

Evaluation of Residual Al^{3+} and Fe^{3+} Concentration in Blended Alum-Ferric Chloride Coagulant Use

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Abstract

The use of aluminum and iron salts as coagulants in water treatment may cause the rise of aluminum and iron ions concentrations in finished water. Approximately drinking water is responsible for nearly five percent of the aluminum ingested by humans although aluminum accumulation in the brain is associated with neurodegenerative diseases. This study aims to evaluate the residual concentration of Al^{3+} and Fe^{3+} upon the use of blended alum-ferric chloride coagulant in clarifier effluent. An experimental study of Jar tests were conducted to evaluate the effect of blended alum-ferric chloride coagulant use on the finished water residual aluminum and ferric ion concentration. All parameters were determined according to APHA standard methods for water and wastewater examination. Data were compiled and analyzed using Microsoft excels 10 and Minitab 16 and presented using tables and graphs. As compared to single coagulant use, 1:1 A-FC (Alum-Ferric chloride) mix-use resulted in an average of 40% aluminum ion concentration and 20% residual ferric ion concentration reduction. At optimal pH and dose, the residual aluminum ion concentration were 0.1 mg/L, 0.06 mg/L and 0.09 mg/L for alum, 1:1 and 3:1 A-FC coagulants, respectively. While, the ferric ion concentration were 0.4 mg/L, 0.32 mg/L and 0.11 mg/L for ferric chloride, 1:1 and 3:1 alum-ferric chloride coagulants, respectively. The study demonstrated a strong correlation ($r=0.93$) between coagulant dose and residual aluminum ion concentration which was statistically significant ($P<0.05$). Also, a strong correlation ($r=-0.97$) was observed between the water pH and residual aluminum ion concentration which was similarly statistically significant ($P<0.05$). The residual aluminum and ferric ion concentrations were greatly reduced when alum and ferric chloride coagulants were used in combinations than used separately. The regular monitoring of chemical residuals in drinking water and control of coagulant conditions should be considered in the water treatment process.

Keywords: Coagulant; Alzheimer's dementia; Water treatment; Aluminum; Iron

Introduction

In the water treatment process, alum and ferric chloride are amongst the extensively used chemical coagulants in many countries. They quickly react with water to give a variety of products containing cationic species, which can neutralize negatively charged particles [1,2]. Although most of the coagulant used is retained in sludge during the sedimentation process, a part of it remains in finished water when important coagulation parameters such as pH, dose coagulant, and raw water turbidity were poorly controlled [3]. The residual Al^{3+} and Fe^{3+} concentration in drinking water distribution lines above the recommended limit are becoming a great concern of drinking water quality as emerging studies are proofing their health concern [4].

High residual aluminum ion concentrations may be deposited inside the drinking water pipelines which can be a source of biofilm consisting mostly of bacteria that further serve as homes for waterborne disease. The deposited coagulant residue can reduce the

efficiency of residual chlorine which safeguards finished water from further contamination [5]. On the other hand, disturbance of these deposits due to water flow rate change may re-suspend aluminum ion levels at the tap which makes the end-user to be dissatisfied as it can cause unwanted taste, odor, color, and turbidity of drinking water [6].

Drinking water is among the source of aluminum ion which can contribute to the ingestion of aluminum. According to the assessment of the American Waste Water Association (AWWA), drinking water provides about five percent of the aluminum ingested by humans [7]. As compared to other routes of exposure, orally swallowed aluminum is intensely poisonous to humans although this element occurs widely in foods, drinking water and many antacid preparations since aluminum is more dissolved in the stomach when the pH is extremely low as the availability of aluminum in elemental form largely depends on the pH [1,7]. Ingested aluminum reaches the blood by absorption and can cross the blood-brain barrier [8], and accumulated in the

brain. Aluminum accumulation in the brain is suggested to be linked with neurodegenerative diseases such as Alzheimer's dementia and dialysis encephalopathy [9-11] hence aluminum in drinking water is considered as a hazard [6]. Hospitals and clinics engaged in treating kidney disease patients may have difficulties with alum treated water. Patients who are exposed to high levels of aluminum in dialysis fluids and medications may develop complications such as dialysis encephalopathy, a progressive form of dementia, and psychosis [11]. Unites States Environmental Protection Agency (USEPA) has established a maximum contaminant level at 0.2 mg/L for aluminum content in drinking water [12]. The assessment of water treatment plant operations is suggested if aluminum residuals exceed 0.3 mg/L in finished water [4].

Similar to aluminum, residual iron is suggested that it can be involved in Alzheimer's disease [13]. As it was cited in Sieliechi JM, et al. [7], Perez and his colleagues have stated that excessive iron in selective regions of the brain may be involved in the neurodegenerative disorders [14]. On the other hand, a high amount of residual iron in drinking water makes the chlorination process to lose its effect, the coloration of water and the metallic tastes of water [7]. It can also serve as food for microorganisms that use a non-carbon source such as iron manganese which facilitates the propagation of pathogenic bacteria. The growth of ferruginous bacteria can form unpleasant thick liquid layers inside pipes and storage tanks [4,15]. The residual iron in finished water can also lead to stains on laundered clothes and plumbing fixtures such as sinks and toilets above certain concentrations [4]. Moreover, excessive ingested iron is poisonous; it can damage the cells of the gastrointestinal tract so that preventing them from regulating iron absorption [16]. From an aesthetic objective, less than 0.3 mg/L content of iron is recommended in drinking water [4].

Minimizing the amount of residual aluminum is essential in water used at hospitals, the food industry, and in drinking water, and also reducing the amount of residual iron is important in water used at the laundry, in drinking water, in water disinfection process as well as in water distribution lines. Hence, in this study, the 1:1 and 3:1 alum-ferric chloride blend in comparison to single alum and ferric chloride usage were investigated to determine their contribution of residual aluminum and ferric ions in drinking water.

Methods

Experimental design

An experimental study was performed in which factors were evaluated under various experimental setup. Two test sets were designed where one set was employed as a control group (Aluminum Sulfate and Ferric chloride) and the other one was the experimental group (1:1 and 3:1 alum-ferric chloride mix). The effect of alum-ferric chloride blend use on treated water residual aluminum and ferric ion concentration was studied using results achieved in an earlier study by Gobena B, et al. [17]. Accordingly, residual aluminum and iron determination were performed under optimal conditions; 25mg/L, 15 mg/L, 15 mg/L and 25 mg/L dosage, and 7,8,8,7 pH, for alum, ferric chloride, 1:1A-FC and 3:1A-FC, respectively. To investigate the residual aluminum relation with the dose of alum use, the coagulant dosages were ranged from 5 to 45 mg/L with an interval of 10mg/L, and the residuals were determined for each dose.

Sample preparation

The synthetic water having 150NTU initial turbidity was obtained by adding clay passed through the sieve no.200 to a certain volume

of deionized water to introduce suspended solids and organic matter [18]. Stock solutions of Aluminum sulfate, $Al_2(SO_4)_3 \cdot 18H_2O$, and ferric chloride, $FeCl_3$ was prepared by dissolving 10.0 grams of alum and/ or ferric chloride into 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 [19]. The 1:1 and 3:1 alum-ferric chloride coagulant combinations were formed by mixing the standard stock solution of alum and ferric chloride to make the required coagulant dosage of alum-ferric chloride blend.

Experimental procedures

The turbidity removal efficiency of study coagulants was determined by the standard Jar test experiments. Each jar was filled with 1000 ml of the sample measured with a graduated cylinder. The pH value of the raw water was adjusted with 0.5N H_2SO_4 and 1N NaOH to the required level [20]. The required coagulant dose designed for each jar was carefully measured into 1000 mL beakers containing 1000mL of synthetic water prepared. The stirrer speed was set at 200 rpm for 1 minute. After 1 minute, the mixing speed was reduced and was set at 20 rpm for 15 minutes. After this period, the stirrer was turned off and the flock was allowed settling for 30 minutes [11,19]. Samples were then withdrawn from where located 20 mm below the water level for analysis [21]. The residual turbidity and pH were measured immediately after the end of the settling time of the Jar test; whereas, residual aluminum and iron were determined after pretreated by acidification at room temperature at pH 1 for two hours [22].

Sample and data analysis

Turbidity was measured by the Nephelometric Method (SM: 2130B) using Turbidity meter HACH Wag-WT3029, the pH was measured by Electrometric method (SM: 4500-H) [23] using a portable pH meter WTW 3310, Al^{3+} determined using the Aluminum method (SM: 8012) [24] and Fe^{3+} determined using Phenanthroline (SM: 3500) [23] method by spectrophotometer. Data were compiled and analyzed using Microsoft excels 10 and Minitab 16 and presented using tables and graphs.

Results and Discussion

Turbidity removal efficiency of different coagulants at different dose

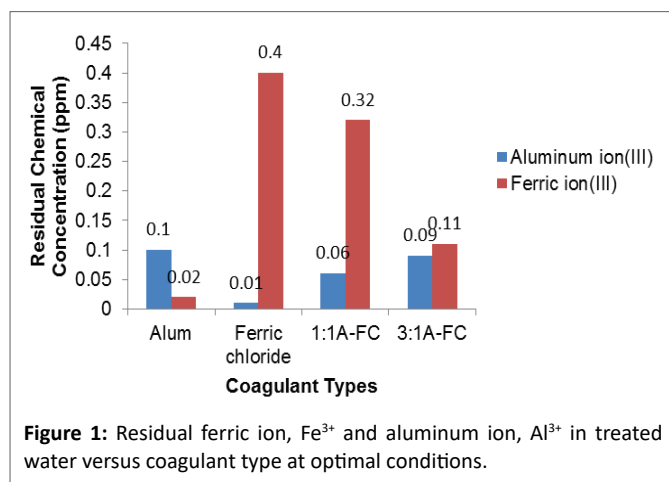
The baseline data from an earlier study showing the turbidity removal efficiency of different coagulants and coagulant combinations were presented in table 1. Accordingly, the finished water residual turbidity of 3.82 NTU (97.45% removal efficiency) was achieved upon the use of 25mg/L of alum. Whereas upon the use of 15 mg/L ferric chloride, the residual turbidity, 2.82NTU (98.12% removal efficiency) was observed. The final pH of finished water upon the use of 5-45mg/L of alum and ferric chloride were in between 6.61-5.35 and 7.25-4.5, respectively. The finished water residual turbidity of 4 NTU (97.34% removal efficiency) upon the 1:1 A-FC coagulant dosage of 15 mg/L and 6.73 NTU (96.57 % removal efficiency) upon 3:1 A-FC coagulant dosage of 25 mg/L were observed. Ferric chloride turbidity removal efficiency was greater than 94% in the 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. The final pH of finished water upon the use of 5-45mg/L of 1:1 A: FC and 3:1 A: FC was in between 6 to 7.5 and 5.73 to 6.75, respectively.

The residual Al^{3+} and Fe^{3+} concentration in finished water at optimal conditions

The residual Al^{3+} and Fe^{3+} concentration in finished water at optimal conditions were presented below. According to this study, upon the use of alum at the optimal dose of 25mg/L and optimal pH of 7, the residual aluminum and ferric ion were 0.1mg/L and 0.02mg/L, respectively. Whilst, upon the use of ferric chloride at the optimal dose of 15mg/L and optimal pH of 8 the residual aluminum and ferric ion, was 0.01mg/L and 0.44mg/L, respectively. However, upon the use of 1:1 alum-ferric chloride combination at the optimal dose of 15mg/L and optimal pH of 8 the residual aluminum and ferric ion, was 0.06mg/L and 0.32mg/L, respectively. Likewise, the residual aluminum and ferric ions were 0.09 mg/L and 0.11mg/L upon the use of 3:1 alum ferric chloride combination at an optimum dose of 25 mg/L and a pH of 7. The results are presented in figure 1.

The lowest aluminum ion concentration was achieved upon ferric chloride use in finished water although it increases the residual ferric ion in finished water to 0.32 mg/L by 95% in comparison to alum use, which is also above the secondary USEPA drinking water regulation of 0.03 mg/L [12]. However, in comparison to alum which has contributed 0.1 mg/L of the aluminum ion at optimal conditions, ferric chloride has reduced the concentration of aluminum ion to 0.01 mg/L by 90% which is by far safe level. The aluminum residual upon ferric chloride use and the iron residual upon alum use appeared may be attributed to the chemical coagulant impurity and the clay employed to make the required initial turbidity of test water.

Comparing the experimental group (1:1 A-FC and 3:1 A-FC use) with the control group (alum and ferric chloride use), the 1:1 A-FC use resulted in a 40 % reduction of aluminum ion concentration as compared to alum use and 20 % reduction of ferric ion concentration as compared to ferric chloride use. Whereas, the 3:1 A-FC use shown a slight reduction (10%) of the aluminum ion as compared to alum use and good reduction (72.5%) of the ferric ion as compared to ferric chloride use. At optimal conditions, the 1:1 A-FC use resulted in acceptable residual aluminum ion and satisfactory residual ferric ion concentration with 97.34% removal efficiency. The concentration of individual coagulant concentration has reduced upon blend use of coagulants which further reduce the amount of coagulant residue in finished water.



The aluminum ion residual concentration was higher when alum was used alone than when alum-ferric chloride 1:1 and 3:1 combinations were used. Miller and his colleagues reported a forty to fifty percent chance of an increase in aluminum concentrations in drinking water plants using aluminum-based coagulants [24,25]. A similar result was observed in the findings of this study. At optimal condition, residual aluminum ion concentration was decreased nearly by two-fold upon 1:1 A-FC without compromising the required turbidity removal efficiency compared to single alum use. While, residual ferric ion concentration was decreased nearly by three-fold upon 3:1 A-FC use, irrespective of required turbidity removal compared to single ferric chloride use.

Effect of coagulant dose on residual coagulant concentration

The findings of this study shows that the residual aluminum ion increases as coagulant dose increases under optimal conditions. The aluminum ion has increased from 0.07 to 0.08 mg/L by 12.5 % upon coagulant dose increase from 5 mg/L to 15 mg/L, increased from 0.08 to 0.1 mg/L by 20 % upon coagulant dose increase from 15 mg/L to 25 mg/L, increased from 0.1 to 0.15 mg/L by 33.33 % upon coagulant dose increase from 25 mg/L to 35 mg/L, and increased from 0.15 to 0.23 mg/L by 34.78 % upon coagulant dose increase from 35 mg/L to 45 mg/L (Figure 2).

The minimum, 0.07mg/L residual aluminum was observed upon the use of the lowest coagulant dose (5mg/L) though the turbidity removal efficiency is below 80%. At 15mg/L coagulant dosage the residual aluminum ion increases to 0.08 mg/L with an increase of 96.2% removal efficiency. Above 25mg/L coagulant dosage, the removal efficiency declines while the residual aluminum concentration continues to increase. In this study as the dosage of chemical coagulant increase, the residuals observed increasing. The residual Al^{3+} ion concentration is directly proportional to the dosage of coagulant. However, at optimum alum dose (25 mg/L) the residual Al^{3+} is relatively low. Upon the use of optimal alum coagulant dosage, 25 mg/L to treat water having a turbidity of 150 NTU the aluminum ion concentration at a solution pH of 6.5 was 0.1 mg/L. The results are presented in figure 2 below.

The presence of aluminum in drinking water can be related to either the addition of aluminum salts in the course of coagulation and flocculation treatment or naturally occurring in raw water due to a low pH ($pH=5.5 \pm 0.5$) value [26]. There are various regulations on

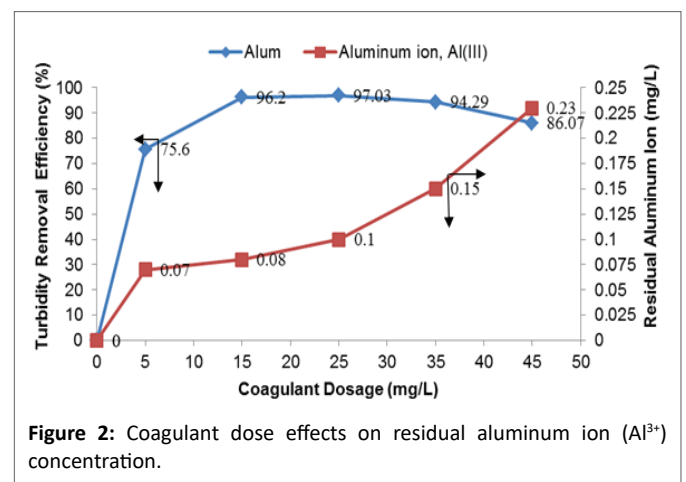


Table 1: Turbidity removal as a function of coagulant dose and type.

Coagulant types	Parameters	Coagulant Dose (mg/L)				
		5	15	25	35	45
Alum ^a	Residual turbidity (NTU)	36.6	5.7	3.82	8.57	20.90
	Removal Efficiency (%)	75.60	96.20	97.45	94.29	86.07
	pH of finished water	6.61	6.35	6.05	5.71	5.35
Ferric chloride ^b	Residual turbidity (NTU)	6.15	2.82	4.95	6.25	8.15
	Removal Efficiency (%)	95.90	98.12	96.70	95.83	94.17
	pH of finished water	7.25	5.91	5.00	4.75	4.50
1:1 Alum-Ferric chloride mix ^b	Residual turbidity (NTU)	35	4.00	13.80	18.90	22.55
	Removal Efficiency (%)	76.67	97.34	90.80	87.40	84.97
	pH of finished water	7.50	7.25	6.80	6.45	6.00
3:1 Alum-Ferric chloride mix ^a	Residual turbidity (NTU)	30.90	5.30	6.73	20.05	46.10
	Removal Efficiency (%)	79.40	96.47	96.57	86.63	69.27
	pH of finished water	6.75	6.58	6.25	6.05	5.73

^a tested at optimal pH 7 of test water,

^b tested at optimal pH 8 of test water and

The initial turbidity of test water for all coagulant was 150NTU.

aluminum in drinking water which can vary for different authorities (Table 2). Aluminum ion concentration in drinking water above 0.3 mg/L poses a health hazard to a human being [6]. World Health Organization suggested several approaches for minimizing residual aluminum concentrations in treated water. These include the use of optimum pH in the coagulation process, avoiding excessive aluminum dosage, good mixing at the point of application of the coagulant, optimum paddle speeds for flocculation, and efficient filtration of the aluminum floc [25,27].

Although there is no health-based guideline value proposed for iron in drinking water [28], this study considered the use of alum in combination with ferric chloride in different ratios as another alternative solution. At optimal conditions, the residual turbidities were below the drinking water standards of USEPA [12] for alum, ferric chloride, and 1:1 alum-ferric chloride which were 3.8mg/L, 2.8mg/L, and 3.9mg/L, respectively. However, when 3:1 alum-ferric chloride was used the residual turbidity was above the standard, 6.7 mg/L. The residual aluminum was 0.1 mg/L upon the application of alum at optimal conditions which is complying with drinking water standards of USEPA, EU, WHO, and AWWA (Table 2).

In another hand, the residual iron was 0.4 mg/L upon the application of ferric chloride at optimal conditions was above the drinking water standards of USEPA [12]. Comparing the results of this study to the residual ferric ion limits set by USEPA [12] and WHO [29], the 1:1 A-FC contribution is above limits; whereas, 3:1 A-FC contribution is below the limits. The fact that this study is discussing the residual coagulant found in the effluent of clarifier the residue has a great chance to be more reduced further in the consecutive treatment process like filtration and any other advanced water treatment processes.

In the drinking water treatment process, the pH of coagulation, and residual turbidity (removal efficiency) were important factors in influencing the residual aluminum concentrations. Hence, this study has tried to demonstrate the relations of parameters as presented in table 3. Accordingly, the coagulant dose and aluminum sulfate turbidity removal efficiency were weakly positively correlated ($r=0.332$) which is statistically insignificant ($P>0.05$). Similarly,

aluminum sulfate turbidity removal efficiency and residual aluminum ion concentration correlation was weak but positively correlated ($r=0.0135$) which is statistically insignificant ($P>0.05$). Whereas the coagulant dose and residual aluminum ion concentration were strongly positively correlated ($r=0.94$) which is statistically significant ($P<0.05$). The correlation coefficient of dose and final pH of the water was strong negative correlation ($r=-0.98$) and statistically significant ($P<0.05$). Similarly, the correlation coefficient of water pH and residual aluminum ion concentration was $r=-0.97$ which also shows a strong negative correlation and statistically significant ($P<0.05$).

As it was reported by Jekel MR [26], there is a correlation between residual aluminum and effluent turbidity (removal efficiency); low effluent turbidity would lead to a reduction in residual aluminum. On another hand, a study of residual aluminum by Driscoll and Letterman revealed that high concentrations of aluminum in drinking water were associated to both raw water concentrations and residual turbidity [22,27]. However, this study, revealed that it is insignificant ($P>0.05$) weak ($r=0.0135$) correlation between aluminum sulfate turbidity removal efficiency and residual aluminum ion concentration. Rather, residual aluminum ion in finished water is strongly correlated to coagulant dose and pH of finished water.

Also the ANOVA was performed at 95% confidence interval to evaluate whether there is a statistically significance difference in the mean residual coagulants among the four coagulants. Accordingly, this study concluded that there is a statistically significance difference in the mean residual aluminum among the four coagulants ($p<0.05$) (Table 4a). Similarly, there is a statistically significance difference in the mean residual iron among the four coagulants ($p<0.05$) (Table 4b).

Conclusions

The study exhibited that the aluminum ion and ferric ion concentration was significantly reduced when coagulants used in combination than used separately. As it was hypothesized at the beginning of the study, blended use of alum and ferric chloride in the ratio of 1:1 and 3:1 has made a balance of residual aluminum ion and ferric ion. Especially, the 1:1 alum-ferric chloride was found to reduce

Table 2: Aluminum regulations in drinking water of different authorities.

S. No.	Jurisdiction	Aluminum Limit
1	Guidelines for Canadian Drinking Water Quality	0.1mg/l
2	USEPA (2018)	0.05 to 0.2 mg/l
3	European Economic Community	0.05 µg/l to 0.2 mg/l
4	World Health Organization	0.2 mg/l
5	AWWA Recommended Operating Level	0.2 mg/l
6	California Code of Regulations	1.0 mg/l

Table 3: Relations of parameters.

Correlation Coefficient Matrix			
	Dose	Alum removal efficiency	Aluminum ion
Alum removal efficiency	0.33, P=0.59	1	
Aluminum ion	0.937, P=0.019	0.013, P=0.983	1
Final pH	-0.983, P=0.003	-0.240, P=0.698	-0.974, P=0.005

Table 4: ANOVA

4a: One-way ANOVA: Residual Aluminum (Al^{3+}) versus coagulant type.

Source	DF	SS	MS	F	P
Factor	3	0.014700	0.004900	34.69	0.000
Error	8	0.001130	0.000141		
Total	11	0.015830			

Table 4: ANOVA

4b: One-way ANOVA: Residual iron (Fe^{3+}) versus coagulant type.

Source	DF	SS	MS	F	P
Factor	3	0.282825	0.094275	341.58	0.000
Error	8	0.002208	0.000276		
Total	11	0.285033			

the residual aluminum and ferric ion concentration in finished water below the limits set by many authorities. The coagulant type used in the coagulation process is an important parameter in determining residual coagulant amounts in finished water regardless of other design parameters. Moreover, it was revealed that the residual aluminum and ferric ion concentration is seemed to be more influenced by the final pH of water followed by the coagulant dose applied. Hence, in water treatment, the optimal conditions should be followed to produce a water that has low residuals. Also, the water treatment plants should switch to using alternative coagulants which contribute less chemical residuals, like blended coagulant use as it has shown promising results.

Abbreviations

A-FC: Alum- Ferric chloride blend; UNEP: United Nations Environment Programme; USEPA: United States Environmental Protection Agency; WHO: World Health Organization; EU: European Commission, AWWA: American Water Works Association;

NTU: Nephelometric Turbidity Unit; SM: Standard Method; EPA: Environmental protection Agency; ASCE: American Society of Civil Engineers

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