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ENHANCED REMOVAL OF RESIDUAL ALUMINUM AND TURBIDITY IN TREATED WATER USING POLYMERS

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Abstract : This study investigated the possibility of reducing the residual aluminum (Al) in the treated water using polymers. Two raw waters (lake and river water) and three kinds of polymers (coagulant, flocculant, and filtration aids) were used for this purpose. This study found that coagulation at the high dose did not necessarily lead to the high concentration of the residual Al in the treated water. The coagulation efficacy was found more important in determining the residual Al than the coagulant dose. The use of a polymer enhanced the removal of turbidity as well as the residual Al. The coagulant aid removed the dissolved Al as well as the particulate Al by helping the coagulation and the solid-liquid separation. The flocculant aid and the filtration aid preferentially removed the particulate Al while helping the solid-liquid separation. The filtration aid reduced the residual Al substantially more effectively than the flocculant aid. The polyamine-based coagulant aid (FL) showed the better performance in reducing the residual Al and turbidity than DADMAC (WT). The cationic flocculant aid with weak charge density and the medium molecular weight (SC-020) showed the best performance in reducing the residual Al.

Key Words : Residual Al, Coagulant aid, Flocculant aid, Filtration aid, Polymer

INTRODUCTION

The residual Al is a by-product of water treatment. Most water treatment plants rely on Al-based coagulants. After these coagulants are added to raw water, some end up in sludge and others remain in the treated water forming the residual Al. Therefore, the residual Al is usually present at the higher concentration in the treated water than in the raw water.[1-3] The residual Al passing through water treatment processes can cause a problem in the distribution system due to post-precipitation. The precipitates increase the turbidity. The increased turbidity then interferes with disinfection. The turbidity can also decrease the water-carrying capacity of the distribution system.[4-6] These problems have led to the regulation of the residual Al concentration in the treated water in Korea (< 0.2 mg/L). There is also a health concern for the residual Al. It has been speculated that the ingestion of the high concentration of Al may be related to the Alzheimer's disease.[7]

There are several methods available for controlling the residual Al. These include;[8]

- pH control
- Use of an alternative coagulant
- Reduction of the Al-coagulant dose using polymers
- Improvement of particulate Al removal efficiency during filtration

The most popular method for controlling the residual Al is the pH control. Since $Al(OH)_3$

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flocs show the minimum solubility at pH 6, the formation of the residual Al can be minimized by adjusting the pH to 6. The benefit of an alternative coagulant is obvious. Since the residual Al results from the use of an Al-based coagulant, the problem can be avoided by using a Fe-based coagulant. Improving filtration performance can also minimize the residual Al. Most of the residual Al consists of the particulate Al, while filtration is very effective in removing the particulate Al. Therefore, improving filtration performance can reduce the residual Al. This method was effectively demonstrated in the plant-scale experiments.[1,9)]

This study targets on the method of using polymers. This method aims to reduce the dose of an Al-based coagulant as much as possible. Since the use of an Al-based coagulant is directly related to the residual Al in the treated water, the more Al-based coagulant that is used, the more probable residual Al that is left behind. This leads to the speculation that reducing the coagulant dose may decrease the residual Al concentration in the treated water. As a result, the proposed method attempts to reduce the coagulant dose using polymers. This method has been previously suggested in literature,[8)] but no experimental data is available. Therefore, this study attempts to check the speculation and subsequently the validity of this method. The enhanced removal of turbidity was also examined because polymer is expected to be helpful in the particle removal.

The objectives of this study can be summarized as follows;

- To investigate whether the high dose of an Al-based coagulant leads to the high concentration of the residual Al,
- To investigate whether polymer can enhance the removal of the residual Al as well as turbidity,
- To compare the effectiveness of different polymer products.

MATERIALS AND METHODS

Raw Water

Two different raw waters were used in this study. "A" and "B" waters were taken from a lake and a river. "B" water is being used as the raw water source for the nearby water treatment plant (BWTP). BWTP produces 270,000 m^3 of the treated water daily, using the processes of pre-ozonation, coagulation, flocculation, sedimentation, filtration, intermediate-ozonation and biological activated carbon. The characteristics of these raw waters are summarized in Table 1. "A" water was high in the alkalinity. Besides the alkalinity, these waters represented the typical qualities found in Korea.

Experiments

PACl (polyaluminum chloride) was used as the primary coagulant in this study because it is currently used at BWTP. The effects of coagulant and flocculant aids were evaluated using the jar tests. The effects of a filtration aid were evaluated using the filtration apparatus, as shown in Figure 1. Rapid and slow mixing was given at 150 rpm for 1 minute and at 30 rpm for 20 minutes. A 30-minute period of settling followed the slow mixing. When a coagulant aid was used, the rapid mixing period was extended to 2 minutes. After the addition of the primary coagulant, 1 minute of rapid mixing was provided. Thereafter, a coagulant aid was injected and the rapid mixing continued for another 1 minute. A flocculant aid was added during the slow mixing period. When the flocs grew to a visible size, a flocculant aid was injected. It took about 5 minutes for the flocs to become visible.

Table 1. Yearly Average Values of the Raw Waters Characteristics Used in This Study

Description	Turbidity, NTU	pH	Alkalinity, mg/L	TOC, mg/L	TDS, mg/L
"A" water	13.2	8.2	110	5 ~ 10	200
"B" water	8.9	7.6	62	5 ~ 10	328

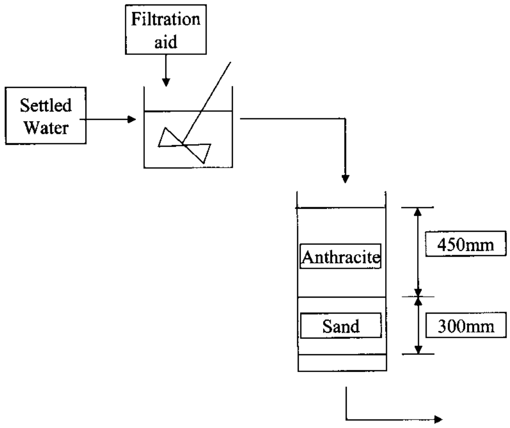

Figure 1. Filtration apparatus used in this study.

The supernatant was taken for the analysis of the residual Al concentration, turbidity, and pH. The sludge was taken to measure its volume. The Eriochrome Cyanine R method was used with a slight modification to measure the residual Al concentration.[10,11] Table 2 shows the evaluation results of the Eriochrome Cyanine R method. As shown in Table 2, this method was reliable. The residual Al was divided into two components: dissolved and particulate Al. The residual Al passing through a 0.45 μm filter paper was taken as the dissolved Al concentration and the difference from the total Al was taken as the particulate Al concentration. Table 2 shows that there was not much difference between filter papers with various pore sizes in their ability to divide the dissolved and the particulate Al concentrations. Therefore, a 0.45 μm filter paper was used in this study.

Polymer

Polymers used in water treatment are generally grouped based on their purposes. They can be used as a primary coagulant, coagulant aid, flocculant aid, filtration aid, or conditioning aid.[12,13] A polymer can be used solely as a primary coagulant or in combination with another primary coagulant during the coagulation step. If a polymer is used in combination with another primary coagulant, it is called "coagulant aid". Once particles are neutralized during coagulation, they need to grow so that they can settle easily. This step is often called "flocculation". A polymer used in the flocculation step in order to improve the floc settleability is called "flocculant aid". Flocculant aid functions by bridging or enmeshing the neutralized particles into larger flocs. When a polymer is added just prior to filtering in order to improve the filtration performance, it is called "filtration aid". A polymer can be also added to improve the sludge dewaterability. A polymer used for this purpose is called "conditioning aid". Since coagulant, flocculant, and filtration aids can all affect the residual Al in the treated water, all of them were used in this study.

The polymer characteristics differ depending on their purposes. A primary coagulant or coagulant aid typically has a low molecular weight with a positive charge. A flocculant, filtration, or conditioning aid usually has a high molecular weight and possesses a positive, negative, or neutral charge. Two different products (FL and WT) were selected as the coagulant aid. The FL was made of polyamines and WT was poly-DADMAC (Diallydimethyl Ammonium Chloride). They were both positively charged. Their molecular weights were within the range of 10^4~10^5. Polyacrylamide products were used as the flocculant and filtration aids. Table 3 shows the six different products of polyacrylamides used in this study. As shown in Table 3, their molecular weights (4×10^6~1.3×10^7) were of a 1~3 magnitude higher than those of FL and WT. These products were selected to include the different charge, molecular weight,

Table 2. Evaluation Results of the Al Analytical Method

Eriochrome Cyanine R method	Dissolved Al concentration	
	Pore size	Measured (0.05 mg/L)
- Recovery 112 %	- 1.20 μm	0.046±0.0071 mg/L
- Precision 4 %	- 0.45 μm	0.044±0.0057 mg/L
	- 0.22 μm	0.052±0.0110 mg/L

Table 3. Polyacrylamide-Based Polymers Used in This Study

Products	Charge type	MW*	Charge density	Viscosity, cp**
SC-312	Cationic	4×10^6	Strong	800
SC-030	Cationic	5×10^6	Medium	800
SC-020	Cationic	6×10^6	Weak	600
SA-407	Anionic	13×10^6	Strong	350
SA-207	Anionic	12×10^6	Weak	200
SN-557	Nonionic	13×10^6	-	50

MW* : molecular weight
cp** : centipoise

and charge density so as to examine their effects.

RESULTS AND DISCUSSIONS

Effects of a Primary Coagulant

Figure 2 shows the effects of a primary coagulant (PACl) on the residual Al concentration and turbidity for "A" water. Since similar results were obtained for "B" water, those results were not included. Figure 2 clearly shows that the coagulant addition initiated the increase of the residual Al concentration. However, the residual Al did not keep increasing with an increasing dose of the coagulant. Once increased, further addition of the coagulant rather decreased the residual Al concentration. This result indicates that the high dose of the coagulant did not necessarily lead to the high concentration of the residual Al. Instead, the residual Al concentration was determined by the efficacy of the coagulation and sedimentation. The minimum concentration of the residual Al was obtained at the optimum coagulant dose in terms of turbidity removal.

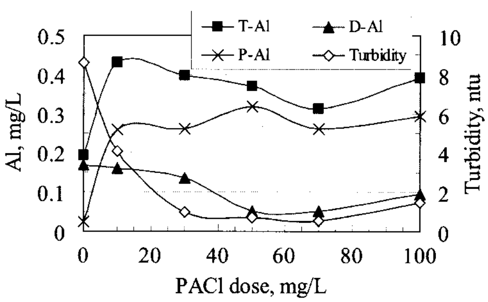

Figure 2. Effects of a primary coagulant on the residual Al concentration and turbidity for "A" water (T-Al: total aluminum, D-Al: dissolved aluminum, P-Al: particulate aluminum).

Figure 2 shows three different residual Al concentrations (total Al, particulate Al and dissolved Al). Each concentration behaved differently with the coagulant addition. The dissolved Al concentration and turbidity showed the similar behavior. The dissolved Al concentration decreased immediately after the coagulant addition. It continued to decrease with an increasing dose of PACl, reached the minimum, and then increased again. The particulate Al showed the opposite pattern. It increased immediately after the coagulant addition and then, changed slightly with an increasing dose of the coagulant.

The behavior of the dissolved Al concentration reflected the coagulation efficacy. The dissolved Al concentration was kept minimal under the optimum coagulation condition. It became high when the coagulation condition deteriorated. The decrease in the dissolved Al concentration with an increasing dose of PACl indicated that the coagulation effectively progressed, approaching the optimum condition. It would appear that the lowest dissolved Al concentration was obtained at the minimum solubility of the $Al(OH)_3$ precipitate. The overdose of the coagulant decreased the pH and deteriorated the coagulation condition. This resulted in an increase in the dissolved Al concentration. The behavior of the particulate Al concentration reflected the efficacy of the solid-liquid separation, (here, sedimentation). Once increased, the particulate Al concentration never decreased because the resulting floc settled poorly.

Coagulant Aid

Table 4 summarizes the effects of a coagulant aid for different source waters: lowly turbid "A" water and highly turbid "B" water. While two different types of coagulant aid (FL and WT) were fixed at 2.0 mg/L, the dose of a primary coagulant (PACl) was varied from 10 to 30 mg/L. Table 4 clearly shows the beneficial effect of the coagulant aid. It reduced the dose of the primary coagulant. The PACl dose of 20 mg/L was required to reduce the settled turbidity to 1.7 NTU without the coagulant aid for "A" water. The use of the coagulant aid reduced the PACl dose (10 mg/L) by half. The settled turbidity of 1.1 NTU (FL) or 1.9 NTU (WT) was obtained with use of the coagulant aid. The coagulant aid was also effective in reducing the residual Al concentration. The residual Al concentration was 0.25 mg/L without the coagulant aid, which exceeded the KDWS (0.2 mg/L). The addition of the coagulant aid successfully reduced the residual Al concentration below the KDWS. Such beneficial effects were also observed for "B" water. This water was difficult to coagulate due to the high turbidity. The minimum settled turbidity obtained by coagulation with PACl alone was 4.6 NTU. The corresponding concentration of the residual Al (0.45 mg/L) was more than twice the Korean Drinking Water Standards. The addition of FL successfully reduced the settled turbidity to 2.6 NTU with the half of the PACl dose (10 mg/L). The coagulant aid also enhanced the removal of the residual Al. The residual Al concentration was reduced from 0.45 mg/L to 0.09 mg/L.

Figure 3 compared the effectiveness of FL and WT in the improvement of the settled water qualities of "A" and "B" waters. These results were based on the settled turbidities of 1.7~2.4 NTU, which are the typical values encountered at the local water treatment plants. Figure 3 clearly shows that the polyamine-based coagulant aid (FL) was better than DADMAC (WT) in the improvement of the settled water qualities for both waters. The charge density might cause the difference. FL possessed higher positive charge than WT, which led to the better performance of FL. Unfortunately, the data of their charge densities are unavailable. The effect of the coagulant aid was also different with the source waters. The reason was not clear, but the more significant improvement was observed at the lake source ("A" water) than at the river source ("B" water). Since FL was found more effective, it was used as the coagulant aid in the following experiments.

Figure 4 shows the general behavior of a coagulant aid. These results are based on the

Table 4. Summary of the Results with the Use of the Coagulant Aid for different source waters

Description	PACl dose, mg/L	"A" water (average pH 7.23, average turbidity 7.4 NTU)		"B" water (average pH 6.94, average turbidity 184 NTU)	
		Turbidity, NTU	Al, mg/L	Turbidity, NTU	Al, mg/L
No polymer	10	4.5	0.32	5.9	0.49
	20	1.7	0.25	4.6	0.45
	30	-	-	5.2	0.48
FL	10	1.1	0.17	2.6	0.09
	20	0.9	0.17	2.6	0.12
	30	-	-	2.8	0.13
WT	10	1.9	0.18	3.2	0.28
	20	1.1	0.20	3.4	0.30
	30	-	-	3.7	0.33

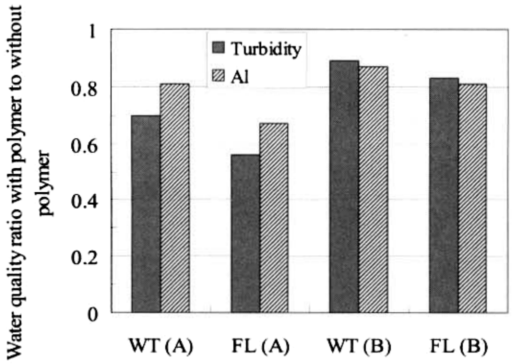

Figure 3. Comparison of FL and WT.

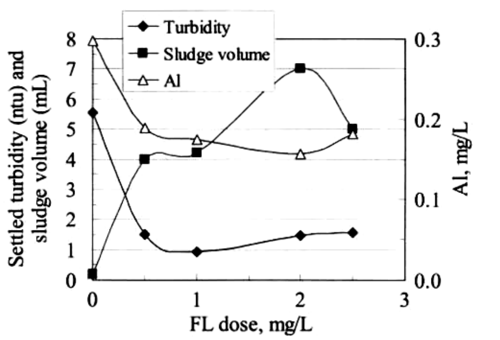

Figure 4. Effects of a coagulant aid on the settled water qualities for "A" water.

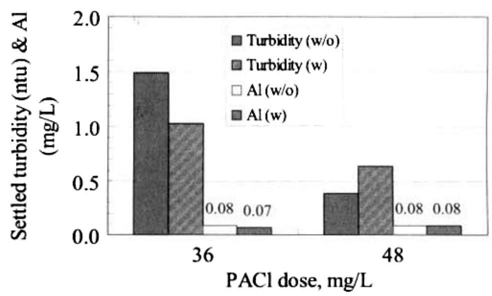

Figure 5. Limitation of the coagulant aid.

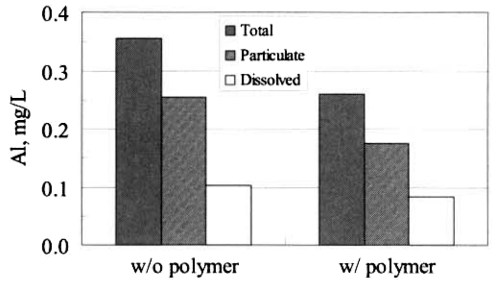

Figure 6. Removal of the particulate and dissolved Al by the use of the coagulant aid.

use of FL in combination with PACl for "A" water. The settled water qualities were monitored while the dose of FL was increased up to 2.5 mg/L at the PACl dose of 10 mg/L. Figure 4 shows that the use of a coagulant aid could improve the settled water quality. No coagulation was achieved with a 10 mg/L addition of PACl. The settled turbidity and the residual Al concentration were high with no sludge formed. The use of a coagulant aid then initiated the coagulation and improved the settled water qualities. The residual Al concentration was reduced along with the settled turbidity. The optimum dose existed. The overdose deteriorated the settled water qualities.

Figure 5 shows the limitation of the coagulant aid. Such beneficial effect was not observed at the optimum coagulation condition. When coagulation was incomplete as indicated by the high settled turbidity (1.5 NTU), the use of the coagulant aid was helpful in the improvement of the settled water qualities. However, the addition of the coagulant aid failed to improve the settled water qualities at the optimum coagulation condition.

The residual Al was then divided into the particulate and dissolved Al in order to identify how the use of the coagulant aid improved the removal of the residual Al. These results are based experiments with "A" water. The primary coagulant was dosed at 10 mg/L and FL was used as the coagulant aid. Figure 6 shows that the coagulant aid targeted both the particulate and dissolved Al, although the removal extent was different depending on the Al type.

The addition of an Al-based coagulant results in the formation of $Al(OH)_3$ floc, which is reflected by the particulate Al as well as turbidity. Not all of the added aluminum goes into the floc due to the solubility. Although most of the aluminum participates in the floc formation, some does not form the floc and remain in the solution. The soluble form of aluminum remaining in the solution is reflected by the dissolved aluminum. As mentioned above, the behavior of the dissolved Al reflects the coagulation efficacy, while that of the particulate Al reflects the efficacy of the solid-liquid separation. The improved removal of the dissolved and particulate Al by the coagulant aid indicates that the coagulant aid helped both the coagulation and the solid-liquid separation. Adding the coagulant aid is like adding more primary coagulant. Therefore, the addition of a polymer brought the coagulation to the optimum condition, which led to the enhanced removal of the turbidity and the dissolved Al concentration. The optimum coagulation condition also helped the small floc to settle. Ideally, all of the floc should settle by gravity. However, only the large flocs settle and small flocs remain in the solution in the practical situation. The use of the coagulant aid helped these small flocs to settle better probably by reducing their surface charges.

Flocculant Aid

The effectiveness of the various products of flocculant aid was compared in Figure 7. These results are based on experiments with "A" water. The primary coagulant of PACl was dosed at 10 mg/L, while various flocculant aids were tested. All products were effective in enhancing the turbidity removal, but their effectiveness on the reduction of the residual Al was minimal. According to Figure 7, the cationic polymers were more effective than the anionic or the nonionic polymers and SC-020 (cationic polymer with weak charge density and the medium molecular weight) was the most effective. The molecular weight became important after the charge type. The polymer with high molecular weight performed better than that with low molecular weight. As mentioned previously, a flocculant aid functions by bridging the neutralized particles into large flocs. It seemed that the particles were still negatively charged after the neutralization, which made the cationic polymer in an advantageous position for the attachment. The polymer with higher molecular weight made the particles to settle better.

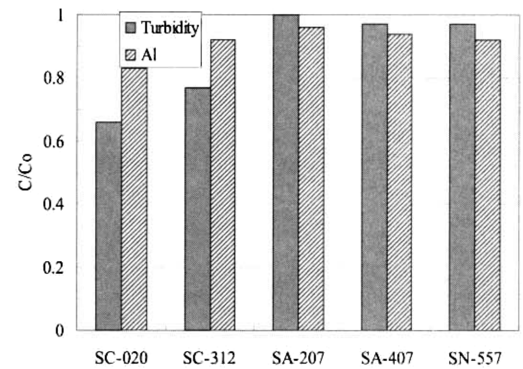

Figure 7. Comparison among various flocculant aids.

Since SC-020 was found the most effective, it was used in the following experiments.

Figure 8 shows the general behavior of a flocculant aid. These results are based on the use of SC-020 for "A" water, which had an average pH of 7.15 and an average turbidity of 4.9 NTU. The dose of SC-020 was increased up to 2 mg/L while the PACl dose was fixed at 10 mg/L. Figure 8 shows that the use of a flocculant aid improved the settled turbidity. The addition of SC-020 reduced the settled turbidity from 3.2 NTU to 1.8 NTU. An optimum dose existed for a flocculant aid as with a coagulant aid. The settled water turbidity did not improve any more after the optimum dose. The use of a flocculant aid was not effective in the reduction of the residual Al, unlike the settled turbidity.

Table 5 shows the effect of a flocculant aid, depending on the water characteristics. Three different characteristics were used; lowly turbid, medium turbid, and highly turbid. Jar tests were

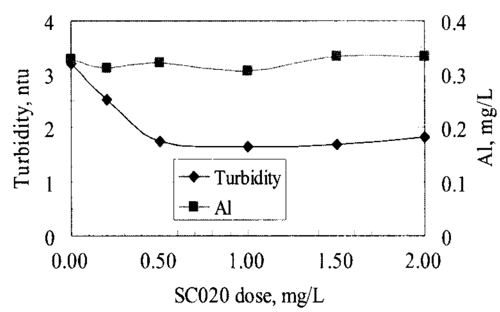

Figure 8. Effects of a flocculant aid on the settled water qualities for "A" water.

Table 5. Summary of the Results with the Use of the Flocculant Aid for "B" water

Description	W/O polymer*		W/polymer**	
	Turbidity, NTU	Al, mg/L	Turbidity, NTU	Al, mg/L
Lowly turbid (Turbidity 8.4 NTU, pH 8.40)	2.0	0.09	1.3	0.10
Medium turbid (Turbidity 27 NTU, pH 6.47)	1.6	0.27	0.8	0.24
Highly turbid (Turbidity 128 NTU, pH 7.90)	5.0	0.35	0.6	0.30

*Without polymer; **With polymer

conducted for each water. The optimum results are summarized in Table 5. Table 5 shows that the use of a flocculant aid improved the settled water qualities, regardless of the water characteristics. However, the improvement extent was different with the water characteristics. The more turbid the water was, the more effective the flocculant aid was. When the raw water was lowly turbid, the flocculant aid reduced the settled turbidity from 2.0 NTU to 1.3 NTU. However, the flocculant aid reduced the settled turbidity of the highly turbid water from 5.0 NTU to 0.6 NTU. Compared to the turbidity, the role of the flocculant aid in the improvement of the residual Al was minimal.

The residual Al was divided into the particulate and dissolved Al in order to identify the portion the flocculant aid preferentially removed. Since the flocculant aid functions by bridging the neutralized particles into a large floc, it was speculated that the flocculant aid would preferentially remove the particulate Al. The speculation was confirmed. Although the result was not shown, the most removal occurred through the removal of the particulate Al.

Filtration Aid

The effect of a filtration aid is shown in Figure 9. It was interesting to note that filtration removed both the particulate and dissolved Al. Since filtration as a good solid-liquid separation process is an effective particle polisher, it was originally expected to remove the particulate Al preferentially. However, the results showed that filtration was also effective in the removal of the dissolved Al. This indicates that post-coagulation might take place during filtration. The dissolved Al might participate in the coagulation or be adsorbed onto the filter media.

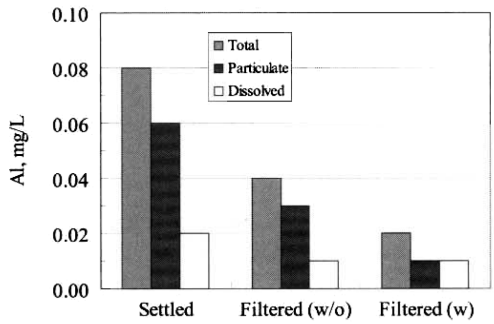

Figure 9. Removal of the particulate and dissolved Al by a filtration aid.

As expected, the filtration aid preferentially removed the particulate Al as the flocculant aid, but the extent of the effect was different. The effect of the filtration aid was more significant, compared to that of the flocculant aid. Although both targeted the same portion of the residual Al (the particulate Al), the filtration aid was substantially more effective in its removal. According to Figure 9, the filtration aid reduced the residual Al by half. On the other hand, the flocculant aid removed only the minimal portion of the residual Al.

Dissolved and Particulate Al

Since the $Al(OH)_3$ flocs formed during the coagulation can be measured by turbidity and the particulate Al concentration, it was speculated that there would be a relationship between the particulate Al concentration and turbidity. This speculation was checked by a linear regression analysis (Figure 10). The regression

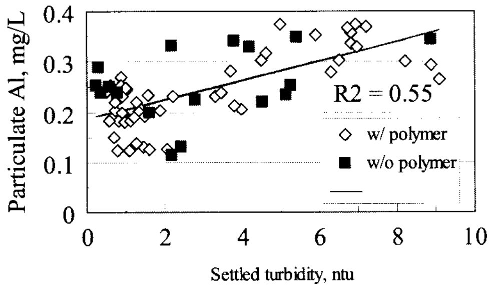

Figure 10. Relationship between the particulate Al concentration and turbidity.

analysis confirmed the relationship between the particulate Al concentration and turbidity with correlation coefficients of 0.55 (R^2). Such a relationship implies that a reduction in the particulate Al concentration can be achieved by improving the turbidity removal efficiency. That was how the polymers used as a coagulant, flocculant, and filtration aids improved the removal of the residual Al. They enhanced the turbidity removal, which then led to an improved removal of the residual Al.

CONCLUSIONS

This study found that the high dose of an Al-based coagulant did not necessarily result in the high concentration of the residual Al in the treated water. Instead, the efficacy of the coagulation and sedimentation was found important to determine the residual Al concentration. The minimum concentration of the residual Al was obtained at the optimum coagulant dose in terms of turbidity removal. This indicates the importance of the coagulation. It is essential to practice the coagulation at the optimum condition to minimize the residual Al concentration.

Regardless of the purpose, the use of a polymer enhanced the removal of turbidity as well as the residual Al. The coagulant aid helped both the coagulation and the solid-liquid separation and subsequently removed the dissolved Al as well as the particulate Al. The flocculant aid and the filtration aid helped the solid-liquid separation, and they preferentially removed the particulate Al. Although both targeted the same portion of the residual Al, their efficacies were different. The filtration aid reduced the residual Al more effectively than the flocculant aid. Therefore, it is recommended to use a filtration aid in reducing the residual Al.

Both the coagulant and flocculant aids reduced the dose of the primary coagulant, while improving the settled turbidity. However, the extent of the improvement was different with the raw water characteristics. When the raw water was difficult to coagulate due to high turbidity resulting from heavy rain, the flocculant aid improved the settled turbidity more effectively than the coagulant aid.

The polyamine product (FL) was found a better coagulant aid than DADMAC (WT) in reducing the residual Al as well as turbidity. The cationic polymer of SC-020 with weak charge density and the medium molecular weight was found the best flocculant aid in improving the settled water qualities.

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