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Enhanced water quality of CSOs with different coagulant treatment

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ABSTRACT

The present-day construction projects on sewer management in Korea use both combined and separated sewer systems. Combined sewer overflows (CSOs) generated from combined sewer systems (CSS) have a large impact on the water bodies. Today, many researches to treat CSOs are being conducted. In this study, the jar test was conducted to determine the applicability of the coagulation–sedimentation process using chemicals to treat CSOs. Stormwater runoff (mixed with sewer) from a catchment area of 998 ha was used as raw water sample for the experiment. Alum, polyaluminum chloride (PAC), and $FeCl_3$ were used as coagulants and the optimum dose was determined. Moreover, the optimum dose of the weighted coagulant additives (WCA) and polymer coagulant were also determined. Based on the TSS removal efficiency, 30 mg/L, 20 mg/L and 38 mg/L of Alum, PAC and $FeCl_3$, respectively, were determined as the optimum dose. The optimum dosage of polymer coagulant was at 4 mg/L, 5 mg/L and 10 mg/L of Alum, PAC and $FeCl_3$, respectively. Proper coagulants, when injected along with WCA and polymer coagulant, were estimated to be 92.2% (alum), 92.7% (PAC), and 87.6% ($FeCl_3$) based on the TSS removal efficiency.

Keywords: Alum; Coagulation; CSOs; $FeCl_3$; PAC; Polymer coagulant; Weighted coagulant additives

1. Introduction

Pollutants accumulated on the urban surface during the dry weather are washed off by stormwater runoff generated during wet weather. Sewer management projects in Korea use both combined and separated sewer systems. Combined sewer overflows (CSOs) are stormwater runoff mixed with sewage that no flow into sewage treatment plant within the combined sewer system (CSS). These are directly discharged into water bodies [1]. In a separate sewer system (SSS), little pollution load is discharged into water bodies during wet weather because stormwater runoff and sewer flow through different pipes. CSS has a large impact on the water bodies because pollutants of sewage deposited during dry weather are discharged with stormwater runoff during wet weather [2,3]. According to the reports of U.S Environmental Protection Agency (EPA), CSOs contain various pollutants, such as organic wastes, bacteria, suspended solids, and so on [4].

On the other hand, in Korea, in order to control CSOs generated during wet weather conditions, the volume of the CSS is designed such that three times the proposed hourly maximum sewage flow occurs during the dry seasons. However, for a stable operation of the sewage treatment plant, the fraction of the inflow that exceeds the design capacity of the sewage treatment plant should be bypassed [1]. CSOs have a major influence on water bodies during wet weather. CSOs should be purified and discharged to manage the water quality of rivers. The research and effort to treat CSOs, therefore, are on demand [5]. In Europe, the processes using coagulation mechanism such as ACTIFLO and DENSADEG have been developed and used to rapidly treat a large amount of CSOs. ACTIFLO and DENSADEG are facilities that treat contaminants using 100–130 μm of micro-sand as Weighted coagulant additives (WCA) [6,7]. Meanwhile, the measures taken

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to remove CSOs in Korea include stormwater tank, swirl regulator, screen filter, stormwater infiltration, real-time control, and so on [8]. Furthermore, the ultra rapid coagulation (URC) process, as a part of the Coagulation–Sedimentation Process (CSP), has been used [9]. Many researchers have reported that CSP is suitable for the treatment of CSOs because a larger percentage of pollutants in CSOs account for the particulate matters [10–12].

This study has focused on determining the applicability of CSP in the treatment of stormwater runoff mixed with sewage discharged from the CSS during wet weather. To determine the optimum coagulant, aluminum sulfate (alum), polyaluminum chloride (PAC), and iron (III) chloride ($FeCl_3$) were used as chemical coagulants in CSP. Jar test was carried out to determine the proper dose of coagulants, WCA, and polymer coagulant.

2. Matrials and method

2.1. Raw water

A series of jar tests were carried out to treat CSOs generated during rainfall, where stormwater runoff mixed with sewage was used as raw water for the experiment. These CSOs were discharged from a catchment area of about 998 ha. The land use type ratios are composed of 52.6% of industrial complex, 32.5% of a residential area, 32.3% of forest area and 2.6% of farmland. And imperviousness ratio of catchment is 78.2%. Raw water was sampled at intervals of 30 min until rainfall ended, and flow weighted composite samples were prepared for the jar tests. These mixed samples were reused three times for analysis. The concentration range of raw water is presented in Table 1. The concentration ranges were measured as follows: 6.8–7.1 hydrogen exponent (pH), 68.3–75.6 NTU Turbidity, 218–7,266 mg/L total suspended solids (TSS), 128–143 mg/L biochemical oxygen demand (BOD), 199–212 mg/L chemical oxygen demand (COD_{Cr}), 42–49 mg/L total nitrogen (TN), and 9.8–11.2 mg/L total phosphorus (TP).

2.2. Jar test and coagulation

Collected raw water was filled in a 1-L beaker and the jar test was conducted. In each jar test, rapid mixing was performed for 3 min followed by a slow mixing of 15 min. After mixing, sedimentation time of 30 min was allowed and the supernatant was sampled to analyze the pollutants. Coagulants used for the jar test include alum (8%), $FeCl_3$(8%), and PAC (10%). Powdered glass, bentonite, and diatomite were used as WCAs and mixed in the ratio of 1:1:2. To increase accuracy of coagulant dose, it was diluted to 200,000 ppm and was used.

Effective grain size of WCAs is 100 μm. Anionic polyelectrolyte diluted to 10,000 ppm, hydrolyze acrylamide, was used as a polymer flocculating agent. 0.1 N H_2SO_4 and NaOH were prepared and injected to control the pH before the coagulant injection. Sufficient alkalinity was provided to all the experiments.

2.3. Analysis of the samples

The samples collected for the coagulant injection and CSP consist of the supernatant, which has been analyzed for water quality. Water quality parameters such as TSS, BOD, COD_{Cr}, TN, TP, total Kjeldahl nitrogen (TKN), phosphate (PO_4^{-3}), pH, and turbidity were analyzed at the laboratory in accordance with the standard methods [13].

2.4. Experimental steps

The experimental procedure followed four steps, which are listed as follows (Table 2). (1) Determine the pH

Table 1
Concentration of raw water

Parameter	pH	Turbidity (NTU)	TSS (mg/L)	BOD (mg/L)	CODCr (mg/L)	TN (mg/L)	NH3-N (mg/L)	TP (mg/L)	PO4-P (mg/L)
Range	6.8~7.1	68.3~75.6	218~266	128~143	199~212	42~49	27~37	9.8~11.2	1.6~2.2

Table 2
Experimental steps conducted in the jar test

Mode	Coagulant	Variable	pH	Coagulant dose (mg/L)	WAC (mg)	Polymer (mg/L)
Mode 1	Alum	pH	5.5~8.5	10	–	–
	PAC		5.5~8.5	10	–	–
	$FeCl_3$		4~7	10	–	–
Mode 2	Alum	Coagulant dose	6.5	12~44	–	–
	PAC		7	12~44	–	–
	$FeCl_3$		6.5	20~56	–	–
Mode 3	Alum	WAC	6.5	30	0.05~5	–
	PAC		7	20		–
	$FeCl_3$		6.5	36		–
Mode 4	Alum	Polymer	6.5	30	0.05~5	1~8
	PAC		7	20		2~9
	$FeCl_3$		6.5	36		4~12

for optimal coagulation reaction by the coagulant type. (2) Determine the optimal coagulant dose under the optimal pH range determined in the previous step. (3) Determine the optimal WCA dose under the conditions of the determined optimal pH and coagulant dose. (4) Determine the optimal polymer coagulant dose under the conditions of the estimated optimal pH, coagulants, and WCA dose. The approximate pH, coagulants, WCA, and polymer coagulant dose range were determined through preparatory experiments.

2.5. pH

Each coagulant has an optimal pH range for the coagulation reaction. Moreover, pH is the predominant variable for the occurrence of the cohesion reaction between the coagulants and the pollutants in raw water. It also has a direct effect on the residual aluminum (Al) concentration in the treatment of CSOs using CSP. This study tried to determine the optimal coagulant pH range for each coagulant type. Therefore, a preliminary research was conducted with a wide range of pH (3, 5, 7, and 9) and the results are shown in Fig. 1. Alum and PAC have the strongest cohesion at pH = 7, while $FeCl_3$ has the lowest turbidity at pH = 5.

The experiments to determine the optimum pH for each coagulant type, depending on the results of preliminary research, was conducted as indicated in the Step 1 of Table 2. An optimum pH of the alum coagulant was achieved by increasing the pH at intervals of 0.5 from 5.5 to 8.5. Another experiment using $FeCl_3$ as coagulant was performed with the pH being 4–7, escalated at intervals of 0.5.

2.6. Coagulant dose

Fig. 2 shows the results of preliminary research on coagulant dose. When alum and PAC were used as coagulants, they showed the lowest turbidity concentration at 20 mg/L. When PAC was used as a coagulant, the turbidity concentration of the treated water decreased, as dosing rate increased. However, when alum was used as a coagulant, the turbidity concentration of the treated water increased, as dosing rate increased. A very small difference was noticed between the $FeCl_3$ dose of 20 mg/L and above. Moreover, the experiment to determine the optimum $FeCl_3$ dose yielded the lowest turbidity concentration, that is, between 20 mg/L and 40 mg/L. It was also observed that the treated water turned yellow, and turbidity concentration increased as coagulant dose increased. On the basis of the results of the preceding studies, an experiment was performed using 12–44 mg/L of alum and PAC and 20–56 mg/L of $FeCl_3$.

2.7. Dose of WCA and polymer coagulant

As shown in the Step 3 of Table 2, the aim of this part of the experiment is to determine the optimum WCA dose to be applied on an optimum pH and coagulant dose obtained in the previous experiment. This was performed by increasing the WCA dose periodically to 0.05 mg, 0.1 mg, 0.3 mg, 0.5 mg. 0.8 mg, 1 mg, 1.5 mg, 3 mg, and 5 mg. A study performed by Yoon et al. [14] shows that the flocculation caused by the injection of WCA had an effect on the sedimentation rate. Accordingly, the sedimentation rate of the flocs was also measured. In order to determine the sedimentation rate of the floc, measurement of the turbidity of the supernatant of the raw water, which was treated by chemical injection followed by agitation and was left aside to settle for 30 min, was taken. On the other hand, the optimum polymer coagulant dose was determined by increasing the input periodically at intervals of 1 mg/L, as shown in Step 4 of Table 2.

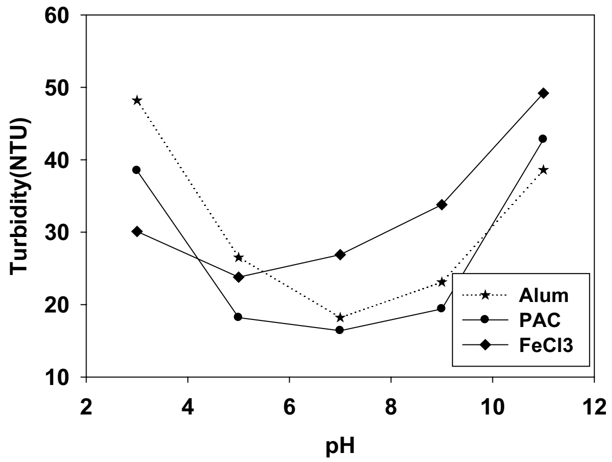

Fig. 1. Result of preceding research to determine optimum pH value.

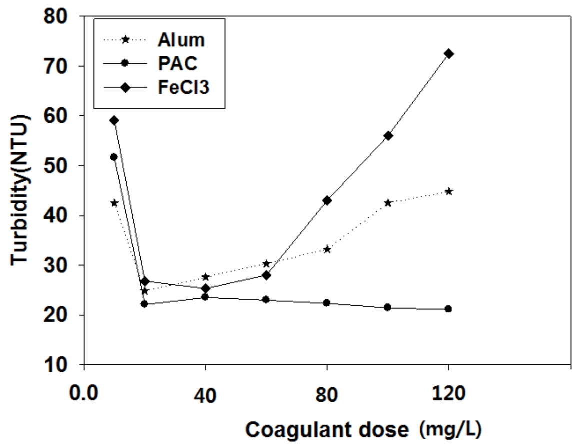

Fig. 2. Result of the preceding research to determine the optimum coagulant dose.

2.8. Coagulant dose curve obtained by TSS concentration variation in raw water

The optimum coagulant dose varies with varying concentrations of the pollutants in the raw water. Accordingly, this study was intended to derive a coagulant dose curve by plotting the variation in the TSS concentration (50–1,000 mg/L) in raw water. With regard to the experimental method, pH of raw water was 6.5 in alum, 7 in PAC, and 6 in $FeCl_3$. This experiment was carried out by injecting coagulants without WCAs and polymer coagulant.

3. Results and discussion

3.1. pH required for optimum coagulation

The experiment was conducted to examine the proper pH that can lead to the coagulation of the pollutants for each coagulant type (Step 1 of Table 2). In case of alum, the pH of raw water was increased from 5.5 to 9.5 at periodic intervals of 0.5. These results are shown in Fig. 3. After the injection of alum, the best removal efficiency was observed between 6.5 and 8. It was also observed that the agglutinates of the contaminants were not formed below a pH of 6.5. A complex compound was formed by its reaction with the organic matters, which is absorbed or removed by aluminum hydroxides. However, it is likely that the contaminants were not well absorbed by alum, because aluminum is not sufficiently hydrated at a low pH [15, 16]. Meanwhile, there was the difference of pH value showed the highest removal efficiency by pollutants type. The lowest concentration of TSS was obtained at a pH of 6.5, while that of BOD was obtained at a pH of 8. The nutrient salts, including TN, TKN, TP, and PO_4^{-3} showed the highest removal efficiency at pH = 6.5.

Therefore, the pH value of 6.5 was selected as the most appropriate pH for the use of alum as a coagulant. However, when PAC was used as a coagulant, there were no significant differences in the treated water quality in the entire pH range (5.5–8.5). pH range for coagulation have wide since pH and alkalinity concentration are not low along with pollutants and PAC that Al ion already combine with OH ion in injecting the coagulants. The highest removal efficiency was observed between 6.5 and 7 values of pH. When $FeCl_3$ was used as a coagulant, at a pH of 6 the water quality was reflected by favorable conditions, such as a TSS concentration of 78.5 mg/L, BOD of 50.6 mg/L, COD_{Cr} of 81.9 mg/L, TN of 27.8 mg/L, and

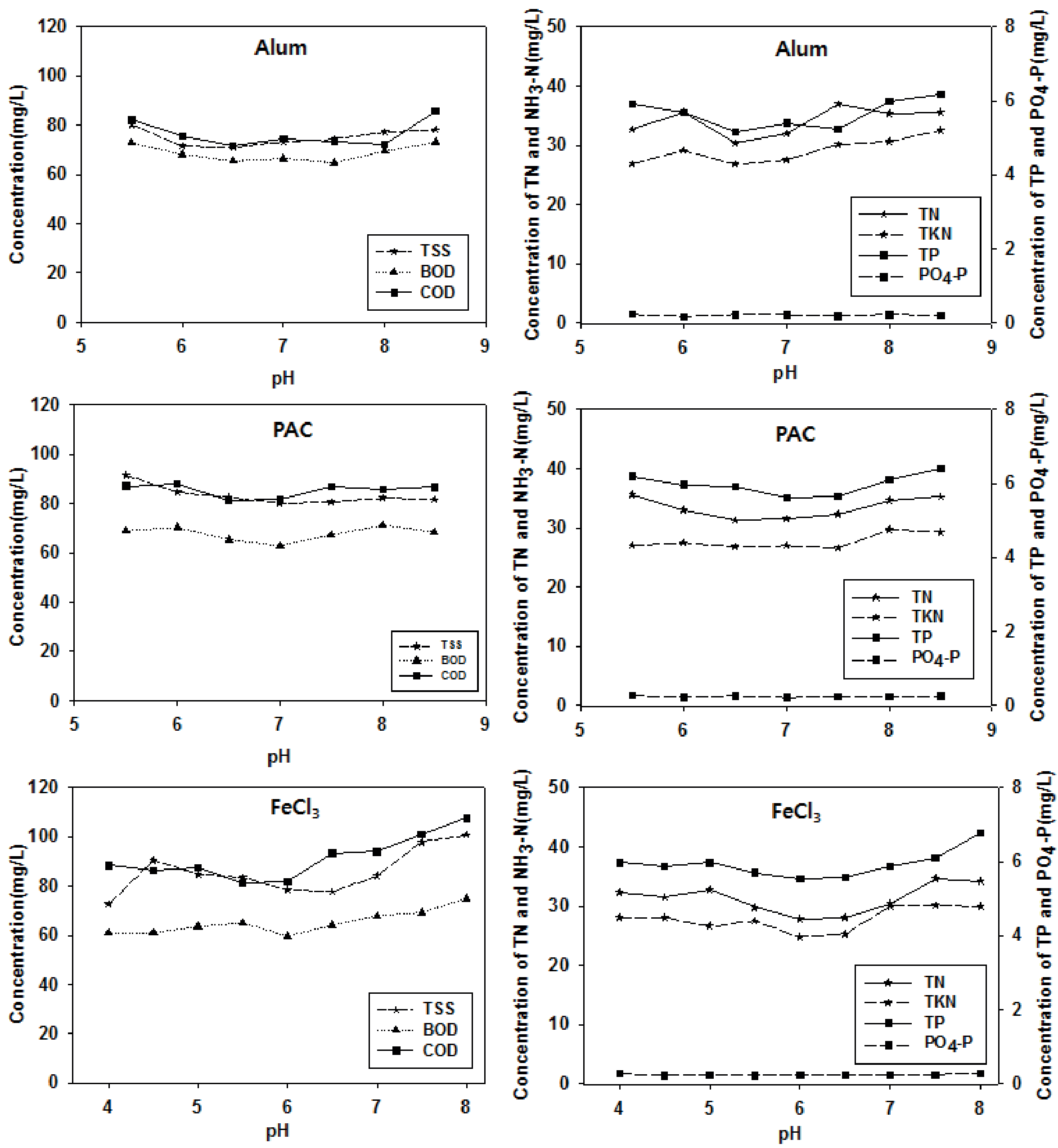

Fig. 3. Experiment result to determine optimum pH.

TP of 5.5 mg/L. Thus, the optimum pH values have been determined as 6.5 for alum, 7 for PAC, and 6 for $FeCl_3$.

3.2. Appropriate coagulant dose

In order to determine the optimum dose of an alum coagulant, its dose was increased periodically at intervals of 4 mg/L from 12 to 44 mg/L. The water quality of the treated water was measured (Fig. 4). When alum was used as a coagulant, the concentration of the pollutants sharply decreased until 30 mg/L of alum was added. However, with higher dosage, there was very slow decrease in the concentration of the pollutants, and reached saturation point during the removal of contaminants. An analysis showed that the TSS treatment efficiency was 82.5% at this point. While using a PAC coagulant, the water quality of the treated water very sharply improved until 20 mg/L was added and saturation was achieved. It was observed that the treatment efficiency was 84.3% at this point. On the other hand, while using $FeCl_3$ the efficiency of coagulation reached its peak at a dose of 36 mg/L. The concentration of BOD and COD_{Cr} started increasing again at a higher dose. TSS removal efficiency was 79.5% at the optimum dose of $FeCl_3$. Therefore, the treatment efficiency of the pollutants according to the optimum coagulant dosage was determined, which follows the order PAC > Alum > $FeCl_3$. Among colloidal materials are repulsive forces because the amount of anion in surface of colloidal materials is relatively more than amount

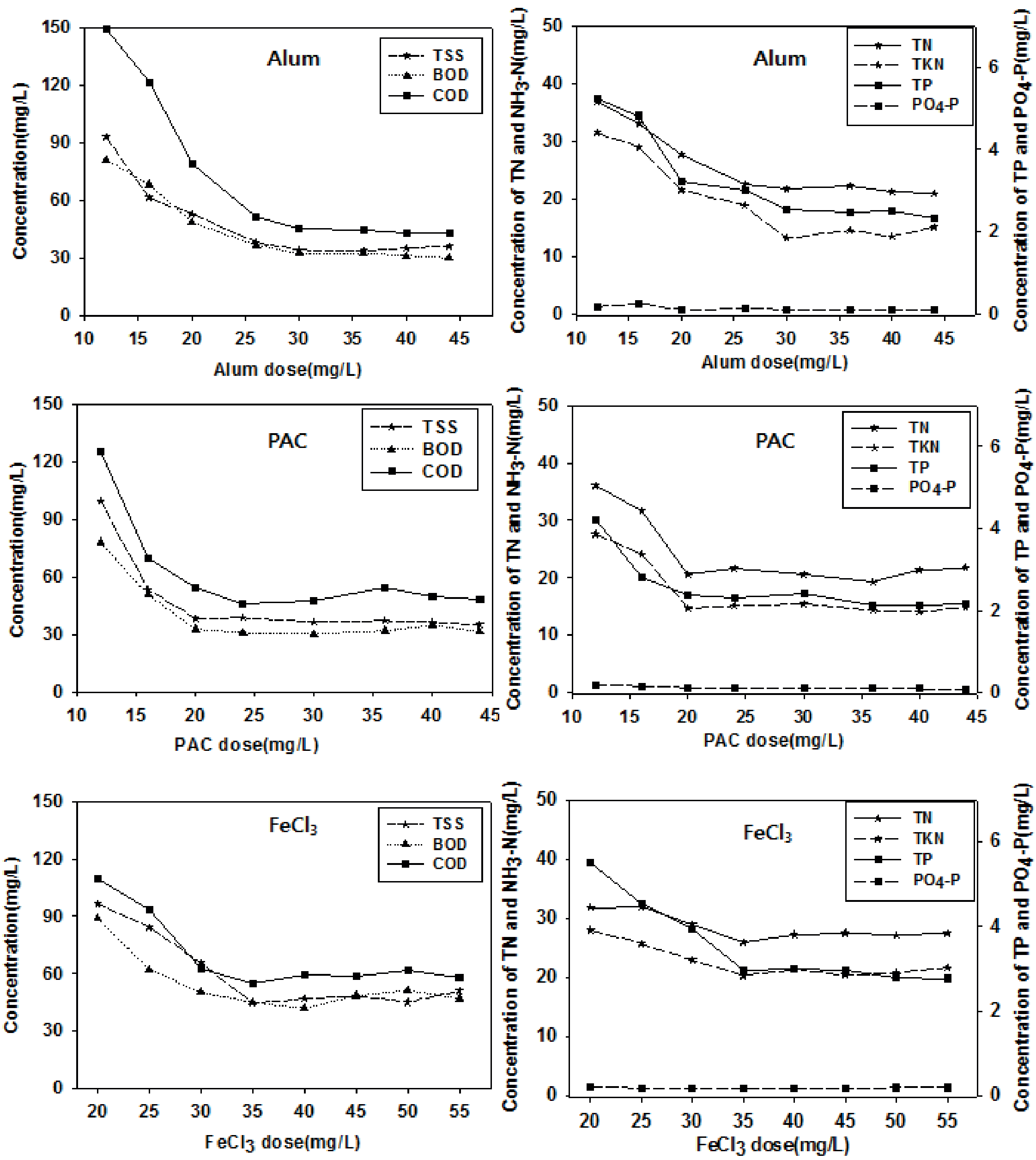

Fig. 4. Experiment result to determine optimum coagulants dose.

of cation. To coagulation of colloidal materials, an anion on colloidal materials surface should be changed to cationic by charge neutralization mechanism. Charge neutralization of an anion is occurred by Al^{3+} and Fe^{3+} ions in Alum, $FeCl_3$ and PAC injected as coagulants. And, $Al(OH)_3$ and $Fe(OH)_3$ generated by injected coagulants have the precipitation characteristic. These molecule adsorb surrounding pollutants over precipitation. These mechanism is called as sweep coagulation. Contaminants removal on this study is happen by two mechanisms mentioned above. Meanwhile, it is well known that PAC is batter coagulants than Alum and $FeCl_3$. The reason is because PAC is combined in advance with multivalent species and charge neutralization occurs more effectively as area ratio by volume of PAC is relatively higher compared with other hydrolysis compounds [17].

3.3. Appropriate WCA dose

WCA enhances the settling of the floc formed during the experiment [18, 19]. As CSOs can be more treated by inducing rapid sedimentation by WCA, WCA regards as the important factor related to the amount of treatment for CSOs. Studies by Park et al. [20, 21] suggest the possibility of regulating the concentration of soluble and insoluble pollutants that exist in the CSOs through coagulation and adsorption by added WCA and alum, re-coagulation and adsorption by the activated surface of the sludge returned as the secondary seed and co-precipitation effect caused by WCA [20, 21]. However, this study shows that the WCA injection causes negligible decrease in the pollutant concentration. Moreover, in case of excess dosing, there was an increase in the concentrations of the pollutants (Fig. 5).

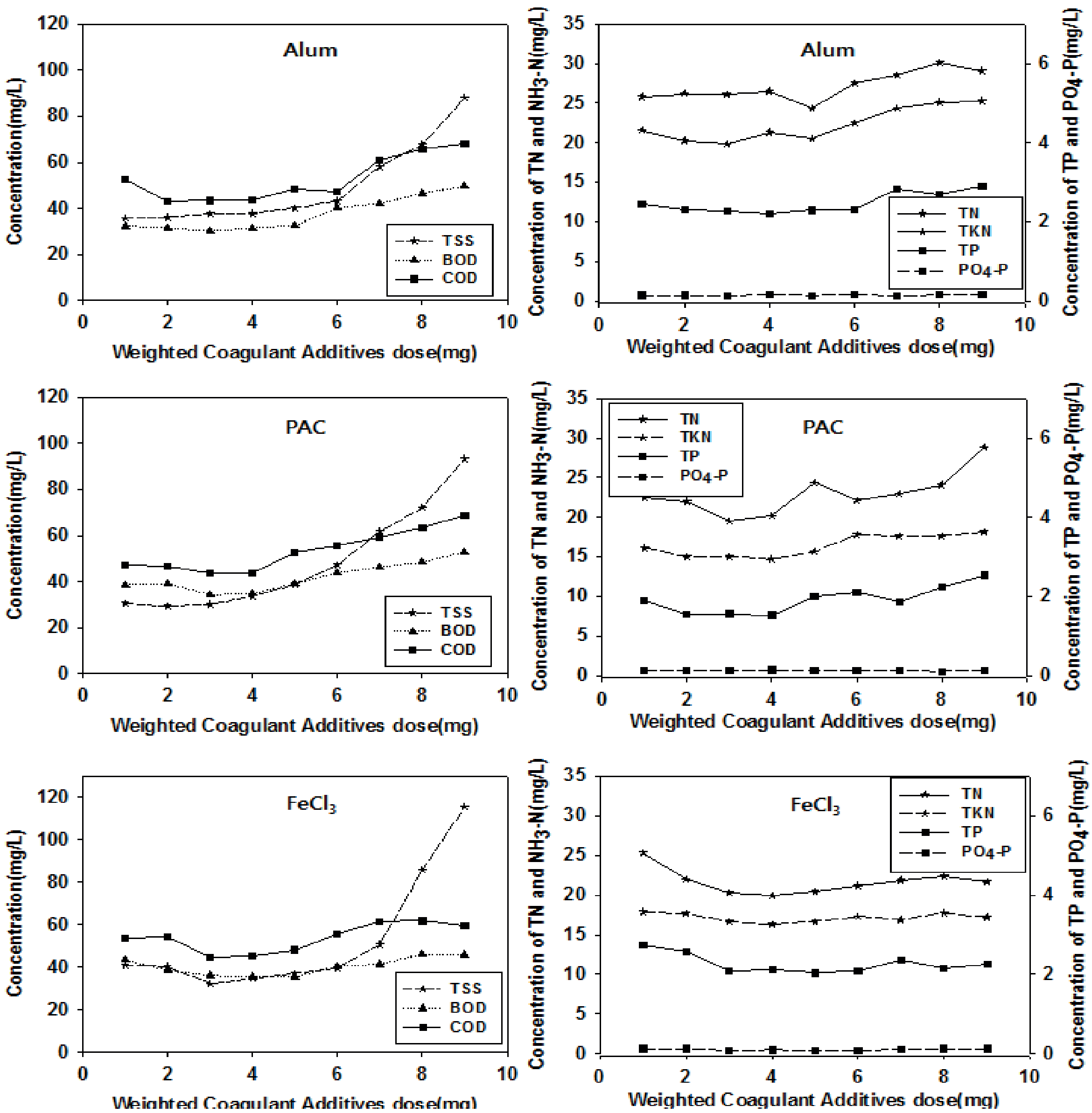

Fig. 5. Experiment result to determine optimum WCA dose.

When alum was injected as a coagulant, the concentration of the treated water was relatively stable at a dose of 1 mg and less; but the concentration of contaminants in treated water increased in a dose of 1 mg and more. PAC showed a stable water quality for treated water up to a dose of 0.5 mg. On the other hand, the use of $FeCl_3$ as a coagulant improved the water quality of the treated water with an increase in the dose of WCA from 0.05 to 0.3 mg, reaching the highest removal efficiency at 0.3 mg. When more than 1 mg WCA was added, the TSS removal efficiency sharply was decreased and the COD_{Cr} treatment efficiency started to lower over a dose of 0.8 mg and more. There were no significant changes in BOD, TN, and TP concentrations in the treated water with an increase in the WCA dose. Fig. 6 shows the turbidity measurements of the supernatant. This was done by plotting the time taken in carrying out an experiment with variation in the dose of WCA. In case of an alum coagulant, sedimentation rate improved until the dose of WCA reached 0.5 mg. The turbidity reduction rate (sedimentation rate) was the highest at this point. On the other hand, for a dose higher than 0.5 mg, the sedimentation rate decreased and the turbidity of treated water increased again. In pollutant concentration analysis by change of WCA dose, pollutant concentration was well treated until WCA was injected with the range from 0.05 to 1 mg/L. In this study, therefore, 0.5 mg that sedimentation ration was the highest an aided economic efficiency was determined as optimum WCA dose. PAC coagulant showed a similar sedimentation rate compared with when alum was used. However, the highest sedimentation rate was achieved when 0.3 mg WCA was added. The optimum WCA dose, therefore, was determined as 0.3 mg. During the use of $FeCl_3$ as a coagulant, the highest sedimentation rate was achieved when 0.1 mg WCA was added. It is likely that the specific gravity of the floc became due to the presence of Fe ion in $FeCl_3$. Therefore, in this case, the sedimentation was rapid even with a very small dose of WCA.

3.4. Appropriate polymer coagulant dose

To determine the optimum dose of a polymer coagulant, the concentration of the treated contaminants with a variety of polymer coagulant dose was analyzed. The concentration of pollutants in the treated water consistently showed a decreasing trend (Fig. 7) when a polymer coagulant was used. However, the decrease in the concentration of the pollutants slowed down after a certain point. This point was achieved at a dose of ≥4 mg/L, ≥5 mg/L, and ≥10 mg/L when alum, PAC, and $FeCl_3$ were used as coagulants, respectively. The concentrations of TSS, COD_{Cr}, TN, and TP at this point are shown in Table 3. TSS removal efficiency was >90%, and it was lowered down to 87% when $FeCl_3$ was used as a coagulant. COD_{Cr} removal efficiency was the lowest (78%) for water treated with an alum coagulant. PAC and $FeCl_3$ treated water had 87% and 80% efficiency of COD_{Cr} removal, respectively. There was a great difference in TN removal efficiency with each coagulant type. PAC-treated water had the highest

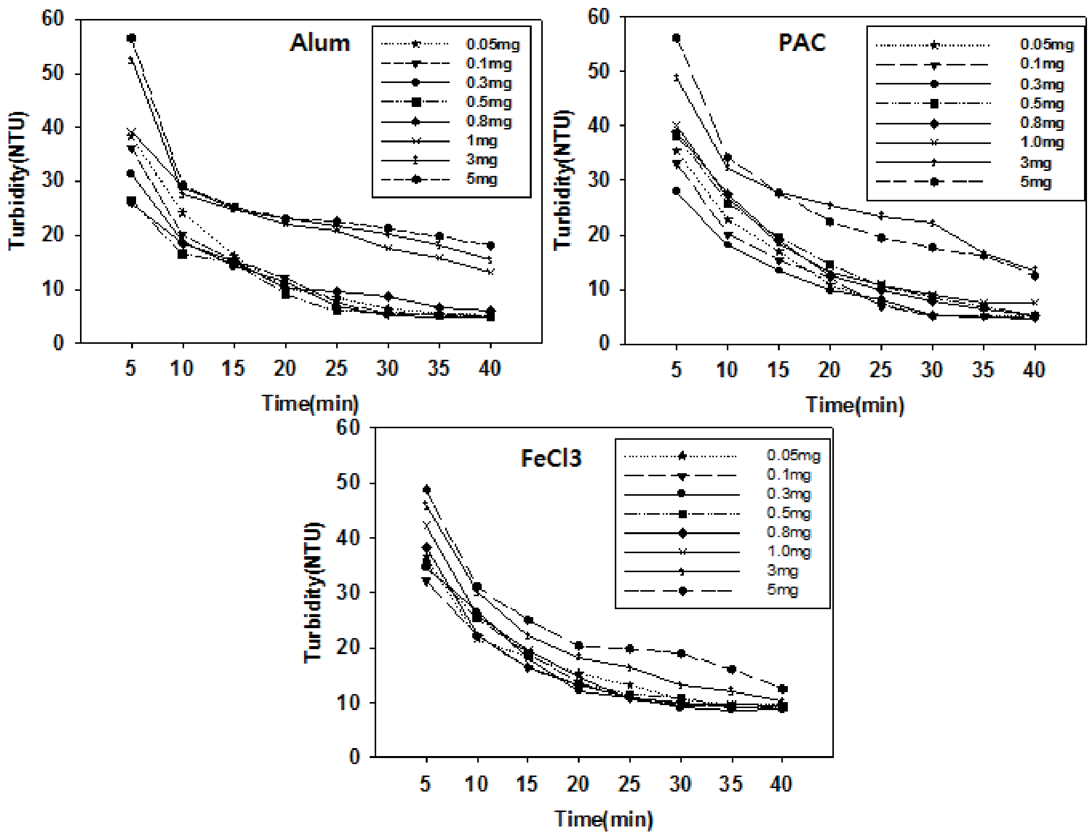

Fig. 6. Sedimentation rate of flocs by weighted coagulant dose.

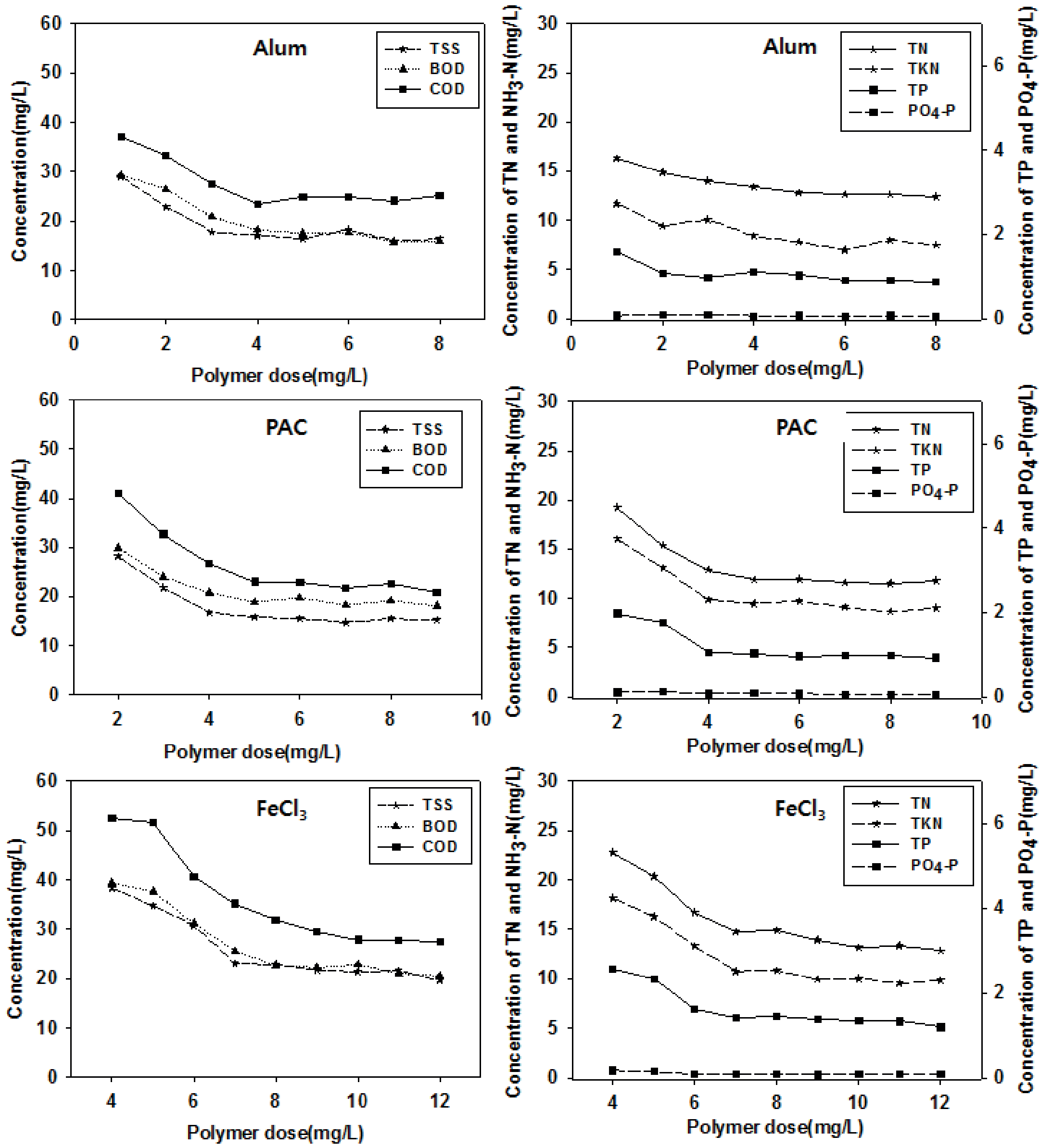

Fig. 7. Experiment result to determine optimum polymer dose.

Table 3
The removal efficiency of pollutants with coagulants

Coagulant	TSS (%)	BOD (%)	COD_{Cr} (%)	TN (%)	TP (%)
Alum	92.2	83.2	78.2	58.3	88.8
PAC	92.7	85.4	87.2	69.5	89.2
$FeCl_3$	87.6	82.4	80.1	45.8	85.5

removal efficiency (69.5%) and $FeCl_3$ treated water had the lowest removal efficiency (45.8%). TP treatment efficiency was similar for the three coagulant types used to treat water. Total treatment efficiency of the coagulants was in the order of PAC > alum > $FeCl_3$, after the injection of all chemicals, including WCA and polymer coagulant.

3.5. Optimum coagulant dose obtained by TSS concentration regulation

The required coagulant dose varies greatly with varying concentration of pollutants in raw water. The optimum dose of alum, PAC, and $FeCl_3$ was measured in terms of TSS concentration variation in raw water. Fig. 8 indicates that a small increase in the dose of PAC was required with increasing TSS concentration. An optimum dose of 35 mg/L of PAC was required to treat raw water with 500 mg/L of TSS concentration, which increased to 71 mg/L for 1,000 mg/L of TSS concentration. On the other hand, a dose of 69 mg/L of $FeCl_3$ coagulant was required for raw water with a TSS concentration of 500 mg/L, which increased to 181 mg/L (twice as high as the required concentration of PAC for the same concentration of TSS) for a TSS concentration of 1,000 mg/L. This shows that a PAC coagulant is more effective and efficient as

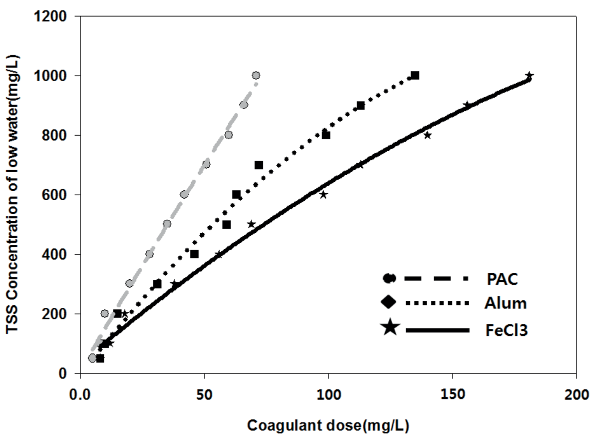

Fig. 8. Coagulant dose curve obtained by TSS concentration in raw water.

compared with $FeCl_3$ with an increasing concentration of pollutant in the raw water to be treated. A PAC coagulant also aids flocculation by cross-linking in addition to the coagulation reaction, unlike the other coagulants used in this study. This is because solids are better adsorbed as cross-linking becomes more active with an increasing quantity of solids. A dose of 59 mg/L for a TSS concentration of 500 mg/L in raw water was the optimum dose of alum, which increased to 135 mg/L for a TSS concentration of 1,000 mg/L. Hence, the required optimum dose of alum was somewhere between those of PAC and $FeCl_3$.

4. Conclusions

(1) When alum coagulant was used, the optimum pH for effective coagulation of flocs was determined as 6.5. For PAC-treated water, the process of coagulation occurred within pH of 6.5–7. The optimum pH of $FeCl_3$ for the coagulation of the flocs was determined to be 6.
(2) Proper coagulant dose to 1 L of raw water was determined to be 30 mg/L for alum, 20 mg/L for PAC, and 36 mg/L for $FeCl_3$. When the optimum coagulant dose was injected, the efficiency of removal of pollutants from raw water of the coagulants was in the order of PAC > Alum > $FeCl_3$.
(3) Injection of WCA has little effect on the pollutant concentration of the treated water. When alum was used as a coagulant, the most rapid sedimentation rate was observed at a WCA dose of 0.5 mg. When PAC was used for coagulation, the appropriate WCA dose was 0.3 mg and 0.1 mg for $FeCl_3$.
(4) Polymer coagulant dose was determined to be 4 mg/L in alum-treated water, 5 mg/L in PAC-treated water, and 10 mg/L in $FeCl_3$-treated water. The definitive TSS removal efficiency of the coagulant types was in the order of PAC > Alum > $FeCl_3$ after all the chemicals were injected. In the experiment to determine the optimum coagulant dose for TSS concentration regulation, a PAC coagulant was the most effective and $FeCl_3$ coagulant was the least effective with increasing concentrations of pollutants to be treated in the raw water.

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