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# Effect of Chitosan as a Coagulant Aid Combined With Poly Aluminum Chloride Removing of Turbidity From Drinking Water

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Chitosan, a biodegradable polymer, is used as an eco-friendly coagulant in a wide variety of applications in water and wastewater treatment. The present study aimed to investigate the effect of chitosan as a coagulant aid combined with poly aluminum chloride (PAC) to enhance coagulating efficiency for bentonite suspensions. A conventional jar test apparatus was used for the tests. The effect of various operational parameters, such as initial pH of the solution (5-9.5), dosage of chitosan (0.5-3.5 mg/L), dosage of PAC (5-35 mg/L) and initial turbidity (50-200 NTU) were investigated. The maximum turbidity removal rates were obtained as pH 8.5 for PAC and pH 7.5 for combined PAC and chitosan (CPC). The coagulating efficiency of bentonite using PAC and CPA was found to decrease with an increase in the pH value of the solutions. The maximum turbidity removal rate was achieved in coagulating by PAC (30 mg/L) alone, and PAC (20 mg/L) combined with chitosan (2.5 mg/L) as coagulant aid with the removal rate of 87% and 96%, respectively. The optimum dosage of chitosan required to obtain the highest removal rate was 2.5 mg/L. Hence, using chitosan as a coagulant aid can not only reduce the required amount of coagulant (35%) but can also enhance the removal turbidity efficiency.

*Keywords:* Water Purification; Chitosan; Coagulants

## 1. Introduction

Generally, source of potable water contains a wide variety of suspended and colloidal materials that may cause turbidity, which is stable and restricts the treatment of raw water. Aesthetics, filterability and disinfection processes are three main reasons for consideration of turbidity in public water supplies (1). As a result, removal of colloidal particles is the first step for treating of raw water. Production of drinking water from most raw water resources usually implicates the use of a coagulation/flocculation process for removal of turbidity (2). This process plays an important role by reducing most impurities such as algae, bacteria, color, organic compounds, clay and silt particles in surface-water treatment (3). Inorganic metal coagulants such as aluminum sulfate and poly aluminum chloride (PAC), $(Al_2 (OH)_n Cl_{6-n})_m$, are most widely used for removal of water turbidity because they are cost effective, easy to handling and availability. However, high level of aluminum remained in the treated water to ensure coagulation efficiency has raised potential public health impacts (4-8). For example, McLachlan discovered that the intake of large quantities of aluminum salt such as aluminum chloride or aluminum sulfate is now controversial due to their possible contribution to Alzheimer disease (9). The research for the best alternative to meet the increasing demand for water quality has become an important area of study. Polymers alone or combined with metal salt coagulants have been applied in the coagulation, and regularly gained popularity in water treatment systems. Synthetic polyelectrolytes produce sludge of better dewatering characteristics. Also, the sludge is smaller in volume than those from conventional coagulation-flocculation process. Conversely, some reports have shown that polymers used in water and wastewater technology may have some disadvantages such as residual monomers, other reactants and reaction by-products that could potentially have negative effects on human health (10, 11). Hence, the increasing demand for environmentally friendly technologies has sparked interest in natural polyelectrolytes, which can replace synthetic flocculants (10). Accordingly, many studies have focused on the use of low-cost biological products (known as biopolymers) such as chitosan, tannins, aqueous extract of the seed of Moringa olefiera and extract of Okra and Nirmali seeds (12). Synthetic biopolymers are safe for human health, biodegradable and have a wider effective dosage range for flocculation of colloidal suspensions (10, 13). The most appropriate cationic biopolymer for an extensive application is chitosan (14). Chitosan as an eco-friendly coagulant and coagulant aid is a linear copolymer of D-glucosamine and N-acetyle-D- glucosamine extracted by means of deacetylation from chitin, which is the most abundant biopolymers in nature after cellulose (10-15). In recent years, chitosan has studied for use as a coagulant for a wide variety of applications in water and wastewater treatment including removal of silt in river (16, 17), fish processing (18), the food industry (19) and azo-dye removal (20). To the best of our knowledge, the removal of colloidal particles from raw water by coagulation process with the application of chitosan as a coagulant aid combined with aluminum sulfate has previously demonstrated (12). However, the application of combined PAC and chitosan as coagulant aid for removal of turbidity from water in coagulation-flocculation process has not been studied.

The purpose of this study was to evaluate the ability of chitosan as an eco-friendly coagulant aid with PAC for the removal of turbidity in coagulation-flocculation process from turbid water. The experimental procedure considered the effect of initial pH, initial dosage of PAC and chitosan separately and the initial turbidity amount.

## 2. Materials and Methods

### 2.1. Preparation of Reagents

Chitosan with minimum of 85% deacetyl was purchased from Sigma Aldrich Chemical Co, (St. Louis, MO, USA). It was ground and sieved using 80 µm mesh size and used to prepare the coagulant solutions. To facilitate the preparation of the 0.5% stock polymer solutions, chitosan was hydrated in deionized water, after which 1% dilute solution of acetic acid was added under agitation (20). Figure 1 shows the chemical structure of chitosan. Bentonite powder was purchased from Haman Trading Company, Iran, added without purification into $NaClO_4$ solution (ionic strength = 10-2N). A $NaClO_4$ solution was used in the preparation to provide background ionic strength in the distilled water.

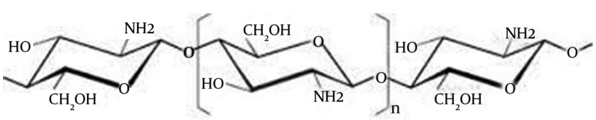



**Figure 1.** Chemical Structure of Chitosan

### 2.2. Procedures

The coagulation-flocculation experiments were performed by jar test apparatus (Phips and Bird, USA). A standardized set of stirring speeds, i.e. 100 rpm and 40 rpm for rapid and slow stirring was done for comparative purposes, respectively. The initial pH of the synthetic water was adjusted to a fixed value depending on the experiments with using (0.1-1 mol/L) $H_2SO_4$ and/or NaOH solutions. For experiments in each run, the homogeneous synthetic turbid water containing different amounts of pH, turbidity, PACl and chitosan was separated into several Plexiglas beakers, each containing, 1000 mL included six paddle stirrer for mixing. The coagulant aid was added one min after the addition of PAC according to Bina et al. (12).

The mixtures were then stirred at high velocity for 1 min, after which the velocity was decreased to 40 rpm and maintained for 30 minutes. After that, the stirring was then stopped and the experiments were performed at room temperature. Following a settling time of 30 min the samples were taken from 3 cm below the top of the settling beaker (5).

### 2.3. Analysis

Turbidity of the samples was measured by the nephelometric method using a turbiditimeter (2100 P, HACH Co) as described in the standard methods (21).

## 3. Results and Discussion

### 3.1. Effect of Influent pH

Since the pH of the aqueous solution has a key role in the coagulation-flocculation processes and its influence on forming different metal hydroxide species (8, 22), the effect of initial pH value of the suspension on the removal efficiency of turbidity was studied in a range of 5-9.5 in coagulation process with chitosan (15 mg/L), PAC (30 mg/L) and CPC (15 mg/L and 2.5 mg/L for PAC and chitosan, respectively) in 50 NTU initial turbidity. Figure 2 shows the turbidity removal efficiencies at different initial pH value. It can be seen that initial pH amounts affects the enhanced coagulation. The results from Figure 2 indicated that in coagulation with PAC alone, when the pH amounts of solution elevates from 4 to 8.5, the removal efficiency of turbidity increased from 40% to a maximum of 87% and then the removal efficiency decreased to 62%, as the pH 9.5. According to Figure 2, as the pH amounts increases from 5 to 7.5, the turbidity removal efficiency increases from 58% to 93% for CPC; however, it decreases from 93% to 75%, when the pH level of solution increases to 9.5. When the pH is lower than 5, the primary hydrolyzates of PAC are some positive hydrolyzates like $Al(OH)_2^+$, $Al(OH)_2^+$, $Al_2(OH)_2^{4+}$ and $Al_3(OH)_4^{5+}$. These positive hydrolyzates are easy to neutralize the exterior negative charges of colloidal material and further destabilize the colloids, and they also benefit the physical or chemical adsorption of the destabilized colloids, which leads to floc growth (23). This can be explained by considering the aluminum species in solution. When the pH is between 6 and 8, there are some high polymeric positive hydrolyzates and $Al(OH)_3$ formed in solution (3-7, 20-22). When the pH is higher than 8, the suspension system is difficult to be destabilized because the hydrolyzates are transformed to $Al(OH)_4^-$ (3-7, 20-25). As Figure 2 shows, in coagulation with chitosan, the highest turbidity removal rate was observed in the solution pH of 6 and the removal rate was decreased with increases and decreases beyond pH 9.5 and 5, respectively. The reactivity of chitosan for coagulation and flocculation of suspended particles results from several mechanisms, i.e. electrostatic attraction, biosorption (correlated to protonation of the amine group of chitosan and chelating capacity due to the high content of hydroxyl groups) and bridging (correlated to the high molecular weight of chitosan). The contribution of each mechanism depends on the pH of the suspension (7). Thereby, this phenomenon can be attributed to the increase in number of protonated amine groups on chitosan at lower pH (8). The results of the recent studies showed that the chitosan, as a primary coagulant, has better performance in acidic solution and needs to be adjusted to the pH values of the solutions (13, 14).

However, our results showed that when chitosan used as a coagulant aid with PAC at the low dosage, an adjustment of pH on turbidity removal rate can be neglected in the water treatment system. Therefore, the optimum pH amount was about 7.5 for CPC and thus the rest of experiment was conducted at the optimum pH amount.

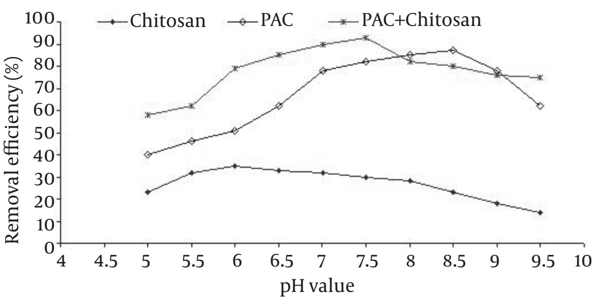
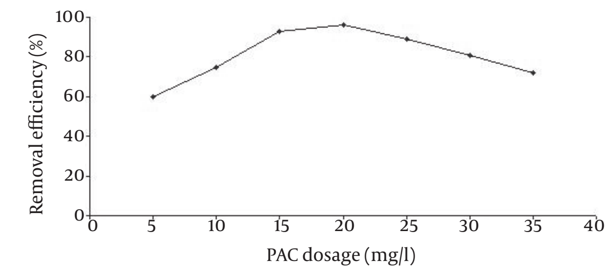



**Figure 2.** Effect of Initial pH on the Turbidity Removal Efficiencies in Different Conditions

## 3.2. Effect of Coagulant Dosage

The optimum dosage of coagulant is one of the most important parameters in coagulation-flocculation process, which determines the optimum operational condition for the performance of metal salt coagulants (14). The poor flocculation performance may be caused by either the insufficient coagulant dosage or overdosing. For that reason, determining of the optimum dosage is important to reduce the chemical cost and sludge formation. The effects of various concentrations of PAC as a main coagulant in the range from 5 to 35 mg/L at constant 2.5 mg/L chitosan dosage were investigated in initial turbidity of 50 NTU. The results are presented in Figure 3 clearly indicated that the coagulation efficiency of PAC within the dosage of 5-35 mg/L at the optimum pH value was significantly dependent on the dosage of coagulant. As shown in Figure 3, the turbidity removal efficiency raises to 96% as the coagulant increases up to 20 mg/L. Moreover, when the coagulant dosage increases to 35 mg/L, the PAC exhibits the serious restabilization, and the turbidity removal rate is only about 72%. As observed from the experiments, restabilization occurs when high dosages of PAC coagulant are used. In this condition, the floc size decreases and floc settling slows down (10). A similar trend in the coagulation process with different coagulants was reported for the turbidity removal rate from raw water by other researchers (11, 24).

**Figure 3.** Effect of Different PAC Dosage on the Turbidity Removal Efficiencies

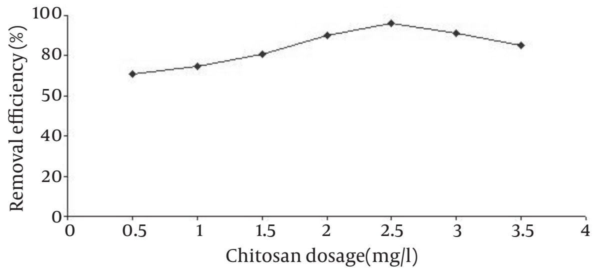



Chitosan = 2.5 mg/L, and Turbidity of 50 NTU.

## 3.3. Effect of Chitosan Dosage

A series of jar tests were conducted to study the coagulation effectiveness for removal of turbidity with chitosan in the range from 0.5 to 3.5 mg/L while other parameters such as turbidity, pH value and PAC dosage were constant. For the assistance of discussion, the optimum chitosan dosage is defined as the point at which the maximum tangent to the coagulation profile occurs. The correlation between the optimum dosage of chitosan and turbidity removal rate is illustrated in Figure 4, which clearly shows that the initial amount of chitosan has considerable effects on coagulating efficiency. From Figure 4, it can be seen that the highest turbidity removal rate was observed in initial chitosan amount of 2.5 mg/L, which residual turbidity reaches to 2 NTU. With an increase of chitosan at 3 mg/L, the turbidity removal rate decreased gradually until residual turbidity rate reaches to 7.5 NTU (85%) at 3.5 mg/L of chitosan. It can be explained based on this hypothesis that chitosan like other polymer additives may act either by bridging and charge neutralization while coagulation process using metal salts is interpreted as being a result of charge neutralization or bulk precipitation. In these mechanisms, chitosan destabilizes the colloidal matter by adsorption of particles with consequent formation of particle-polymer-particle bridges (11, 24). Hence, chitosan could be produced the flocs of better quality, namely larger flocs and faster settling velocity (6, 14). However, restabilization of colloidal particles occurs through the presence of chitosan at high dosage level. Similarly, Yang et al showed that different dosage of chitosan in the chitosan-PAC composite coagulant affects the removal efficiency of chemical oxygen demand and suspended solid from water (22). Also, another study showed that, compared to chitosan and PAC alone, chitosan combined with PAC is more efficient and economical for removing of turbidity in turbid water (13). Ahmad et al. reported that the initial amount of chitosan has a significant effect on the turbidity removal rate in which, by increasing the chitosan from 0.3 to 10 g/L, the turbidity of treated solution increase from 10 to 12 NTU (14).

**Figure 4.** Effect of Different Chitosan Dosages on the Turbidity Removal Efficiencies




PAC = 20 mg/L, pH = 7 and 50 NTU.

### 3.4. Effect of Different Initial Turbidity

We found that, in coagulation process the turbidity amount of turbid water is one of the most important parameters to determine not only the turbidity removal efficiency but also the coagulant dosages. For this reason, the effect of initial amount of turbidity i.e. 50, 100 and 200 NTU when PAC used as coagulant at optimum parameters was determined. Moreover, the effects of initial turbidity i.e. 50, 100 and 200 NTU in CPC was demonstrated where PAC and chitosan used as coagulant and coagulant aid, respectively, which the results presented in Figure 5. As seen in this figure, when PAC used as a coagulant, the turbidity removal efficiency was gradually decreased from 86% to 75% while the initial turbidity was raised from 50 to 200 NTU in the constant amount of PAC dosage and pH value. Also, the results shown in Figure 5 clearly indicated that initial amount of turbidity has affected the turbidity removal rate when chitosan used as coagulant aid combined with PAC; however, the variation of the turbidity removal efficiency was lower than coagulation using PAC alone. For example, in the coagulation process using CPC 96%, 95% and 93% of the turbidity have removed when the initial amount of turbidity were 50, 100 and 200 NTU, respectively.

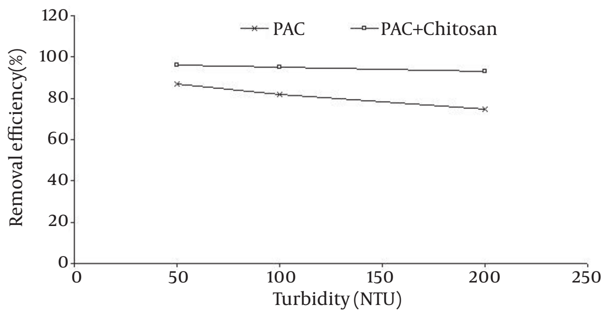



**Figure 5.** Effect of Initial Turbidity on the Turbidity Removal Efficiencies in Coagulation Using PAC and CPC in Optimum Condition

## 4. Conclusion

In the present study, a batch jar test experiments were done to assess the coagulation efficiency of PAC and CPC for removing of turbidity from turbid water. Addition of chitosan as a coagulant aid could significantly reduce the PAC dosage required for complete turbidity removal in which the residual turbidity dropped to 5 NTU as initial turbidity amount of 100 NTU without filtration, which meet the Iran drinking water standard (25). Flocculants dosage at the optimum pH found for PAC was 30 mg/L, while adding chitosan as a coagulant aid reduced the required dosage of PAC near 35%; thereby, reducing costs of treatment. The maximum coagulation of turbidity were achieved by CPC at the dosage of 20 mg/L for PAC and 2.5 mg/L for chitosan (96%) compared with PAC alone at the dosage of 30 mg/L at pH 7.5 (87%). Among the seven dosage of chitosan used for coagulation at 20 mg/L concentration of PAC, 2.5 mg/L chitosan was more effective for enhancing the coagulation efficiency. Also, the results indicated that the coagulation of turbidity using chitosan or/and PAC were very sensitive to pH and the optimum pH values were 8.5 and 7.5, respectively.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## References


1. Sawyer CN, McCarty PL, Parkin GF. *Chemistry for environmental engineering and science*. Boston: London: McGraw-Hill; 2003.
2. Lee PC, Gau SH, Song CC. Particle removal of high-turbidity reservoir water by electro-aggregation. *J Environ Eng Manag*. 2007;**17**(5):371.
3. Shi B, Wei Q, Wang D, Zhu Z, Tang H. Coagulation of humic acid: The performance of preformed and non-preformed Al species. *Colloid Surface A*. 2007;**296**(1–3):141–8.
4. Libecki B, Dziejowski J. Optimization of humic acids coagulation with aluminum and iron (III) salts. *Polish J Environ Stud*. 2008;**17**(3):397–403.
5. Seidmohammadi A, Faradmal J, Hosinzade E, Asgari G. Removal of turbidity and humic acid using chitosan as coagulant aid: modeling with artificial neural network. *Environ Eng Manag J*. (**In press**)
6. Seidmohammadi A, Asgari G, Samadi MT, Ahmadian M, Poormohammadi A. Removal of humic acid from synthetic water using chitosan as coagulant aid in electrocoagulation process for Al and Fe electrodes. *Research J Chem Environ*. 2014;**18**(5):19–26.
7. Seidmohammadi A, et al. [Electrocoagulation and electrocoagulation/flotation processes for removing high turbidity from surface water using Al and Fe electrodes]. *Water Wastewater J*. 2013:62–9.
8. Zhao S, Gao B, Wang Y, Yang Z. Influence of a new coagulant aid-Enteromorpha extract on coagulation performance and floc characteristics of aluminum sulfate coagulant in kaolin–humic acid solution treatment. *Colloid Surface A*. 2013;**417**(0):161–9.
9. McLachlan DRC. Aluminium and the risk for alzheimer's disease. *Environmetrics*. 1995;**6**(3):233–75.
10. Bolto B, Gregory J. Organic polyelectrolytes in water treatment. *Water Res*. 2007;**41**(11):2301–24.
11. Renault F, Sancey B, Badot PM, Crini G. Chitosan for coagulation/flocculation processes – An eco-friendly approach. *Eur Polym J*. 2009;**45**(5):1337–48.
12. Bina B, Mehdinejad MH, Nikaeen M, Movahedian Attar H. Effec-

tiveness of chitosan as natural coagulant aid in treating turbid waters. *Iranian J Environ Health Sci Eng*. 2009;**6**(4):247–52.
13. Huang C, Chen S, Ruhsing Pan J. Optimal condition for modification of chitosan: a biopolymer for coagulation of colloidal particles. *Water Res*. 2000;**34**(3):1057–62.
14. Ahmad AL, Sumathi S, Hameed BH. Coagulation of residue oil and suspended solid in palm oil mill effluent by chitosan, alum and PAC. *Chem Eng J*. 2006;**118**(1–2):99–105.
15. Crini G, Badot PM. Application of chitosan, a natural aminopolysaccharide, for dye removal from aqueous solutions by adsorption processes using batch studies: A review of recent literature. *Prog Polym Sci*. 2008;**33**(4):399–447.
16. Divakaran R, Sivasankara Pillai VN. Flocculation of kaolinite suspensions in water by chitosan. *Water Res*. 2001;**35**(16):3904–8.
17. Divakaran R, Sivasankara Pillai VN. Flocculation of river silt using chitosan. *Water Res*. 2002;**36**(9):2414–8.
18. Gregory J. Rates of flocculation of latex particles by cationic polymers. *J Colloid Interf Sci*. 1973;**42**(2):448–56.
19. Chi FH, Cheng WP. Use of Chitosan as Coagulant to Treat Wastewater from Milk Processing Plant. *J Polym Environ*. 2006;**14**(4):411–7.
20. Szygula A, Guibal E, Arino Palacin M, Ruiz M, Sastre AM. Removal of an anionic dye (Acid Blue 92) by coagulation-flocculation using chitosan. *J Environ Manage*. 2009;**90**(10):2979–86.
21. APHA. AWWA. WEF . *Standard methods for the examination of water and wastewater*. 21st ed. Washigton, D. C.: American Public Health Association; 2005.
22. Yang ZL, Gao BY, Yue QY, Wang Y. Effect of pH on the coagulation performance of Al-based coagulants and residual aluminum speciation during the treatment of humic acid-kaolin synthetic water. *J Hazard Mater*. 2010;**178**(1-3):596–603.
23. Zhang P, Wu Z, Zhang G, Zeng G, Zhang H, Li J, Song X, Dong J, et al. Coagulation characteristics of polyaluminum chlorides PAC-Al30 on humic acid removal from water. *Sep Purif Technol*. 2008;**63**(3):642–7.
24. Oladoja NA, Aliu YD. Snail shell as coagulant aid in the alum precipitation of malachite green from aqua system. *J Hazard Mater*. 2009;**164**(2-3):1496–502.
25. Iranian Institute of Standard and Industrial Researches . Water Quality Company. Iran; Teran: 1993. Available from: http://www.isiri.org.