. Among the various sectors, laundry is classified as the sector that demands the highest volumes of water that generates a proportional amount of complex effluents with high toxicity and recalcitrance.[7,22,36–40]

Due to the diversity of chemicals added to the washing processes, hospital laundry effluents may contain soap,[29,41] detergents,[41–44] surfactants,[41,45,46] sodium hypochlorite,[47,48] hydrogen peroxide,[7,49,50] peracetic acid,[49–51] softeners,[52,53] neutralizing additives,[54] chlorine,[43,44,48] adsorbable organic halogens (AOX),[44,55] nitrogen, phosphorus,[7,37,56] and heavy metals[57–60] that give these residues the power to exercise less biodegradable characteristics to the effluent generated by the hospital units.[9]

However, tissues from different areas such as the operating room, intensive care unit, hospitalization, hemodialysis, imaging, emergency room, among others, are sources of dirt such as blood, pus, medication residues, secretions and excretions,[61–64] which can contain pathogenic bacteria,[49,51,71,54,61,65–70] fungi or viruses.[56,72–74] Besides, a high concentration of particulate material, organic matter, proteins, starch, oils and greases[40,75] can be found.

The correct disposal of hospital wastewater must be done in order to comply with environmental legislation and minimize the impacts on watercourses after its ejectment. In this sense, adequate treatment for hospital wastewater is always a necessary solution. Various methods are used to treat effluents. Coagulation and flocculation,[76–79] ion exchange,[80,81] precipitation,[82] adsorption,[83,84] biological[85,86] and advanced oxidation[87–89] process are used to remove colloidal particles in the wastewater.[90,91] Among all treatment methods, the coagulation/flocculation (C/F) process is one of the oldest[92] and most essential treatment methods for most water and sewage treatment.[93–95]

A coagulant-flocculant (C-F) promotes the junction of colloidal and other particles suspended in a liquid forming larger particles (or flakes) to promote the settling of impurities from the stable suspension.[94,96–99] Due to this characteristic, high efficiency in reducing turbidity and pollutants can be achieved.[76,98,100–104] In general, inorganic C-Fs are more commonly used for this purpose,[105–109] just as synthetic polymers have also been applied. Both have low cost and good efficiency.[110,111]

Although cheap and effective, inorganic and synthetic coagulants have distinct disadvantages. Among them includes limited availability in certain regions; it is not biodegradable; large chemical doses are necessary for the treatment of eutrophic waters, and a massive amount of chemical sludge is produced.[79,98,112–114] Also, its residues cause harmful effects for both animal species[115–117] and public health.[115,118–121] Hereupon, the total or partial replacement of the traditionally used compounds, with natural and biodegradable natural substances, is a solution that is being much discussed in the literature.[122–130]

Chitosan (CS) offers several advantages over traditional compounds, including wide availability (higher after cellulose), cost-benefit, respect for the environment, atoxicity, biodegradability, biocompatibility, bioactivity, solubility in weak acids, sensitivity at pH, better biosorption, they do not produce secondary pollution, they are produced from renewable organic biomass, it allows the reuse of sludge as an agricultural fertilizer, among others.[123,131–135] In the same line, hydroxyapatite (HA), a calcium phosphate-based[136] biomaterial[137] is an up-and-coming candidate for water treatment and environmental remediation[138–141] due to its good thermal stability,[142] acid-base properties,[143] high porosity[144] and ion exchange capacity.[145,146] Moreover, it is biocompatible,[147,148] non-toxic,[149,150] anti-inflammatory,[147] chemically inert[143,151] and derived from renewable biomass.[152,153] HA is also known as a powerful adsorbent,[154–157] widely available and at low cost[158] compared to others such as quartz, fluorite and calcite.[140]

QS alone or HA associated with QS can be a promising substitute in C/F processes due to its potential viability in treatment without presenting any health threat,[102,121,138,139,144,155,159,160] unlike inorganic and synthetic compounds that, among other problems, can cause Alzheimer's disease.[119,121,161,162]

In this context, a study was developed to assess the performance of QS and HA in the treatment of wastewater from hospital laundries, to reduce the toxic load of discharge into the sewer, using C/F techniques to promote the optimized reduction of turbidity and apparent colour and indirectly mitigate environmental pollution caused by hospital units.

2. MATERIALS AND METHODS

2.1. Characterization of the study area

The General Hospital of Palmas (GHP) has 472 beds, located in Palmas, the central region of the state of Tocantins, Brazil. On average, approximately 376 m^3/day of wastewater is generated by the hospital. It is estimated that the hospital's laundry produces about 156 m^3/day of effluents, resulting from washing 5435 kg/day of textile items, which represent about 42% of the hospital's wastewater volume. This amount of sewage is discarded in the public sewage network after being partially treated by an internal sewage treatment plant, equipped with coarse solids retainer (grating), followed by an upward flow anaerobic reactor and percolating filter. This system was installed to remove only coarse solids and organic matter.

2.1. Collection and characterization of effluents

2.2.1. Sample collection

The effluent samples for carrying out the tests were collected directly in the outlet pipe of the washing machines, chosen at random, following the hygiene and safety standards of the HGP laundry. No synthetic effluents were used. Depending on the degree of soiling, the clothes are separated for washing in two programs – slight and heavy. Figure 1 shows the details the collection process until laboratory packaging, as well as the types of chemicals added to each stage. The "x3" indicates the number of times that the volume (1200 mL) was collected in each step, i.e., in triplicate.

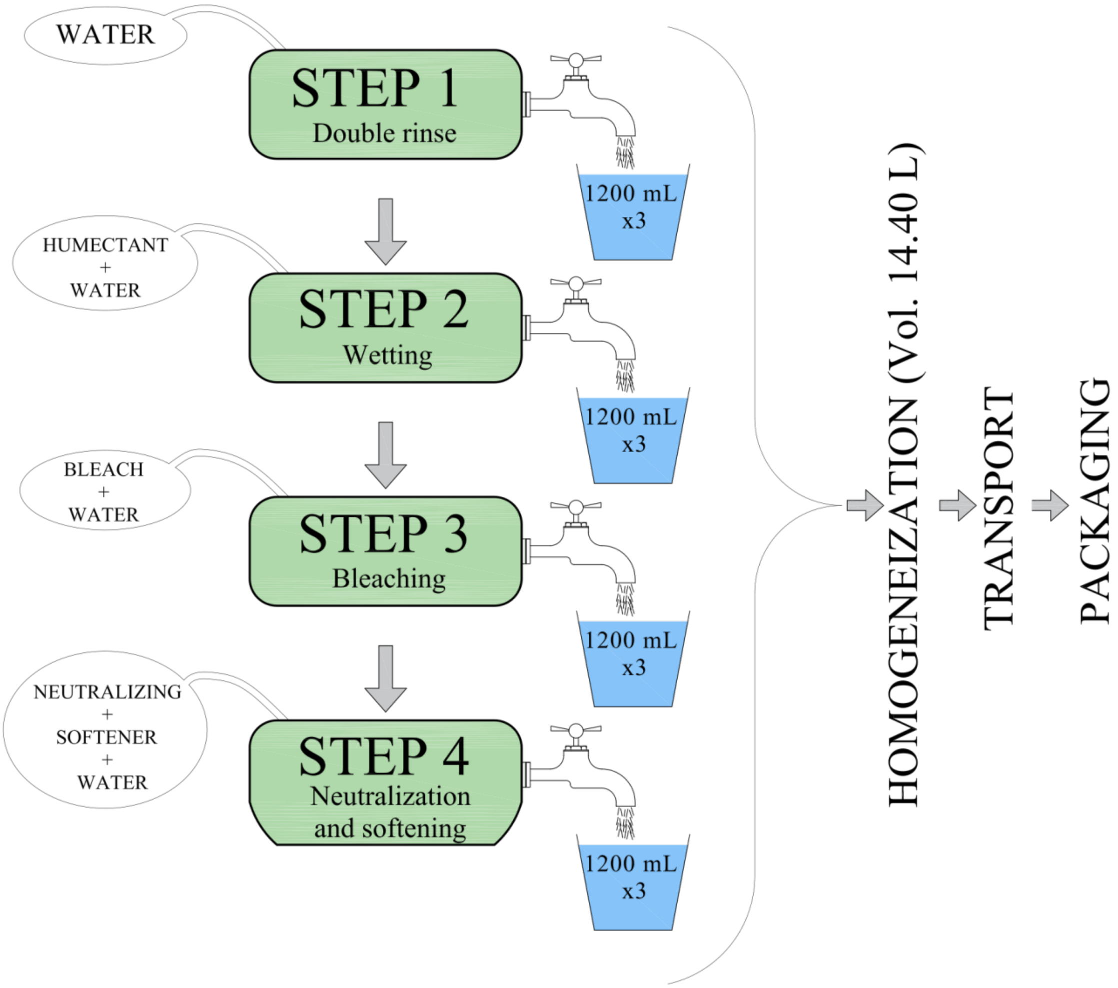


Figure 1 – Schematic representation of the sample collection process and main additives added to each stage of the heavy washing process.

2.2.2. Characterization of raw effluent

The effluent was characterized at the Research Laboratory for Environmental Chemistry and Biofuels (LAPEQ) and the Environmental Sanitation Laboratory (LABSAN) – both at the Federal University of Tocantins (UFT). Physical, chemical and biological analyzes were taken into account only for the washing steps in which more chemicals are added. In Table 1 it is possible to check the chosen parameters, the technique used and the respective laboratory in which they were performed.

Table 1 – Parameters of the Initial Characterization Associated with the Technique and Its Respective Laboratory.

Analytical parameters	Technique (APHA 2005)[163]	Laboratory
Chemical Oxygen Demand (mg/L)	Spectrophotometry	LABSAN
Biochemical Oxygen Demand (mg/L)	Differentiation	LABSAN
Total coliforms (MPN/100 mL)	Colilert	LABSAN
Escherichia coli	Colilert	LABSAN
Electric conductivity (µS/cm)	Potentiometry	LAPEQ
Turbidity (NTU)	Nephelometry	LAPEQ
Total dissolved solids (PPM)	Potentiometry	LAPEQ

Apparent colour (Pt/L)	Spectrophotometry	LAPEQ
True colour (Pt/L)	Spectrophotometry	LAPEQ
Oils and greases (mg/L)	Solvent extraction	LAPEQ
Ph	Potentiometry	LAPEQ
Total nitrogen (mg/L)	Differentiation	LAPEQ
Total phosphorus (mg/L)	Spectrophotometry	LAPEQ
Total hardness (mg/L)	Titrimetry	LAPEQ
Total alkalinity (mg/L)	Titrimetry	LAPEQ
Manganese (mg/L)	Spectrophotometry	LAPEQ
Zinc (mg/L)	Spectrophotometry	LAPEQ
Chrome (mg/L)	Spectrophotometry	LAPEQ
Aluminum (mg/L)	Spectrophotometry	LAPEQ
Fixed suspended solids (mg/L)	Calcination	LAPEQ
Organic suspended solids (mg/L)	Calcination	LAPEQ
Total suspended solids (mg/L)	Calcination	LAPEQ

2.3. Materials and equipment

Two types of natural chitosan-based C-Fs were used, a gel and a biocomposite, both in the form of stock solutions. The compound in gel form was the one first studied by Martins,[164] in which the best CS solution found was the formulation entitled "K10G" and object of the patent BR 102016005006-5.[131] It was prepared by dissolving 1.0444 g of CS in 100 mL of acetic acid (1%). This mixture was subjected to magnetic stirring for 15 minutes under heating. After that period, 34 mL of glycerol and 206 mL of water were added. Stirring and heating were continued for another 50 minutes. Then, the procedures were interrupted and the final product was stored at room temperature. The final concentration of the CS gel was 10.44 mg/mL.

The second C-F, a biocomposite produced by Araújo Júnior et al.[165] based on hydroxyapatite/chitosan (HA/CS), obtained from uçá crab shells (*U. cordatus*) acquired from the Filé do Mangue micro-company. According to the authors, obtaining the biocomposite proceeded as follows: after the meat was extracted, the shells were crushed, washed with drinking water and exposed to sunlight for seven days. The shells were dehydrated in the oven at 60 ° C for 4 hours to remove the water, and then they were ground for 2 hours in a ball mill. This powder was washed with 99.7% ethanol, 99% sodium hydroxide, both from Alphatec®, and water to remove proteins and lipids. By adding sulfuric acid (0.5%, Alphatec®) to the powder of the shells, the HA bioceramics and the CS biopolymer were extracted with a weight ratio of approximately 1:0.25 (HA:CS).[165]

HA and CS are often insoluble in water.[159,166–168] Then, to improve solubility, the HA/CS biocomposite was transformed into a stock solution. In this sense, due to its practicality and economy, since no magnetic stirring is required for dissolution, the preparation procedure Divakaran and Pillai[169] was chosen. It was prepared by mixing 200 mg of the HA/CS biocomposite in 10 mL of 0.1 M hydrochloric acid and set aside for two hours until completely dissolved. The solution was diluted in 100 mL of distilled water to produce 20 mg of HA/CS per mL of stock solution.

The C/F tests were carried out in a six-axis multiple stirrer units with stainless steel blades arranged inside 2 L jars (Jar-test model PoliFloc III – rectangular blades: 75 mm × 25 mm – from PoliControl®, São Paulo, Brazil).[170] A digital thermo-hygrometer was used (mod. TH50, from Incoterm®) to monitor the room temperature in the execution of the experiments.

2.4. Coagulation/flocculation tests with chitosan

2.4.1. Experimental procedure

The efficiency of natural C-Fs was assessed at two pH levels (6 and 8) and the ability to reduce turbidity and apparent colour (control parameters). Preliminary experiments showed that dosages of the K10G gel below 40 mg/L required more than 24 hours of sedimentation time to return palpable results and dosages above 100 mg/L of HA/CS caused an increase in initial turbidity. Based on initial data, the values of several factors for the execution of the study were established, which are shown in Table 2.

The isoelectric point of CS is around pH 8.70.[171] When the pH rises to values higher than this, CS becomes insoluble in an aqueous medium. As a consequence, its main C/F mechanisms are considerably impaired. Souza[172] points out that the addition of CS in effluents with a pH above 9, in addition to not observing the formation of flakes, the treatment was ineffective. Thus, before adding coagulant, the pH was adjusted according to Table 2.

Table 2 – Parameters of the experimental procedure, their respective levels and baseline references.

Study factors	Levels		Reference
	Gel K10G	HA/CS	
pH	6	6 e 8	
Concentration of biocomposite (Jar 1)	50 mg/L	50 mg/L	
Concentration of biocomposite (Jar 2)	60 mg/L	60 mg/L	
Concentration of biocomposite (Jar 3)	70 mg/L	70 mg/L	Preliminary experiments
Concentration of biocomposite (Jar 4)	80 mg/L	80 mg/L	
Concentration of biocomposite (Jar 5)	90 mg/L	90 mg/L	
Concentration in the control jar	0	0	
Room temperature	26 ºC	26 ºC	Di Bernardo, Dantas e Voltan (2013)[173]

Ph adjustment time	30 s	30 s	Di Bernardo, Dantas e Voltan (2013)[173]
Ph adjustment gradient	100 s^{-1}	100 s^{-1}	Di Bernardo, Dantas e Voltan (2013)[173]
Coagulation mixing time	2 min	2 min	Saritha, Srinivas, Srikanth e Vuppala (2017)[174]
Coagulation mixing gradient	80 s^{-1}	80 s^{-1}	Saritha, Srinivas, Srikanth e Vuppala (2017)[174]
Flocculation mixing time	20 min	20 min	Saritha, Srinivas, Srikanth e Vuppala (2017)[174]
Flocculation mixing gradient	20 s^{-1}	20 s^{-1}	Saritha, Srinivas, Srikanth e Vuppala (2017)[174]
Sedimentation time	8 h	8 h	Preliminary experiments

The volume of two litres of collected effluent was added to the six jars. A standard sample was taken to measure turbidity, apparent colour and pH before the start and at the end of each experiment. Different amounts of K10G gel and HA/CS were added to the jars and kept under agitation in the Jar-test, obeying the levels established in Table 2, according to the standard procedure of the American Society for Tests and Materials (ASTM).[175] A blank experiment was also carried out simultaneously in the absence of C-F to assess the natural decantation of the suspension under similar experimental conditions. Figure 2 schematically details the execution of the experimental procedure.

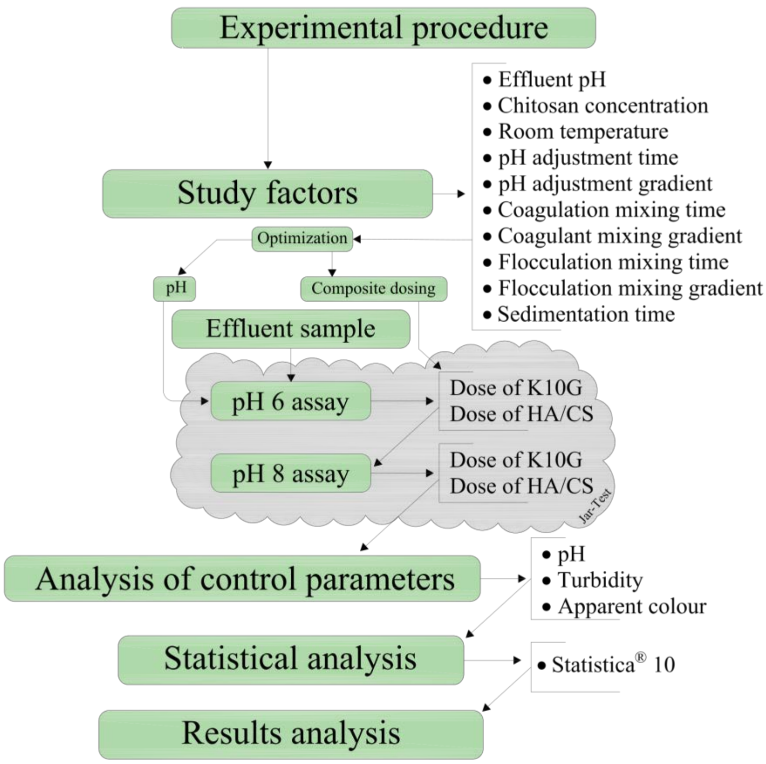


Figure 2 – Schematic representation of the experimental procedure.

2.4.3. Data analysis

The effluents of the studied laundry, without the addition of C-F, showed a small reduction in the values of the control parameters (on average 16%). In this sense, the percentage of reduction in the control parameters was calculated, taking into account their respective value in the control jar, according to Equation 1.

$$Percentual\ Reduction = \left(\frac{V_j - V_{jc}}{V_{jc}}\right) \times 100 \qquad \text{(Eq. 1)}$$

Onde:

Where:

V_{jc} and V_j represent the values of the control parameters in the control jar and the others jar in the test jar, respectively.

2.4.2. Statistical analysis

In environmental studies of real effluents, sample degradation is a matter of great concern when long periods of experimentation are needed to determine the best conditions for a treatment process.[7] In this sense, in addition to carrying out the experiments with a maximum of 24 hours after collection, each procedure and control parameter was repeated five times. The mean value and standard deviation of the five repetitions were calculated. Statistical analyzes, graphs and tables were performed using the Statistica® 10[176] software (5% significance level) aided by Microsoft Excel® 2010.[177]

In both software, histograms were elaborated to analyze the normality of the data. For data sets *N*>50 and *N*<50, the Kolmogorov-Smirrnov & Lilliefors and Shapiro-Wilk's parameters were taken into account, respectively. In both statistical analyzes, the data distribution was normal. Therefore, two parametric methods were used to statistically assess the significant difference (p <5%) of each factor (C-Fs and pH) at different levels: Analysis of Variance (ANOVA) of repeated measures and factorial ANOVA. After both ANOVA tests, the Tukey test was used to show the best level for each factor.

3. RESULTS AND DISCUSSIONS

3.1. Characterization of laundry effluents

The composition of the effluents produced by the laundry is different from those generated by other sectors of the hospital. Several physical-chemical and biological characterizations of these effluents were carried out. Before calculating the mean and standard deviation of the parameters, the results were separated taking into account effluents collected in periods when the dosing of chemicals in the machines was carried out manually and automatically. These data and the discharge limits for effluents from Brazil (National Environment Council – CONAMA) and Europe (European Economic Community – EEC) are shown in Table 3.

Table 3 – Characterization of Laundry Effluents from Hospital Geral de Palmas and Effluent Discharge Limit from Brazil (CONAMA) and Europe (EEC).

Analytical parameters	Mean ± standard deviation (manual dosage)	Mean ± standard deviation (automatic dosage)	EEC 91/271[178]	CONAMA 430[179]
Chemical Oxygen Demand (mg/L)	149 ± 109.56	1288.5 ± 88.5	125	–
Biochemical Oxygen Demand (mg/L)	70 ± 28.71	296.05 ± 17.75	–	120
Total coliforms (MPN/100 ml)	13.1 ± 8.85	2419.6 ± 0	–	–
Escherichia coli	–	248.1 ± 0	–	–
Electric conductivity (µs/cm)	6583.33 ± 7751.94	831.04 ± 827.96	–	–
Turbidity (NTU)	53.5 ± 7.84	29.1 ± 8.9	–	–
Total dissolved solids (PPM)	3290.87 ± 3876.41	1119.5 ± 286.5	–	–
Apparent colour (Pt/L)	179.33 ± 34.5	136.5 ± 25.5	–	–
True colour (Pt/L)	97.33 ± 38.66	33 ± 12	–	–
Oils and greases (mg/L)	153.73 ± 153.28	–	–	–
pH	10.96 ± 2.69	12.24 ± 0.27	6.0 – 9.0	5.0 – 9.0
Total nitrogen (mg/L)	9.66 ± 5.55	13.05 ± 3.42	10	20
Total phosphorus (mg/L)	–	18.45 ± 6.05	1	–
Total hardness (mg/L)	10.78 ± 0.94	8.69 ± 0.96	–	–
Total alkalinity (mg/L)	1210 ± 1006.21	158 ± 56	–	–
Manganese (mg/L)	0.23 ± 0.15	0.09 ± 0.03	–	–
Zinc (mg/L)	0.03 ± 0.02	0.01 ± 0	–	–
Chrome (mg/L)	0.28 ± 0.14	0.01 ± 0	–	–
Aluminum (mg/L)	0.01 ± 0	0.01 ± 0	–	–
Fixed suspended solids (mg/L)	0.53 ± 2.78	13.6 ± 12.4	–	–
Organic suspended solids (mg/L)	44.27 ± 4.59	–	–	–
Total suspended solids (mg/L)	44.8 ± 7.35	–	35	–

Several authors have reported high polluting loads in hospital effluents.[9,10,28–34] However, in Tab. 2 it is possible to observe a robust eutrophic load in the effluents of the studied laundry, which, in turn, are mixed with the effluents of the hospital. In this laundry, high levels of chemical oxygen demand – COD (1288.50 mg/L) and biochemical oxygen demand – BOD_5 (296.05 mg/L) were found, whose high values are well above those found in other studies.[180,181] Concerning the European directive EEC 91/271, COD values are extrapolated more than ten times. On the other hand, DBO_5 values exceed the legislation (CONAMA 430 and CEE 91/271) by almost 2.5 and 12 times, respectively.

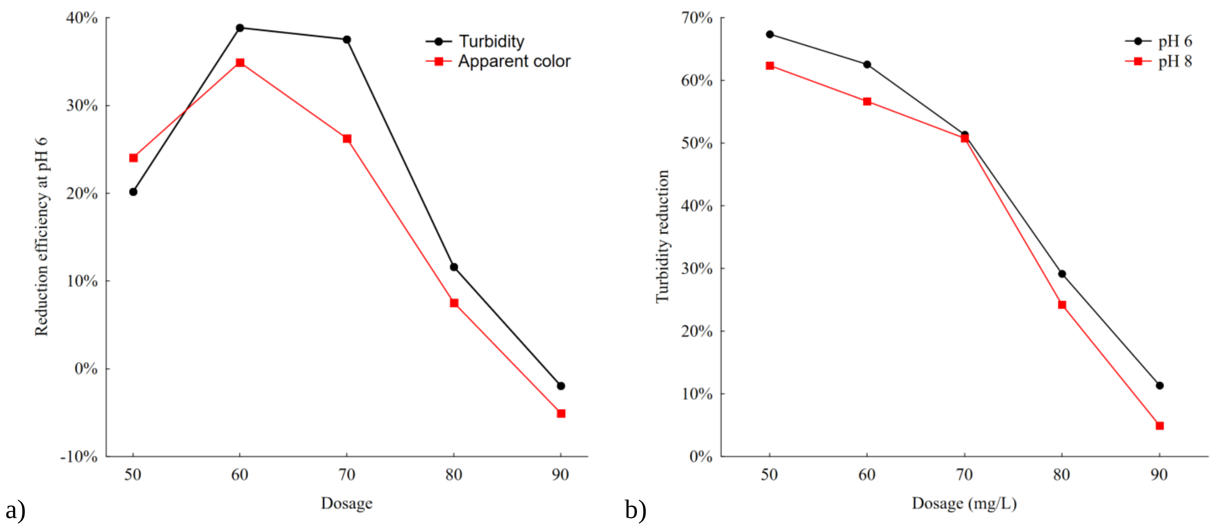
Tab. 3 also shows a pH that is highly alkaline – in line with the high total alkalinity (1210 mg/L) – and is being launched in disagreement with both laws. The turbidity values are high, probably due to the presence of particles, such as blood and cotton fibre.

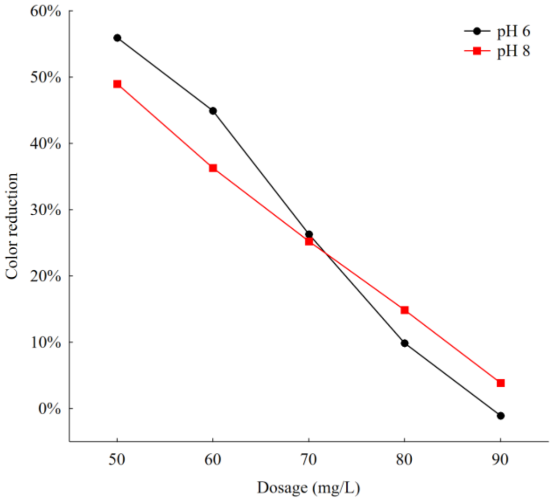
Regarding the nutritional load, the liberation of nitrogen into the sewage is slightly above the limit established by EEC 91/271 and as Brazilian legislation is less demanding, the disposal does not exceed the value allowed by CONAMA 430. However, the values of discarded phosphorus are 18 times higher the limit established by the EEC.

The load of pathogens was also high and the incidence of *E. coli* indicates contamination of wastewater by human faecal matter. Several studies have reported the high incidence of these coliforms in wastewater from hospital laundries.[37,66,75] In this sense, the scientific community reports several cases of infection in hospital laundries: *Salmonella*,[65] rotaviruses, *Clostridium difficile*,[49,51,61] influenza virus (H1N1),[73] *Streptococcus spp.*,[182] *Enterococcus spp.*,[70,71] *Acinetobacter spp.*,[69] *Staphylococcus spp.*,[68,183] *Pseudomonas*,[67] *Bacillus spp.*[54] and hepatitis A virus.[72] Besides, workers at a North American cooperative that washes the tissues of 40 hospitals were infected with the new COVID-19.[74]

3.2. Evaluation of the coagulation and flocculation process

Despite being highly polluting, inside a hospital, these effluents can be treated and reused at a non-potable level and have the potential to reduce water consumption in these institutions, as well as avoid their direct disposal in the untreated urban sewage network.[7] For this purpose, several composite samples were submitted to C/F under different conditions of pH and dosage of C-Fs. The results of the study are shown in Figure 3. All reduction efficiencies (in percentage) are related to the control jar and negative values indicate that the dosage of that jar caused an opposite effect (increase in turbidity/apparent colour above that presented in the control jar).





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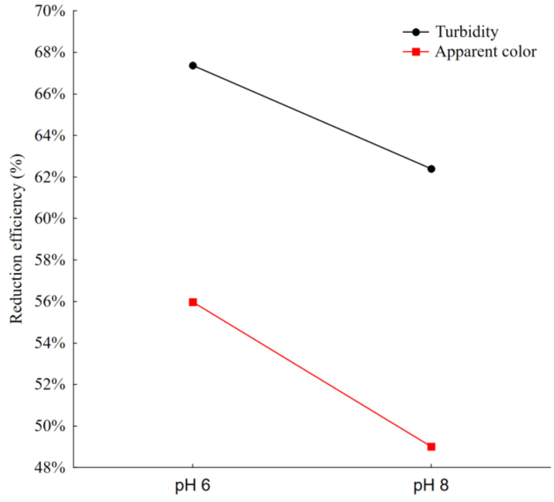


d)

Figure 3 – a) Turbidity and apparent colour reduction efficiency using K10G gel at pH 6; b) Turbidity reduction efficiency using HA/CS at pH 6 and 7; c) Efficiency of apparent colour reduction using HA/CS at pH 6 and 7; d) Comparison between turbidity reduction efficiency and apparent colour at different pH levels for the optimal dosage (50 mg/L) of HA/CS.

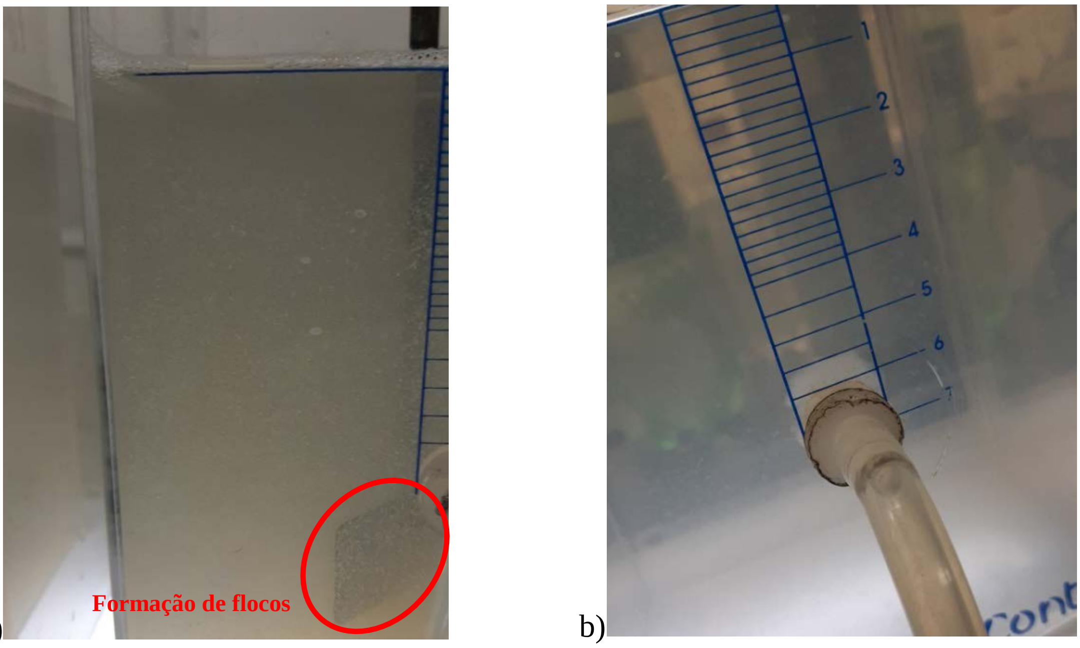
Fig. 3a points out that C/F with K10G at pH 6 provided maximum reductions of approximately 35% and 40% in apparent colour and turbidity parameters, respectively, with an optimal dosage of 60 mg/L. Martins[164] found better results using it in the treatment of bovine slaughterhouse effluents, with a high organic load and oils and greases (> 600 mg/L). Although the effluents in this study have reasonable amounts of oils and greases, it was observed that the effluents studied by Martins[164] have different characteristics – they can contain four times more oils and greases than those from hospital laundries. Also, during the laundry washing processes, a large part of the organic matter is eliminated in the first rinses and as shown in Fig. 1, to the next steps, chemicals of low biodegradability (such as surfactants) are added. The scientific literature[36] reports that only C/F with CS is not enough to remove surfactants and, in general, adsorption improves the process. Souza[172] tested the removal of surfactants in hospital laundry effluents by C/F processes with CS and two other C-Fs. However, it was not successful. Due to the low performance in this study, no experiments were performed using the K10G gel at pH 8.

On the other hand, Fig. 3b and 3c show that HA/CS was considerably more efficient (≅55%) than K10G gel, with a slightly lower dosage and similar sedimentation time. This C-F achieved maximum reductions of about 67 and 55% for turbidity and apparent colour, respectively. Generally, due to the improvement of adsorbent properties, the association of HA and CS has been shown to be more effective in treating effluents than with CS alone.[141,144,155,184] Herewith, for this study, it is likely that the better performance of the HA/CS composite compared to the K10G gel is due to the powerful adsorption activity that HA promotes.

There is a precise dosage of C-F for significant flake formation to occur due to its cationic nature.[185] In this sense, a trend is observed in Figs. 3a, b and c: there is a strong relationship between C-F dosage and efficiency of reducing control parameters – the higher the dosage, the lower the efficiency of the C/F process. A well-known mechanism in C/F processes is the formation of a polymer bridge that, in turn, causes destabilization, formation of dense flakes [109,186,187] that, consequently, increase the sedimentation and solid-liquid separation rates.[188] However, an overdose of C-Fs can result in re-stabilization because it becomes difficult for the extended polymer molecule to find empty sites available for adsorption.[122,189] Thus, it is likely that the aforementioned strong relationship can be explained by the saturation of the polymer bridge caused by the overdose of C-F.

Regarding the influence of pH on C/F, in general, Fig. 3b, 3c and 3d point out that the experiments carried out with pH 6 were slightly more efficient than with pH 8. However, with this slight difference, it is not possible to confirm a significant difference (p <5%) between these two pH levels in the treatment of effluents by C/F with the HA/CS composite. However, this adjustment is necessary due to the low performance of the treatment without adjusting the pH (11.20).

In agreement with these results, the scientific literature[171,190] reports that, in general, at pH close to 6, CS offers less turbidity/apparent colour and the increase in pH also causes a slight increase in residual turbidity. Another factor that may be weakly influencing the better performance of the C/F treatment at pH 6 is that the zeta potential of the CS surface in acidic environments is usually positive due to the protonation of the amino groups ($–NH_3^+$) in these conditions.[191,192] On the other hand, impurities usually have a negative charge.[76,77] Thus, the electrostatic interaction of the negatively charged pollutants in contact with the positive charges of the QS causes the agglomeration of particles, formation of flakes and the consequent general improvement of the process. In this sense, the fact that the QS is in a smaller proportion (about 4 times) in the composite, may be the cause of the insignificant influence. Figure 4 shows the formation of flakes into jars with and without the addition of HA/CS (control jar), at pH 6.



a) b)

Figure 4 – Flakes formation in the jar of the test jar (experiment nº 9). Coagulation/flocculation with (a) and without the addition of hydroxyapatite/chitosan (b) both when initiating sedimentation (pH 6).

If the coagulation reaction occurs under non-optimized pH conditions, the quality of the treated and filtered water can be degraded by high concentrations of the C-F employed.[189] In this sense, a fact that drew attention was the high addition of acidifier to the jars – average doses of 9.75 mL (pH 6) and 7.78 mL (pH 8) – to optimize the pH of the experiments. However, the pH control in this study was fundamental, since initial tests showed low performance not only of the K10G gel, but also of the HA/CS in the clarification of the raw effluent without pH correction.

As mentioned, the optimal dosage of the HA/CS composite was 50 mg/L. When comparing this dosage in the treatment of hospital laundry effluents with dosages of commercial C-Fs (Table 4), the HA/CS in dosages eight times lower produces similar reductions in apparent color and turbidity. That indicates that CS associated with HA is more efficient than the C-Fs compared.

Tabela 4 – Comparison Between the Dosages of Aluminium Sulfate, Aluminum Polychloride (PAC), Tanfloc SG and Hydroxyapatite/chitosan (HA/CS) in the Reduction of Turbidity and Apparent Colour.

Coagulant/flocculant (optimal dosage)	% Turbidity reduction ± standard deviation	Source
Aluminium sulfate (400mg/L)	86,4 ± 0,5	Souza (2012)[172]
PAC (400mg/L)	85,3 ± 0,5	
Tanfloc SG (400mg/L)	76,9 ± 0,8	
HA/CS (50mg/L)	67,4 ± 3	this study
Coagulant/flocculant (optimal dosage)	**% Colour reduction ± standard deviation**	**Source**
Aluminium sulfate (400mg/L)	63,2 ± 2,6	Souza (2012)[172]
PAC (400mg/L)	73,6 ± 1,3	
Tanfloc SG (400mg/L)	52,7 ± 2,6	

HA/CS (50mg/L)	56 ± 5,7	this study

3.3. Statistical analysis

The ANOVA test of repeated measures proved the hypothesis that the compounds used promoted a statistically significant improvement ($p < 5\%$) in the control parameters compared before and after the addition of C-Fs. The factorial ANOVA test proved the hypothesis that both C-Fs cause significantly different effects in the C/F process (at pH 6). The comparison between the different pH values of HA/CS did not show palpable levels of significance. However, the dosage strongly influenced the C/F process. The Tukey test showed that the C-Fs have less turbidity and apparent residual color with dosages of 50 and 60 mg/L for HA/CS and K10G, respectively.

4. CONCLUSION

When compared to the CS gel K10G, the HA/CS composite is a significantly more efficient C-F that, in turn, promotes the efficient C/F of hospital laundry effluents at considerably lower dosages than some commercial C/F available. Although it has not been investigated here, the literature points out that HA is a powerful adsorbent[141] and, in general, when combined with CS, it has the potential to improve the treatment of effluents,[144,155,184] a detail that may explain the better performance of the HA/CS composite in this study. Due to the low efficiency of the K10G gel at pH 6, it is suggested that such C-F is not the most suitable to promote C/F in hospital laundry wastewater.

Using the HA/CS, from a statistical point of view, reducing the pH from 8 to 6 did not promote improvement in the results. Thus, due to the high volume of hydrochloric acid added to reduce the pH to 6, it appears that the treatment at pH 8 using the HA/CS composite is the most efficient. Because, in addition to consuming less acidifying, it promotes reductions in the values of control parameters statistically equal when compared to C/F at pH 6. Although CS presents better results at pH close to 6, due to the presence of amino groups, it is in lower proportion in the HA/CS compound and, in general, it could have caused a weak improvement in the treatment. The optimal dosages of the K10G gel and the HA/CS composite were 60 and 50 mg/L, respectively.

ABBREVIATIONS

AOX	adsorbable organic halogens
ANOVA	Analysis of Variance
ASTM	American Society for Tests and Materials
BOD_5	biochemical oxygen demand

C/F	coagulation/flocculation
C-F	coagulant-flocculant
COD	chemical oxygen demand
CONAMA	National Environment Council
CS	chitosan
EEC	European Economic Community
GHP	General Hospital of Palmas
H1N1	influenza virus
HA	hydroxyapatite
HA/CS	hydroxyapatite/chitosan
K10G	K10G gel
LABSAN	Environmental Sanitation Laboratory
LAPEQ	Research Laboratory for Environmental Chemistry and Biofuels
MPN	most probable number
NTU	nephelometric tubidity unit
PAC	aluminium polychloride
PPM	parts per million
UFT	Federal University of Tocantins

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