



Investigating the feasibility of removing the rapid mixing unit in conventional surface water treatment and its effect on turbidity removal

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ABSTRACT

Coagulation and flocculation processes play a major role in surface water treatment. The aim of this study was to eliminate the rapid mixing unit in the water treatment plant. This experimental study was conducted on turbid water. Turbid water was synthesized by kaolin powder. The conventional jar test method was used. The flocculation and sedimentation processes were performed on the turbid water without a rapid mixing unit for getting the new optimal condition. When the PACl coagulant was used alone and in conjunction with chitosan, the percentages of turbidity removal in low, medium and high turbidities were obtained as 86.7%, 95.8%, 97.8% and 86.67%, 95.73%, 98.26%, respectively. When the rapid mixing unit was emitted, the efficiency of turbidity removal in the low turbidity was reduced from 5.26% to 21.73%. But, in higher turbidity in two states (presence and absence of the rapid mixing units) it did not have a significant difference. This study showed that the removal of the rapid mix unit on the removal efficiency of turbidity in the low turbidity is effective, but does not have an effect on higher turbid water. Also, using PACl in conjunction with chitosan was effective in the removal efficiency and to reduce residual aluminum.

Key words: coagulation, coagulants, jar test, rapid mix unit, turbidity

HIGHLIGHTS

- To decrease the cost of water treatment.
- Changing the process related to coagulation and flocculation stages.
- To decrease the time and the consumed coagulant.
- To increase efficiency of turbidity removal.
- Simplicity of process.

1. BACKGROUND

Surface waters contain particulates of inorganic and organic origin. Inorganic particulate constituents, including clay, and mineral oxides, typically enter surface water by natural erosion processes. Organic particulates may include viruses, bacteria, algae, protozoan cysts and oocysts. Removal of colloidal particles is one of the most important steps in water purification processes in coagulation, flocculation and deposition processes. This step is required because they can reduce the clarity of water to unacceptable levels and have toxic compounds adsorbed to their external surfaces. The most method common used to remove turbidity are coagulation/flocculation and sedimentation processes. The coagulation process is the rapid and uniform diffusion of chemicals (coagulants) into the raw water through the creation of intense turbulence, which is generally defined as 'rapid mixing', in the process of purification of the turbid waters, and the removal of parameters such as turbidity, color, infectious agents and toxic constituents in raw water (Crittenden *et al.* 2012). One of the important coagulants is polyaluminum chloride (PACl). The major part of polyaluminum chloride ($\text{Al}_2(\text{OH})_6\text{XCl}_x\text{YH}_2\text{O}$) is the Al_{13}^{+7} ion and its difference from aluminum sulfate is due to the type of aluminum structure present in them. The characteristic of PACl is that, besides chloride, it contains some of the hydroxide ions, and these hydroxide ions are formed by the formation of small polymer complexes of Al in PACl. So, due to the best fit of the electric charge to the radius, such polymer structures have a better effect on the stability of colloids than single Al^{+3} molecules in the alum. Polyaluminum chloride produces more positive charges in water. It works better than alum in

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the coagulation process of turbid waters with medium to high turbidity and also produces higher density sludge (Malakutian *et al.* 2011).

It is also interesting to note that coagulation is the process of decreasing or neutralizing the electric charge on suspended particles, or zeta potential. Similar electric charges on small particles in water cause the particles to naturally repel each other and hold the small, colloidal particles apart and keep them in suspension. The coagulation–flocculation process neutralizes or reduces the negative charge on the particles. This allows the van der Waals force of attraction to encourage the initial aggregation of colloidal and fine suspended materials to form microflocs (Ebeling *et al.* 2004). Chitosan is an abundantly available low-cost bio-polymer for turbidity removal that can be obtained from natural resources. It can use for different industries and water treatment too. This cationic polymer is a good alternative for the coagulation process instead of metal coagulants. In practice, coagulant aids are often necessary to further enhance the coagulation performance and guarantee a low concentration of residual Al in water when PACl is employed as the primary coagulant (Zhang *et al.* 2018).

With a strong emphasis on coagulation theory, some clarifier reactors have not been rapid mix units and in contrast in removal of turbidity by flocculating and settling process had a good efficiency. Kawamura wrote that in the enhanced coagulation and sweeping coagulation processes, rapid mixing has little effect on water quality. Some of the parameters such as local conditions, raw water characteristics, pressure drop and cost are important for designing rapid mixing systems (Kawamura 2000). Since no research has been done on the feasibility of rapid mixing unit removal in the water treatment process and its effect on turbidity removal efficiency, therefore, considering the costs of energy supply, the costs of construction, operation and maintenance of equipment of the rapid mixing unit, we decided to investigate the possibility of eliminating the rapid mixing unit in water treatment plant for treatment of turbid waters.

2. MATERIALS AND METHODS

In this semi-experimental study, kaolin powder was purchased from Merck Co., Germany. Three synthetic turbid water samples were prepared from kaolinite powder as low turbidity, medium turbidity and high turbidity.

2.1. Preparation of synthetic water

Preparations of turbid water samples were performed with kaolin powder according to Muyibi and Mehdinejad (Muyibi & Evison 1996; Mehdinejad & Bina 2018). 10 g of kaolin was suspended in 1 L of distilled water. The suspension was stirred vigorously for 1 h at 200 rpm to disperse kaolin. The suspension was allowed to stand for 24 h for complete hydration of the kaolin. This kaolin suspension was used as the stock solution for preparation of synthetic water samples. Three groups of different turbidities were prepared, including low turbidity (10–20 NTU), medium turbidity (100–120 NTU) and high turbidity (200–220 NTU). According to Table 1, alkalinity and hardness [$\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ (magnesium hardness) and $\text{CaCl}_2 \cdot 5\text{H}_2\text{O}$ (calcium hardness)] were added to water samples to produce water similar to natural water. The hardness and alkalinity of the samples were measured after the addition of kaolin over a day's interval. A stock solution of alkalinity was prepared using 16.8 g Na_2HCO_3 (Merck) and 1 L distilled water and resulting in a concentration of 10,000 ppm of alkalinity (Muyibi & Evison 1996). The hardness and alkalinity of the treated water samples were determined by the titrimetric method with EDTA (Method 2340-C in Standard Method) and by the titrimetric method with sulfuric acid (Method 2320-B in Standard Method) in the end of tests (APHA 2012).

Table 1 | Composition of synthetic water

Component	Chemical formula	Concentration (mg/l)
pH	–	7 ± 0.5
Magnesium hardness	$\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$	50
Calcium hardness	$\text{CaCl}_2 \cdot 5\text{H}_2\text{O}$	50
Alkalinity	Na_2HCO_3	100

2.2. Preparation of polyaluminum chloride (PACl) solution

Firstly, 10g of PACl powder with digital scales weighted and mixed with 1,000 ml of distilled water in a Jouleen balloon. This mixture was shaken well until all PAC particles were dissolved. Each 1 ml of the resulting solution contained 10 mg of PACl.

2.3. Preparation of chitosan solution

Chitosan (deacetylated chitin; poly- [1- 4] - β - glucosamine) with minimum %85 deacetyl prepared from crab shells was obtained from GMA Chemical Company. 100 mg of chitosan powder was accurately weighed and mixed with 10 mL of 0.1 M HCl solution, and kept aside for about one hour to dissolve. It was then diluted to 100 mL with distilled water to obtain a solution containing 1.0 mg chitosan per mL of solution (Mehdinejad *et al.* 2009; Abdullah & Jael 2019).

2.4. Experimental procedures

A standard jar test apparatus, the Phipps and Bird six-paddle stirrer at six beakers of 1,000 ml volume was employed for the tests. The jars are provided with a sampling port, 10 cm below the water line, which allows for repetitive sampling with minimal impact on the test (Ebeling *et al.* 2004). All experiments were performed in two stages. In the first stage, the process of turbidity removal was performed with the presence of the coagulation stage and in the second stage without the coagulation stage. The coagulation was performed in six beakers of 1,000 ml volume with rapid mixing at 120 rpm for 60 s and then the flocculation step with slow mixing at 40 rpm for 7.5 min and 20 rpm for 7.5 min was carried out. The time of settling process at all of tests was 20 min in final. In this study, the optimal PACl concentration and the optimal concentration of PACl in conjunction with chitosan were determined in two states of with and without rapid mix unit in jar tests.

In this study, pH values were used in range from 7 to 7.5 according to past research (Mehdinejad *et al.* 2009). Turbidity removal efficiency in all of tests was measured in all of synthetic water samples (low turbidity, medium turbidity and high turbidity). The turbidity and pH of the samples was measured by a turbidimeter (DR2100P, HACH) and by pH meter HACH model, respectively.

2.5. Statistical analyses

The data presented in the figures are the arithmetic mean values of the duplicate measurements. The data was analyzed by SPSS (Version 18) software. $P < 0.05$ were considered statistically significant. Independent sample t-test was used to evaluate differences between the means. Correlation between two variables was analyzed using bivariate analysis of variance.

3. RESULTS AND DISCUSSION

3.1. Determination of optimum dose of PACl with using a rapid mix unit

The first, the initial turbidities of turbid water samples were measured. pH was adjusted about 7–7.5 in all of tests. For determining of optimum dosage of PACl, experiments were conducted by adding dosages of PACl between 0 and 45 mg/L in three groups of present synthetic water. The obtained results are shown in Figure 1. As shown, optimum dose of PACl was obtained 35, 30 and 30 mg/l in low turbidity, medium turbidity and high turbidity, respectively. The percentage of turbidity removal in low, medium and high turbidities was % 86.7, %95.8 and %97.8, respectively. In blank samples, the percentage of turbidity removal was %3.6, %15.8 and %40.4 in low, medium and high turbid water, respectively. Residual turbidity was obtained in accordance with drinking water standards and was less than 5 NTU (APHA 2012). PACl was hydrolyzed better than alum coagulant and produced more positive charges in suspension. The use of this coagulant often leads to lower aluminum in treated water in comparison with alum. PACl consumes lower alkalinity and as well, lower sludge produce in comparison with alum. These characteristics of PACl caused this coagulant to be used more in the water treatment process. The solubility of alum in water is strongly dependent on the pH. At higher pH (7–7.5), the efficiency of polyaluminum chloride is lower than that of alum due to the lower number of positively charged monomers (Al^{+3} , SO_4^{-2}) in solution by polyaluminum chloride. The obtained results by Mehdinejad *et al.* showed that the amount of released aluminum ions in treated water by coagulation and flocculation process have been in the range of 0.02–0.1 mg /l and 0.02–0.04 mg/l by using alum and polyaluminum chloride coagulants, respectively. In polyaluminum chloride coagulants, most of the aluminum ions are in the form of oligomers and polymers, and there is only a small proton of aluminum similar to aluminum sulphate (alum) in monomeric form. This

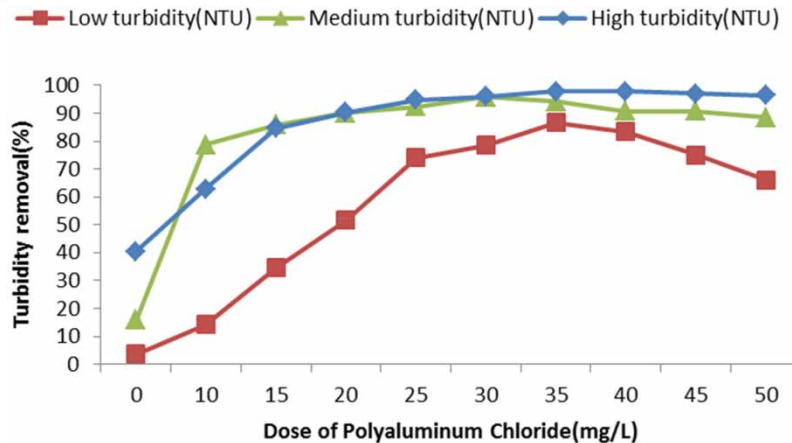


Figure 1 | Determination of optimum dose of polyaluminum chloride: (a) low turbidity = 30 NTU; (b) medium turbidity = 120 NTU; and (c) high turbidity = 225 NTU (pH = 7–7.5; rapid mix time = 1 min, shaking rate = 120 rpm; flocculation1 step: slow mix time = 7.5 min, shaking rate = 40 rpm; flocculation2 step: slow mix time = 7.5 min, shaking rate = 20 rpm; settling time = 30 min).

causes a large difference between aluminum sulphate and polyaluminum chloride compounds. By neutralizing the negative charges of colloidal particles by polyaluminum chloride, the possibility of monomeric aluminum is less due to the turbidity of the water and the amount of charge neutralization, and therefore in the treated water in the form of residual aluminum is less apparent. This phenomenon is the reason for the technical superiority of polyaluminum chloride in flocculation over alum that due to the less number of available monomers, it consumes less alkalinity.

In this coagulation process, colloids destabilization and adsorption are a predominant mechanism for the removal of turbidity by PACl. As shown in Figure 1, the removal percent increase as the PACl dose increases until reach to optimum dose. After getting to optimum dose, efficiency of turbidity removal decreased. Floccs formed by PACl coagulant were stable and the breakage of floccs seldom appeared during the growth period. One of the most important reasons for this phenomenon can be restabilization of destabilized floccs in suspension. Additionally, our study showed that efficiency of turbidity removal was affected from initial turbidity and with increasing of turbidity, efficiency of turbidity removal was increased. These results were in agreement with the performed studies by Ebrahimi and Zhang (Ebrahimi *et al.* 2014; Zhang *et al.* 2018).

As seen in Figure 1, at medium and high turbidity, the removal efficiency of turbidity improved significantly and there was a significant correlation between turbidity removal in low turbidity with medium and high turbidity ($P < 0.05$).

3.2. Optimization of PACl in conjunction with chitosan as coagulant aid with using rapid mix unit

In order to measure the turbidity removal, the jar tests were accomplished. The results of use of PACl in conjunction with chitosan are shown in Figure 2. The optimum doses of PACl in conjunction with chitosan were obtained as 10 and 0.5 mg L⁻¹ at low turbidity, 10 and 0.5 mg L⁻¹ at medium turbidity and 10 and 1 mg L⁻¹ at high turbidity. The best of turbidity removal efficiency were obtained 86.7%, 95.7% and 98.3% at low, medium and high turbidities, respectively. These results showed that the obtained residual turbidities in all the turbidity levels were the below 5 NTU and were in accordance with drinking water standards (US EPA 2009; 816-F-09-004). As expected, the highest removal efficiency occurred at high turbidity. Furthermore, using chitosan coagulant as a coagulant aid, the amount of polyaluminum chloride was reduced compared to using polyaluminum chloride alone. This phenomenon showed that the use of chitosan can not only help reduce the consumption of PACl but can also greatly reduce the turbidity of water. Generally, in the coagulation process, turbidity and alkalinity are two important factors, especially in low turbidity. The colloids destabilization and adsorption are a predominant mechanism for the removal of turbidity by PACl. In the coagulation process, the use of PACl alone in low turbidity, due to the lack of high colloidal particles and low negative charge, means the removal of turbidity is difficult. Due to the properties of chitosan, the mechanism of action of this substance is the neutralization of the ions charges and bridging between particles. For this reason, due to the bridging nature of this coagulant, when used with PACl, in addition to neutralizing the charge, bridging between particles also occurs. Therefore, the use of PACl in combination with chitosan has increased turbidity removal efficiency. Many studies have been

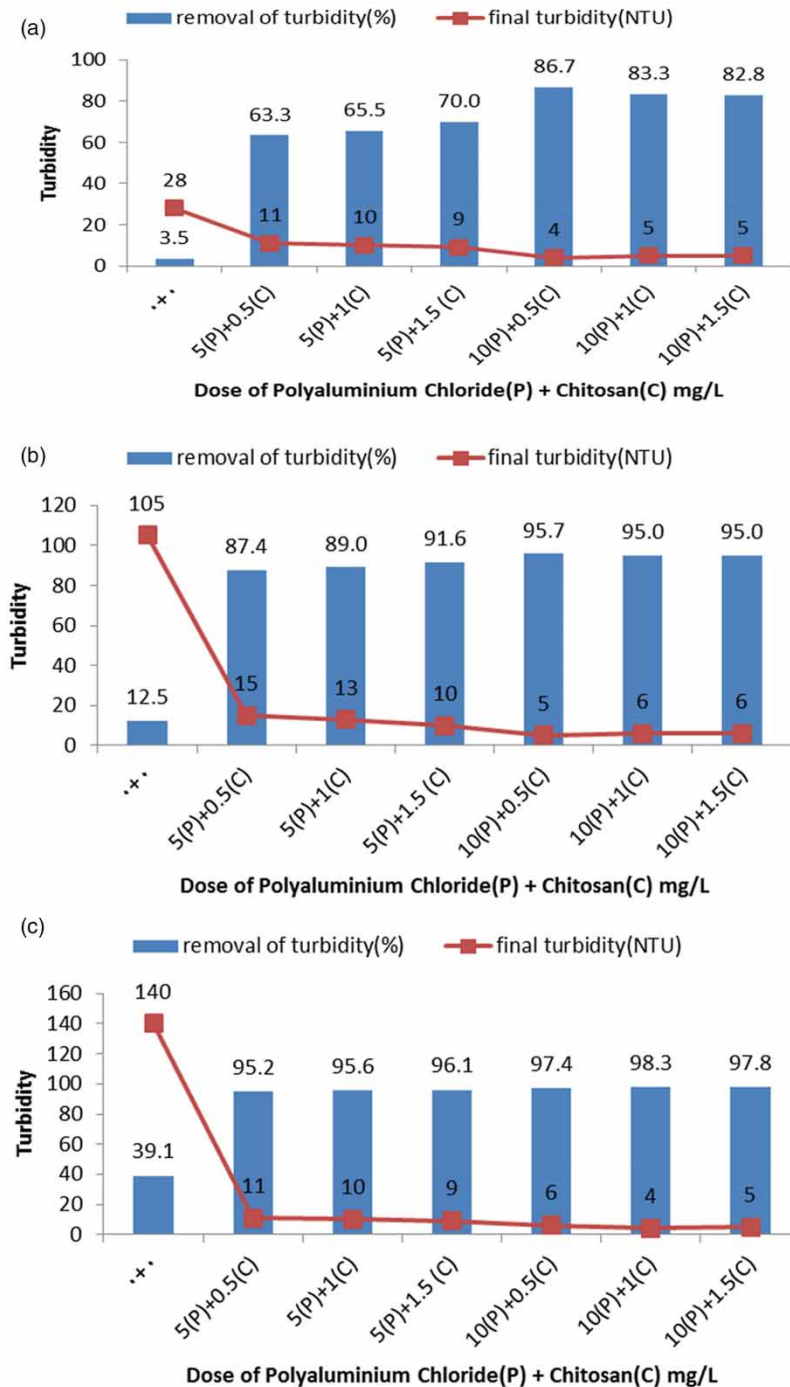


Figure 2 | Determination of optimum dose of polyaluminum chloride in conjunction with chitosan: (a) low turbidity = 30 NTU; (b) medium turbidity = 120 NTU; and (c) high turbidity = 230 NTU (pH = 7–7.5; coagulation step: shaking rate = 120 rpm; rapid mix time = 1 min, flocculation1 step: shaking rate = 40 rpm; slow mix time = 7.5 min; flocculation2 step: shaking rate = 20 rpm; slow mix time = 7.5 min, settling time = 30 min).

done in this regard and their results confirm our findings. On the other hand, using chitosan can create heavier flocs that can be effective in reducing settling time. Previous studies have illustrated that the use of chitosan together with an Al-based salt/polymer is effective for removing turbidity (Zemmouri *et al.* 2012; Zhang *et al.* 2018). As can be seen in Figure 2(a)–2(c), the mechanism for removing turbidity and determining the optimal dose in these conditions does not have a special order and in many cases, by increasing the coagulant and coagulation aid, the efficiency of turbidity removal has decreased. Our results showed that the use of these two substances had decreased the PACl consumption by up to 50% (Figures 1 and 2). These results were in agreement

with another study by the author in 2009. In that study, the use of chitosan as a coagulant aid in combination with alum reduced the required dosage of alum between 50 and 87.5% (Mehdinejad *et al.* 2009).

3.3. Comparison of turbidity removal with using the rapid mixing unit and without the rapid mixing unit by using PACl alone

The purpose of the rapid mixing stage is to provide uniform and fast coagulant dispersion in the water and to destabilize the particles. Therefore, if we intend to eliminate the rapid mixing unit, the conditions of the flocculation and settling process must change. For this purpose, the flocculation process was performed with the following conditions. By eliminating the rapid mixing unit, the flocculation process was performed with a mixing intensity of 40 rpm at 7.5 min followed by a mixing intensity of 20 rpm at 7.5 min. The time of settling process in all tests was 20 min in final. The optimum doses of PACl used were 35 mg/L at low turbidity, 30 mg/L at medium turbidity and 30 mg/L at high turbidity. According to Table 1, in the low and medium turbidities, the removal percentage of turbidity in the absence of the rapid mix unit decreased from 86.7% to 73.8% and 95.8% to 91.8%, respectively. But, in high turbidity, the efficiency of turbidity removal was almost constant. The obtained results at this stage showed that the presence of the rapid mix stage in high turbidities has little effect on turbidity removal. Due to the aforementioned facts, the rapid mix unit in high and medium turbidities with the higher confidence percent, in comparison with low turbidities, can be eliminated in the water treatment process. The cost benefits analysis of building a rapid mixing unit depends on many factors such as the quantity of influent water, tank volume and agitator and other instruments. Generally, the cost of building a rapid mixing unit is equivalent to 2,40,000 \$ in Iran (assuming that : LPCD = 150 and population = 1 million), which is about one-fifth the cost of building a slow mixing unit in Iran country. As is seen, in this country, this cost is very high and this can be spent on other matters in the WTPs.

The results of the statistical analysis showed that significant differences were also not observed in medium and high turbidities on the removal turbidity when the process was used in two modes, using a rapid mix unit and without using a rapid mix unit ($p > 0.05$). But in low turbidities, a significant difference in removing turbidity in the coagulation and flocculation process ($p < 0.05$) was observed. Furthermore, it is not recommended that the rapid mix unit is eliminated for turbid waters with low turbidities.

3.4. Determination of the optimum time in the slow mix unit in the absence of rapid mix unit in water treatment process

The result of Table 2 indicated that in the normal condition of the jar test (including coagulation, flocculation and settling processes), the percentage of turbidity removal in different turbidities was 86.7%, 95.7% and 97.8%, respectively. In addition, the percentage of turbidity removal in different turbidities without the rapid mix unit was 73.8%, 91.88% and 97.17%, respectively.

Table 2 | Comparison of turbidity removal in water treatment process using the rapid mix unit and without the rapid mix unit in low, medium and high turbidities

Type of turbidity	Dose of PACl (mg/l)	Primary turbidity (NTU)	^a Percentage of turbidity removal with rapid mix unit	^b Percentage of turbidity removal without rapid mix unit	P value	Percentage of changes (%)
Low turbidity	35	28	86.7 ± 1.6	73.8 ± 2.6	$P < 0.05$	-13
Medium turbidity	30	119	95.8 ± 3.4	91.88 ± 4.7	$p > 0.05$	-4
High turbidity	30	226	97.8 ± 2.3	97.17 ± 1.07	$P > 0.05$	-0.7

^aWith rapid mix unit.

^bWithout rapid mix unit.

In this section, the main goal is to eliminate the rapid mixing unit; therefore, new optimal conditions for slow mixing time and intensity in the flocculation process must be considered. In this step, the optimal dosages obtained in different turbidities in the previous step (Figure 2) were used to obtain the optimal times for these doses, which include 10 mg L⁻¹ as PACl and 0.5 mg L⁻¹ as chitosan at low turbidity, 10 mg L⁻¹ as PACl and 0.5 mg L⁻¹ as chitosan at medium turbidity and 10 mg L⁻¹ as PACl and 1 mg L⁻¹ as chitosan at high turbidity. The flocculation process was performed at slow mixing speeds of 40 and 20 rpm and settling time of 20 min including: (5 min stage at 40 rpm, 5 min stage at 20 rpm), (7.5 min stage at 40 rpm, 7.5 min stage at 20 rpm), (10 min stage at 40 rpm, 10 min stage at 20 rpm). In all stages, the settling time and pH were 20 minutes and 7 ± 0.5, respectively.

As shown in Figure 3, the highest turbidity removal (98.7%) was obtained in high turbidity at the optimum time of 7.5 min stage at 40 rpm, 7.5 min stage at 20 rpm and the lowest turbidity removal was achieved at the optimum time of 5 min stage at 40 rpm, 5 min stage at 20 rpm in low turbidity. By comparing the turbidity removal efficiency at different times according to Figure 3, the best optimum mixing time was 7.5 minutes. Bivariate analysis of variance has showed significant differences in turbidity removal with the changes of times in the slow mixing process between low turbidity with medium turbidity and high turbidity ($P < 0.05$).

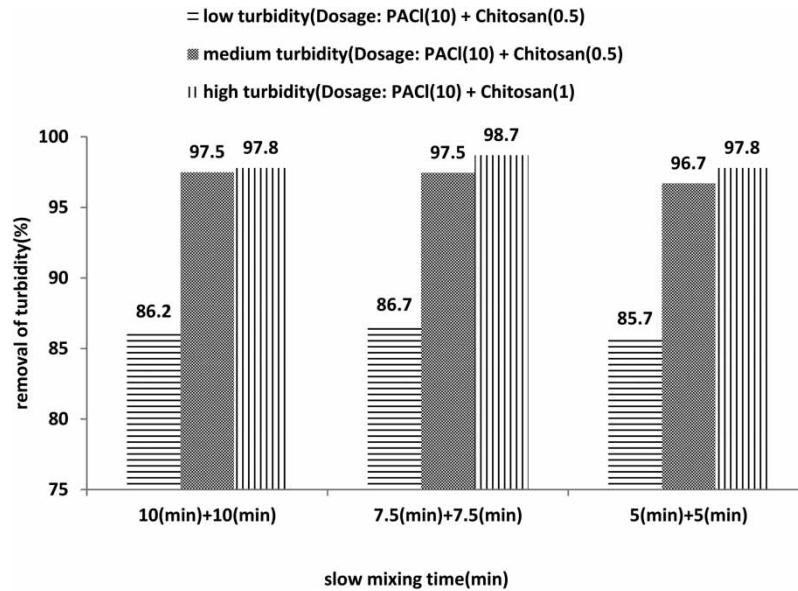


Figure 3 | Determination of the optimum time in the slow mix unit in absence of rapid mixing unit (pH = 7–7.5; flocculation1 step: shaking rate = 40 rpm; flocculation2 step: shaking rate = 20 rpm; settling time = 30 min).

3.5. Determination of residual aluminum in treated water

The amount of residual aluminum was determined in two stages, including: using PACI alone and using PACI in conjunction with chitosan. PACI coagulant when used alone, the amount of residual aluminum in the treated water in low, medium and high turbidities were obtained as 1.1 mg/L, 1.7 mg/L and 2.1 mg/L, respectively. Residual total Al^{+3} first decreased at lower dosage and then increased when the dosage further increased. According to the EPA national secondary drinking water regulation, the maximum contaminant level range is between 0.05 and 0.2 mg/L for residual Al^{+3} (US EPA 2009; 816-F-09-004). This means that the use of PACI leads to an amount of residual aluminum in treated water after the coagulation/flocculation process that was not in agreement with USEPA standards. But the use of PACI in conjunction with chitosan as a coagulant aid reduced the amount of aluminum ions (Al^{+3}) in the treated water according to Figure 4. As can be seen, residual Al^{+3} ions

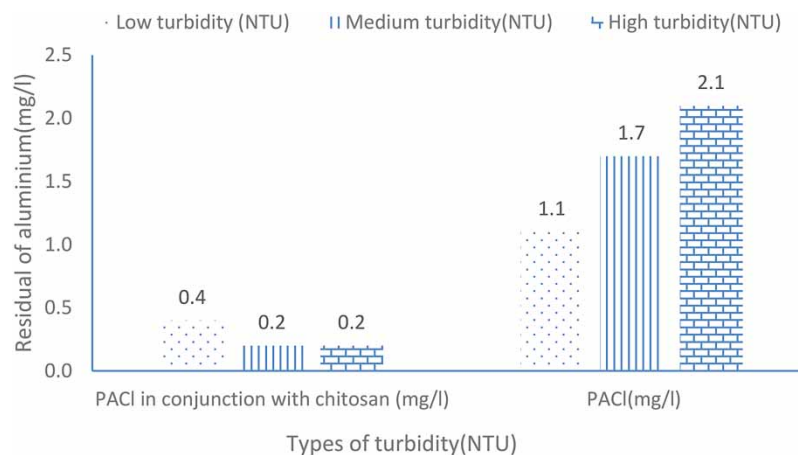


Figure 4 | Residual aluminum in turbid waters by using PACI and PACI in conjunction with chitosan.

decreased obviously when initial PACl in conjunction with chitosan increased. This phenomenon was related to hydrolysis of these coagulants. In fact, chitosan as a coagulant aid effected the performance of coagulation and prevented an increase in the dose of PACl. Chitosan is a natural linear polymer, which contains reactive functional groups, including an amino group and both primary and secondary hydroxyl groups at the C-2, C-3 and C- 6 positions (Cheng *et al.* 2005). The amine group can provide positive charge and bind with metals such as Al via the electron doublet (Kumar *et al.* 2014). Besides, the high content of the hydrophilic hydroxyl group in chitosan can promote the chelation of cations such as Al^{3+} via hydrogen-bonding interactions. This can have a huge impact in reducing the damage caused by aluminum in the water, and especially in incidence of diseases such as Alzheimer's, Parkinson's and multiple sclerosis. These result agree with results obtained by Mehdinejad *et al.* (Mehdinejad *et al.* 2009), who reported a decrease in the residual Al^{3+} in treated water when chitosan as a coagulant aid in the coagulation-flocculation process was used.

CONCLUSION

The following conclusions were drawn from this study:

- Removals of colloidal particles by PACl in conjunction with chitosan were more than when PACl was applied alone.
- The more dose of PACl coagulant was consumed in comparing with PACl in conjunction with chitosan for removing of turbidity
- The obtained results showed that the removal of the rapid mixing unit has little effect on the removal of turbidity in high and medium turbidities. However, in low turbidity, the percent difference in turbidity removal was about 13%.
- It is not recommended that the rapid mix unit is eliminated for treatment of turbid water with low turbidities in water treatment plants.
- Without the rapid mix unit, the optimum time obtained was 7.5 min at 40 rpm and 7.5 min at 20 rpm for the flocculation unit. This result showed that the detention time in the flocculation unit could be less than the standard limit (30–60 min). This result can help in designing rapid mix and flocculation tanks in terms of detention time and construction.
- Without the rapid mix unit, a decrease in the residual Al^{3+} in treated water was observed to the limit of USEPA standards when we used chitosan as a coagulant aid in the coagulation-flocculation process.
- In the coagulation process, not having a rapid mix unit can reduce the economic cost of building a treatment plant.
- Generally, the cost of building a rapid mixing unit is equivalent to one-fifth of the cost of building a slow mixing unit in the country of Iran. As is seen, in this country, this cost is very high and can be spent on other matters in the WTPs.
- Without the rapid mix unit, the costs will be lower and operation problems will reduce. This study can be a new primary idea in design of WPTs and can improve the treatment process.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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