

Improvement in the coagulation performance by combining Al and Fe coagulants in water purification

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Abstract—This study evaluated the beneficial effects on the coagulation process of combining coagulants with Fe and Al in the removal of turbidity and DOC (dissolved organic carbon), and proposed conditions for proper operation of a coagulation process. In addition, the floc characteristics and sludge dewater ability were evaluated. Blended coagulants were more effective than single coagulants at lower concentrations in water purification. The optimal blended ratio for the removal of DOC was 1.45 mM as Al/mM as Fe. Also, blended coagulants were less affected than single coagulants by pH, temperature, and rapid mixing intensity. Based on the results of the change in the apparent molecular weight distribution (AMWD) of DOC in raw and treated water by coagulation, low molecular weight organic matters were removed by 40.7% for the blended coagulant (1.45 mM as Al/mM as Fe). Blended coagulants formed larger flocs than single coagulants did. At pH 6.0, floc strength and sludge dewaterability were both improved.

Key words: DOC, Turbidity, Coagulation, Water Treatment, Floc, Blended Coagulant

INTRODUCTION

Surface water is the primary source of water supply in Korea. However, the quality of the surface water is usually deteriorated due to various factors, such as natural and artificial pollutants [1]. Thus, it is necessary to identify an effective method for eliminating such pollutants. In particular, untreated organics in the water treatment contribute to the development of THMs (trihalomethanes) that are known as carcinogens and possibly promote bacterial regrowth in the distribution system [2-5].

The removal of turbidities during conventional water treatment has been studied in depth. While the removal of turbidity is still considered critical, the removal of organics has become increasingly important in the management of residuals after water treatment. Methods to remove organic matters during water treatment include ozone oxidation, GAC (granular activated carbon), and BAC (biological activated carbon), that are known as advanced water treatment [6,7]. However, optimization and efficiency improvements in the conventional water treatment process are required to support the establishment of additional facilities and to hold down relatively high operation costs and introduce new processes.

The natural organic matter (NOM) in natural water significantly influences the behavior of treatment processes such as oxidation, coagulation and the use of disinfectants. NOM in raw water is considered as the precursor of DBPs (disinfection by products), which results when a disinfectant is applied in a water treatment [8]. Conventional coagulation strategies which have been employed for turbidity removal should be re-evaluated to meet the required water standard for DBPs. Rates of ultraviolet (UV) absorbance and DOC

could be used as surrogate parameters to understand DBPs formation. The level of total organic carbon (TOC) was a good indicator to estimate the concentration of THMs [9]. A good correlation was found between THM formation potential (THMFP) and UV absorbance at 254 nm in raw water [10]. The greater level of UV absorbencies reflects a higher aromatic content and greater molecular size [11].

The efficiency of coagulation is influenced by certain factors, such as pH, temperature, alkalinity, coagulant type, and mixing intensity [12-17]. Many researchers have recommended a pH adjustment method to enhance the coagulation efficiency [18-21]. Temperature is also a critical factor in the removal of turbidity [21]. Dharmappa et al. focused on rapid mix parameters as a critical parameter for optimizing the coagulation process [22]. Specific values were suggested for the optimal detention time and intensity of rapid mix by Mhaisalkar et al. [15]. Sludge dewater ability is known as one economic factor in water treatment that affects the condition of pH, temperature, viscosity and so on. The physical characteristics related to coagulation processes include floc intensity, density, floc size and shape, and so on. It is used as a microscopic method, Coulter counter, and laser diffraction techniques for measuring floc size [23-26].

Coagulation for removal of DOC and turbidity has been reported to be improved when the coagulant is made with a one to one blend of alum and ferric chloride compared to a single coagulant. Morris and Knocke reported that ferric chloride was less influenced at a low temperature compared with alum, and they suggest the combination of alum and ferric chloride as a potential method [14]. However, Johnson and Amirtharajah reported the coagulation performance was not evidently enhanced by the combination of alum and ferric chloride [27].

The main objective of this research was to identify the effect on coagulation of a combination of ferric chloride and poly aluminum

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chloride (PAC). This work investigated physical and chemical factors, such as pH, temperature, and G value, that affect coagulation performance in the elimination of turbidity and DOC, and proposed proper operational conditions for the application of the blend coagulant. The obtained results have been compared to those of single applied coagulants.

MATERIALS AND METHODS

1. Sample Preparation

The sample used in this study was collected by using a grab sampling method at a point in the Han River located 150 m from Young-dong bridge. The mean level of DOC and turbidity was 3.45 mg/L and 5.32 NTU, respectively. The fraction of molecular weight below 1,000 daltons was 51% while the fraction of high molecular weight over 30,000 daltons was 13%. It is well known that it is difficult to remove the range over 30,000 daltons by using a coagulation process [28]. The specific ultraviolet light absorbance (SUVA) value was 0.9.

2. Experimental Procedure

Analytical grade PAC (poly aluminum chloride, Backkwang Chemical) and FeCl_3 (ferric chloride, Wako Chemical) were selected as single coagulants in this study. Blended coagulants were prepared with a mixture of PAC and Ferric chloride in the ratios of 1.45, 0.48, and 0.16 mM Al/mM Fe and labeled as the blend coagulant I, II, and III, respectively.

The experiment was performed in a Jar tester (Model 1416, Dongyang Scientific) with rectangular impellers (length 8 cm, width 2 cm). The Jar was fabricated by using acryloyl and with the following dimensions: showed an 11.5 cm of length, 11.5 cm of width, and 21 cm of height. The volume of the jar was 2 L. A Jar test was performed as follows: rapid mixing at 120 rpm for 1 min, flocculation at 20 rpm for 15 min and settling of 30 min. Supernatant samples were taken at the end of process and were tested for DOC, UV254, and turbidity.

3. Analytical Method

An ultrafiltration fraction system (Model 8200, Amicon) consisting of 200 mL UF cell and clamp was used to analyze the molecular weight distribution of organics in the sample. Membranes applied in this system were manufactured by using cellulose acetate with a diameter of 62 mm. This study defined molecular weight under 1 K as low molecular weight (LMW), between 1 K and 10 K as medium molecular weight (MMW), and over 10 K as high molecular weight (HMW). Samples were supplied to membranes after applying a filtration process by 0.45 μm membranes and the system was pressurized to 55 psi with pure nitrogen gas. It was agitated by using a magnetic stirrer to reduce concentration polarization. Samples were taken in vials pretreated with acid and heat. The first 10 mL sample was discarded to avoid concentration differences due to membrane washing.

A CST (capillary suction time) system consists of a sludge container with a diameter of 1.8 cm of diameter and a height of 2.5 cm and timer. Moisture in the sludge is absorbed into filter paper (Whatman No. 17) by capillarity and its spreading velocity is measured. The sludge was concentrated for 2 hr at 20 °C and 7 mL of sludge was taken for measuring CST [29].

DOC was analyzed with a TOC analyzer (TOC 5000, Shimadzu).

Samples were filtered by using a 0.45 μm pore size filter before DOC measurement. Also, 1 N HCl was added in the sample and then it was measured. The pH was analyzed by a Corning pH probe and meter (Model 250). The alkalinity was measured by a standard method of 2320 [30]. The value of UV254 was analyzed with a UV visible spectrophotometer (UV1601, Shimadzu) after filtering the sample through a 0.45 μm pore size filter. Turbidity was measured with a HACH 2100A. A microscope (Axioplan 2 imaging, Carl Zeiss) was used to observe the size and shape of floc.

RESULTS AND DISCUSSION

1. Comparison of Coagulants Efficiency

Fig. 1 illustrates the variation in DOC and turbidity using PAC and ferric chloride with pH 7.2 at 20 °C. The concentration of DOC and turbidity decreased continuously until the dosage of 10 and 7.5 mg/L for the single ferric chloride and PAC, respectively. The highest removal efficiency in DOC was 35.2% and 36.3% while the ferric chloride and PAC were dosed at 10 and 7.5 mg/L. The highest efficiency in turbidity was shown as 77.0% and 88.8% when 7.5 mg/L of ferric chloride and 5 mg/L of PAC were dosed, respectively. The highest removal efficiency in UV254 was 41.8% and 43.8% when 7.5 mg/L of ferric chloride and 5 mg/L of PAC were dosed, respectively.

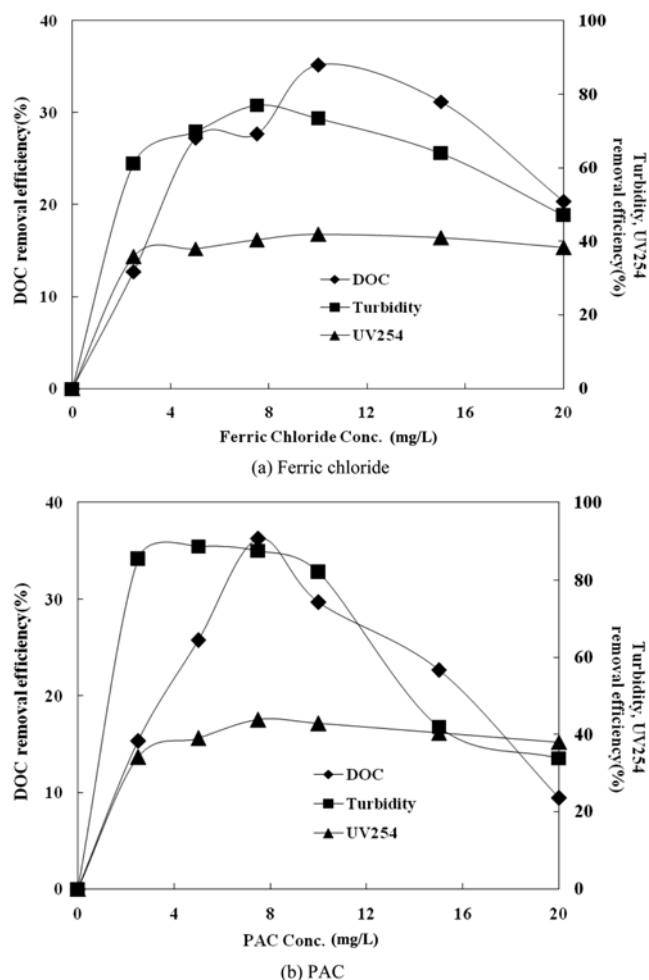


Fig. 1. Removal of DOC, turbidity, and UV254 using ferric chloride and PAC as a function of coagulant concentration.

when the ferric chloride and PAC were dosed at 10 and 7.5 mg/L.

It was observed that the PAC removed DOC and turbidity more efficiently than the ferric chloride. This agrees with the findings of Furrey et al. [24]. It was clear the optimum coagulant dosage for removing DOC required a higher concentration level compared to that of the turbidity.

The highest levels for DOC removal efficiency were found to be 43.6, 41.2, and 38.9% for the blend coagulant I, II, and III (1.45, 0.48, 0.16 mM Al/mM Fe, respectively) at 5 mg/L (Fig. 2). The levels for removal efficiency for UV254 were found to be 53.7, 50.0,

and 46.2% for the blend coagulant I, II, and III at 5 mg/L. The highest levels for turbidity removal efficiency were 91.4%, 90.8%, and 89.5% for the blend coagulant I, II, and III when the dosage was 2.5 mg/L, significantly lower than that of the DOC. Blend coagulant I was superior in the removal of DOC and turbidity in the range from 2.4% to 4.7% compared to blend coagulant II and III. This means that the removal performance of the blend coagulants was elevated when the ratio of Al to Fe was high. It was apparent that the coagulation efficiency was enhanced due to the use of combined coagulants with PAC and ferric chloride. The removal efficiency increased in the range from 7.3% to 8.4% in DOC and 2.6% to 14.4% in turbidity compared to the use of PAC or ferric chloride only.

It should be noted that the optimum dosage of blend coagulants was found to be approximately half that of the single ferric chloride, which shows additional advantages for using blend coagulants with Al and Fe. In other words, it might be possible to reduce the amount of sludge after completing coagulation and sedimentation processes by using the blend coagulants.

2. The Variation of Molecular weight Distribution by Coagulation

This study evaluated the variation in molecular weight distribution with ferric chloride, PAC, and blend coagulants in the removal of DOC at two pH conditions, 6 and 7.5 (Table 1 and 2). The pH in the sample was controlled by a 0.1 N sulfuric acid and caustic soda solution. Coagulants were injected as 10, 7.5, and 5 mg/L of ferric chloride, PAC, and blended coagulant, respectively, and the optimum coagulant concentration for removing DOC is presented in Fig. 1 and 2. Samples with a pH of 6 removed DOC more efficiently, from 3 to 10% more than a pH of 7.5.

28.6% and 15.1% of the LMW portion (under 1 K) of DOC was eliminated by applying coagulants with PAC and ferric chloride at

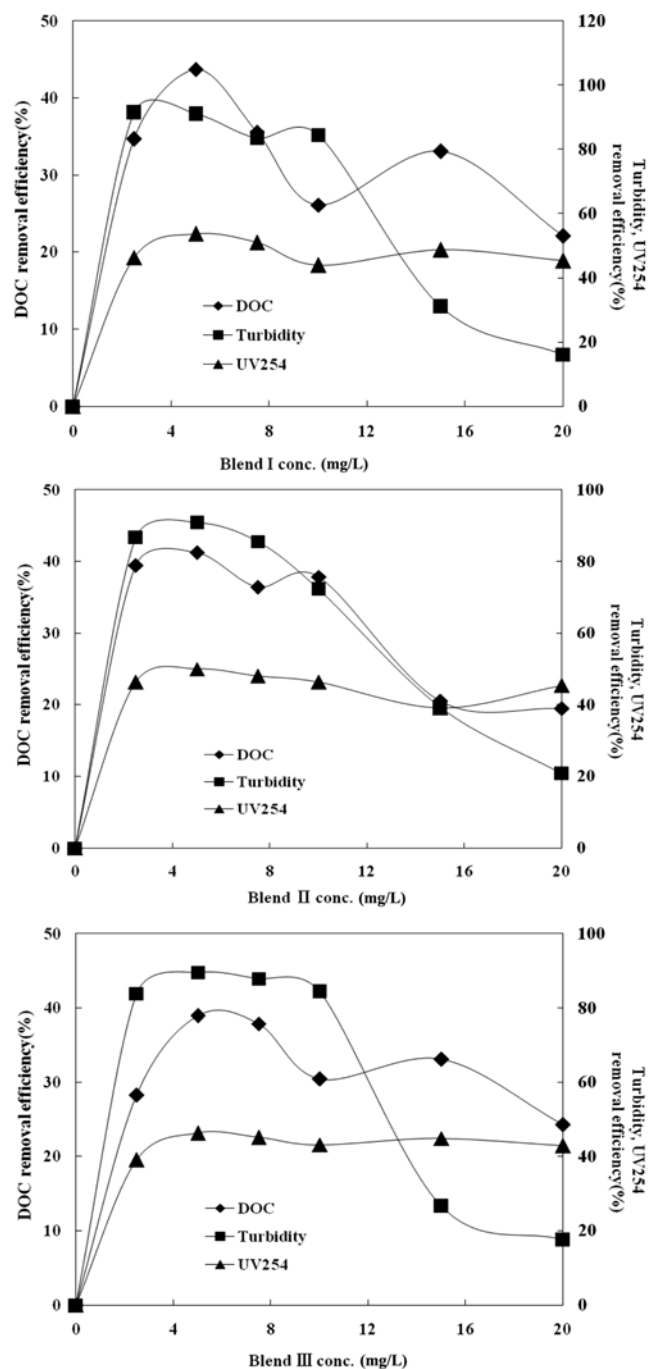


Fig. 2. Removal of DOC, turbidity, and UV254 using blend I, II, and III as a function of coagulant concentration.

Table 1. DOC removal efficiency based on the molecular weight distribution with the coagulation at pH 7.5 (unit: %)

MW fractions (Daltons)	FeCl ₃	PAC	Blend I	Blend II	Blend III
<0.5 K	0.78	24.0	38.8	34.1	31.0
0.5-1 K	41.43	37.1	40.0	17.1	17.1
1-3 K	52.3	38.4	55.8	44.2	24.4
3-10 K	29.1	58.2	29.1	61.8	72.7
10-30 K	64.7	47.1	76.5	76.5	82.3
>30 K	78.1	31.3	78.1	78.1	90.6

Table 2. DOC removal efficiency based on the molecular weight distribution with the coagulation at pH 6 (unit: %)

MW fractions (Daltons)	FeCl ₃	PAC	Blend I	Blend II	Blend III
<0.5 K	3.88	27.1	40.3	34.9	33.3
0.5-1 K	42.9	42.9	41.4	20.0	18.6
1-3 K	53.5	50.0	57.0	46.5	31.4
3-10 K	61.8	63.6	32.7	63.6	78.2
10-30 K	64.7	52.9	76.5	82.4	82.4
>30 K	81.3	37.5	81.2	84.4	90.6

Table 3. Variation in DOC, turbidity according to the pH and temperature

(unit: %)

Coagulants	Temp.	pH 5		pH 6		pH 7		pH 8	
		DOC	Turbidity	DOC	Turbidity	DOC	Turbidity	DOC	Turbidity
Ferric chloride	5 °C	28.9	66.7	31.3	71.9	33.1	74.7	32.6	73.7
	20 °C	36.4	77.7	38.5	82.7	37.0	78.6	35.7	76.5
	35 °C	37.7	80.3	39.8	83.2	38.5	81.4	37.0	77.8
PAC	5 °C	28.9	79.7	30.5	82.9	33.1	83.8	32.0	83.2
	20 °C	35.9	86.6	38.5	89.4	37.7	87.7	35.7	85.8
	35 °C	38.2	88.3	40.1	90.1	38.8	89.0	37.7	86.4
Blend	5 °C	39.5	85.5	41.9	87.0	43.2	88.3	40.1	87.7
	20 °C	44.7	89.4	47.0	91.1	45.5	90.3	44.7	88.8
	35 °C	48.1	90.5	49.6	91.6	47.8	90.9	47.5	89.2

the condition of pH 7.5. 43.3% and 46.1% of the MMW (between 1 K and 10 K) in DOC was removed with ferric chloride and PAC respectively. In the range of the HMW (over 10 K) 73.5% and 36.7% was removed with ferric chloride and PAC, respectively.

The DOC removal in the LMW range was found to be as 17.6% and 32.7% for ferric chloride and PAC at pH 6. In the MMW range, DOC removal was found to be 56.7 and 55.3% and in the HMW range, 75.5 and 42.9% at pH 6. The removal efficiency in the range of the MMW increased from 9.2% to 13.1% and HMW increased from 2% to 6.2% when the value was held at pH 6 instead of pH 7.5.

The DOC removal efficiency was 46.3%, 42.7%, and 40.1% for the blend coagulant I, II, and III, respectively, at pH 7.5. Also, it was 48.1%, 45.0%, and 43.4%, respectively, at pH 6 with a high level of 2.3% and 3.3% compared to pH 7.5. It was 33.7%, 28.1%, and 26.1%, respectively, for the blend coagulant I, II, and III in the LMW. Also, it was 45.4%, 51.1%, and 43.3%, respectively, for the MMW and was 77.6%, 77.6%, and 87.8%, respectively, for the HMW at pH 7.5. It was found to be 40.7%, 29.6%, and 28.1%, respectively, for the blend coagulant I, II, and III for the LMW. At pH 6, it was 47.5%, 53.2%, and 43.3%, respectively, for the MMW and was 79.6%, 83.7%, and 87.8%, respectively, for the HMW. The removal efficiency at pH 6 showed higher values of 7%, 2.1%, and 6.1% than that at pH 7.5. It was clearly observed that the portion of HMW was significantly removed when blended coagulants were applied.

The highest elimination rate in the LMW range was 40%. However, for the MMW and HMW ranges, the highest elimination rate was about 80% during the coagulation. These results showed a consistency with the report of Amy et al. [31] who showed that the removal was achieved by applying coagulants for the materials in the range of 5 K to 10 K, but that it was not easy to eliminate the matter in a range of less than 1 K.

The DOC removal efficiency for the blend coagulant I (1.45 mM Al/mM Fe) was recorded as 40.7% at pH 6, while the highest removal efficiency was as 32.7% in the application of single ferric chloride and PAC at pH 6 in the LMW range. The DOC elimination rate at pH 6 was greater than at pH 7.5 with the use of blended coagulants for the LMW and MMW ranges. The HMW range showed 77.6% and 87.8%, while the removal efficiency of DOC in the scope of LMW decreased when blend and coagulants were used.

3. Effect of pH and Temperature on Removal of DOC and Turbidity by Coagulation

The variation of residual DOC, turbidity with the influence of pH and temperature when ferric chloride, PAC, and blend coagulant I were applied at the optimum concentration determined by the previous experiment is given in Table 3. The removal of turbidity evidently decreased at 5 °C compared to other temperature conditions. The floc size was small when it was observed with the naked eye at 5 °C, and the formation of floc was difficult to identify during the process of slow agitation. This was consistent with the report of Morris and Knocke [1984] that showed a decrease in the turbidity removal and smaller flocs under low temperature with alum as a coagulant. It was observed that ferric chloride was less sensitive in the changes in temperature than was PAC. The DOC and turbidity removal efficiency decreased according to the decrease in temperatures. At pH 7, the removal efficiency at low temperature was better than that at pH 6 due to the increase in the solubility of metals [32].

The DOC removal efficiency was 33.1%, 33.0%, and 43.0% for ferric chloride, PAC, and blend coagulant I, respectively, at 5 °C with pH 7. It was 38.4%, 38.4%, and 47.0%, respectively, at 20 °C with pH 6. Also, it was 39.7%, 40.1%, and 49.7%, respectively, at 35 °C with pH 6. These results showed the DOC removal efficiency for blend coagulant was better than for a single coagulant at all water temperature. So, it might be possible that the decreased coagulation efficiency can be complemented by using the blend coagulant with Al and Fe in winter time.

The turbidity elimination rate was 74.0%, 83.8%, and 88.3% for ferric chloride, PAC, and blend coagulant I, respectively, at 5 °C and pH 7. Also, it was 82.7%, 89.3%, and 91.1%, respectively, at 20 °C and pH 6 and 83.2%, 90.1%, and 91.6% for ferric chloride, PAC, and blend coagulant I, respectively, at 35 °C and pH 6.

The floc size and type in various coagulants were observed in this study. Fig. 3 shows an image of flocs with coagulants. The floc size and shape possibly affected the treated final water quality, the efficiency of succession processes, such as sedimentation process and filtration process, and the dewater ability of sludge. The size of PAC was comparatively small. The floc sizes associated with the selected coagulant in this study were 76.5 µm for blend coagulant III, 61.5 µm for blend II, 60.7 µm blend I, 54.8 µm for ferric chloride and 50.8 µm for PAC. It appeared that the floc size in blend coagu-

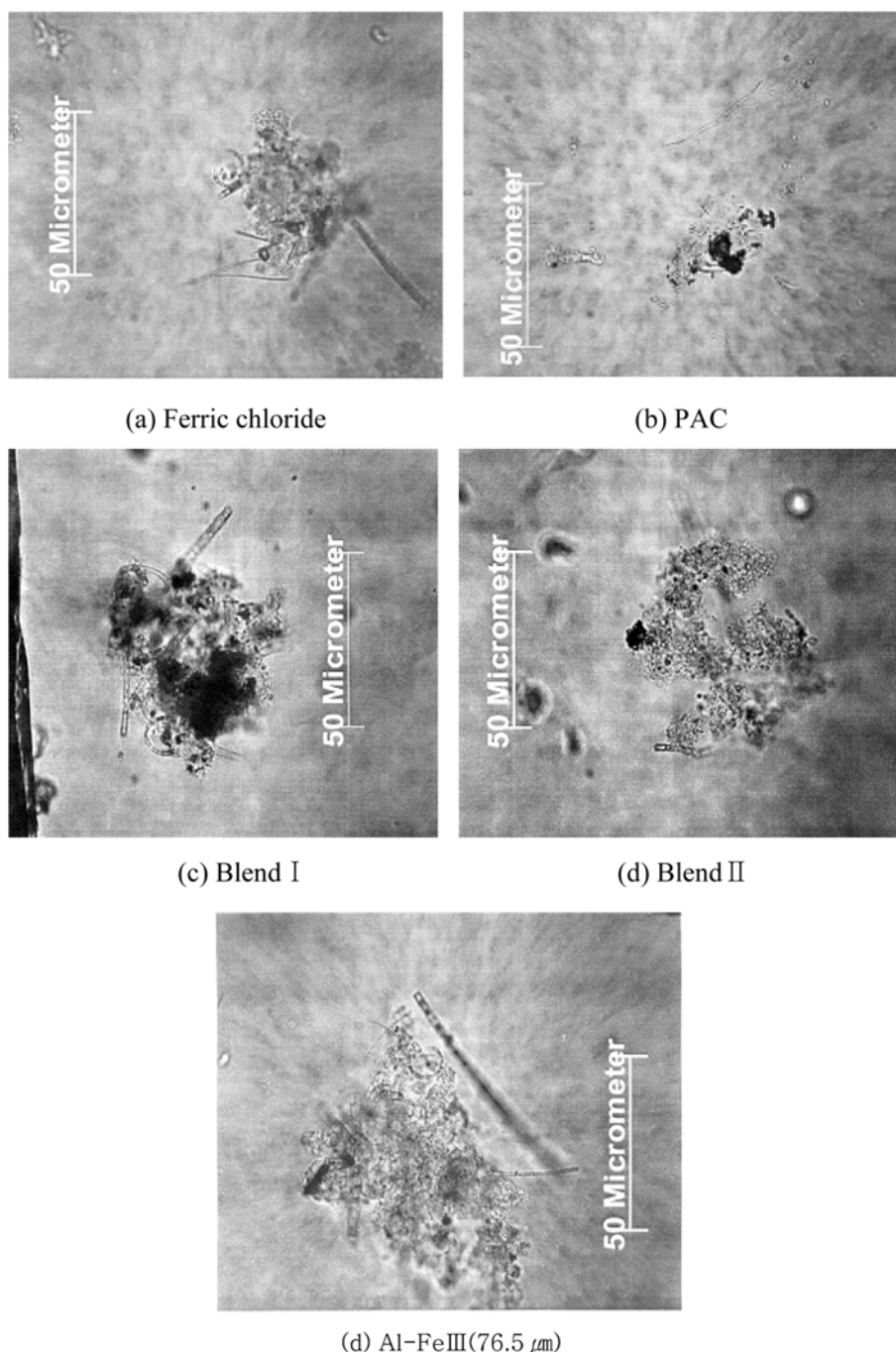


Fig. 3. Floc structure and size.

lants was bigger than that in single coagulants.

4. Comparison of the Optimal G Value and Break-up Intensity of Floc

This experiment investigated the removal efficiency in turbidity, DOC, and UV254 on G (velocity gradient) value and identified the suitable range for G values for each coagulant. Fig. 4 illustrates the variation in DOC, turbidity, and UV254 at different G values. The G value was changed between 300 and 1,300 sec^{-1} for the rapid mixing and controlled at 17 sec^{-1} for the slow mixing applied in this study. The level of DOC decreased drastically over 500 sec^{-1} . However, it increased again over 900 sec^{-1} . The highest DOC removal

in FeCl_3 , PAC, blend coagulant I, blend coagulant II, and blend coagulant III was 36.1, 40.3, 46.6, 42.5, and 39.9%, respectively, at G values between 700 and 900 sec^{-1} . The optimum G values for DOC removal appeared to be 900 sec^{-1} for the PAC, 700 sec^{-1} for the ferric chloride and blend III and 900 sec^{-1} for blends I and II.

The optimal mixing intensity to eliminate the turbidity was 700 sec^{-1} . The best removal efficiency in turbidity at 700 sec^{-1} was 82.8%, 90.8%, 94.8%, 93.2%, and 92.3% for the ferric chloride, PAC, blend coagulant I, blend coagulant II, and blend coagulant III. The optimum G ranges for UV254 were between 700 and 900 sec^{-1} similar to the result for DOC. The optimal removal of UV absorbance was

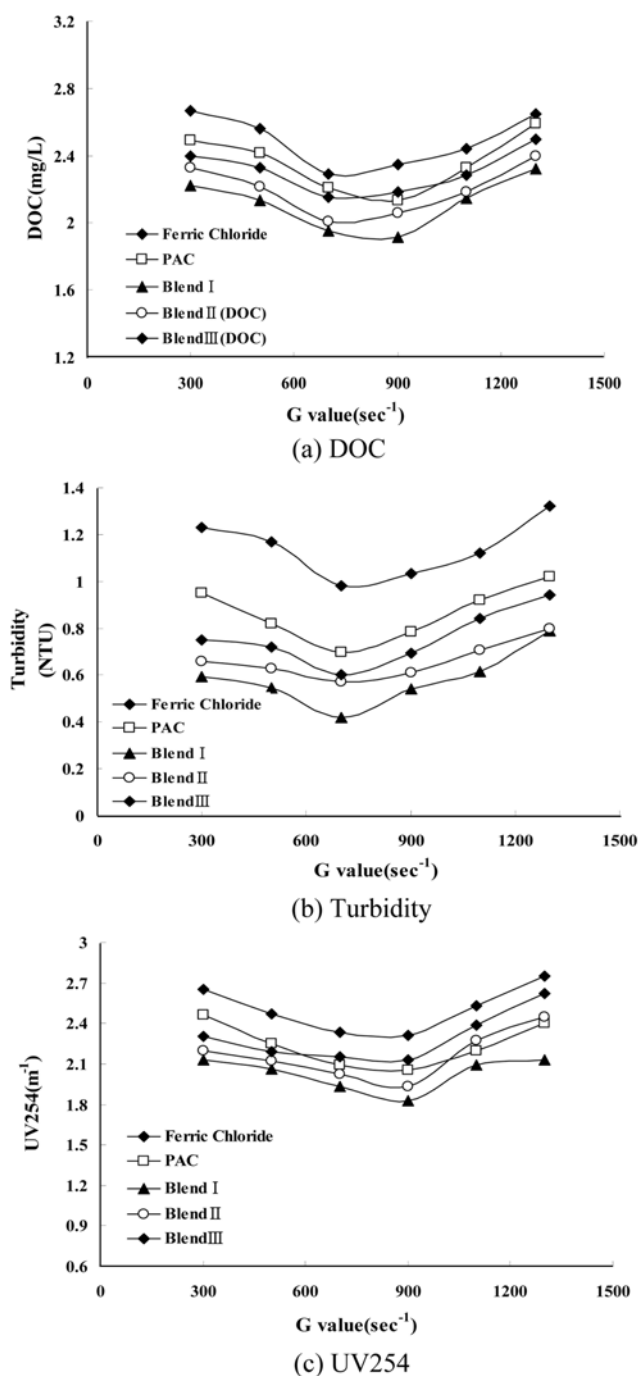


Fig. 4. Effect of the rapid mixing velocity gradient on the DOC, turbidity, and UV254 removal efficiency.

observed at the G value of 900 sec^{-1} . The value of UV254 increased over 900 sec^{-1} . The best removal rates of UV254 were 54.7%, 55.8%, 64.1%, 62.2%, and 58.2% for the ferric chloride, PAC, blend I, blend II, and blend III. Also, it was observed that blend I showed the fairest efficiency in DOC and UV254 among selected coagulants at the optimal G condition. In addition, the optimal G values of DOC and UV254 were higher than that of the turbidity for rapid mix.

Fig. 5 illustrates the intensity of the floc break-up according to the coagulants selected in this study. The floc formed by coagulants is usually weak and can be easily broken by stress. When flocs are

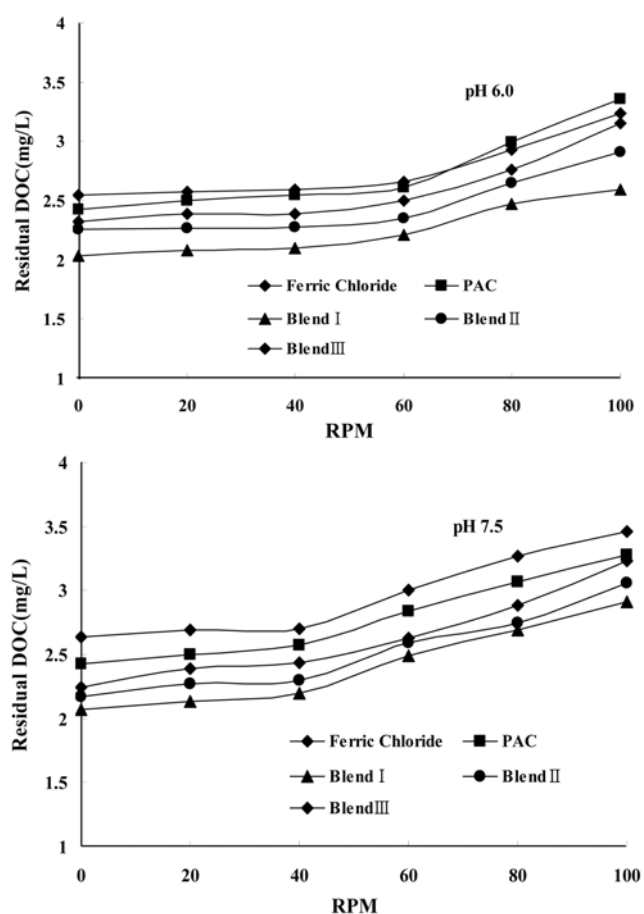


Fig. 5. Variation in DOC on the intensity of floc break-up at different pH values.

moved to the basin of sedimentation after flocculation, they can be easily broken by a type of fluid shearing force or bumps on the pipe wall. The flocs were broken at a mixing intensity between 20 and 100 rpm for 3 min after applying coagulation and sedimentation processes consecutively after which the supernatant was analyzed.

The levels in DOC continuously increased according to the increase in the agitation intensity. The organics seceded from floc might suspend in water. The residual DOC concentration rapidly increased over 60 rpm at pH 6 for all selected coagulants and significantly increased over 40 rpm at pH 7.5. Accordingly, it was thought that the intensity of the floc could be reinforced by controlling the condition of pH in the coagulation process applied in these results. The blended coagulant showed the higher floc break-up intensity compared to the use of single coagulants.

5. Comparison of the Sludge Dewater Ability

Fig. 6 shows the evaluation of the sludge dewater-ability on G value at different pH conditions. The sludge was taken after applying coagulation and sedimentation processes and was concentrated during 2 hr in which the CST was measured. The G values were controlled as 300, 700, and $1,100 \text{ sec}^{-1}$ to evaluate the effect of sludge dewater-ability on G values. The determination for the range of suitable mixing intensity affected not only the coagulation efficiency but the sludge dewater-ability.

The best dewater-ability was shown at the G value of 700 sec^{-1} .

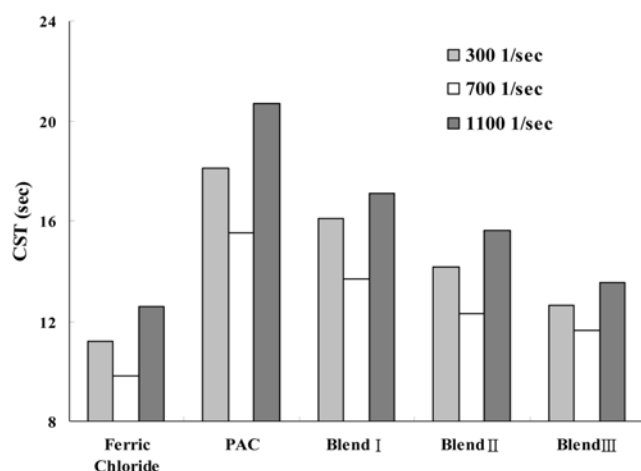


Fig. 6. Variation in CST on G values.

Also, the lowest dewater-ability was shown at the level between 13.58 sec and 26.68 sec at G 1,100 sec^{-1} . It is observed that the ability of settling decreased and pin flocs were formed at a high G value. The CST in ferric chloride was 11.07 sec, which represented the best dewater-ability in selected coagulants at the G value of 700 sec^{-1} . The CST of blend I, blend II, and blend III were 13.67, 12.27, and 11.64 sec, respectively, and the blended coagulants demonstrated better dewater-ability than the PAC. The higher order in CST was PAC, blend I, blend II, blend III, and ferric chloride. It was observed that dewater-ability was improved when the ratio of Fe increased in the blended coagulants.

Fig. 7 shows the CST value according to pH levels, such as 5, 6, and 7. The dewater-ability showed the lowest level and appeared in the range between 13.79 and 25.12 sec at pH 5. The CST in ferric chloride, PAC, blend I, blend II, and blend III was 9.24, 15.02, 13.51, 12.03, and 11.22 sec, respectively, at pH 6, and it showed a larger dewater-ability at pH 6 than at other pH conditions.

CONCLUSIONS

This study investigated the advantage of combining coagulant

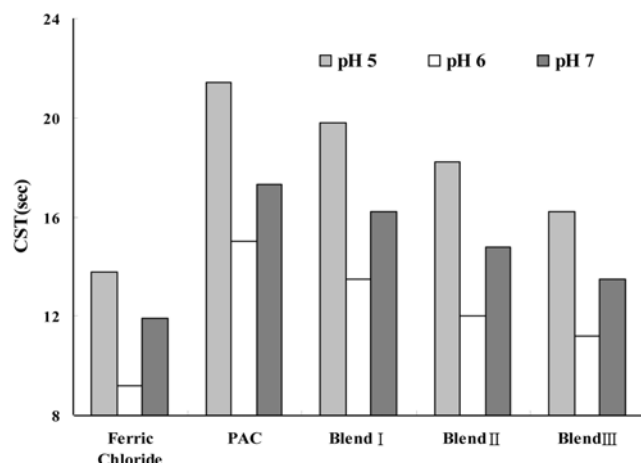


Fig. 7. Variation in CST on pH configuration.

with PAC and ferric chloride and attempted to optimize operational conditions for applying it in a coagulation process. To accomplish this goal, we evaluated the variation in DOC and turbidity to find affecting factors in physical chemical parameters, such as coagulation pH, mixing intensity, floc structural characteristic, and sludge dewatering-ability, and compared the coagulation efficiency of single coagulants (PAC, ferric chloride) with that of the combined coagulants with PAC and ferric chloride based on the dosage of 1.45, 0.48, and 0.16 mM as Al/mM as Fe.

The DOC and turbidity removal efficiency in such combined coagulants applied in this study was found to be 7.3-8.4% and 2.6-14%, higher than that of single coagulants. It appeared the optimum blend ratio was 1.45 mM as Al/mM as Fe when PAC and ferric chloride combined. 26.2% to 33.7% of the organic matter in the low molecular weigh distribution (less than 1,000 daltons) was removed by using the combined coagulant, although its removal by coagulation is usually considered difficult. The optimal G value of DOC and UV254 removal was higher compared with that of turbidity for the rapid mix and optimal G value ranged between 700 and 900 sec^{-1} .

The optimum dosage of combined coagulants was determined to be approximately half the dosage used for single ferric chloride to eliminate DOC. The coagulant dosage in DOC elimination needed a higher value than that of the turbidity. The dewater-ability was improved when the ratio of Fe increased in the blend coagulants. The intensity of floc break-up was enhanced 1.5 times when pH was controlled at pH 6 and low floc intensity for PAC was reinforced by combination with ferric chloride.

The decrease in the coagulation efficiency in DOC and turbidity appeared in a low temperature condition. The coagulation efficiency in blended coagulants was found to be better than that of single coagulants for all water temperature conditions. The floc size in blend coagulants was larger than that of single coagulants. The data obtained in this study indicated the coagulation efficiency could be enhanced by using the blend coagulant.

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