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Application of dissolved air flotation (DAF) with coagulation process for treatment of phosphorus within permeate of membrane bioreactor (MBR)

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ABSTRACT

It is well known that eutrophication is one of the main issues with regard to environmental pollution and that phosphorus is the most critical among various nutrients. Consequently, effluent water quality of wastewater treatment plants has been recently reinforced in Korea. Many studies have been made on membrane bioreactor (MBR) processes, some of the more advanced wastewater treatment processes. However, MBR processes are relatively vulnerable to biological phosphorus treatment in comparison with conventional activated sludge processes. Alternately, dissolved air flotation (DAF) processes were applied as post-treatment of MBR processes in order to decrease T-P concentration of the final effluent. It was observed that T-P removal efficiency was 51.2% and that T-P concentration of the final effluent ranged from 0.49 to 1.62 mg/L with addition of 15 mg/L of alum to the rapid mixing reactor of DAF processes. The next step includes the addition of anionic polymers and bentonite to slow and rapid mixing tanks of DAF processes, respectively, to improve phosphorus removal efficiencies. Although T-P removal efficiency increased significantly from 51.2 to 78%, T-P concentration of effluent was observed between 0.38 and 0.96 mg/L. Lastly, the MBR process was controlled by adding a low concentration of alum to an aerobic tank and influent under 1 ± 0.3 mg/L of T-P. While T-P removal efficiency was around 80% in both 7 and 10 mg/L of alum, most measured values were under 0.2 mg/L except for several days of measurement. This indicates the potential of DAF processes for removing phosphorus as post-treatment of MBR.

Keywords: Dissolved air flotation (DAF); Membrane bioreactor (MBR); Phosphorus; Coagulant; Coagulation/flocculation

1. Introduction

The membrane bioreactor (MBR) has gained increasing popularity in municipal wastewater treatment due to its potential for high-quality effluent and its small footprint through a combination of membrane filtration and biological treatment [1–3]. The application of MBR is expected to continuously increase in capacity and broaden in various categories due to more stringent regulations and water reuse initiatives [4,5]. An advantage of MBR has been reported for efficient nitrogen removal due to prolonged sludge

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retention times (SRT), which are favorable conditions for heterotrophic bacteria [6,7]. However, long SRT has a negative influence on phosphorus removal since phosphorus accumulating organisms (PAOs) with P luxury uptake must be removed from the reactor in order to eliminate phosphorus [8].

The concentration of phosphorus is low compared with organic matter and nitrogen in wastewater, but its influence is important as an essential, often limiting, nutrient for plants and microorganisms. So, phosphorus is the most critical factor of eutrophication in most cases. As a result, in Korea, requirements for effluent water quality of wastewater treatment plants were strengthened to between 0.2 and 0.5 mg/L from 2012 according to region. In China, a country with many environment-related problems, increasingly stringent phosphorus regulations are expected for wastewater, aiming for a total phosphorus limit of 0.5 mg/L [9]. Phosphorus removal methods can be divided into enhanced biological phosphorus removal (EBPR) processes and physicochemical methods using metal salts, and many studies have been carried out to increase removal efficiency [10–12].

Al or Fe(III) is often used in pretreatment or post-treatment processes to maintain acceptable T-P concentrations. In the case of MBR processes, metal salts are sometimes added in the bioreactor directly for removal of phosphorus [13–16]. Although it is indisputable that coagulant dosage is efficient for removing phosphorus, it is controversial whether or not metal salts are effective for controlling membrane fouling in MBR. Some studies have reported that alum- or iron-based coagulants improved filterability of activated sludge due to an increase in floc size and a reduction in soluble microbial products (SMP) [17–19]. In contrast, other studies have shown that added metal salts can be major foulants and that elimination of these foulants from membranes is not easy [20,21]. These contradictory results may be caused by differing experimental conditions, in particular, the dosage of added coagulants. Appropriate dosing is helpful for fouling control, whereas the addition of excessive chemicals for phosphorus removal can cause severe fouling [22–24]. Even though injection of coagulants into the reactor is very simple and effective for phosphorus removal, it is not easy to comply with strict water quality regulations by simply adding coagulants into the reactor, and metal salts have the potential to cause serious fouling.

In this study, removal of phosphorus from permeate water is performed by MBR through dissolved air flotation (DAF) with microbubbles and chemicals. DAF is applied widely as an alternative clarification method for sedimentation in drinking water treatment, pretreatment of industrial wastewater and solid–liquid separation in activated sludge processes [25–27]. A previous report indicated that DAF was more efficient than sedimentation for removing turbidity and particles [28]. While there are many reports regarding phosphorus removal through the addition of chemicals and application of DAF in wastewater treatment [29–32], very few attempts have been made at DAF with chemicals with regard to permeate water of MBR. So, this study examines the removal of phosphorus through DAF with coagulants such as alum, anionic polymer, and bentonite considering properties of permeate water of MBR.

2. Material and methods

2.1. Laboratory-scale test (jar test)

A jar test was carried out to determine the proper dosage of coagulant (alum) and coagulant aid (anionic polymer; A331P, SNF Korea) for efficient phosphorus removal through DAF. The jar test was carried out in six beakers (1 L) simultaneously, and alum or bentonite was added under stirring after raw water (1 L) was poured into each beaker. Coagulant aid was added using slow mixing for good flocculation. After rapid mixing for 1 min at 250 rpm and slow mixing for 5 min at 50 rpm, the supernatant was obtained from a point located about 2 cm below the surface of the liquid. A disposable syringe was used during sampling to minimize errors, and chemicals for controlling pH were not used. Phosphorus and turbidity were analyzed by Standard Methods and 2100 N Turbidimeter, respectively, (HACH).

Wastewater without pretreatment was used as raw water in the first jar test. Orthophosphate-P was measured to find the optimal alum concentration that enables sufficient physicochemical bonding between orthophosphate-P and aluminum. Turbidity was checked as an indirect index of the potential for flotation. In the second jar test, wastewater with 0.45 um filtration was used to evaluate phosphorus removal efficiency in water without particulates.

2.2. Pilot plant test conditions and operation

As shown in Fig. 1, the DAF process was composed of rapid mixing for coagulation, slow mixing for flocculation, and flotation separation to remove flocs and produce treated water through an outflow pipe at the end of the flotation tank. The MBR process, performed prior to the DAF process, consisted of anoxic and aerobic tanks, and produced about 100 tons of water a day. The characteristics of MBR

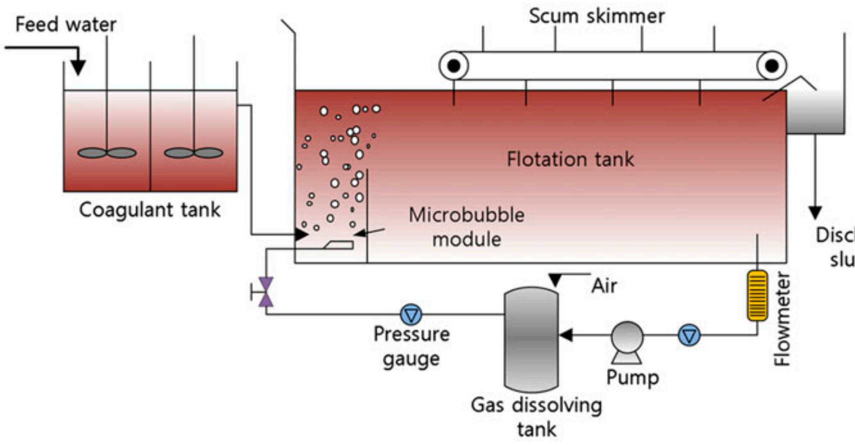

Fig. 1. The illustration of a pilot-scale DAF process operated in this study.

effluent are shown in Table 1, and rapid mixing took place for about 5 min at an agitation intensity of 215 s^{-1}, followed by slow mixing for about 10 min at an agitation intensity of 37 s^{-1}. Alum and bentonite were added into the rapid mixing tank, and anionic polymer was injected into the slow mixing tank for efficient coagulation and flocculation. Bentonite dosage concentration was fixed at 2 mg/L, similar to the effluent from conventional activated processes, since phosphorus was removed effectively in chemical DAF processes for which raw water with a small or considerable amount of suspended solids was used. Hydraulic retention time (HRT) of the flotation tank was about 20 min, and suspended solids on the surface of the solution were removed by a scum skimmer periodically.

3. Results and discussion

3.1. *Jar test for alum concentration determination*

Coagulant dosage plays an important role in determining coagulation efficiency in the coagulation and flocculation processes. Especially, coagulant conditions are directly relevant to removal efficiency in the case of phosphorus treatment due to the electrochemical combination of aluminum or ferric ions of coagulants with orthophosphate-P. Fig. 2 shows the effects of different dosages of alum as the sole coagulant on the removal of orthophosphate-P and turbidity from wastewater without pretreatment. The results demonstrate that orthophosphate-P and turbidity removal increased significantly as the dosage of alum increased by 20 mg/L. It was observed that removal of orthophosphate-P increased sharply up to 10 mg/L. On the other hand, while there was not a considerable change in orthophosphate-P removal with increasing doses of alum above 20 mg/L, removal efficiency of turbidity declined. This may be a result of re-suspension of solids at this concentration. When the dosage of alum was 20 mg/L, the highest turbidity removal efficiency was achieved, and the concentration was 3.35 NTU with 97% removal efficiency. In the case of

Table 1
The quality of MBR effluent

Section	Maximum	Minimum	Average
BOD_5 (mg/L)	2.8	0.6	1.3
COD_{Cr} (mg/L)	30.1	10.1	17.6
T-N (mg/L)	15.2	7.9	9.7
NH_4-N	5.4	0.1	1.4
NO_3-N	9.5	2.9	6.0

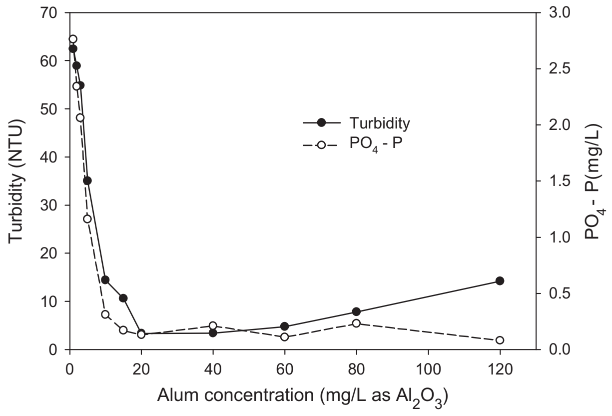

Fig. 2. Rejection of PO_4-P and turbidity according to alum concentration.

phosphorus, although the lowest concentration was observed at an alum concentration of 120 mg/L, the optimal injection concentration was about 20 mg/L considering coagulant consumption. Target substances must be large enough to float up to the surface of solution [33,34]. Additionally, alum physicochemically removes phosphorus on the principle of direct adsorption of phosphate ions on hydrolysis products, sweep floc, and the formation of insoluble salts with aluminum [35]. The optimum alum concentration seems to be between 10 and 20 mg/L for effective DAF operation.

3.2. Effect of polymer coagulant aid through jar test

Permeate water from the MBR process has almost no particulate compounds due to perfect solid–liquid separation. Although this enables excellent water quality, it works against phosphorus removal by DAF processes since appropriate flocs are favorable to substances flotation by microbubbles. Fig. 3 illustrates the T-P concentration of supernatant depending on anionic polymer concentration with constant alum (10 mg/L) and bentonite (2 mg/L). Removal efficiency using only alum was about 63% when T-P of the raw water decreased from 3.78 to 1.4 mg/L. When anionic polymers of 0.6 and 1.2 mg/L were added, removal efficiency increased by 15 and 20%, respectively, compared to that without anionic polymer, and T-P concentration was above the intensified regulation at 0.83 and 0.63 mg/L, respectively. Additionally, although the dosage of anionic polymers was increased above 1.8 mg/L, there was no improvement in phosphorus treatment, and T-P concentration of supernatants was higher than that of the 1.2 mg/L dosage. Similar results can be found in a previous report dealing with wastewater treatment using various polarized polymers [36].

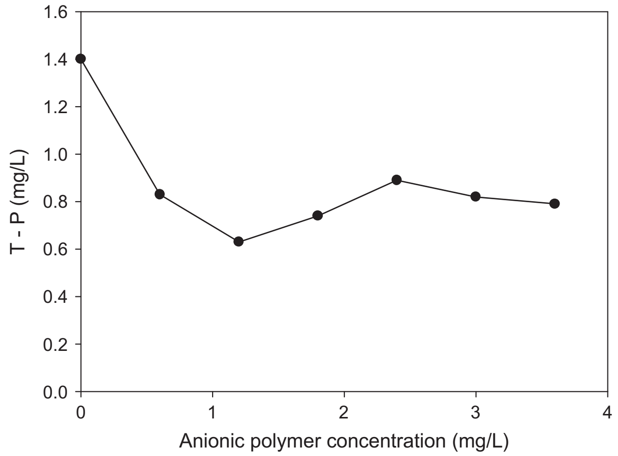

Fig. 3. Rejection of T-P according to anionic polymer concentration.

Several previous studies have mentioned that anionic polymers with metal salts produced bigger and more compact flocs due to the strong bonds of polymer chains [36–38], a positive result for flotation by microbubbles. As shown in Fig. 4, it was observed that floc size grew bigger as the amount of anionic polymers increased. It is notable that while the flocs increase with increasing dosages of anionic polymer, T-P removal efficiency showed a different trend, as shown in Fig. 3. This result may be attributed to properties of soluble phosphorus combined with aluminum physicochemically, unlike particulate causing turbidity. In other words, there can exist soluble phosphorus that is joined with aluminum but kept afloat in the solution.

3.3. Phosphorus removal by DAF using only alum

Fig. 5 describes the T-P concentration of raw water (MBR effluent) and treated water (DAF effluent) in the DAF process using only alum for about 35 d, and resulting T-P removal efficiencies are shown. When alum was added at concentrations of 10 and 15 mg/L, average T-P removal efficiencies were 41 and 51.2%, respectively. The final DAF effluent had concentrations ranging from 0.74 to 2.02 mg/L and from 0.49 to 1.62 mg/L, respectively, and T-P concentration was fluctuating with water quality of the MBR effluent. This process does not seem to be effective for phosphorus treatment despite the use of chemicals compared with biological treatment. However, orthophosphate-P results measured after 0.45 um filtration showed that average values of influent and effluent of DAF processes were 2.73 and 0.61 mg/L, resulting in 77% removal efficiency. This means that physicochemical coagulation between alum and orthophosphate-P was achieved sufficiently, but solid–liquid separation in the DAF process was not accomplished well. However, a previous report indicated that removal of phosphorus by the DAF process with chemicals was successful [32]. The reason for these differing results, despite the use of the same DAF process, may be that while treated water from SBR has some suspended solids, there are few particulates to be bonded to coagulants and followed by combination with microbubbles in permeate water of MBR processes. Additionally, it was reported that bigger particles require more microbubbles [39], so floc size is a considerable factor for successful DAF processes.

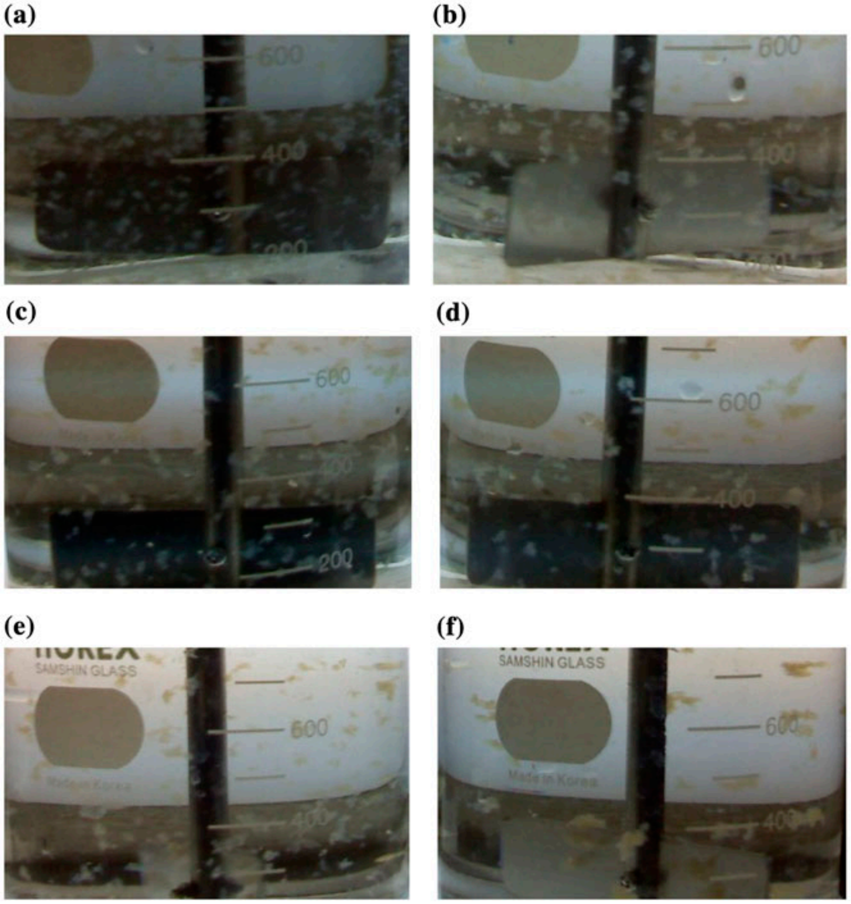

Fig. 4. Coagulation/flocculation according to anionic polymer concentration (a) 0.6 mg/L, (b) 1.2 mg/L, (c) 1.8 mg/L, (d) 2.4 mg/L, (e) 3.0 mg/L and (f) 3.6 mg/L.

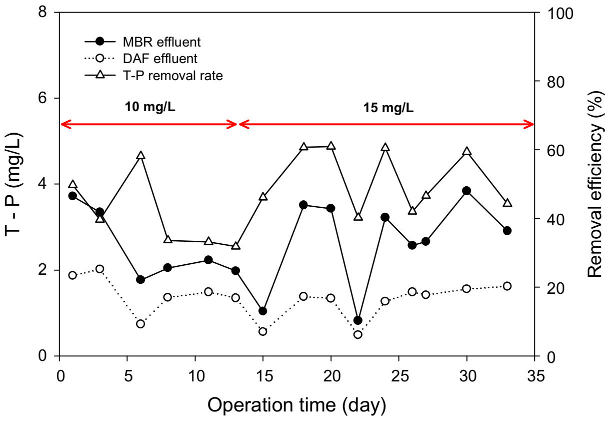

Fig. 5. T-P removal effect in DAF processes with only alum.

3.4. *Phosphorus removal by DAF process with alum, anionic polymers, and bentonite*

In Section 3.3, removal of phosphorus was attempted through the chemical DAF process, a conventional method for wastewater treatment. However, we could not obtain acceptable results despite sufficient bonding between alum and orthophosphate-P since target substances were not separated from the mixed liquor. Therefore, in this section, supplemental bentonite and anionic polymers were added to increase the phosphorus removal efficiency of the DAF process. The bentonite played the role of a nucleus in effluent of SBR processes for coagulation, and anionic polymers help increase floc size in order to assist floatation with microbubbles.

Fig. 6 is a graph showing phosphorus removal efficiency of the DAF process with bentonite (2 mg/L) and anionic polymers (1.2 mg/L) depending on alum concentration for about 55 d. The average T-P removal efficiencies were 75 and 67% for 10 and 15 mg/L alum addition, respectively. Removal efficiency of 10 mg/L was better than that of 15 mg/L due to the fact that the difference in alum concentration did not have a big impact on T-P removal, and MBR effluent T-P concentration was relatively high in the 10 mg/L alum

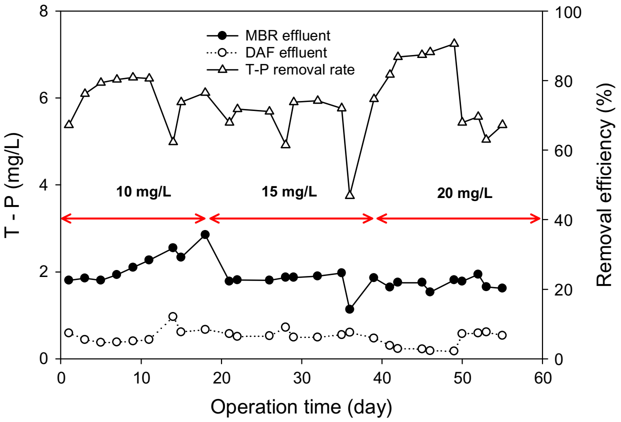

Fig. 6. T-P removal effect in DAF processes with coagulant aid.

sample. As a result, phosphorus removal efficiencies improved by between 16 and 34% from addition of bentonite and anionic polymers. However, T-P concentration of effluent was seen between 0.38 and 0.96 mg/L, demonstrating the difficulty in complying with enhanced regulations about phosphorus. In the case of 20 mg/L, phosphorus removal efficiency was around 78%, and it was possible to obtain effluent with a phosphorus concentration under 0.2 mg/L. Despite high removal efficiency, T-P concentration of treated water could be determined as acceptable or not since phosphorus regulations vary according to region. Furthermore, excessive alum was used for removing phosphorus. It is well known that coagulants such as alum, iron, calcium, and magnesium are effective chemicals in wastewater treatment [40]. In other words, the key point of chemical coagulation is the reduction of coagulant dosage in order to reduce subsequently generated waste sludge, which intensifies the problems of operation cost and environmental pollution.

While the dosage of alum increased from 10 to 20 mg/L, phosphorus removal efficiency did not improve in spite of a doubled concentration of coagulant. This indicates that an increase in coagulant concentration is not efficient in DAF processes to deal with MBR permeate water, and the critical alum concentration for maximum phosphorus removal efficiency is around 10 mg/L.

3.5. Phosphorus removal using DAF with injection of alum into MBR

In Section 3.4, we determined the presence of a limit to the increase in phosphorus efficiency through the addition of alum. Therefore, a low concentration of alum (around 2 mg/L) was injected directly into the aerobic tank of the MBR process to obtain an effluent T-P concentration under 0.2 mg/L without an increase in treatment efficiencies of the DAF process. In case of chemical phosphorus control in MBR processes, it is difficult to decrease the phosphorus concentration of effluent under 0.2 mg/L, but it is possible to obtain permeate water below 2 mg/L, which meets past regulations [41]. So, T-P concentration of influent of the DAF process could be under 1 ± 0.3 mg/L due to alum injection into the MBR reactor. As shown in Fig. 7, 7–10 mg/L alum were added to the rapid mixing reactor for about 30 d. Phosphorus removal efficiencies were about 80% for alum concentrations of both 7 and 10 mg/L, and T-P concentration of effluent was mostly under 0.2 mg/L. Although there were several days for which T-P concentration of effluent was above 0.2 mg/L, a stable water quality was observed.

Phosphorus removal efficiencies notably increased by about 8% compared to results from Section 3.4 despite use of the same alum concentration. It was difficult to demonstrate the cause of this result due to the limited data in this study. However, it is likely that there may exist aluminum or aluminum hydroxide that did not combine with orthophosphate-P in MBR effluent, and the presence of these aluminum compounds could influence the T-P removal efficiencies of DAF processes. Previous studies regarding chemical phosphorus treatment have made attempts to elucidate the chemical removal of phosphates through adsorption by ligand exchange and the negative charge per P atom [42,43].

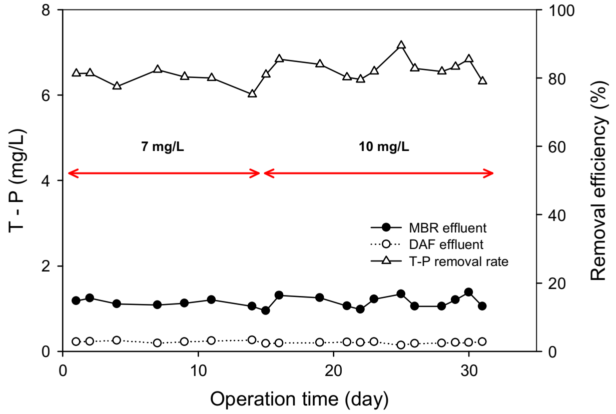

Fig. 7. T-P removal effect in DAF processes after MBR phosphorus control.

4. Conclusions

MBR processes have many benefits from perfect solid–liquid separation compared to conventional activated sludge processes. However, long SRT, one of the advantages of MBR processes, works against biological phosphorus removal such that it is difficult to comply with strict phosphorus regulations. Therefore, the removal of phosphorus from permeate water of MBR processes was examined, and results are summarized as follows.

1. In the jar test for determining the appropriate amount of coagulant, the optimal dosage of anionic polymers was about 1.2 mg/L to form flocs for floating with microbubbles.
2. Although the dosage of alum increased by 15 mg/L in the pilot-scale DAF process with only alum, T-P removal efficiency was about 51.2%. On the other hand, orthophosphate-P decreased by 77% due to successful physicochemical coagulation.
3. In case of alum with anionic polymers and bentonite, T-P removal efficiencies were around 78% or 1.5 times more than those of only alum. However, it is impossible to comply with intensified regulations using these operating conditions of DAF processes.
4. Lastly, a small amount of alum (2 mg/L) was added into the aerobic tank of MBR processes so that influent T-P concentration of DAF processes was around 1 ± 0.3 mg/L. Phosphorus removal efficiencies were above 80% in 10 mg/L of alum despite the small dosage. T-P concentration was mostly under 0.2 mg/L with the exception of several days of measurement. Results suggest that DAF processes are worth considering as a means for removal of phosphorus from effluent of MBR processes.

Acknowledgment

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