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Study of the chitosan performance in conjunction with polyaluminum chloride in removing turbidity from Ahvaz water treatment plant

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ABSTRACT

Chitosan is a biodegradable cationic polymer derived from the deacetylation of chitin. This study aims to investigate the effects of chitosan as a coagulant aid for the improvement of a polyaluminum chloride coagulant in removing the turbidity from drinking water. This study was conducted in the laboratory using a jar test in the water treatment plant of Ahvaz, Iran. Experiments were conducted based on various variables such as concentration of coagulant, pH, and different concentrations of chitosan as a coagulant aid. After preparation in a jar test, samples were mixed fast at 120 rpm for 1 min and slowly at 40 rpm for 20 min and remained under stable conditions for 30 min to allow for settlement. The optimal pH for the removal of the turbidity was pH 8. The optimal dosage of polyaluminum chloride combined with chitosan was 5 and 0.02 mg/L, respectively. Under optimal conditions, the use of chitosan could reduce the concentration of polyaluminum chloride by approximately 50%. In addition, particles formed in flocculation by chitosan were coarser and settled faster. Results indicated good performance of chitosan as a coagulant aid for the removal of the turbidity from drinking water.

Keywords: Coagulation; Turbidity; Chitosan; Polyaluminum chloride; Water treatment

1. Introduction

Water contaminated naturally or by human intervention must be treated by different processes to be turned into drinking water. Common processes for water treatment include grit removal, coagulation, flocculation, sedimentation, purification, and disinfection. Coagulation is a process during which insoluble particles called colloids, which are the main factors contributing to turbidity, stick together and form coarser particles and settled particles. The water treatment process is impossible without using particles that increase the sedimentation rate of colloids [1]. Different coagulants and coagulant aids are used in the process of coagulation. Coagulants include materials

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which are used for the destabilization of particles, making them cling together. Coagulant aids are used for increasing the density of particles that are stuck together and help to speed up deposition [2]. Aluminum and ferrous salts are the most common coagulants used for water and wastewater treatment. However, during recent years, new types of coagulants called "Inorganic Polymer Flocculants (IPFs)" have been prepared using common ferrous and aluminum salts. Polyaluminum chloride (PACl) is one of the most common types of IPF and is more applicable than other types [3,4]. Hydrated aluminum chloride, or PACl, is mineral macromolecule, the monomers of which are a two-core complex of aluminum. This compound, when in low concentrations in aqueous solutions, comprises a multi-core complex which causes the unique capability of this coagulant in the coagulation process. This material has a polymeric structure with the formula $(Al_3(OH)_{b-x}\,Cl_x{\cdot}YH_2O)_z$ and is produced by reacting aluminum hydroxide with hydrochloric acid as given below:

$$2Al(OH)_3 + nHCL \rightarrow Al_2\,(OH)_nCl_{6-n} + H_2O \quad (1)$$

Value Z is a variable between 12 and 18, but for suitable formulation in 95% of compounds, Z is equal to 15.

Aluminum salts leave remnants in water, thus causing high volumes of sludge, the excavation of which causes serious environmental damages [5]. According to Finkel et al., of seven patients affected by encephalopathy dialysis, there was a significant correlation between increased concentrations of aluminum in the brain tissues with concentration of this element in the dialysis fluid [6]. To minimize any potential risk that may arise as a result of residual aluminum in the treated water where aluminum coagulants have been used, it is necessary to optimize the water treatment processes in order to reduce the rate of residual aluminum to the lowest possible level.

In recent years, there has been a plethora of research on natural coagulants, including chitosan application, in order to attempt to remove the complications that relate to chemical coagulants [7]. Chitosan is one of the most important chitin derivatives because of its distribution of acetyl groups from chitin. Chitin has a crystalline, hard, and white structure and is abundantly found in the shells of crustaceans, in insects and in the mycelium of fungi. Chitosan is made up of a chitin where more than 50% of its acetyl groups have been removed [8,9]. Fig. 1 illustrates the structure of chitosan. Chitosan is a nontoxic, biodegradable, and sustainable material

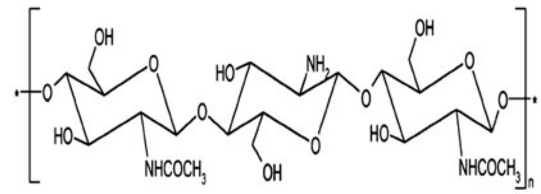

Fig. 1. Chitosan structure.

with antibacterial properties. It is a cationic linear polyelectrolyte with a high positive charge [10–12].

Since many materials come with negative charges, the positive charge of chitosan can mix with the negative surfaces of these materials and neutralize them. Due to its high number of $-NH_2$ groups, chitosan can react with negatively charged colloids [13]; it is therefore a suitable and effective coagulant for a wide variety of suspensions because of its excellent properties such as biodegradability, biocompatibility, and nontoxicity, as well as its polyelectrolyte properties [14].

Mahdinejad et al. studied the performance of chitosan and coagulant protein *Moringa oleifera* as coagulant aids combined with alum to remove the colloidal particles *E. coli* and *Streptococcus faecalis*. Optimal concentration of alum for low, average, and high turbidities is 40, 20, and 20 mg/L, respectively, and optimal pH is 7–7.5. Applying chitosan could reduce the concentration of the alum coagulant from 50 to 87.5% in different turbidities and leave the concentration of residual aluminum below 0.2 mg/L. The optimal concentration of alum when combined with chitosan for low, average, and high turbidities was 10 and 1 mg/L maximum turbidity for the removal of up to 74.3%, 5 and 0.5 mg/L maximum turbidity for the removal of up to 96%, and 5 and 0.5 mg/L maximum turbidity for the removal up to 98.2% [15]. Liu et al. studied the effect and mechanism of chitosan as a coagulant aid for removing the turbidity and organic materials in drinking water. The optimal concentrations were 35 mg/L of PACl, 0.15 mg/L of chitosan, and the optimal pH level was 7.5 [16]. Wang et al. studied the influence of chitosan as a coagulant aid on the shape and stability of flocculation. Under optimal conditions for the lessening of turbidity, the dosages of ferric chloride and chitosan were 29 and 0.1 mg/L, respectively. Flocculation formed by chitosan was coarser with higher sedimentation rates thus resulting in higher integrity flocculation [17]. Roussy et al. studied the influence of chitosan on coagulation and flocculation of bentonite particles. Using 0.1 mg/L of chitosan, turbidity was reduced by more than 95% for distilled water under pH 5 and treated water under pH 7. Distilled water under pH 7 required higher dosages for clearing the turbidity. Deacetylation by chitosan 89.5 and 95% with higher molecular weights indicated best result of turbidity removal [18]. Table 1 illustrates general specifications of chitin and chitosan.

This study aims to investigate and evaluate the efficiency of using chitosan as a coagulant aid combined with a coagulant-like polyaluminum chloride for the removal of the turbidity from Karoon river, Iran, as well as its influence on the coagulation process and sedimentation.

2. Materials and methods

2.1. Chemicals and tools

In this study, the commercial grade of PACl (30% w/w Al_2O_3) (FalizanTasfieh Co), chitosan powder with deacetylation degree of 85% (Sigma Chemical Co), NaOH 0.02 N, sulfuric acid 0.02 N, ascorbic acid, sodium aluminum acetate, and eriochrome cyanine R were used. The effects of various parameters were determined by carrying out a jar test procedure (using a JLT6 instrument). Measurement of pH was carried out using a 340ipH meter (WTW-Germany). Other analytical devices included a turbidity meter, model 2100 N (HACH), a TOC meter, model TOC-VCSH (SHIMADZU) and spectrophotometer, model DR/5000 (HACH).

2.1.1. Preparation of polyaluminum chloride solution

Polyaluminum chloride, 1,000 ppm, was prepared by dissolving polyaluminum chloride powder into distilled water at a rate of 1 g per liter.

2.1.2. Preparation of chitosan solution

To prepare the chitosan solution, 100 mg of chitosan powder was accurately weighed and dissolved in 10 mL of hydrochloric acid. This solution was then dissolved in distilled water to reach a volume of 100 mL. Any 1 mL of this solution contained 1 mg of chitosan [19].

2.2. Methodology

This study was conducted in the laboratory using a jar test device for the water treatment of Ahvaz, Iran. In the experiment, beakers with a capacity of 1,000 ml were used and were filled with 1,000 ml of water. Sulfuric acid and sodium hydroxide were used to obtain the desired pH. In order to determine the optimal pH of PAC, a PAC with constant concentration of 10 mg/L was added to all samples. Jar tests were then conducted on samples with the pH levels of 5, 5.5, 6, 6.5, 7, 7.5, 7.78, 8, 8.5, and 9. To study the concentration of the coagulant PAC, the jar test was carried out on water samples under optimal pH conditions as determined in the previous stage (pH 8) under different concentrations of PAC (3–50 mg/L), thus giving the optimal concentration of PAC. After determining the optimal concentration of PAC, in order to determine the optimal concentration of coagulant chitosan, a constant concentration of PAC (10 mg/L) was added to the water samples. By adding the optimal concentration of PAC to the water samples, a fast mixing process with a rate of 120 rpm was carried out for 1 min. Once the solutions were mixed, different dosages of chitosan were added as a coagulant aids (0.02–10 mg/L) to the water samples. This was mixed slowly for 20 min at a reduced rate of 40 rpm. After determining the optimal concentration of chitosan as a coagulant aid, in order to determine the optimal concentration of PAC together with chitosan as a coagulant aid, a constant concentration of chitosan (0.02 mg/L) and different values of coagulant PAC (1–20 mg/L) were added to the samples. Following this, jars were removed from devices to allow the formed coagulations to deposit for

Table 1
Physical properties of chitin and chitosan

Parameter	Chitin	Chitosan
Empirical formula of biopolymer unit	$C_8H_{13}NO_5$	$C_5H_{11}NO_4$
Polymerization degree	1,600–1,800	600–1,800
Molecular weight	(1–5) × 10^5	(1–5) × 10^5
Moisture (%)	2–10	2–10
Nitrogen (%)	6–7	7–8.4
Viscosity of biopolymer solution in acetic acid 1%(cp)	Insoluble	200–3,000
Dissociation constant (K_a)	6–7	6–7
Deacetylation (%)	10–15	70–85
X-ray peak	8° 58′–10° 26′ 19° 58′–20° 00′	8° 58′–10° 26′ 19° 58′–20° 00′

30 min. Final samples were obtained with a pipette 5 cm under the surface of the water in order to determine the turbidity, alkalinity, pH, TOC, and residual aluminum [20].

All tests were conducted in accordance with standard methods for water and wastewater tests. Turbidity was evaluated using a turbidity meter HACH, model 2100 N, and according to nephelometric method. Aluminum was measured using spectrophotometer Dr-5000 (3500-Al B. Eriochrome Cyanine R Method). TOC was measured using TOC meter, SHIMADZU, model TOC-VCSH (5310-B High Temperature Method) and alkalinity was measured by 2320 B. Titration and pH were measured using a digital pH meter, Model 340i (WTW). In addition, in order to prove the repeatability of the method of analysis, some samples were prepared two or three times for analysis. The total number of samples was 232. Figures were prepared using Microsoft Excel and data were analyzed using SPSS Software.

3. Results and discussion

This section shows the results of the determination of the optimal pH of PAC, the optimal concentration of coagulant and PAC, optimal concentration of chitosan as a coagulant aid, as well as the optimal concentration of PAC together with chitosan as a coagulant aid. For further review, some of these results were compared with the results of other studies.

3.1. Determining optimal pH of polyaluminum chloride

Any coagulant has an optimal pH in which the coagulation process for a given concentration of coagulant agent is conducted in the least amount of time with maximum efficiency. Therefore, it is necessary to determine this factor in order to ensure a strong coagulation performance. According to the authors, the only reliable method to determine a proper pH range in coagulation is by trial and error in conjunction with using laboratory results [21].

Average input turbidity during this stage of sampling has been calculated in the range of 33–34 NTU. During this time, average output turbidity during this step of sampling is between 2.08 and 6.81 NTU.

Fig. 1 indicates the influence of the pH level on the turbidity clearance by PAC. As indicated in the figure, the removal of the turbidity using PAC is very sensitive to pH changes, such that maximum average percentage of removal of the turbidity is 93.9% (with a confidence level of 95% in pH 8). In this range of pH, the deposits formed during the process and/or the same hydrolysis products of PSC have min solubility. According to the ANOVA test ($p < 0.05$), there is a significant difference between the pH level and the turbidity removal percentage. Another point to consider is that the levels of flocculation in the optimal pH solution, in comparison with other pH levels, were significantly high. According to Fig. 1, in acidic pH levels, turbidity rates decreased. For this reason, charge neutralization may not be considered as the only factor to affect flocculation. It seems that in this case, flocculation in the range of the optimal pH occurs mostly by trapping the particulates in the polymeric chains of the coagulant. This is called a sweeping coagulation mechanism and works by trapping the particles in sediment. Bina et al. also confirmed that the best condition for the coagulation of PAC is in pH 8 [22].

3.2. Determining the optimal concentration of the coagulant polyaluminum chloride

The average input turbidity during this stage of sampling has been calculated in the range of 44–45 NTU. At this time, average output turbidity was in the range of 0.89–10.64 NTU. Fig. 2 illustrates the effect of the concentration of polyaluminum chloride on the turbidity removal at optimum pH.

The results indicated that generally, by increased concentration of PAC, the turbidity removal rate increased such that the maximum average of turbidity removal percentage, in confidence level of 95%, for coagulant dosages of 10 and 30 mg/L, was 93.98 and 97.58%, respectively. According to the ANOVA test ($p < 0.05$), there was a significant difference between the coagulant dosage of PAC and the turbidity removal percentage. In the determination of the optimal concentration of a coagulant, different factors,

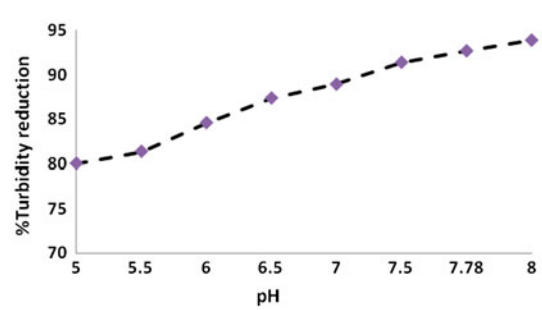

Fig. 2. Changes in turbidity removal percentage in comparison with the pH changes of the coagulant PAC. Ambient conditions, initial concentration of PAC: 10 mg/L, initial turbidity: 33–34 NTU, temperature: 24.5.

such as environmental regulations and coagulant price, are the most influential.

For turbidity removal using PAC and according to Fig. 2, it seems that removal rate of 93.95% is an eigenvalue or on the other hand a critical point in this figure. For this reason, and according to this figure, the optimum concentration of PAC for turbidity removal is 10 mg/L. Conversely, results indicated that mechanisms causing less residual turbidity in the water depend on the concentration of coagulant used and the pH level of the water. Under such conditions, the dominant mechanisms were absorptive, sweeping, or a combination of both [23,24].

3.3. Determining the optimal concentration of chitosan as a coagulant aid

The average input turbidity at this stage of sampling was calculated in the range of 62.67–132.6 NTU. At this stage, output turbidity in the sample with dosages of PAC between 0 and 10 mg/L with chitosan as a coagulant aid was in the range of 0.53–2.47 NTU. Fig. 3 illustrates the results. As indicated in Fig. 3, adding chitosan as a coagulant aid has increased the efficiency of the process. In the best state, i.e. when the concentration of PAC and chitosan is 10 and 0.02 mg/L, respectively, the turbidity removal percentage is 99.16%. According to ANOVA test ($p < 0.05$), there is a significant difference between chitosan as a coagulant aid and the turbidity removal percentage. The final turbidity of the samples with chitosan as a coagulant aid is in the range of 0.02–0.8 mg/L, which was lower than control sample. Under the higher dosages with chitosan as a coagulant aid within the range of 1–10 mg/L, the final turbidity was higher than the control sample, but was less than input turbidity. The reason for this behavior is that up to 1 mg/L, positively charged chitosan neutralizes the negative charges of the colloids and under such concentrations, the charge of the solution is neutralized.

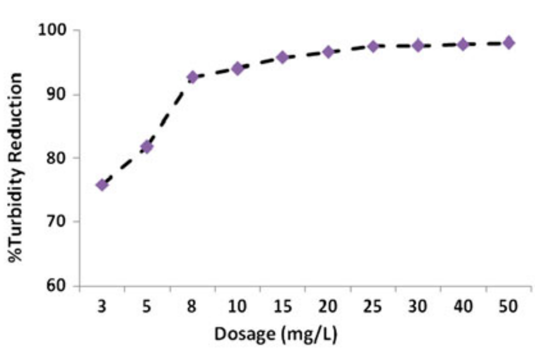

Fig. 3. Trend of changes in the turbidity removal percentage compared with dosage changes of the coagulant PAC. Ambient conditions, pH 8, temperature = 23.9, initial turbidity = 44–45 NTU, fast mixture speed = 120 rpm.

Additional injected chitosan either resulted in additional positive charges, or oppositely, resulted in reversed potential and increased secondary turbidity. Therefore, the optimal dosage for chitosan as a coagulant aid is 0.02 mg/L. Nonod et al., as well as other researchers, concluded a higher optimum dosage of chitosan in their works because of the higher turbidity of raw water [24]. Additionally, flocculation formed by chitosan was coarser than the control sample (10 mg/l PAC) with faster sedimentation speed.

During their coagulation process, Divakaran and Pillai achieved 80–95% of removal for initial turbidity between 10 and 160 NTU. Their optimal pH for removing the turbidity was 7–7.5 and their optimal dosage of chitosan was 0.5 mg/L. Under higher concentrations of chitosan, they showed stabilized particulates. Flocculation gave rise to coarser particles and faster sedimentation speed due to bridging mechanism between particles [19].

3.4. Determining the optimal concentration of PAC combined with co-coagulant chitosan

Coagulant aids such as activated silica, clay, and polyelectrolytes are usually used to achieve a greater efficiency in the process of coagulation and flocculation, as well as a reduction in the amount of coagulant needed and to form a stronger flocculation process with higher sedimentation [25].

Average input turbidity during this stage of sampling has been calculated in the range of 46.8–70 NTU. A total of 0.02 mg/L of chitosan used with PAC dosages between 1 and 20 mg/L gave an average output turbidity range between 0.64 and 7.98 NTU. Fig. 4 shows these results. The type of relation between additional PAC dosages and efficiency of turbidity removal in 0.02 mg/L is nonlinear and has a relation coefficient (R^2) between both variables of 0.9986. According to the figure, the increased dosage of PAC at this stage meant that the efficiency of turbidity removal increased from 1 to 5 mg/L and achieved 96.59% removal. However, amounts above 5 mg/L led to a slower increase. 5 mg/L is therefore considered to be the optimal point.

Results of the ANOVA test with a confidence level of 95% ($p < 0.05$) indicated a significant difference between the injected PAC and turbidity removal percentage. Results indicated that the combination of the two coagulants, PAC and chitosan, has the ability to remove the turbidity under the standard limit of 5 NTU. A combination of metal salts and polymers

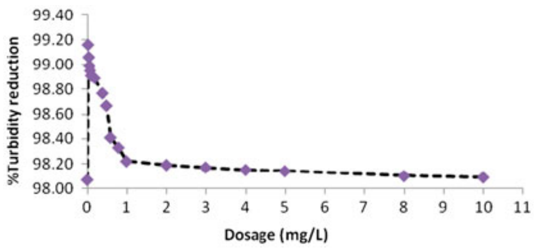

Fig. 4. Trend of changes in turbidity removal percentage compared with changes in co-coagulant chitosan. Ambient conditions, initial concentration of PAC: 10 mg/L, initial turbidity: 62.67–132.6 NTU, pH 8, temperature = 24.7, mixture speed = 120 rpm.

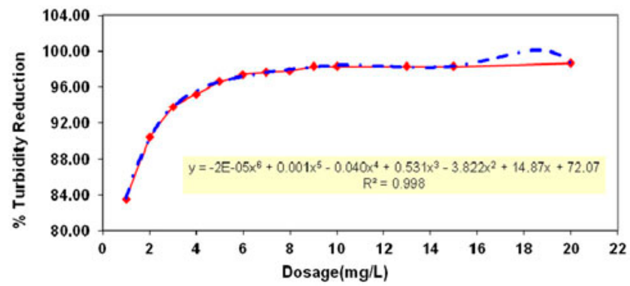

Fig. 5. Changes of the turbidity removal percentage compared with changes in the PAC dosage. Ambient conditions, initial concentration of chitosan: 0.02 mg/L; initial turbidity: 46.8–70 NTU; temperature: 26.2; fast mixing speed: 120 rpm.

may be a promising method for the treatment of extremely turbid raw water. Hu et al. used the chitosan and aluminum salt to treat surface water. They found that coagulation with the chitosan + aluminum salt mixture coagulant gave a lower residual turbidity and produced a lower volume of sludge. Consequently, the residual aluminum concentration, which could lead to Alzheimer's disease, can also be reduced significantly after the addition of chitosan [26].

Huang et al. indicated that the best result of turbidity removal was obtained under conditions where the optimal dosage of PAC was 4 mg/L and chitosan dosage was 1 mg/L [27]. The combining of chitosan with PAC in a 1:1 mass ratio significantly enhanced the coagulation process in a study carried out by Chen and Chung. They found that the highest floc settling velocity and coagulation efficiency occurred at a dosage of 5–6 mg/L of the coagulant mixtures [28]. In similar research, Chung et al. indicated that chitosan/PAC water-soluble coagulant mixtures illustrated much broader ranges of optimal concentrations for turbidity reduction than PAC alone [29]. Laboratory observations also indicated that flocculation occurring from the use of chitosan as a coagulant aid under optimal conditions, in comparison with the use of PAC alone, gave coarser particles and required less time for sedimentation. Initial tests indicated that 0.5 mg/L of polyelectrolyte forced 95% of flocculation deposits in two minutes. While if the coagulant is used separately, only 50% of flocculation deposits occurred in the same amount of time [30].

Roussy et al. indicated that the dominant mechanism for the removal of the turbidity by chitosan (due to higher number of amine groups and production of higher positive charge under neutralized, and to some extent acidic conditions) is charge neutralization [18]. Applying such materials has no effect on the alkalinity and pH of sample water before or after the jar test. Results of ANOVA test ($p > 0.05$) did not indicate a significant difference between the injected PAC (together with chitosan as a coagulant aid) and output alkalinity rate. Fig. 5 indicates the changes of input and output alkalinity according to the added PAC dosage and output alkalinity in the fast mixing stage. The relation between increased PAC dosage and output alkalinity in a dosage of 0.02 mg/L of chitosan is nonlinear, with an equation of six degree, and a relation coefficient (R^2) between both variables equal to 0.9885. Results of this study conformed to those of Divakaran and Pillai [19].

Results indicated that when PAC is used individually for the removal of turbidity, the aluminum residues in the treated water samples were between 0.08 and 1.4 mg/L, which was higher than the standard limit for drinking water as determined by the United States Environmental Protection Agency (USEPA) (Fig. 6). According to the USEPA, maximum aluminum content in drinking water is between 0.05 and

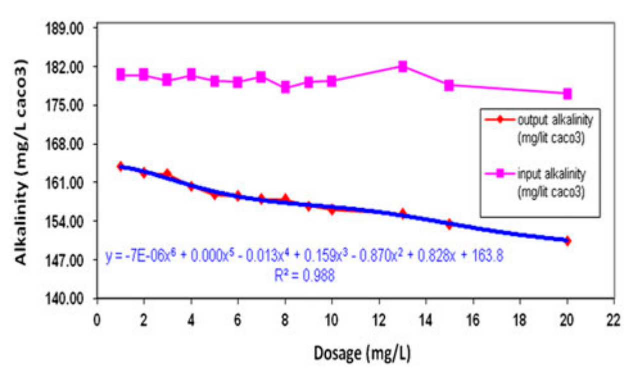

Fig. 6. Changes of the input and output alkalinity compared with changes in the PAC dosage added during the fast mixing stage. Ambient conditions, initial concentration of chitosan: 0.02 mg/L; initial turbidity: 46.8–70 NTU; pH 8; temperature: 26.2; fast mixing speed: 120 rpm.

0.2 mg/L [31]. While using chitosan as a coagulant aids, the aluminum residues in treated water samples were between 0.006 and 0.057 mg/L. Results indicated that higher concentrations of aluminum residues in treated water were caused by improper control of pH levels in nonoptimal conditions. This was important as the best way to reduce aluminum residues in treated water and to achieve optimal conditions in the coagulant's concentration and pH [32]. Using chitosan as a coagulant aid, while reducing the remaining turbidity to under 5 NTU, may reduce PAC usage by up to 50% under optimal conditions. Such a reduced use of PAC in the water treatment process, besides the reduction in the cost of purchasing the coagulant, may also reduce residual aluminum to below the standard level of 0.2 mg/L in treated water. Results of the application of chitosan as a coagulant aid in the coagulation process indicated that under optimal conditions, the content of TOC released is 0.56 mg/L. According to the TOC content obtained by chitosan, it was apparent that with an increased concentration of this material, the changes of TOC in treated water were ignorable, and under optimal conditions, the TOC content obtained was less than the control sample (0.78 mg/L). Laboratory observations and the efficiency of turbidity removal indicate that the presence of organic carbon had no considerable effect on the behavior during coagulation. Results of this study conformed to the studies of Mahdinejad et al. conducted on chitosan used as a coagulant aid [15].

4. Conclusion

In this study, it was found that pH was a very effective factor in the removal of turbidity and it was necessary for the pH to be adjusted for the optimal usage of the coagulant. The optimal pH for the removal of the turbidity with PAC was equal to 8. An increase in the concentration of PAC gave rise to an increase in turbidity removal. The investigation showed that the optimal concentration of PAC for turbidity removal was 10 mg/L. When the PAC concentration was adjusted to 10 mg/L, adding 0.02 mg/L of chitosan to the process, maximum turbidity removal was achieved at 99.16%. Under higher concentrations of chitosan, particles stabilized again. The optimal concentrations of PAC mixed with chitosan were 5 and 0.02 mg/L, respectively. The application of chitosan as a coagulant aid caused a reduction of aluminum residues in treated water to below the standard level (0.2 mg/L) as well as a reduction of PAC usage as a coagulant by up to 50%. Using chitosan also had no effect on pH and alkalinity.

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