

# **Connotation of Natural Coagulants on Total Organic Carbon content of water**

VaraSaritha<sup>1</sup>, N.Srinivas<sup>2</sup> and BhavyaKavitha Dwarapureddi<sup>2</sup>

Assistant Professor, Department of Environmental Studies, GITAM Institute of Science, GITAM University,  
Visakhapatnam, Andhra Pradesh, India<sup>1</sup>

Department of Environmental Studies, GITAM Institute of Science, GITAM University, Visakhapatnam, Andhra  
Pradesh, India<sup>2</sup>

Research Scholar, Department of Environmental Studies, GITAM Institute of Science, GITAM University,  
Visakhapatnam, Andhra Pradesh, India<sup>2</sup>

**ABSTRACT:** Coagulation process in drinking water Treatment is of such importance that a great deal of studies were carried out on Optimization of working parameters for coagulation and removal of turbidity from raw water source. The aim of the present study is to test the efficiency of natural coagulants, sago and chitin for removing turbidity and decreasing organic materials from water. It was an empirical and analytic study using Jar Test apparatus. To perform qualitative and quantitative analyses, samples with 70 and 150 NTU turbidities, respectively, were tested with the coagulants. The study advocated that natural coagulants were highly efficient in removing turbidity up to 99% for higher turbid samples. In terms of removal of Total Organic Carbon (TOC), Chitin was equally proficient as alum compared to sago.

**KEYWORDS:** turbidity, natural organic matter, coagulation, total organic carbon.

## **I. INTRODUCTION**

Particles are ubiquitous in all natural waters. Their origins, compositions and concentrations vary widely. They can be contaminants or associated with contaminants and therefore need to be removed. One of the most basic processes in the treatment of raw source waters to meet drinking water standards is the solid/liquid separation process to remove particulate material. Particulate material originating in raw water or contributed by addition of treatment chemicals is physically separated from source water during drinking water treatment by clarification and filtration processes. These processes target not only removal of particulate material itself but also contaminants that are associated with the particulate material. Clays, sands, colloids, and so on all may comprise typical particulates to be removed; however, removal of other particle classes, such as microorganisms and particulate forms of natural organic matter (NOM), is beneficial for efficient treatment (Kwok-Keung et al., 2011).

In view of growing awareness of pollution problems, dispersal of organic contamination in the environment is becoming a matter of concern. Ever increasing use of chemical and related compounds in each and every field of industry and agriculture summons an urgent need of method for their effective removal from water and wastewater (Patel and Vashi, 2013). Removal of organic constituents from natural waters has always been of concern to water treatment. Conventionally water treatment goals have included removal of color, taste and odor producing compounds, the standards for which are based on esthetic considerations.

Coagulation takes place by the neutralization of charges on the electronegative colloids particles by this ionized centre or by hydrogen among the charged surfaces. Electro static cross linkage also binds the linear chain together (Patel and Vashi, 2013). These colloids may include organic particulate and inorganic particulate. The role of NOM, particularly

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the pool of DOC, in exerting a coagulant demand and the need to remove this material from water has become more important. The coagulants are used not only destabilize colloidal particles, but also to remove NOM. The removal of NOM from water can occur by direct precipitation of the NOM or by adsorption of NOM onto precipitate metal hydroxide. Particles in water may be mineral (clay, aluminum and iron oxides and hydroxides, asbestos, silica) or organic (viruses, bacteria, protozoa, cysts, algae). Existing primarily as colloidal suspensions, these suspensions are stable such that the particles have a slow rate of particle aggregation or flocculation. The cause of particle stability may be i) electrostatic repulsive interactions due to diffuse electrical double layers ii) hydrophilic effects due to bound water at particle surfaces and iii) steric effects due to adsorbed macro molecules (Edzwald, 1993).

The effectiveness of a coagulant may depend on several factors including coagulant type and dosage, mixing conditions, pH, alkalinity, temperature as well as the presence of divalent cations and concentrations of destabilizing anions (bicarbonate, chloride, sulphate etc.) (Duan and Gregory, 2003; Matilainen et al., 2010; Verma et al., 2012). In the present study we have focused on the effect of type and dosage of coagulant and pH.

Natural coagulants have been reported to have several other advantages compared to synthetic coagulants such as alum and ferric chloride, in that, they produce much lower sludge volume and are safe to humans. Ghebremichael(2004) investigated that the sludge produced from *Moringa oleifera* coagulated turbid water is only 20–30% that of alum. Litherland, Katayon et al., and Sanghi et al., showed that the residue of alum in water may be carcinogenic. Natural coagulants are biodegradable and cost effective for developing countries since they can be locally grown and have a wider effective dosage range for flocculation of various colloidal suspensions (Sanghi et al., 2006; Madhukar and Yogesh, 2013)

The mechanisms associated with different natural coagulants are varied as well (Babu and Chaudhuri, 2005). It is imperative for relevant stakeholders to fully comprehend the technicalities involved when considering the coagulants for rural, domestic or industrial water treatment. Focusing on these issues, the current investigation is directed to assess the potential of natural coagulants in removing turbidity and along with the trends of Total Organic Carbon contents before and after coagulation so that environmental specialists can tailor its usage for a myriad of water contaminants.

## II. MATERIALS AND METHODS

### Preparation of Synthetic Turbid water samples:

The natural turbidity of raw water varies from 10 to 500 NTU for maximum period. So the experiments were carried out for water samples of 70 and 150 NTU turbidity values. Turbid water samples were prepared by using Bentonite clay. 5 gm of Bentonite clay was mixed to 500ml distilled water. Mixed clay sample was allowed for soaking for 24 hrs. Suspension was then stirred so as to achieve uniform and homogeneous sample. Supernatant was withdrawn. Turbidity was determined and portions of suspension were diluted to desired turbidity values. Resulting suspension was found to be colloidal and used as stock solution for preparation of turbid water samples. Everyday stock sample of Bentonite clay was diluted by using distilled water to get desired turbidity (Gidde et al., 2008).

### Coagulants used in the study

The natural coagulants used in the study are chitin and sago.

Chitin is a natural polymer. Chitin is the second most abundant polysaccharide in nature (after cellulose). At least 10 giga tons of chitin are synthesised and degraded each year in the biosphere. Chitin is present in nature usually complexed with other polysaccharides and with proteins. It is a renewable resource and is isolated from crab and shrimp waste. It is used for waste water clearing among many other applications. Employed as a chelation agent, chitin and its derivatives are used for treating drinking water by separating organic compounds and heavy metals, and for treating sewage by precipitating certain anionic wastes and capturing pollutants such as DDT and PCBs (polychlorobenzene). The Environmental Protection Agency (EPA) has already approved the use of chitosan in water at concentrations of up to 10 mg per litre (greatvistachemicals).

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Indian sago starch extracted from Tapioca roots finds its application not only as a food but also numerous commercial applications. The sago starch is a cheap, easily available, biodegradable and a versatile polymer. Indian sago starch is extracted from *Manihotesculenta* belonging to family Euphorbiaceae. This sago is native to Brazil, Amazon, Colombia, Venezuela, West Indies, Cuba, and Puerto Rico. In India it was introduced in later part of 19<sup>th</sup> century. Kerala, Andhra Pradesh and Tamil Nadu are the major producers of sago starch. Tapioca root is the basic raw material for sago and starch. These roots contain about 5-13% starch content, 60-70% moisture content, 7-12% protein (32-35% total carbohydrates) and fat present in trace amount. Indian Tapioca roots are known to have 30-35% starch with appreciable amount of calcium and vitamin-C (Sabuindia, 2013; RenuYadav and GarimaGarg, 2013).

## Experimental procedure:

A conventional jar test apparatus was employed for the process of coagulation and flocculation. All tests were carried out with 1 L samples in 2-L beakers. Beakers were filled with 1000 ml of the synthetic water, and placed on each slot in a jar tester. Coagulant was added into each beaker at various doses and agitated at two different mixing speed and time: 100 rpm for 1 min and then reduced to 20 rpm for 30 min. The coagulant dose was added at four concentrations i.e 0.1, 0.2, 0.3 and 0.4 mg/l. The coagulation was performed at three different pH conditions by adding 0.1 M H<sub>2</sub>SO<sub>4</sub> and 0.1 M NaOH in all coagulation tests. After sedimentation for 20 min, an aliquot of 10 ml was sampled from the mid depth of the beaker and residual turbidity was determined. Turbidity measurements were conducted using nephelometric turbidimeter (ELICO, CL52D). The pH values of samples were measured using pH meter (ELICO, L1 126).

## III. RESULTS

In the present study the efficiency and optimization of the coagulants and the total organic carbon in the treated water was studied. Synthetic water was treated with different concentrations of the natural coagulants whose efficiency was compared with that of alum.

The initialization of the experiments was carried out with doses of 0.5, 1.0, 1.5 and 2.0, of which 0.5 resulted in good turbidity removal with formation of less concentration of suspended solids in the settled sludge (4.61, 7.01 and 9.37mg/500ml by alum, sago and chitin respectively) and higher turbidity removal. Taking this into consideration we progressed to test the efficiency of the coagulants with reduced concentrations. Hence in the next level of experiments the coagulant dose was taken as 0.25, 0.5, 0.75 and 1.0. A specific tendency of increased concentration of suspended solids in the settled sludge was observed with increasing concentration of the coagulant by all the three coagulants whereas the reduction in turbidity was contrary, i.e it increased with decreasing dosage (4.81, 6.08 and 6.48mg/500ml with alum, sago and chitin respectively).

Since the prime objective of the study was to determine the lowest concentration of coagulants that can result in efficient reduction of turbidity, in the next stage we tested the doses of coagulants with 0.05, 0.1, 0.15 and 0.2 (with only 0.05 difference between concentrations). These concentrations have resulted in interesting results with increased suspended solids in the settled sludge as the concentration increased from 0.05 to 0.2 by alum and chitin, whereas sago has shown a converse trend of decreasing suspended solids in the settled sludge with increasing concentration (5.93, 6.01 and 5.28mg/500ml by alum, sago and chitin respectively) Fig - 1.

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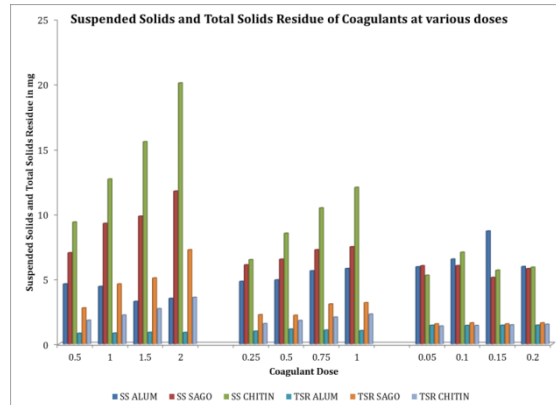


Fig – 1: Comparison of Concentration of suspended solids in the settled sludge and Total Solids Residue from Test Coagulants at various doses

Similar results were observed with the Total solids residue. Remarkable result observed in the study is that least sludge was obtained by alum at higher dosages of coagulant that is 0.5, 1.0, 1.5 and 2.0 (4.61, 4.42, 3.27 and 3.5mg/500ml respectively) with least suspended solids in the settled sludge at 1.5 dosage (3.27mg/500ml). Whereas the natural coagulants have shown decreasing concentration of suspended solids in the settled sludge with decreasing dosage of coagulant.

Turbidity reduction by the coagulants was in the order of chitin>alum>sago. Chitin showed reduction in turbidity both at higher and lower doses of the coagulant. Infact the highest reduction in turbidity was seen at dose of 1.5mg. All the coagulants were efficient at the doses of 1.5 and 2.0mg doses of coagulant. On a general note the reduction in turbidity was enhanced with reduction in coagulant dose. Even though chitin and other coagulants were efficient at higher doses the noteworthy point is that they were equally efficient at lower doses. Fig - 2

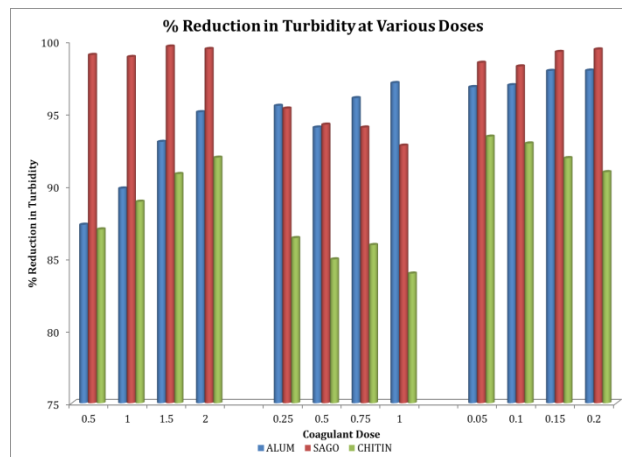


Fig – 2: % Reduction in Turbidity by Coagulants at various doses

These verdicts have stimulated us to take up the lower doses to be optimized doses for further studies. The next stair of study included testing the optimized doses at different turbidities that are generally present in surface waters, in this study the lower and higher turbidities are taken as 70 and 150 NTU with pH conditions ranging from 6, 7 and 8, which is the characteristic of surface water and also is the governing factor for efficiency of coagulants.

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Beyond their advantages over Alum, natural coagulants have also limitation. For instance they increase organic load in the water which tend restablization to occur. With this point of view, the present study has focused on the total organic content of the waters treated with natural coagulants in comparison with alum.

Turbidity reduction with all the test coagulants was observed to be good with highest reduction achieved to be 99.93 by alum at pH 6, coagulant dose 0.05 and initial turbidity 150NTU and lowest with alum pH 6, coagulant dose 0.15 and 70NTU initial turbidity. Alum has shown greater efficiency with all doses at pH 7 and least efficiency at pH 6. A noteworthy result obtained is that alum has shown a large variation in turbidity reduction with various doses, while natural coagulants were much stable at varied conditions applied like coagulant dose and particularly at varied pH. While all the coagulants were efficient in turbidity removal with higher initial turbidity i.e 150NTU their efficiency reduced with reduced initial turbidity of 70NTU (Fig - 3).

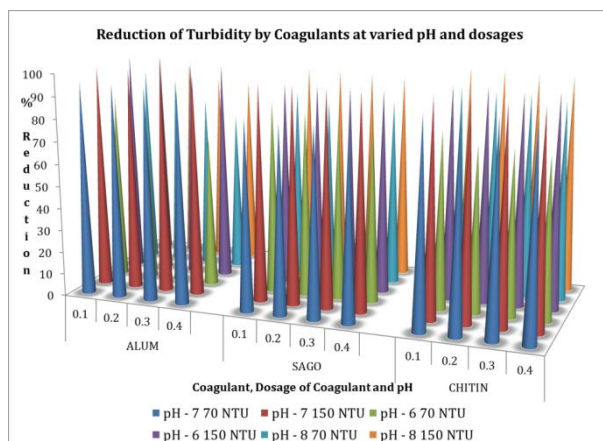


Fig – 3: % Reduction in Turbidity by Coagulants at varied parameters of pH, coagulant dose and initial turbidity

The individual turbidity reductions of the coagulants at varied pH, initial turbidity, coagulant doses and their respective total organic carbon (% reduced) are as follows (Fig - 4):

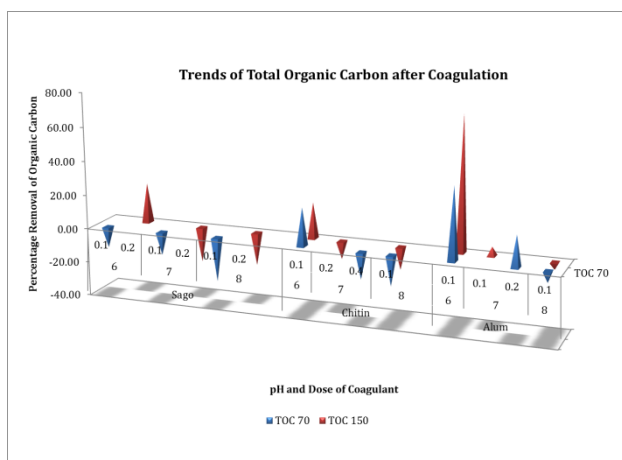


Fig – 4 Trends of Total Organic Carbon Reduction by Coagulants (%)

### Alum:

At pH 6 the greater reduction of turbidity with initial turbidities of 70 and 150NTU are 83.36% and 99.93% respectively with 0.05mg coagulant dose, their respective Total Organic Carbon reduction is 42.11% and 77.93%.

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96.14% and 99.84% turbidity reduction was observed at pH 7 with coagulant dose of 0.1 and 0.05 with initial turbidities of 70 and 150NTU respectively and the successive TOC reductions are 18.22% and 5.2%. 0.05mg of coagulant dose resulted in 90.85% and 94.78% turbidity reduction at pH 8 with 70 and 150NTU initial turbidity, the TOC reduction was observed to be -6.6307 and -4.29%.

## **Chitin:**

At pH 6 the greater reduction of turbidity with initial turbidities of 70 and 150NTU are 76.97% and 96.18% respectively with 0.05mg coagulant dose, their respective Total Organic Carbon reduction is 22.24% and 20.96%. 93.93% and 96.97% turbidity reduction was observed at pH 7 with coagulant dose of 0.2 and 0.05 with initial turbidities of 70 and 150NTU respectively and the successive TOC reductions are -15.7% and -10.45%. 0.05mg of coagulant dose resulted in 92.62% and 98.5% turbidity reduction at pH 8 with 70 and 150NTU initial turbidity, the TOC reduction was observed to be -17.82% and -13.2%.

## **Sago:**

At pH 6 the greater reduction of turbidity with initial turbidities of 70 and 150NTU are 85.4% and 97.5% respectively with 0.05mg and 0.1mg coagulant dose, their respective Total Organic Carbon reduction is -12.3% and 23.30%. 86.39% and 92.3% turbidity reduction was observed at pH 7 with coagulant dose of 0.1 and 0.05 with initial turbidities of 70 and 150NTU respectively and the successive TOC reductions are -13.5% and -20.5%. 0.05mg of coagulant dose resulted in 85.59% and 94.1% turbidity reduction at pH 8 with 70 and 150NTU initial turbidity, the TOC reduction was observed to be -25.82 and -19.28%.

## IV. DISCUSSION

### **Effect of Coagulant Dose**

Coagulation dosage is one of the most important factors that have been considered to determine the optimum condition for the performance of coagulants in coagulation and flocculation. Essentially, insufficient dosage or overdosing would result in the poor performance in flocculation. Therefore, it is significant to determine the optimum dosage in order to minimize the dosing cost and sludge formation and also to obtain the optimum performance in treatment (Patel and Vashi, 2013).

The pattern of turbidity removal is characterized by a gradual increase with decreasing concentration of coagulant, approaching a maximum of about 99% and an average of 90%, with removal remaining in-complete at high coagulant dosage.

Studies by MohdAriffin, have shown that the percentage of reduction for COD and turbidity has decreased when the chitosan dosage concentration was in the range of 30 mg/l to 66 mg/l. This poor performance was due to the phenomenon of excess polymer is adsorbed on the colloidal surfaces and producing restabilized colloids. Thus, there were no sites available on the particle surfaces for the formation of interparticle bridges. The restabilized colloidal particles can become positively charged and cause the electrostatic repulsion among the suspended solids.

Polyelectrolytes used in the study are advantageous over chemical coagulants because they are safer to handle, require less doses where they are quite efficient and are easily biodegraded.

### **Effect of pH**

Trials with different coagulant concentrations were carried out, varying pH conditions between 6 and 8. With the change of the pH, the removal of turbidity changed little for natural coagulants but varied greatly or efficiency decreased with alum.

Mohd Omar Fatehahet.al., 2013 from their studies stated that the natural coagulants employed in the study have similar coagulation characteristics with the commercial coagulants. However, the natural coagulants possess better metal adsorption capability than the commercial coagulants. A 3 level factorial experimental design was used in the



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Response Surface Methodology (RSM) analysis and indicated that starches are capable to remove TS from the semiconductor wastewater and the removal performance were almost similar to alum and higher than PAC.

Their studies have also shown that the surface structure of tapioca flour is spherical and truncated in shape, contrast spots were identified, which proved the presence of elements in the tapioca starch coagulant. The EDX analysis shows the presence of sodium and aluminium on the surface of the coagulated sludge. However, the contrast spots on the surface of the sludge are most probably silicon particles. The highest TS removal were estimated by potato flour (90%) followed by tapioca flour (89%), alum (78%), corn flour (77%), rice flour (73%), PAC (53%) and glutinous rice flour (17%).

Domard et al., 1989 pointed out that there are 90 % of the functional group of  $\text{NH}_2$  on chitosan surface has been protonated at pH 4, and gradually reduced to about 50% as pH increased to 6. Therefore, the positive charges on the chitosan surface will significantly decrease as solution pH increased, so the contribution by the charge neutralization of the chitosan to destabilize the particles becomes less important as pH increased. The properties of chitosan, including its cationic behavior and molecular weight, may be used both for charge neutralization (coagulating effect for anionic compounds) and for particle entrapment (flocculating effect). Moreover, based on observation, the floc produced by chitosan appears rapidly at pH 4 and form a large size, which can be easily settled (MohdAriffin).

## Trends of Total Organic Matter

Sufficient coagulant must be added to satisfy the charge demand of raw water for the effective treatment of NOM. NOM removal will be less at higher pH with all coagulants. The required coagulant doses for NOM removal will also probably increase as the water temperature decreases (Indiana Garcia, 2005).

TOC does not provide an indication of the aromaticity, aliphatic nature, functional group chemistry, or chemical bonding associated with natural organic molecules. The reactivity of chemical bonds and functional groups is probably a significant factor in explaining why different waters with the same TOC concentration can give different DBP concentrations under identical disinfection conditions and bromide levels (Indiana Garcia, 2005).

A comparison of alum and iron salt used as coagulant for the removal of natural organic matter showed that alums remove more NOM than iron salts due to the pH used at the coagulation process (Garcia and Moreno, 2005). Bina et al., 2010 applied MOCPC as coagulant aid in water treatment processes and showed that TOC concentrations at all of experiments were between 0.5 to 0.65 mg/L and were less than the background TOC levels. The results showed that MOCPC could be used as natural coagulant aid for drinking water treatment with the lowest risks of organic release.

The coagulation process with alum as the sole coagulant is capable of achieving significant organic removal. The removals were found to be strongly pH and coagulant dose dependent. The pH of the water during coagulation has profound influences on effectiveness of coagulation for organic removal. Organic removal is much better in slightly acidic condition. The optimum pH for alum coagulation is influenced by the concentration of organic matter in the water. For water of higher organic content the optimum pH is displaced to slightly more acidic values (AWWA Committee Report, 1979). An adequate amount of alkalinity in raw water is required during the flocculation process for two reasons i) the need for a buffer capacity in order to avoid a steep drop of pH due to alum addition and ii) a sufficient supply of bicarbonate to form metal hydroxide (Kawamura, 1996; Prakash Raj, 1997).

Best NOM removal by alum and natural coagulants in the study were at pH 6 and coagulant dose of 0.1mg/l. Thus conventional coagulation practices may provide excellent organic removal if the coagulant dose and pH condition are adjusted into the optimum range. The influence of pH was very great. (Semmens and Field, 1980).

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## IV. CONCLUSION

World population increasing year to year and reaching 7 billion in 2012 whereas an access of getting pure water remains a problem especially for people who live in developing countries. The severity is much observed in rural dwellers who is their source of drinking water is surface (raw) water which is not purified and this results for transmission of waterborne diseases.

To reduce such problems and the strong push to meet the drinking water needs of the developing world have led to the recent growing interest in using plant based natural coagulants and disinfectants and plant species showed promising result in coagulating and disinfecting raw water.

Efficiency of Natural Coagulants in the present study can be concluded as follows:

- Concentration of Suspended Solids in settled sludge obtained was less natural coagulants with decreasing doses of coagulants, which will require less coagulant and yet efficient in removal of turbidity.
- Total Solids Residue increased with reduced coagulant dose by natural coagulants resulting in solids that can be easily handled.
- pH and Coagulant dose has profound effect on removal of Natural Organic Matter which was surrogated through TOC analysis.
- TOC removal was obtained maximum 50% for adsorption with AC, clay and modified clays.
- Best NOM removal by alum and natural coagulants in the study were at pH 6 and coagulant dose of 0.1mg/l.

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