Evaluation of Mix-Chemical Coagulants in Water Purification Process

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Abstract*— Coagulation and flocculation followed by sedimentation are the customarily used unit operations in conventional water treatment process. Usually the process of coagulation is carried out using different metal salts such as aluminum and iron oxides. Therefore, this study was aimed at evaluating the performance of mix-chemical coagulants in water purification process. An experimental comparative study was done by evaluating controlled factors under various experimental setups. Jar tests were conducted to assess the efficiency of alum and ferric chloride coagulants in dual (1:1 and 3:1 alum to ferric chloride) combinations as well as separately. ANOVA tests were performed to select the best performing coagulant using Minitab version 16. The highest percentage TDS removal performance of 55.8%, 72.6%, 81.4% and 81.4% were exhibited for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination. And the highest percentage COD removal performance of 71%, 58.1%, 63.6%, and 50.9% was demonstrated for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination, respectively. The highest percentage turbidity removal performance shown by alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination were 98.7%, 99.1%, 98.7% and 97.8%, respectively. The 1:1 alum-ferric chloride coagulant combination shows highest (80.8%) concurrent TDS, COD, and turbidity average removal. The result of this study indicated that 1:1 alum-ferric chloride was found the most suitable coagulant to perform the coagulation process for the removal of COD, TDS and turbidity simultaneously. The use of optimized alum-ferric chloride combinations as a coagulant is preferable to single coagulant use if appropriately managed.

Keywords*— *Optimization; Coagulant; Alum-ferric chloride mix; TDS; COD; Turbidity

I. INTRODUCTION

Suspended particles can be simply removed by usual physical treatment. Unlike suspended particles, dissolved molecules cannot be removed by conventional physical treatment. Thus, the removal of colloids is the most complicated feature in conventional water treatment [1].

Turbidity can cause infection or gastrointestinal irritation which may pose a hazard to human health. Apart from the health point of view, the potability of water less than a certain degree of clarity is considered to be suspicious by the general community. Moreover, turbid water supplies are intolerable to many industrial consumers: for instance, process water used in food and paper industries is required to have a high degree of clarity to assure adequate final product quality [2].

Dissolved materials cannot be seen and don't contribute to the clarity of the water but they can result in colour problem in the water. Despite that the major source of drinking water is surface water, some surface water can contain such a high concentration of dissolved humic and fulvic acids, that they resemble the colour of black tea. Hence such water would be unacceptable as a purified drinking water [3].

The most common way to remove particulate matter from surface waters is by coagulation followed by sedimentation and/or filtration [4]. In this process, coagulants are added which will at first cause the colloidal particles to become destabilized and bunch together. When pieces of floc bunch together, they may form larger, heavier flocs which settle out and are removed as sludge [5]. The most commonly used commercial coagulants in water purification are aluminum and iron salts [6]. In water treatment using coagulants, considering potential additional costs and trade-off related to each particular coagulant is obligatory. When we compare alum versus ferric chloride, alum was shown to have the higher cost relative to the ferric chloride [7]. The complex forms of aluminum coagulant usually cost twice as much as alum because they are derived from these salts [8]. The cost of coagulants accounts for about 5% of the price of the drinking water produced [9].

Many findings have been reported on various contaminant removal using alum and ferric chloride for a long time. However, in this study the mix-coagulants, namely 1:1 and 3:1 alum-ferric chloride combinations in comparison to single alum and ferric chloride usage were investigated with the aim of determining their capabilities to reduce turbidity, total dissolved solid and chemical oxygen demand of drinking water. The interest of this study lies in the theory that the mix-chemical coagulant removal capacity will be different from the single coagulant. The result of this study can be used as bench mark in investigating a middle ground between aluminum and iron coagulant pitfalls by evaluating different alternatives of coagulant usage in drinking water treatment purification in order to intensify contaminant removal efficiency.

II. MATERIAL AND METHODS

A. Study design

An experimental comparative study where controlled factors were evaluated under various experimental set up. Two test sets were designed where one set was employed as a control group and the other one was experimental group.

B. Sample preparation

For reagent preparation, deionized water was used throughout the study for the sake of quality assurance. The samples were defined as follows: 30, 150, 300 and 500NTU. These levels were obtained by adding clay passed through the sieve no.200 to a certain volume of deionized water in order to introduce suspended solids and organic matter [10].

C. Coagulant stock preparation

Aluminum sulfate, $Al_2(SO_4)_3.18H_2O$ and ferric chloride, $FeCl_3$, was used as a coagulant. Stock solutions were prepared by dissolving 10.0 grams of alum and/ or ferric chloride in to 1,000 mL deionized water, in which 1 mL applied on a sample of 1000 mL represents a concentration of 10 mg /L when added to 1,000 mL of water to be tested [11]. The 1:1 and 3:1 alum-ferric chloride coagulant combination were formed by mixing the standard stock solution of alum and ferric chloride to make the required coagulant dosage in mix-coagulation experimental study.

D. Experimental procedures

Each jar was filled with 1000 mL of sample measured with a graduated cylinder. The coagulant dose destined for each jar was carefully measured into 1000 mL beakers. The stirrer speed was set on 200 rpm for 1 minute. After 1 minute, the mixing speed was reduced and was set on 20 rpm for 15 minutes. After this time period, the stirrer was turned off and flock allowed settling for 30 minutes [12]. Samples were then withdrawn 20 mm below the water level for turbidity, TDS and COD removal determination [13].

Optimization of pH and coagulant dose

A known volume of prepared alum or ferric chloride solution was added to jars containing 1000 mL of raw water at different pH values adjusted with 0.5N H_2SO_4 and 1N NaOH. To optimize the pH of the coagulation process, Jar tests were conducted over the pH range of 4 - 9 and constant coagulant dosages of 15 mg/L. Similarly, to investigate the optimum coagulant dose, the pH value of the raw water was maintained at an optimum pH as determined above and the coagulant dosages ranged from 5 to 45 mg/L [14].

Optimal initial turbidity determination for optimal pH and coagulant dosage

A known volume of prepared alum or ferric chloride solution was added to the jars with 1000 mL of raw water. To investigate suitable initial turbidity of the optimized pH and coagulant dosage, the pH value of the raw water and coagulant dosage was maintained at an optimum as determined above where as the raw water initial turbidity is ranged from 30 – 500 NTU.

Determination of simultaneous removal capacity of test coagulants

The real samples were taken on three different days using Jerry can to get different water sample characteristics. The physicochemical analyses of real sample were performed. Then, the simultaneous removal capacity of test coagulants was evaluated for turbidity, TDS and COD at the optimal conditions.

E. Data quality assurance

Extensive quality control measures were implemented throughout this study. Quality control measures for laboratory data collection were performed according to the Standard Methods for the Examination of Water and Wastewater [15]. Triplicate measurements were taken to assess the consistency of the precision of the analytical instrumentation. Duplicates were done for each Jar test run as well as for each analysis.

F. Sample and data analysis

Throughout this study COD test was performed by HT COD-Test LCK 214 method, turbidity was measured by Nephelometric Method using Turbidity meter HACH Wag-WT3029, TDS was determined by Gravimetric Method (SM: 2540) and pH of samples was measured by using a portable pH meter WTW 3310.

The removal efficiency was calculated by the following formula:

$$\text{Removal percentage} = \frac{C_0 - C}{C_0} \times 100$$

Where, C_0 and C = Turbidity, TDS and COD contents of wastewater (mg/L) before and after coagulation treatment, respectively. Data was compiled and analyzed using Microsoft Excel version 10 and Minitab 16 and presented using graphs.

III. RESULTS AND DISCUSSION

A. Turbidity removal efficiency at different pH and coagulant type Abbreviations and Acronyms

Alum turbidity removal efficiency in the pH range of 5-8 was greater than 95%, while at pH 4 and 9 it was below 90%. The maximum turbidity removal performance of ferric chloride was at pH 5 (96.4%) and pH 8 (98.1%). The 1:1 alum-ferric chloride combination has highest (97.5%) turbidity removal efficiency at pH 8 followed by 95.1% removal efficiency which was observed at pH 6. While, 3:1 alum-ferric chloride coagulant combination highest turbidity removal efficiencies were 97.37% at pH 7 and 96.7% at pH 8. The optimum pH for alum and 3:1 alum-ferric chloride combination was 7 at concentration of 25 mg/L. And for ferric chloride and 1:1 alum-ferric chloride combination it was 8 at concentration of 15 mg/L. Fig. 1 gives percentage turbidity removal efficiency at constant dose of 15 mg/L and initial turbidity of 150 NTU for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations as a function of solution pH, respectively.

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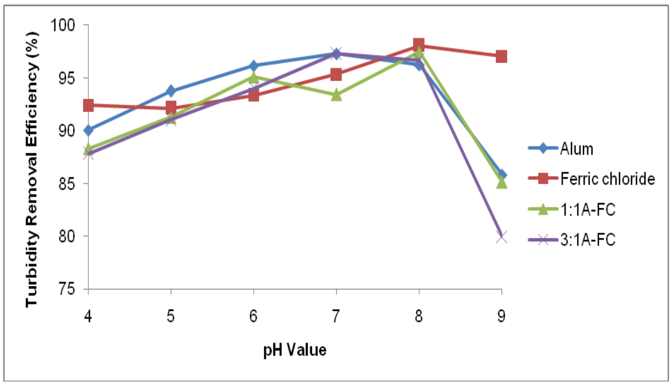

Fig. 1. Percentage turbidity removal efficiency at constant dose and initial turbidity as a function of solution pH

B. Turbidity removal as a function of coagulant dose and type

The maximum turbidity removal efficiency was 96.7% at dosage of 25 mg/L, 98.2% at dosage of 15 mg/L, 97.5% at dosage of 15 mg/L and 96.7% at dosage of 25 mg/L for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination, respectively. Ferric chloride turbidity removal efficiency was greater than 94% in 5-45 mg/L coagulant dose range. However, relatively lower turbidity removal efficiency (<87%) was observed for alum, 1:1 and 3:1 alum-ferric chloride combinations at a dosage of 5 mg/L and 45 mg/L. Therefore, the optimum coagulant dose for alum and 3:1 alum-ferric chloride combination was 25 mg/L whereas the optimum coagulant dose for ferric chloride and 1:1 alum-ferric chloride combination was lower than alum, 15 mg/L. The results are presented in Fig. 2.

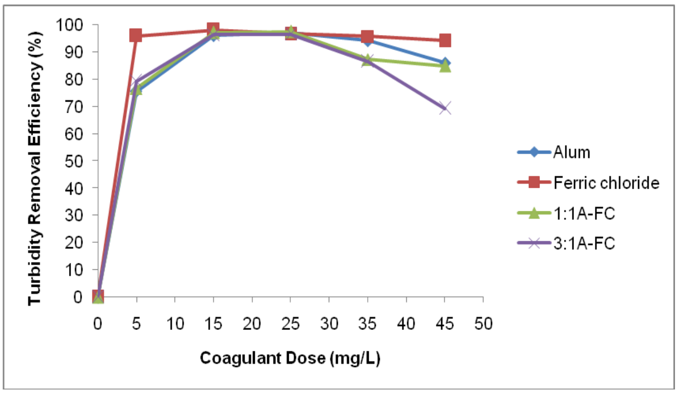

Fig. 2. Percentage turbidity removal as a function of coagulant dose at constant initial turbidity of 150NTU and optimum pH

C. Coagulant dose effect on treated water pH

Fig. 3 shows that an increase in the coagulant dose is associated with a decrease in the solution pH. The initial pH of solution at which ferric chloride and 1:1 alum-ferric chloride combination dose effect on final solution pH studied were 8. The initial pH of solution at which alum and 3:1 alum-ferric chloride combination dose effect on final solution pH studied were 7. As coagulants were added at a concentration of 5-45 mg/L the final pH value of the solution decreased in proportional to the dose increment rate.

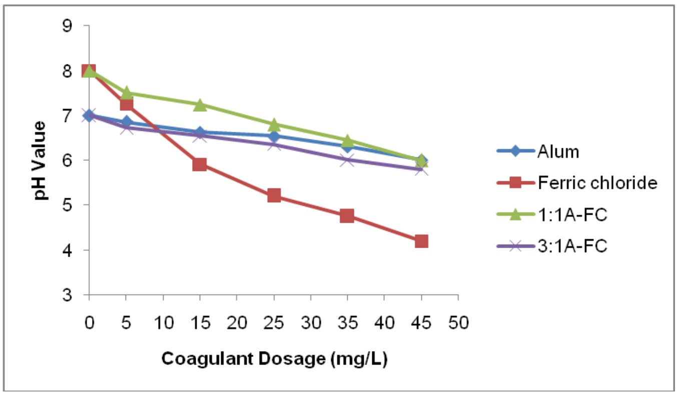

Fig. 3. Effect of coagulant dose on treated water pH

D. Effect of initial concentration of turbidity on coagulation process

The optimum dosages and pH of coagulants used in this study were tested for their effectiveness in varying initial turbidity. Accordingly, 25 mg/L alum coagulant concentration was effective over 150-300 NTU which has an efficiency of greater than ninety seven percent. Ferric chloride coagulant dose of 15 mg/L was effective through 30-500 initial turbidity at efficiency of greater than eighty six percent. The highest removal efficiency of ferric chloride was 99.06% at coagulant dose of 15 mg/L at initial turbidity of 300 NTU. At lowest (30 and 150 NTU) initial turbidities 1:1 alum-ferric chloride coagulant combinations was more effective than the rest three coagulants having efficiency of 87% and 98%, respectively. The optimized doses of alum and 3:1 alum-ferric chloride coagulants combination were overdosing for synthetic water having initial turbidity of 30 NTU which require much less concentration to destabilize colloidal. Fig. 4 gives percentage turbidity removal at optimal dose 25 mg/L, 15 mg/L, 15 mg/L, and 25 mg/L and optimal pH of 7, 8, 8 and 7 for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations as a function of initial turbidity, respectively.

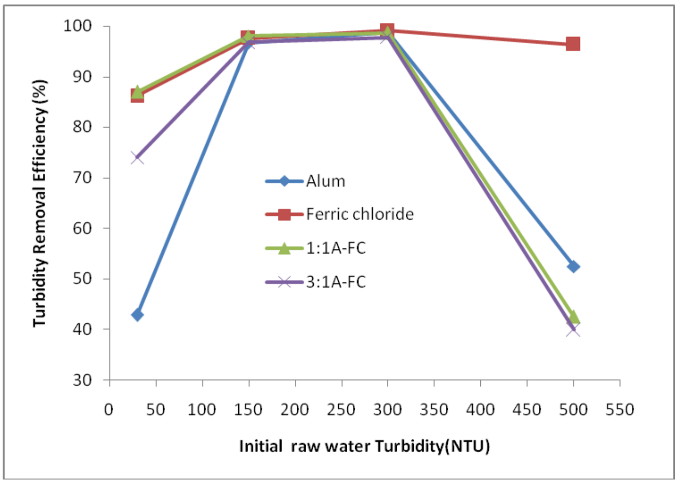

Fig. 4. Percentage turbidity removal at optimal dose and pH as a function of initial turbidity

The best performance of alum was observed at pH 7 over the selected turbidity range but its performance decreased to some extent at pH values of 4 and 9. The coagulation efficiency of alum at pH 6 and 8 was almost close to that of pH 7. The highest turbidity removal was attained at pH 7 when 25 mg/L alum concentration was used except for initial turbidity of 30 NTU and 500 NTU. At optimal dose and pH, turbidity removal efficiency of alum was 42.8%, 96.7%, 98.7% and 52.5% for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU, respectively. Overdosing was observed for low initial turbidity (30 NTU) when the optimal dose (25 mg/L) which was gained at initial turbidity of 150 NTU was used. Turbidity removal efficiency was slightly decreased by increasing alum concentration from 35 to 45 mg/L which may be related to charge reversal and destabilization of colloidal particles due to overdosing as suggested by Stumm and O'Melia (1968) [16]. The 1:1 alum-ferric chloride combination shows highest (94.82%) average turbidity removal and lowest standard deviation of 5.58 than the rest three coagulants. There was statistically significant difference (P<0.05) between means of coagulants removal efficiency at varying initial turbidity.

According to the finding of this study, the turbidity removal efficiency was varied by pH, alum dose and initial turbidity of water. The obtained result was in accordance with Baghvand *et al*. (2010) [13] and Volk *et al*. (2000) [17] which suggested that the pH of coagulation and dose of coagulant were influential parameter affect turbidity removed. The Baghvand *et al*., (2010) [13] study indicated that the initial turbidity of water was also affecting the turbidity removal efficiency. Under dosing was observed when 5 mg/L alum was used. The added coagulant dose was not much enough to form floc which can be categorized under zone 1 type of coagulation as stated by Shammas 2002[1].

The best performance of ferric chloride was observed at pH 8 and 15 mg/L dosage. The optimal ferric chloride coagulant dose 15 mg/L was used for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU. But this dose causes under dosing for raw water having initial turbidity of 500 NTU and overdosing for raw water having initial turbidity of 30 NTU. In this study, turbidity removal efficiency of ferric chloride was relatively stable at all dosages which were in the range of 94% -99%.

At optimal pH and dose, turbidity removal efficiency of ferric chloride coagulant combination was 86.17%, 97.63%, 99.06% and 96.35% for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU, respectively. The highest (99.06%) removal efficiency of ferric chloride was shown at dose of 15 mg/L which is at initial turbidity of 300 NTU. The optimized dose of ferric chloride coagulant was overdosing for synthetic water having initial turbidity of 30 NTU. Therefore, optimal dosage of 15 mg/L ferric chloride can be selected over the applied range of turbidity except for initial turbidity of 30 NTU which require much less concentration to destabilize colloidal. Based on ANOVA done, overall initial turbidity range (30-500 NTU) ferric chloride coagulants has much higher average turbidity removal efficiency (94.8%) and lower standard deviation (5.6) than the rest three coagulants used in this study which have less than 81.6% removal efficiency and greater than 26.6 standard deviation. The difference between means of coagulants removal efficiency was statistically significant (P<0.05).

The best performance of 1:1 alum-ferric chloride combination was observed at pH 8 over selected range of turbidity but its performance decreases at pH 4 and 9 which is below 90%. The coagulation efficiency of 1:1 alum-ferric chloride at pH 6 and 8 was almost close to each other. The highest turbidity removal was attained at pH 8 when 15 mg/L alum-ferric chloride 1:1 coagulant combination was used. Overdosing was observed for low initial turbidity (30 NTU) when the optimal dose (15 mg/L) gained at initial turbidity of 150 NTU was used. Turbidity removal efficiency was decreased by increasing coagulant concentration from 25 to 45 mg/L. At optimal pH and dose, turbidity removal efficiency of 1:1 alum-ferric chloride coagulant combination was 87.03%, 98.03%, 98.67% and 42.5% for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU, respectively. For 1:1 alum-ferric chloride coagulants combination the highest turbidity removal efficiency, 98.03% at standard deviation of 0.042 and initial turbidity of 300 NTU was observed. On the contrary, the lowest efficiency (42.5%) was shown at standard deviation of 0.42 and initial turbidity of 500 NTU which is due to under dosing of the optimal dose determined.

The best performance of 3:1 alum ferric chloride combination was observed at pH 7 and dosage of 25 mg/L which was similar optimal pH and dose for alum. The optimal coagulant dose 25 mg/L was used for initial turbidity of 30 NTU, 150 NTU, 300 NTU and 500 NTU. But this dose shows under dosing for initial turbidity of 500 NTU where as overdosing for initial turbidity of 30 NTU.

E. *Concurrent removal of contaminants*

The Jar tests experiment done to verify the optimal doses and optimal initial turbidity using real water sample indicated the following results. The initial TDS, COD and turbidity concentration of raw water was 565mg/L, 149mg/L and 173.6NTU, respectively. The highest percentage turbidity removal of alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination were 98.7%, 99.1%, 98.7% and 97.8%, respectively. The highest percentage removal of COD, TDS and turbidity were 71%, 55.8% and 97.4% for alum; 58.1%, 72.6%, and 97.6% for ferric chloride; 63.6%, 81.4% and 97.3% for 1:1 alum-ferric chloride; 50.9%, 81.4% and 97.4% for 3:1 alum-ferric chloride coagulants. The ferric chloride coagulant demonstrated highest (94.8%) average varying initial turbidity removal. Whereas, 1:1 alum-ferric chloride coagulant combination shows highest (80.8%) concurrent TDS, turbidity and COD average removal. Fig. 5 gives percentage turbidity, TDS and COD simultaneous removal from medium (173.6 NTU) initial turbidity real water at optimal dose of 25 mg/L, 15 mg/L, 15 mg/L and 25 mg/L for alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride combinations, respectively.

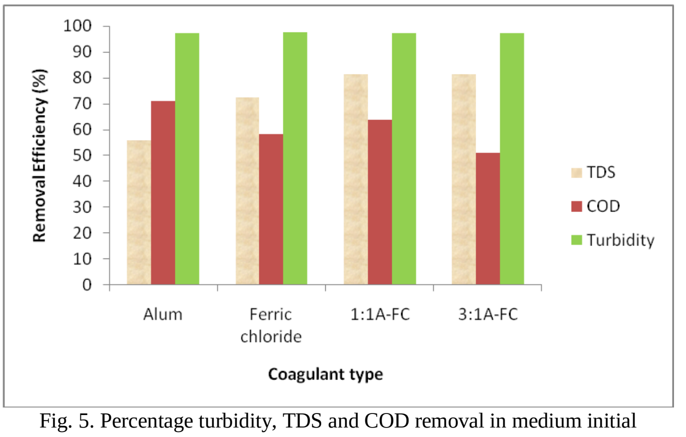

Fig. 5. Percentage turbidity, TDS and COD removal in medium initial turbidity at optimal dose

According to the statistical tests analysis ferric chloride coagulant demonstrated highest (94.8%) average varying initial turbidity removal. Whereas, 1:1 alum-ferric chloride coagulant combination shows highest (80.8%) concurrent TDS, turbidity and COD average removal. It can be concluded that in the analysis table of four coagulants ferric chloride and 1:1 alum-ferric chloride combination are the most suitable material to perform the coagulation process. This is because of their lower standard deviation and higher average removal efficiency than the other coagulants in concurrent removal of contaminants. Even though ferric chloride performed better turbidity removal than 1:1 alum-ferric chloride combination in wider initial turbidity range, it was observed that it cause red color water at low and high initial turbidity treatment.

According to the optimal rate of alum, ferric chloride and 1:1 alum-ferric chloride combination which is 15 mg/L, 25 mg/L and 15 mg/L, respectively. 1:1 alum-ferric chloride combination was economically suggested for doing chemical treatment. In this statistical test, alpha was 0.05. The optimal pH to use ferric chloride was 8 and for aluminum sulfate it was 7. Turbidity removal efficiency was sufficient to meet national drinking water limits of WHO (5 NTU) at optimum alum, ferric chloride and 1:1 alum-ferric chloride combination dose for waters with initial turbidity of 150-300 NTU.

As presented in Fig. 5, the turbidity and TDS removal efficiency for 1:1 alum-ferric chloride combination was higher than the rest coagulants used in this study. However, alum shows a highest COD percentage removal efficiency (71%). All four coagulants applied in this study show almost similar efficiency at removing turbidity although it is better removed by ferric chloride. The TDS removal efficiency of alum was relatively lower (55.8%) than other coagulants. The 1:1 and 3:1 alum-ferric chloride combination coagulants have better TDS removal efficiency (81.42%) than either alum (55.7) or ferric chloride (72.566%). The COD removal efficiency of alum, ferric chloride and 1:1 alum-ferric chloride combination were in a range of 60-70%. While, 3:1 alum-ferric chloride combination shows lower removal of COD (40.94%).

V. CONCLUSION

In this study the Jar test experiments were performed on low to high turbidity waters to find optimal conditions at which better turbidity removal performance of alum, ferric chloride, 1:1 and 3:1 alum-ferric chloride coagulants. The synthetic water was used to determine the optimum pH and coagulant concentration ranges required to achieve the maximum turbidity removal percentage and it was later verified with real water.

The coagulation experiments using aluminum sulfate, ferric chloride, 1:1 and 3:1 alum-ferric chloride combination indicated that coagulation process effectively removes turbidity from water using 15-25 mg/L dose of the coagulants. The alum-ferric chloride 1:1 and 3:1 combinations were more effective in the removal of TDS removal. The optimum pH range for turbidity removal was found 7, 8, 8 and 7 for alum, ferric chloride 1:1 and 3:1 alum-ferric chloride combinations, respectively. The selected optimal doses were more effective for medium (150 NTU-300 NTU) than low (30 NTU) and high (500 NTU) initial raw water turbidity.

According to the optimal rate of alum, ferric chloride and 1:1 alum-ferric chloride combination which is 15 mg/L, 25 mg/L and 15 mg/L, respectively, indicated that alum-ferric chloride combination is economically suggested in water treatment at optimal conditions assuming equal coagulant cost.

The optimized alum-ferric chloride combination can be an alternative to remove many contaminants simultaneously. Currently, the use of mix-chemical coagulant is not at a stage where it can be implemented at full scale. Application of alternative coagulant combinations to meet allowable limits of contaminant should be further studied.

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