



The 4-stage anoxic membrane bioreactor for simultaneous nitrogen and phosphorus removal, and its strengths and weaknesses

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ABSTRACT

In the laboratory-scale research, anoxic membrane bioreactor (MBR), in which a membrane is submerged in the second anoxic reactor, and aerobic MBR, where a membrane is installed in the aerobic reactor, were operated simultaneously to observe advantages and disadvantages of the 4-stage anoxic MBR process. The advantages observed were as follows: (1) nitrogen concentration in effluent (5.2 mg N/L) superior to that in aerobic MBR (7.1 mg N/L) and (2) efficient utilization of carbon source to remove nutrients (2.46 g SCOD_{utilized}/g [N_{denitrified} + P_{released}]). In contrast, the disadvantages of this process were as follows: (1) relatively higher phosphorus concentration in effluent (0.9 mg/L) than in aerobic MBR (0.5 mg P/L), (2) 20% lower membrane permeability, and (3) 25% lower sludge settleability.

Keywords: Anoxic membrane bioreactor; Enhanced biological phosphorus removal; Simultaneous nitrogen and phosphorus removal

1. Introduction

A survey by Pagilla and Urgan-Demirtas [1] on nutrient removal at wastewater treatment plants (WWTPs), in which achieved either very low total nitrogen (TN) and/or total phosphorus (TP) effluents (below 5 mg N/L and/or 0.5 mg P/L), indicated that the combinations or modifications of various technologies (e.g. BNR, MBR, filtration, and chemical P) could be used to meet very low concentration requirements in the effluent. To achieve very high-quality effluent, a conceptual agenda known as the “Limit of Technology

(LOT)” has been formulated by Water Environment Federation [2,3]. The LOT in WWTP is loosely defined as plants meeting either 3 mg/L TN or 0.1 mg/L in the final effluent. The LOT concept has been further expanded to achieve high-quality effluent without chemicals or filtration [4].

In 2011, average effluent TN and TP concentrations from WWTPs in Korea with biological nutrient removal (BNR) were 12.7 and 1.0 mg/L, respectively [5]. N removal appears to be a key consideration for enhancing the effluent quality. Chemical treatment easily enhances the P removal efficiency, while an unfavorable C/N/P ratio and temperature variation could hamper the N removal. In addition, chemical P

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removal in biological plants might result in problems involving operational complexity and cost increases.

The utilization of denitrifying phosphorus accumulating organism (DPAO), which can simultaneously remove N and P in the anoxic condition, could be an alternative to achieve the LOT in BNR plants. The recent research on DPAO has emphasized several scientific points, such as the microbial community and metabolic pathway in the anaerobic–anoxic SBR system [6–10]. It is known that the observation of anoxic P uptake with denitrification by DPAO was apparent in the BNR process [11–14], although the microbial community of DPAO was not clearly isolated and identified.

The use of MBR has rapidly increased worldwide in recent years. The major role of membrane separation in biological plants seems to be as a replacement for the secondary clarifier for the membrane, which is used to solve the sludge settling problems. Drastically reduced solid and organic concentrations in the effluent are advantageous for water reuse, but the energy-intensive operation and membrane fouling remain technical challenges. The advantages of using membranes in the biological nutrient removal system have not been fully explored. In particular, the role of membrane installation in the anoxic zone requires further investigation. In this study, two unique 4-stage step-feed MBR systems (one is the anoxic MBR, and the other is the aerobic MBR) were operated to analyze their performance under laboratory conditions, and evaluate the advantages and disadvantages of the anoxic MBR compared to the ordinary aerobic MBR.

2. Materials and methods

2.1. Process description

The schematic diagrams of the 4-stage aerobic MBR (a) and anoxic MBR (b) are illustrated in Fig. 1. In the anaerobic (AN)—anoxic (1st AX)—anoxic (2nd AX)—aerobic (OX) flow scheme, the submerged-type membrane (PTFE, membrane area = 0.09 m²) was placed in the 2nd AX zone (denoted the anoxic MBR), while the membrane that was submerged in the OX zone (called the aerobic MBR) was used as the control unit. The 1st AX was operated to cultivate DPAO, and the role of the 2nd AX was to denitrify the remaining NO₃⁻-N using carbon energy in a step-feed system. Nitrate recycle from OX to the 1st AX varied within 100–200% of the influent Q, and denitrified sludge recycle from the 2nd AX to AN was 30–50% of the influent Q. The flow rate of nitrified recycle was controlled to optimize the NO₃⁻-N concentration as the electron acceptor of DPAO in the 1st AX zone. The

total HRT was 7.5 h. In the anoxic membrane system, N₂ gas was used for both mixing and fouling prevention.

2.2. Sewage characteristics and reactor operation

Table 1 presents characteristics of sewage, which were taken from a primary clarifier of a WWTP. The 4-stage aerobic and anoxic MBR systems were operated to four phases with various sludge recycle ratios. The variation of the recycle ratio aimed to find out the optimum condition for DPAO growth because a maximum condition of stored internal carbon energy as well as sufficient supply of NO₃⁻-N is the key to cultivate DPAO under anoxic conditions. Average VFA COD concentration was 39.8 mg/L. MLSS concentration maintained at the range from 4,000 to 6,000 mg/L for the overall experimental period. Average SRT in 4-stage aerobic MBR and anoxic MBR was 24.2 and 26.8 d, respectively.

2.3. Analytical methods

The analysis of pH, COD (total, soluble), NH₄⁺-N, TP, TSS, VSS, and SVI was performed in accordance with Standard Methods [15]. NO₂⁻-N, NO₃⁻-N, and PO₄³⁻-P were measured with ion chromatography (IC-80, Dionex). The analysis of the TN was conducted by a DR4000 (HACH Co.). Samples were taken regularly from each reactor during the experiment periods. VFA was analyzed by an HPLC (Agilent Technologies 1200 series) equipped with an ultraviolet detector (210 nm) and an Aminex HPX-87H column after pretreatment with a 0.45 m GF/C membrane filter. A batch test was conducted for measuring SPRR, SPUR, and SDNR using 150 mL of anoxic sludge and 200 mL of aerobic sludge. For the batch experiment, 200 mg/L of acetic acid was added as an external carbon source. To determine the anoxic and oxic P uptake rate, phosphate was completely released under anaerobic conditions for 2 h. The particle sizes in the aerobic MBR and anoxic MBR were measured using a particle size analyzer (Micro-P, MALVERN, USA).

3. Results and discussion

3.1. Sludge settling characteristics with solid and organic removal

Fig. 2 shows the solids concentration of the influent and effluent in the 4-stage aerobic MBR and anoxic MBR. The influent VSS/TSS ratio was 0.85 in both MBR systems. The variation in the recycle ratios

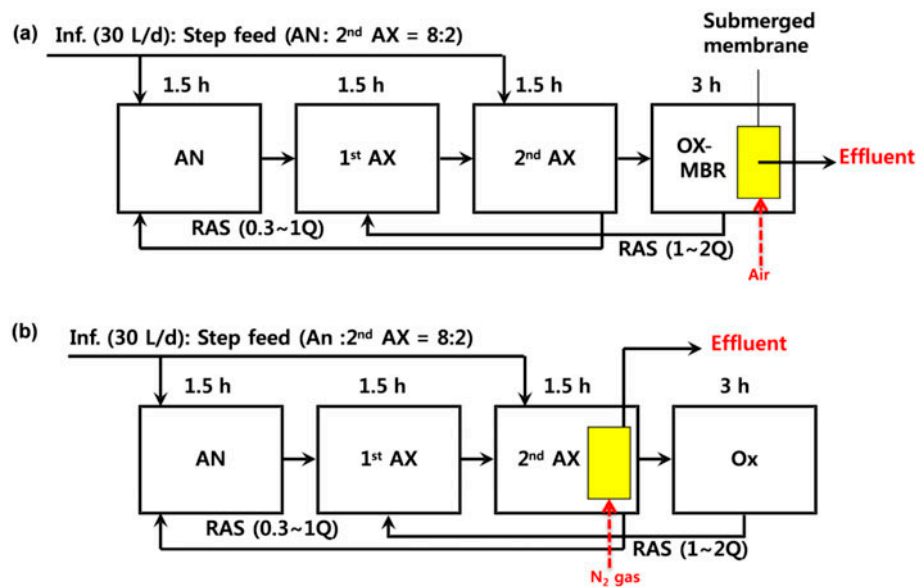


Fig. 1. Schematic diagrams of (a) aerobic MBR and (b) anoxic MBR.

Table 1
Influent sewage characteristics and operating conditions

Parameters	Phase 1 Nitrified recycle*: 2Q Anoxic recycle**: 1Q	Phase 2 Nitrified recycle: 2Q Anoxic recycle: 0.5Q	Phase 3 Nitrified recycle: 1Q Anoxic recycle: 0.5Q	Phase 4 Nitrified recycle: 1Q Anoxic recycle: 0.3Q	
Operating days (d)	121	35	72	69	
SS	TSS (mg/L)	70–320 (135)	50–227 (108.5)	43–118 (72.2)	36–127 (91)
	VSS (mg/L)	68–280 (128)	53–227 (104)	35–122 (66.1)	30–108 (79.1)
COD	TCOD (mg/L)	170–390 (284)	94–454 (242)	102.3–273 (201)	145–349 (255.8)
	SCOD (mg/L)	40–128 (92.9)	59–115 (86.3)	54–168 (111.2)	61–140.8 (98.4)
N	VFA (mg/L)		17.0–58.2 (39.8)		
	TN (mg/L)	23–50 (36.1)	25–38 (31.3)	25.5–38 (35.9)	27.5–34 (30.8)
P	NH ₄ -N (mg/L)	12.9–29.2 (22.7)	13.2–24 (19.3)	13.8–26.6 (20.4)	7.5–26.4 (19.5)
	TP (mg/L)	2.7–5.5 (3.9)	2.8–4.0 (3.3)	1.6–5.1 (3.4)	3.5–5.8 (3.8)
SCOD/N/P ratio	PO ₄ -P (mg/L)	1.8–4.4 (3.0)	1.2–3.0 (2.8)	0.5–3.8 (2.1)	1.4–4.3 (2.9)
		23.8:9.3:1	26.2:9.5:1	32.7:10.6:1	25.9:8.1:1
Influent flow rate (L/d)	30				
MLSS (mg/L TSS)	Aerobic MBR	3,280–4,750 (3,940)	2,560–5,320 (4,050)	3,280–5,560 (3,870)	4,960–7,570 (5,560)
	Anoxic MBR	2,310–3,840 (3,330)	2,950–4,810 (3,850)	3,270–5,140 (4,180)	4,380–7,010 (5,784)
SRT (d)	Aerobic MBR	8.9–37.5 (20.6)	5.2–53.8 (24.8)	15.8–32.5 (21.5)	18.5–68 (29.3)
	Anoxic MBR	9.4–37.5 (23.4)	12.5–40.8 (25.8)	10.5–35.8 (20.6)	20–56.8 (30.2)

*Recycle from Ox to 1st AX.

**Recycle from 2nd AX to AN.

resulted in varying reaction residence times. It is important to process variables in biological systems with an ordinary settling tank in which solids–liquid separation is of critical importance. However, the effluent solids concentration is not a technical concern

in the MBR system, as shown in Fig. 2. The effluent TSS concentration was close to zero during the experiment because of the membrane separation. The supremacy of the membrane separation often overlooked the importance of the sludge properties in the

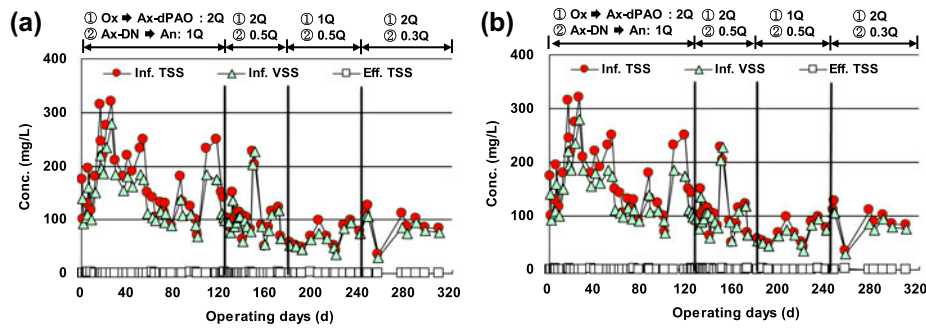


Fig. 2. Influent and effluent solids concentration in (a) the 4-stage aerobic MBR and (b) the 4-stage anoxic MBR.

membrane operation, especially in the fouling prevention. The sludge settling properties often surrogate the overall characteristics of the biological sludge.

The typical settling characteristics (Fig. 3(a)) and particle size distribution (Fig. 3(b)) in typical sludge samples taken from the 2nd AX and OX zone in the anoxic MBR and the OX zone in the aerobic MBR are illustrated in Figs. 3 and 4. In the BNR system, sludge settling characteristics are an important factor in determining the effluent quality. However, most MBR systems did not monitor the biological property of the sludge itself because of the powerful separation ability of the membrane. As shown in Fig. 3, the sludge settling characteristics measured by the sludge volume index (SVI) were different for the two systems. The SVI values of the OX zone in the 4-stage aerobic MBR and anoxic MBR were 79 and 106 mL/g, respectively. The initial settling velocity of OX sludge in the aerobic MBR was faster than OX and AX sludge in the anoxic MBR (Fig. 3(a)). The sludge settleability in the aerobic MBR was better than in the anoxic MBR. A particle size distribution test was conducted to investigate the cause of the different settling characteristics between the aerobic and anoxic MBR. The particle size at the maximum volume (4.76%) and the mean particle size (150.8 μm) in the aerobic MBR were larger than those in the anoxic MBR (Fig. 3(b)).

In the anoxic MBR, the aeration rate for nitrification was 2 L/min air, while N₂ gas was purged at the rate of 2 L/min N₂ gas to prevent fouling in the anoxic membrane. In the aerobic MBR, however, 2.5 L/min air was provided for aeration and fouling prevention. The particle size seems to affect the settleability in the anoxic MBR, although the effluent SS concentration was almost zero in both the aerobic and anoxic MBRs. These results were in agreement with the observations in study [16], who reported that the particle size was inversely proportional to the SVI. We also confirmed that the smaller floc size was related to a higher fouling tendency because the pore blocking resistance was increased [16–18]. These results indicate that sludge settleability is significantly related to the membrane fouling.

3.2. Membrane fouling of the anoxic MBR

The membrane fouling in the anoxic zone seems to be a significant factor affecting operation. For instance, in a comparative study on an identical sequencing batch reactor (SBR)-like MBR operated under oxic and anoxic conditions, Yun et al. [19] reported that the anoxic MBR exhibited severe fouling because of the rapid increase in the TMP. These researchers claimed

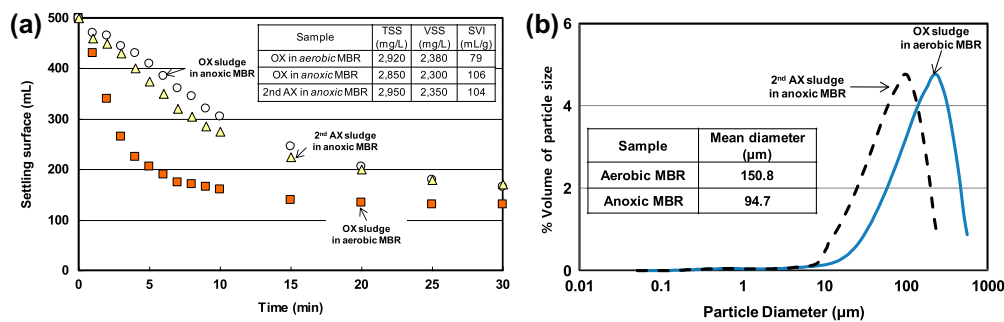


Fig. 3. The results of (a) the SVI test and (b) the particle size distribution in the aerobic MBR and anoxic MBR.

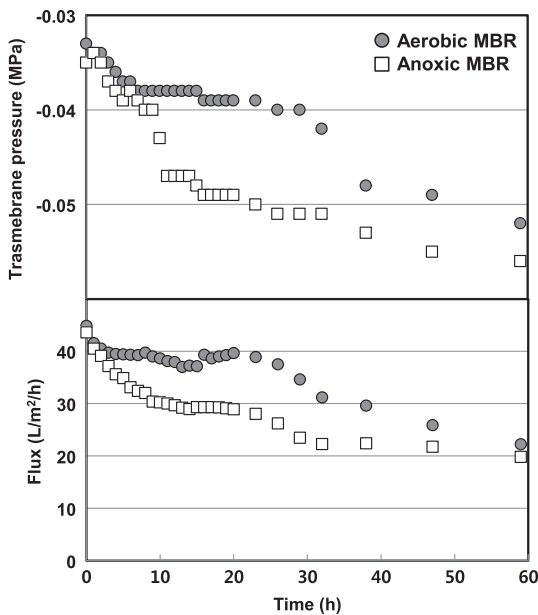


Fig. 4. The comparison of membrane filterability between the aerobic and anoxic MBRs.

that the rate of membrane fouling for the anoxic MBR was five times faster than that of the aerobic MBR, which was fed with synthetic wastewater. If the membrane installation in the anoxic zone suffers from severe fouling, it cannot be used in practice.

However, the membrane fouling in this experiment was not as severe as that in previous study by Yun et al. [19]. Fig. 4 shows the TMP and membrane flux in the aerobic and anoxic MBRs in the current study. The average flux in the aerobic MBR was 36.9 L/m²/h, while the flux in the anoxic MBR was 29.9 L/m²/h. The arrival time limiting the TMP (−0.05 MPa) in the anoxic MBR was approximately two times faster than that in the aerobic MBR. The TMP and flux for the initial 10 h decreased at the same rate in both the anoxic and aerobic MBRs, but the permeate of the anoxic membrane rapidly dropped within the following 10 h. Sato and Ishii [20] reported that SCOD had the largest effect on membrane fouling, indicating that rapid drop on TMP and flux at initial 10 h was significantly related to the SCOD inflow by step feed system to the anoxic reactor of the anoxic MBR.

After 35 h, the gap in the TMP and flux between the anoxic and aerobic MBRs was narrowed down. Unlike the single MBR process examined by Yun et al. [19], the continuous flow BNR–MBR system seemed to have less influence due to anoxic-mediated membrane fouling. Although Yun et al. [19] reasoned that the nature of the biofilm was due to the different DO responsible for the poor fouling in the anoxic membrane, this does not seem applicable in our study. The

anoxic membrane operation in this study is therefore tolerable in terms of flux compared with the aerobic operation. Further investigation on the sludge characteristics is required to reduce the adverse anoxic fouling in the BNR–MBR system.

3.3. Effluent water quality

Fig. 5 presents the average concentration of influent and effluent for various operating conditions. The SCOD concentration of effluent in the aerobic MBR was similar to that in the anoxic MBR. Effluent TN in the anoxic MBR was close to 5 mg/L in Phase 4. Although the anoxic MBR could produce relatively lower TN effluent than the aerobic MBR (average 7.1 mg/L), the effluent from the anoxic MBR contained more NH₄⁺-N (69% of the effluent N). Very low DON was measured in the anoxic MBR compared with the aerobic MBR (Fig. 6). Dissolved organic nitrogen (DON), which is not converted to inorganic N, is difficult to remove in WWTPs [21]. WWTPs have not considered DON removal in the BNR process because DON represents a small portion of effluent. For WWTPs facing stringent effluent TN regulations, the portion of DON in the effluent could be a critical factor. Pagilla et al. [21] reported that DON constituted 9–50% of effluent TN. Thus, the anoxic MBR may contribute to reducing the DON concentration of the effluent. Effluent TP in the aerobic MBR was achieved at nearly 0.5 mg/L in Phase 3. The aerobic MBR produced a higher quality effluent TP compared to anoxic MBR because the aerobic MBR utilized phosphorus-accumulating organisms (PAOs) in the OX zone.

3.4. The effect on recycle ratio

Fig. 7 shows the effect on effluent TN and TP of variations in the nitrified and anoxic recycle ratios. As the nitrified recycle ratio was decreased from 2Q to 1Q, the effluent TN concentration increased and the effluent TP declined because the amount of NO₃-N recycled to the 1st AX zone increased and the potential for denitrification increased (Fig. 7(a)). When the denitrified recycle ratio was decreased from 1Q to 0.3Q, the effluent TN and TP concentrations decreased together. In particular, the TP concentration was significantly reduced because the amount of NO₃-N recycled to the AN zone decreased (Fig. 7(b)). The tendency of effluent TN and TP in the anoxic MBR was similar to that in the aerobic MBR (Fig. 7(c) and (d)). The results show that TN removal was significantly related to the nitrified recycle ratio and that the effluent TP concentration was proportional to the anoxic recycle ratio.

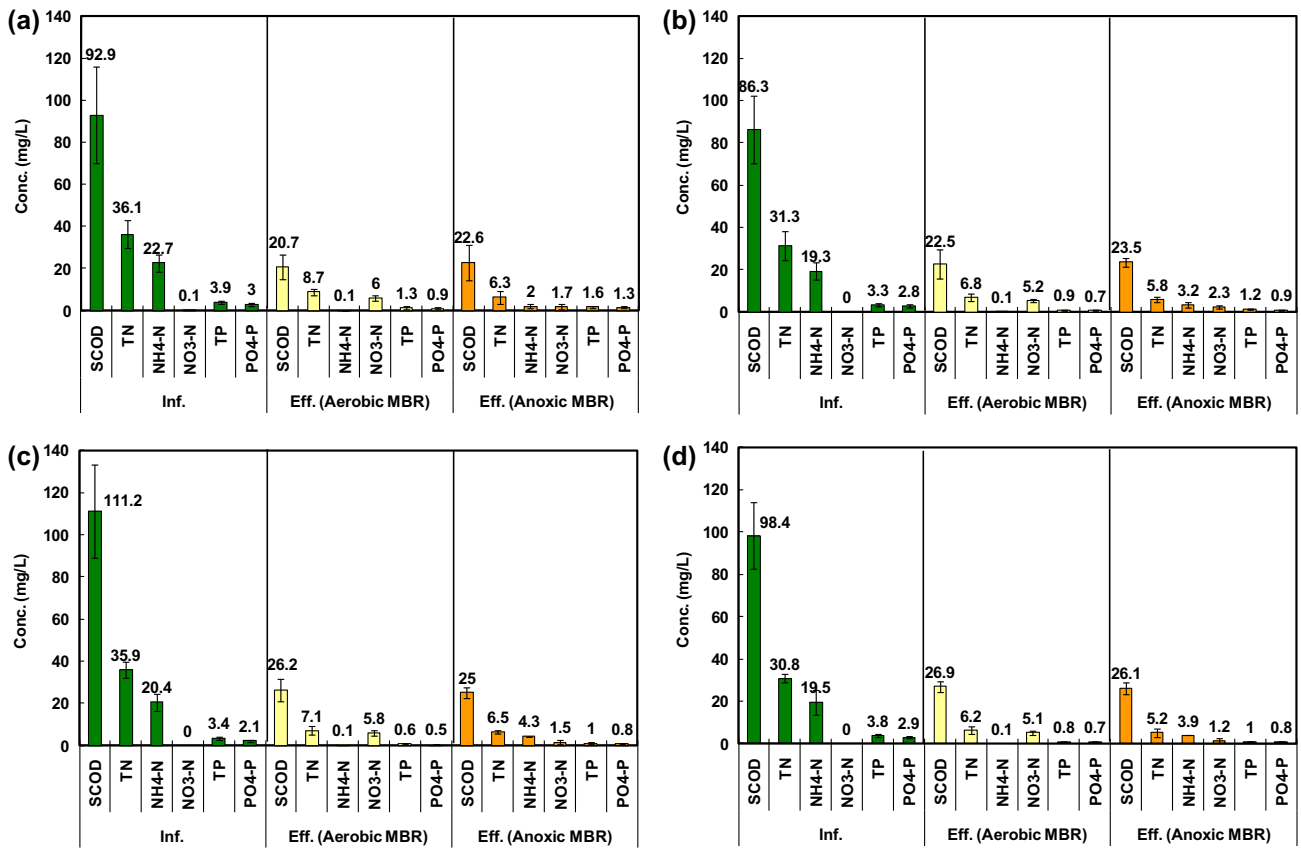


Fig. 5. Influent and effluent concentration of 4 operating phases: (a) Phase 1, (b) Phase 2, (c) Phase 3, and (d) Phase 4.

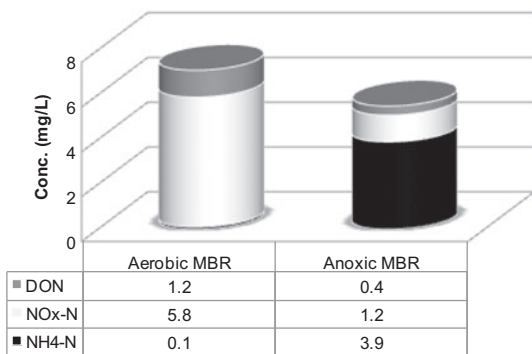


Fig. 6. The effluent characteristics of the TN.

3.5. Carbon requirement for N and P removal

Fig. 8 represents calculated mass balance in 4-stage aerobic MBR and anoxic MBR. In aerobic MBR, 2,670 mg SCOD was fed to the AN zone. About 54.3% of total SCOD was consumed so that 189 mg P was released and 10 mg NO₃⁻-N was also denitrified at anaerobic condition (Fig. 8(a)). The

amount of carbon utilization for P release was 7.7 g SCOD_{utilized}/g P_{released}. After anaerobic condition, 80 mg NO₃⁻-N was reduced as an electron acceptor of DPAO when 111 mg was reduced as an electron acceptor-P was accumulated in 1st AX zone. The amount of PO₄³⁻-P accumulated for unit PO₄³⁻-N reduction was 1.4 g P_{uptaked}/g N_{denitrified}. The ratio between anoxic P uptake and oxic P uptake was 1.7:1, indicating that denitrification as well as P uptake in the 1st AX zone was successfully achieved by DPAO. Almost full nitrification occurred in OX zone (Fig. 8(b)).

In anoxic MBR, most of the SCOD (1,357 mg, 50.8% of influent) was removed with 201 mg P release at anaerobic condition (Fig. 8(c)). The amount of carbon utilization for P release was 6.8 g SCOD_{utilized}/g P_{released}, which was lower than the result of the aerobic MBR. The amount of PO₄³⁻-P accumulated for unit NO₃⁻-N reduction was 2.1 g P_{uptaked}/g N_{denitrified} (Fig. 8(d)). It means that DPAO might contribute towards PO₄³⁻-P accumulation with denitrification in anoxic condition. The amount of carbon requirement for both N and P removal in aerobic and anoxic MBR

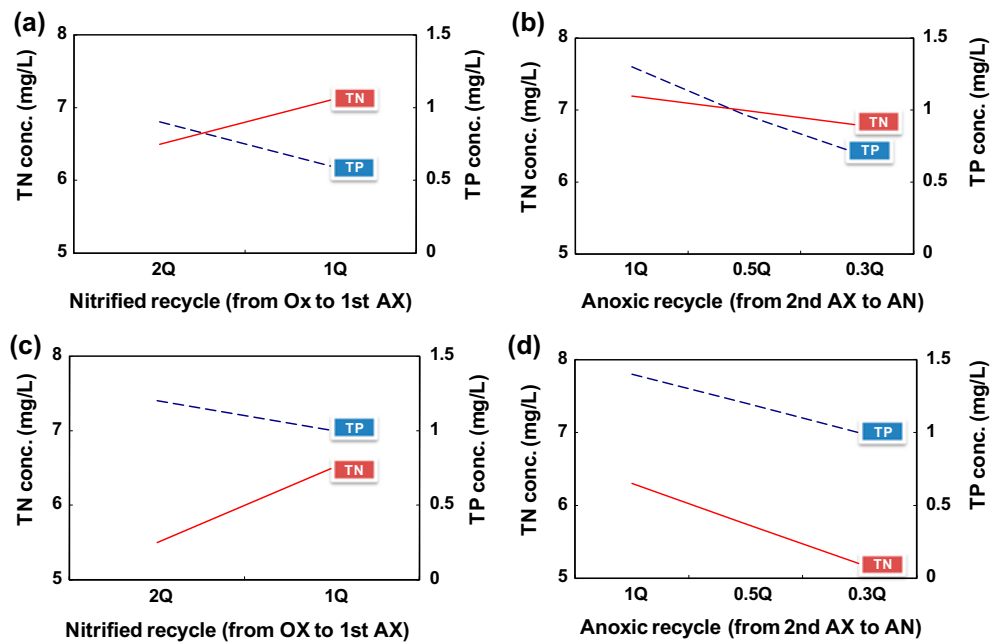


Fig. 7. The effect of the recycle ratio. (a) Nitrified recycle and (b) anoxic recycle in the aerobic MBR and (c) nitrified recycle and (d) anoxic recycle in the anoxic MBR.

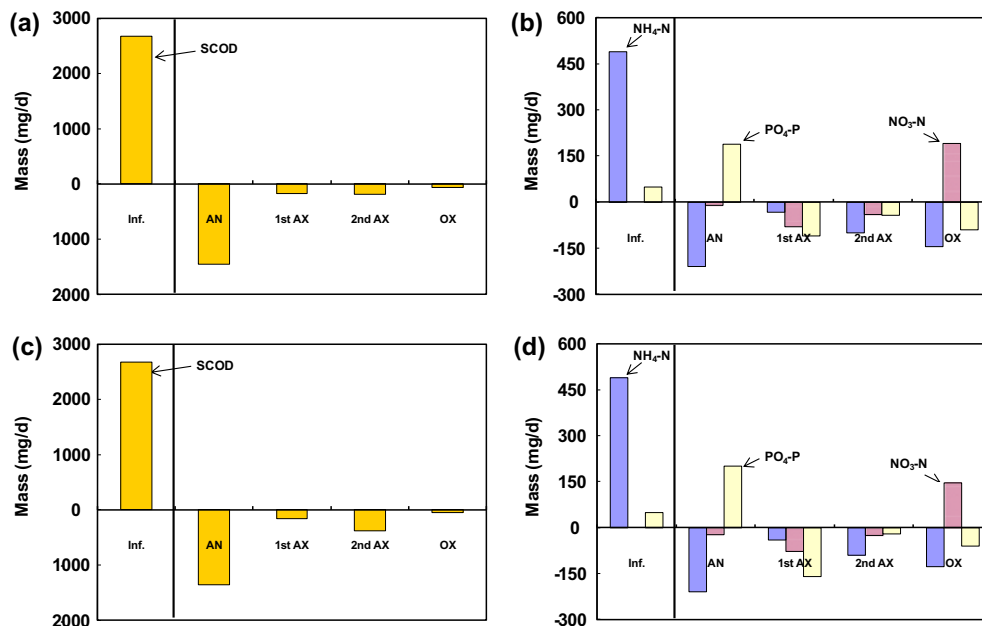


Fig. 8. Mass balance of (a) SCOD, (b) NH₄⁺-N, NO₃⁻-N, and PO₄³⁻-P in aerobic MBR, (c) SCOD, and (d) NH₄⁺-N, NO₃⁻-N, and PO₄³⁻-P in anoxic MBR.

was calculated at 2.54 and 2.46 g SCOD/g (N_{denitrified} + P_{released}), respectively. The amount of carbon utilization in various DPAO systems such as DEPHANOX and A₂N was in the range from 3.1 to

3.6 g SCOD/g (N + P) [22–25]. These results show that DPAO could effectively utilize carbon energy (= COD) in influent.

4. Conclusions

A novel 4-stage anoxic MBR was developed to achieve the performance of the LOT levels for sewage under laboratory conditions, and its performance was compared with an aerobic MBR as a control unit. From the results of SVI and the particle distribution, the sludge settleability in the aerobic MBR was better than that in the anoxic MBR, although the SVI value (104 mL/g) of the anoxic MBR was in the range of properly settling sludge. Particle size seems to affect the settleability in the anoxic MBR. The average flux of the anoxic MBR (29.9 L/m²/h) was approximately 4/5 of that of the aerobic MBR (36.9 L/m²/h). The anoxic membrane operation is tolerable in terms of flux compared with the aerobic operation.

An acceptable effluent TN concentration in the anoxic MBR was successfully achieved (nearly 5 mg/L) because of the lower NO₃⁻-N and DON concentrations in the effluent. However, the NH₄⁺-N concentration was another source of effluent N that needed to be lowered. In contrast, the aerobic MBR produced a higher quality effluent TP (0.5 mg/L) than did the anoxic MBR (0.9 mg/L) without chemical treatment. The effect on the recycle ratio shows that the effluent TN concentration correlated with the nitrified recycle ratio, while the effluent TP was proportional to the anoxic recycle ratio. The amount of carbon requirement for both N and P removal in aerobic and anoxic MBR was calculated in 2.54 and 2.46 g SCOD/g (N_{denitrified} + P_{released}), respectively. The anoxic MBR facilitated an enhanced N removal process under an unfavorable C/N/P ratio, but the improvement of P removal was required to meet the stringent water quality standard without chemical P removal. The trade-off between the aerobic and anoxic MBR may offer a selective option for the existing state of affairs.

Acknowledgment

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