



## Treatment of highly turbid water using chitosan and aluminum salts

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### ABSTRACT

The high turbidity in surface water may make it difficult for water treatment plants to supply drinking water. Chitosan, a natural linear cationic polymer, and aluminum chloride, a metal salt, and the mixture of the two coagulants were used to treat highly turbid raw water in this study according to the residual turbidity, sludge volume and residual aluminum concentration. The residual turbidity was less than 50 NTU but the sludge/water volume ratio was over 150 mL/L after aluminum salt coagulation (135 mg/L as Al), which could stop the sedimentation process. The amount of sludge produced after chitosan coagulation (5 mg/L) was only about 1/5 of that for aluminum coagulation for the similar turbidity removal. Chitosan coagulation, however, still has two problems that need to be solved. First, the residual turbidity of treated water is still too high for sand filtration. Second, the colloid particles may restabilize if chitosan is overdosed. Adding a comparative low dosage of aluminum salt (13.5 mg/L as Al) with chitosan can successfully solve both of the problems. The sludge volume ratio only increased slightly and the residual turbidity was less than 10 NTU. Moreover, the restabilization of colloids did not occur. The residual aluminum concentration, which could lead to Alzheimer's disease, can also be reduced significantly after addition of chitosan.

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### 1. Introduction

The turbidity of river water in Taiwan and other tropical areas occasionally rises to over 10,000 NTU during typhoons, hurricanes, or rainstorms. This makes it difficult for water purification plants to supply useable water. A lack of water supply may cause sanitary problems and interfere with manufacturing activity which could lead to huge economic damage. How to treat highly turbid water efficiently and economically, therefore, is an important issue for environmental engineers.

Coagulation with metal salts, such as aluminum or iron salts, can effectively increase the size of the particles in water, after which they can be removed by sedimentation. This process, however, may produce large amounts of sludge because of the formation of metal hydroxides if large amounts of metal salt are added to treat high turbid water. The water purification process sometimes has to stop in Taiwan due to too much sludge in the sedimentation tank during typhoons. Moreover, the residual aluminum concentration in the water may increase due to the increase of aluminum dose. It may raise the risk of Alzheimer's disease [1].

Synthetic polymers, such as poly acryl-amide and poly diallyl-dimethyl ammonium chloride, are alternative coagulants used to remove turbidity in water. They can reduce the turbidity of water but do not increase the volume of sludge because they do not generate metal hydroxides. However, health concerns related to the release of carcinogenic oligomer [2–5] restricts their use.

Chitosan, which is a natural linear cationic polymer generated by extensive deacetylation of chitin, is a bio-degradable coagulant and has been used to treat colloidal particles [6–14], COD [15–20], metal ions [16,21–24], humic acids [25], cryptosporidium cysts [26] and other bio-microspheres [27]. It has similar properties to synthetic polymers but has a relatively low toxicity for humans and aquatic species. Moreover, it contains amino and hydroxyl functional groups, which show significant adsorption capacity for various pollutants.

Divakaran and Sivasankara Pillai [8] have successfully used chitosan to reduce the turbidity of water. The turbidity removal efficiency was over 90% in their study but the turbidity of the raw water was lower than the target water in this study. Sekine and coworkers used chitosan to treat turbid river water [11]. Their results showed that the turbidity removal efficiency was good when the turbidity was under 1000 NTU, but the coagulation process failed if the turbidity was over 1000 NTU. The use of mono-coagulants, such as chitosan, therefore, may not be a good solution for highly turbid water purification. New coagulants or coagulation

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processes need to be investigated to solve these problems. This study tried to use chitosan with aluminum salt to form a mixture coagulant to treat highly turbid water (over 1000 NTU). The residual turbidity, sludge volume, and residual aluminum concentration were investigated after sedimentation to access the applicability of the coagulant mixture. Finally, a coagulation test using the raw water of Zhi-tan water purification plant during typhoon Megi was performed to examine the effectiveness of the combined procedure.

## 2. Materials and methods

### 2.1. Preparation of coagulants

Chitosan powder purchased from Wako Pure Chemicals (Tokyo, Japan) was dissolved in 1.0% chloride acid solution by stirring overnight at 150 rpm to make 1.0% (w/w) stock solution. Aluminum salt stock solution was prepared by dissolving 133.5 g of aluminum chloride ( $\text{AlCl}_3$ ) in 1 L deionized water ( $[\text{Al}^{3+}] = 1.0 \text{ M}$ ). Prior to the coagulant mixture experiments, both of the two stock solutions were added to the deionized water and carefully mixed with magnetic stirring to prepare a coagulant mixture solution of desired concentration.

### 2.2. Preparation of model water

Model waters with the desired turbidity were prepared by mixing given amounts of clay or bentonite (Hayashi Co., Japan) with deionized water.  $\text{NaHCO}_3$  was added to produce the alkalinity at 50 mg/L as  $\text{CaCO}_3$ .

### 2.3. Coagulation test

A jar test apparatus was used for the coagulation and flocculation experiments. For all experiments, the pH of the suspension was adjusted by adding a 0.1 M NaOH or 0.1 M HCl ( $\text{pH} = 7.5 \pm 0.1$ ). After the coagulants were added, the water was rapidly mixed (at 100 rpm) for 3 min followed by 20 min of slow mixing (at 30 rpm). After standing for 30 min, the supernatant from the top 5 cm of the suspension was withdrawn for turbidity and residual aluminum measurement.

### 2.4. Analytical measurements

The turbidity of each sample was measured by a turbidity meter (HACH Ratio, USA). The sludge volume was measured using a quiescent column. Residual aluminum ions were analyzed by a TJA IRIS Advantage/1000 Radial ICP spectrometer calibrated against a CLARITAS certified reference solution (SPEX Certi Prep, Inc). A particle size analyzer (Model Zetasizer 3000, Malvern Instrument Ltd., Worcester, UK) was used to determine the particle size distribution before and after coagulation.

## 3. Results and discussion

### 3.1. Jar test with different coagulants

Figs. 1 and 2 show the variations of residual turbidity and the water-sludge volume ratio after aluminum and chitosan coagulation, respectively. The residual turbidity decreased with increasing coagulant dosage for aluminum coagulation but had an optimum dosage (5 mg/L) for chitosan coagulation. This is due to the different mechanisms of the two coagulants. The mechanisms for aluminum coagulation are compression of the electrical double layer, charge neutralization and flocs sweeping. The main mechanism

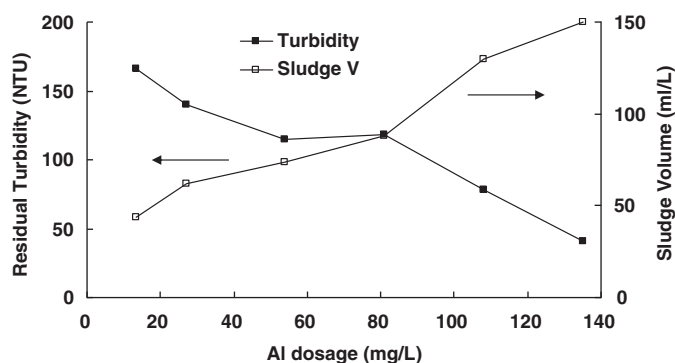


Fig. 1. Variation of residual turbidity and sludge volume per liter of water after coagulation with aluminum chloride. (Initial turbidity = 10,000 NTU, settling time = 30 min.)

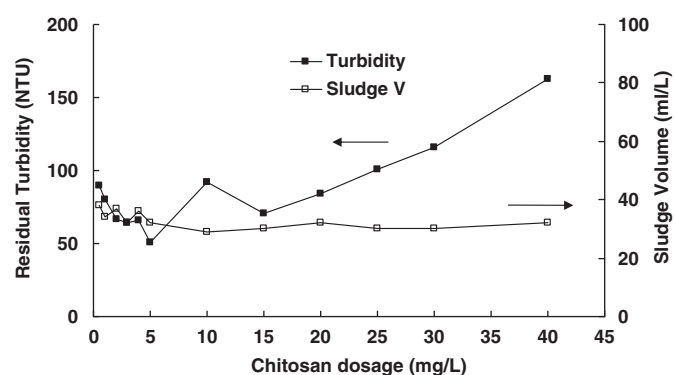
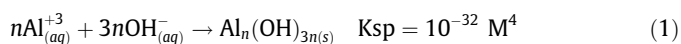


Fig. 2. Variation of residual turbidity and sludge volume per liter of water after coagulation with chitosan. (Initial turbidity = 10,000 NTU, settling time = 30 min.)

in this study should be flocs sweeping because the aluminum hydroxide must appear at high aluminum dosage. The addition of large amounts of aluminum salt can produce aluminum hydroxide flocs (as shown in reaction (1)) which settle by gravity, within a reasonable time.



These coagulant flocs then collide with and drag colloids down with them. The more flocs produced, the better the turbidity removal. The removal of turbidity, therefore, increased with increasing aluminum dosage. The addition of chitosan; however, did not produce hydroxide flocs. The functional groups on the chitosan combine with the active sites of particles and the interaction of a single molecule with many particles produces a bridging effect. This combines them into larger particles which settle under the action of gravity.

When a larger amount of chitosan was added; however, the effect of bridging coagulation became relatively weak and the colloids restabilized, possibly due to steric hindrance and repulsion between charged polymers [28,29]. Similar phenomenon also occurred in previous studies which use chitosan to treat river water [6,8,14]. The optimum dose of chitosan (1.0–2.0 mg/L) in their works were less than that (5.0 mg/L) in this study. This phenomenon may be due to the higher turbidity. Chatterjee et al. [30] observe the same optimum dose of chitosan and the initial turbidity in their work was also extremely high ( $\text{SS} = 5.0 \text{ g/L}$ ).

The amount of sludge produced by aluminum coagulation was proportional to the amount of aluminum salt added. Although aluminum hydroxide floc can sweep the colloids in the water, the floc itself is a kind of sludge. The sludge/water volume ratio may rise to 15% if the dosage used for the same turbidity removal as chitosan is

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