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Effect of dissolved oxygen on the performance of a step-feed hybrid MBR process treating synthetic domestic wastewater

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ABSTRACT

An innovative step-feed Anaerobic-(Oxic/Anoxic)^n-Membrane Bioreactor [An-(O/A)^n-MBR] process was developed to treat synthetic domestic wastewater. Performance of the lab-scale system was investigated at different dissolved oxygen (DO) concentration (0.4–2.4 mg/L) in the aerobic tank of the multiple A/O zone. The results showed that, under the conditions imposed, the DO level has little influence on chemical oxygen demand removal and the removal efficiency was more than 94% throughout the operation. However, DO levels have great influence on nitrogen and phosphorus removal. Better nitrification efficiency could be obtained when DO was in the range of 0.8–2.4 mg/L; the ammonia removal efficiency was more than 99%. High total nitrogen (TN) and total phosphorus (TP) removal performance can be obtained when DO was in the range of 0.8–1.2 mg/L; the average removal efficiency was 74.8 and 71.4%, respectively. In this condition, DO can meet the demand of nitrification and phosphorus uptake simultaneously, and the simultaneous nitrification and denitrification occurred in the aerobic tanks under lower DO concentration. Meanwhile, since the DO circulated from the aerobic tank to the anoxic tank decreased, denitrification was enhanced and the nitrate quantities in the sludge recycle system decreased, resulting in the decrease of carbon substrate competition between denitrification and phosphorus release in the anaerobic zone. Ultimately, the performance of TN and TP removal was enhanced.

Keywords: Dissolved oxygen (DO); Step-feed; Nitrogen and phosphorus removal; Membrane bioreactor; Nitrification/denitrification

1. Introduction

Nitrogen (N) and phosphorus (P) are the key nutrients causing eutrophication in waterways [1]. In the past decades, a number of biological nutrient removal (BNR) processes had been developed [2–5]. Technically, the developed BNR processes, including anaerobic, anoxic, and oxic phases, could be divided into two types according to the implementing approaches of the three phases: the temporal BNR process and the spatial BNR process. The temporal BNR process, such as the sequenced batch bioreactors, achieved the anaerobic, anoxic, and oxide conditions by arranging them temporarily in a single reactor. As compared to the conventional activated sludge systems, the temporal BNR system had many advantages such as smaller footprint, improved nitrogen and phosphorus removal, less bulking, and flexible operation mode [6]. However, since all the phases occurred in a single reactor, the temporal BNR process always operated in a discontinuous flow system. In a continuous flow system, the spatial BNR process, such

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as anaerobic/anoxic/oxic (AAO) process, was widely used throughout the world. This type of BNR process assigned different zones for each period (with sludge recycling serving as inoculums) to obtain the three conditions spatially. However, most of the spatial BNR process required additional energy for internal nitrified liquor circulation or addition of external carbon substrate for denitrification in anoxic zones, which led to the increase of the operational cost.

Step-feed anoxic/oxic-activated sludge process (SAOASP) was one of the most practical methods to solve these problems because of its elimination of internal recycling and optimizing organic carbon utilization for denitrifcation [7–9]. The SAOASP processes were widely studied for nitrogen removal. To improve the phosphorus removal, SAOASP system was improved by adding an anaerobic zone before the multiple stages of aerobic–anoxic zones [10].

For decentralized, sensitive and yet unsewered areas, membrane bioreactor (MBR) technology could provide an elegant, robust, and cost-effective treatment solution to achieve high effluent standard. When combined with enhanced biological phosphorus removal (Bio-P) and/or phosphorus coprecipitation (Co-P), high and stable phosphorus removal can be expected [11]. However, and in contrast to conventional activated sludge plants, process optimization still has to be done.

On the basis of above knowledge, the aim of the current work, therefore, was to develop a step-feed An-(O/A)^n-MBR [Anaerobic-(Oxic/Anoxic)^n-Membrane Bioreactor] process for nutrient removal. This process, combining SAOASP with membrane separation, was composed of an anaerobic reactor, a multiple phases of aerobic and anoxic zones in sequence followed by a continuous aerated MBR. Different dissolved oxygen (DO) concentration (0.4–2.4 mg/L) in the aerobic tank of the multiple A/O zone were investigated on a lab-scale system. Nutrient removal performance was compared at the different conditions imposed.

2. Materials and methods

2.1. An-(O/A)^n-MBR system

The experimental setup was shown in Fig. 1. The lab-scale An-(O/A)^n-MBR system was composed of an anaerobic reactor, a multiple phases of aerobic and anoxic zones in sequence (multiple A/O zone) followed by a continuous aerated MBR. The working volume for individual reactors was 14.6 L (anaerobic zone), 34.6 L (multiple A/O zone), and 23.3 L (Membrane zone), respectively. The water depth in each reactor was about 44.6 cm. The multiple phases of aerobic and anoxic zones consisted of four uniform aerobic compartments and four uniform anoxic compartments, the aerobic and the anoxic compartments were arranged alternately, and the volume ratio of aerobic zone to anoxic zone was 2:1.

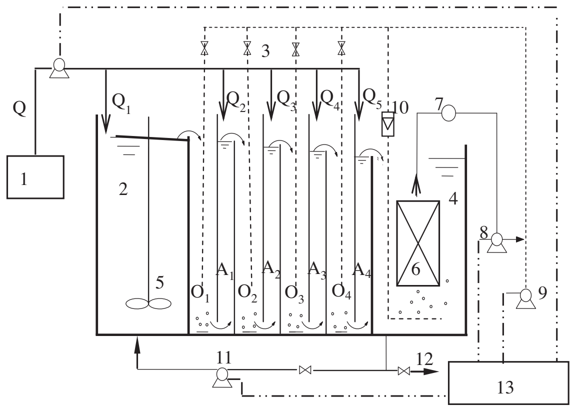

Fig. 1. Schematic diagram of the An-(O/A)^n-MBR system. (1) Wastewater reservoir; (2) anaerobic reactor; (3) multiple phases of aerobic and anoxic zones; (4) MBR tank; (5) agitator; (6) membrane module; (7) pressure gauge; (8) peristaltic pump; (9) air blower; (10) air flow meter; (11) return sludge; (12) excess sludge; (13) PLC system (O—aerobic compartment, A—anoxic compartment).

The synthetic influent (Q) was fed into the system by step-feeding mode. The first wastewater flow (Q_1) supplying nutrients for micro-organisms growth and carbon for phosphorus release was fed into the anaerobic reactor while the second flow (Q_2) combined with the third flow (Q_3), the forth flow (Q_4), and the fifth flow (Q_5) were fed into the anoxic zone by stepwise feeding. For the duration of the experimental period, the DO concentrations in the aerobic tanks of the multiple A/O zone were controlled at different levels by adjusting the valves of aeration pipes.

A microfiltration hollow fiber membrane module (MOTIMO, China) was immersed in the MBR tank. The membrane module had an effective filtration area of 1.0 m^2 and its nominal pore size is 0.22 μm. An air diffuser was installed underneath the membrane module to provide air at 0.5 m^3/h. The bubbling air was used to serve for three purposes, providing oxygen for the micro-organisms in aerobic tank, mixing the aerobic tank, and removing of cake deposition on the membrane surface. The mixed liquor at the bottom of MBR tank was recycled to the anaerobic reactor continuously at a rate of $0.75 * Q$. According to the results of previous studies [12], the membrane flux was maintained at 10 L/(m^2 h). To alleviate membrane fouling, the membrane was operated in an intermittent mode (10 min suction and 2 min rest). The transmembrane pressure (TMP) was measured using a pressure gage, and the membrane was cleaned when TMP reached 30 kPa.

2.2. Influent quality

Synthetic wastewater, composed of glucose (250 mg/L), soluble starch (150 mg/L), NH_4Cl (100 mg/L), KH_2PO_4(20 mg/L), $NaHCO_3$ (100 mg/L), and mineral solution (Mg, Ca and Fe), was stored in a wastewater container and was fed to the lab-scale system throughout the operation. The initial influent contained 340.5–419.0 mgCOD/L, 31.5–35.6 mgTN/L, 27.4–32.7 mg NH_4–N/L, and 3.9–5.7 mg TP/L. The pH value was about 7.3.

2.3. Operation

In this study, the synthetic wastewater was fed into the system with a flow rate of 0.20 m^3/d, 70% of the influent (Q_1) was fed to the anaerobic zone and the rest were fed to the bottom of the four anoxic compartments in the multiple A/O zone ($Q_2 = Q_3 = Q_4 = Q_5$). In the aerobic tanks of the multiple A/O zone, the air flow rates were adjusted according to the DO levels required in different operation runs. In this study, four different DO levels (2.0–2.4 mg/L, 1.3–1.7 mg/L, 0.8–1.2 mg/L, and 0.4–0.6 mg/L) in the aerobic tanks were implemented by adjusting the valves of aeration pipes. The total hydraulic retention time was 8.70 h. The mixed liquor suspended solids (MLSS) concentration in the multiple A/O zone was kept at 3,800–4,500 mg/L and the excess sludge was withdrawn periodically to keep the sludge retention time (SRT) at about 60 d. The operation temperature was at 26.5–32.5℃ during the operation. Table 1 shows the operation conditions of these experiments.

2.4. Analytical methods

All the results presented were obtained from the An-(O/A)^n-MBR system at its steady state. The samples taken from bioreactors were filtered using 0.45 μm filter paper. DO concentration was measured using the DO meter (WTW Oxi 340, Germany). The pH was measured using the pH meter (PHSJ-4A, China). Particle size distributions (PSDs) in mixed liquor were analyzed using a Laser Particle Size analyzer (WICS-50, ANKERMID, Dutch). Measurement of chemical oxygen demand (COD), MLSS, total nitrogen (TN), oxidized nitrogen (NO_3^-–N and NO_2^-–N), ammonium nitrogen (NH_4–N), orthophosphate concentration (ortho-P), and total phosphorus (TP) followed standard methods [13].

Table 1
Specifications of the experimental conditions

Conditions	Run1	Run 2	Run 3	Run 4
Experimental period (d)	1–21	22–45	46–66	67–87
DO in the aerobic tanks (mg/L)	2.0–2.4	1.3–1.7	0.8–1.2	0.4–0.6
SRT (d)	60			

3. Results and discussion

Performances of COD, TN, and TP removal in the An-(O/A)^n-MBR system are presented in Table 2.

3.1. COD removal

Daily COD variation of the influent and effluent was studied during the operation. It seemed that DO in the aerobic tanks of the multiple A/O zone had little influence on the organic pollutants removal in the An-(O/A)^n-MBR system. As shown in Table 2 and Fig. 2, although the influent COD fluctuated from 340.5 to 419.0 mg/L, the COD removal efficiency was high and stable, and was 94.7–97.7%, 95.2–97.6%, 94.3–98.1%, and 94.7–97.5% in Run 1, Run 2, Run 3, and Run 4, corresponding to 8.8–20.2 mg/L, 8.9–17.7 mg/L, 6.8–21.7 mg/L, and 9.9–19.7 mg/L in the effluent. The effluent COD level in the An-(O/A)^n-MBR process was sufficient to meet the standard of water reclamation in China.

In this study, the feed wastewater was stored in a 150 L container. To prevent the possible degradation of readily degradable soluble matter (COD) occurring within the container, the container was rinsed thoroughly every morning and then the fresh synthetic wastewater was poured into the container. Therefore, the high COD removal might be attributed to the growth of high biomass concentration (3,800–4,500 mg/L) in the An-(O/A)^n-MBR system and the efficient utilization of organic compounds in the anaerobic reactor for phosphorus release and in the anoxic zones for denitrification. In addition, perfect retention of the suspended COD and biomass by membrane filtration also guaranteed a low level of COD concentration in the effluent [14].

3.2. Nitrogen removal

Effects of DO levels in the aerobic tanks of the multiple A/O zone on the TN removal and on the

Table 2
Characteristics of nutrient removal in each experiment (with standard deviations in parentheses)

Items	Run 1 (DO = 2.0–2.4 mg/L)			Run 2 (DO = 1.3–1.7 mg/L)			Run 3 (DO = 0.8–1.2 mg/L)			Run 4 (DO = 0.4–0.6 mg/L)		
	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)
COD	388.3 (19.4)	15.5 (3.3)	96.0 (0.9)	372.6 (19.1)	13.7 (3.0)	96.3 (0.8)	373.2 (17.5)	14.0 (4.6)	96.3 (1.2)	378.6 (21.5)	13.7 (3.1)	96.4 (0.9)
TN	32.0 (1.7)	14.7 (1.8)	54.2 (4.2)	31.9 (1.3)	9.17 (1.48)	71.2 (4.4)	31.7 (1.7)	8.0 (1.41)	74.8 (4.6)	32.1 (1.2)	14.0 (1.9)	57.1 (6.3)
TP	4.3 (0.4)	2.1 (0.4)	51.3 (4.6)	4.4 (0.5)	1.4 (0.2)	68.5 (5.6)	4.4 (0.5)	1.2 (0.2)	71.4 (4.6)	4.6 (0.4)	1.8 (0.3)	60.9 (5.3)

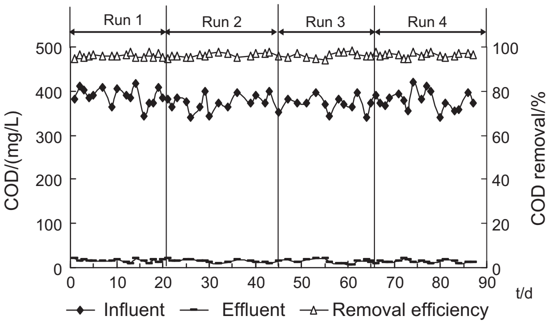

Fig. 2. Effect of DO concentration on COD removal.

nitrogen component in the effluent are presented in Figs. 3 and 4, respectively.

In the An-(O/A)^n-MBR process, the special process configuration allowed the nitrification stream flow from oxic tank of one pass to anoxic tank of next pass straightly in the multiple A/O zone, so the alkalinity consumed in nitrification process could be partially offset in the following denitrification process, achieving a relatively stable pH variation (6.8–7.4) of the mixed liquor in the multiple A/O zone, so it could be inferred that pH was not a limiting factor in the nitrogen removal process.

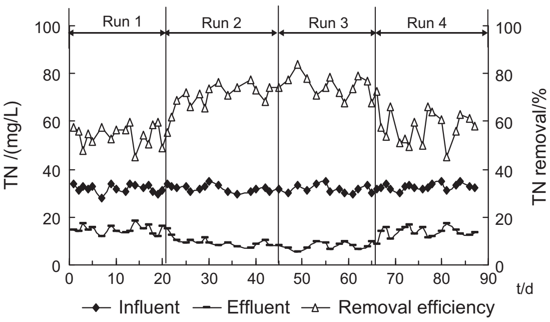

Fig. 3. Effect of DO concentration on TN removal.

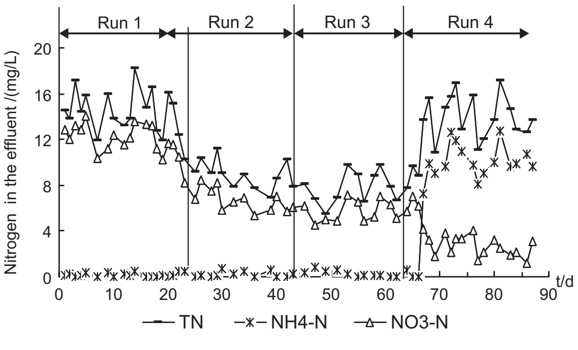

Fig. 4. Effect of DO concentration on nitrogen composition in the effluent.

In Run 1, as shown in Fig. 3 and Table 2, the DO in the aerobic tank in the multiple A/O zone was set at 2.0–2.4 mg/L, poor TN removal performance was obtained and the removal efficiency was in the range of 45.1–59.4% (only 54.2% on average). Data obtained in Run 1 also showed that little ammonia nitrogen was detected and it even became undetectable in the effluent (shown in Fig. 4), implying that nitrification was almost perfectly completed in the An-(O/A)^n-MBR process. Therefore, the efficiency of TN removal would mainly depend on the degree of the denitrification in the system. In this study, since the influent quality was controlled at a steady state (as shown in Table 2), the composition of organic matter, as well as the carbon to nitrogen ratio (C/N), was kept at a relatively stable level. Therefore, it could be concluded that influent quality had no great effect on nitrogen removal in this experiment. Then, the poor denitrification performance in Run 1 might be caused by two factors. (1) High DO levels increased oxygen quantity circulated from the aerobic tank to the anoxic tank, so more organic compounds in the anoxic tank were consumed by aerobic bacteria. As a result, the denitrification could not be achieved completely because of deficiency of organic substances. (2) High DO levels destroyed the microenvironment in the aerobic tank in which the simultaneous nitrification and denitrification (SND) occurred under lower DO concentration. Therefore, the denitrification capacity was decreased accordingly.

When DO was set at 1.3–1.7 mg/L in Run 2, nitrate concentration in the effluent decreased obviously and TN removal increased to 71.2% on average. When DO was controlled at 0.8–1.2 mg/L (Run 3), ammonia nitrogen concentration in the effluent was still at a low level, indicating that the DO level imposed in Run 3 could still meet the DO needs of nitrification. Meanwhile, nitrate concentration in the effluent continued decrease and the average TN removal increased to 74.8%. When DO was decreased to 0.4–0.6 mg/L (Run 4), ammonia nitrogen concentration in the effluent increased rapidly, the nitrite nitrogen was lower than 0.5 mg/L and TN removal was below 60% accordingly, obviously indicating that the DO level imposed in Run 4 could not meet the oxygen requirement for nitrobacteria and nitrification was inhibited.

To evaluate the nitrogen removal mechanism in the An-(O/A)^n-MBR system, the typical variations of TN, ammonia nitrogen and nitrate nitrogen in each tank at different DO settings were studied. Samples for dynamic studies were collected regularly from different zones of the reactors. The data presented in Fig. 5 are obtained at 17 d, 40 d, 61 d, and 83 d in Run 1, Run 2, Run 3, and Run 4, respectively. As illustrated in Fig. 5(a), TN concentration in each tank gained a stable level in Run 1, a decrease trend in Run 2 and Run 3, and an increase trend after the tank O_3 in Run 4. Nitrate nitrogen level was high in each tank in Run 1, as shown in Fig. 5(b), and its concentration in anoxic tank accumulated along the bulk liquor flow, indicating poor denitrification performance achieved under the higher DO level imposed in Run 1. Compare to Run 1, performance of nitrogen removal was better in Run 2 and Run 3. The nitrogen compounds in the effluent existed mostly in the form of nitrate nitrogen, showing that nitrification was almost perfectly completed. Moreover, TN concentration in the aerobic tanks was lower than that in the previous anoxic tanks, indicating the SND occurred in the aerobic tanks. As presented in Fig. 5(c), there was a high level of ammonia nitrogen in each tank in Run 4, and its concentration in aerobic tank accumulated along the bulk liquor flow, demonstrating poor nitrification performance achieved under the low DO level imposed in Run 4. Studies showed that endoge-

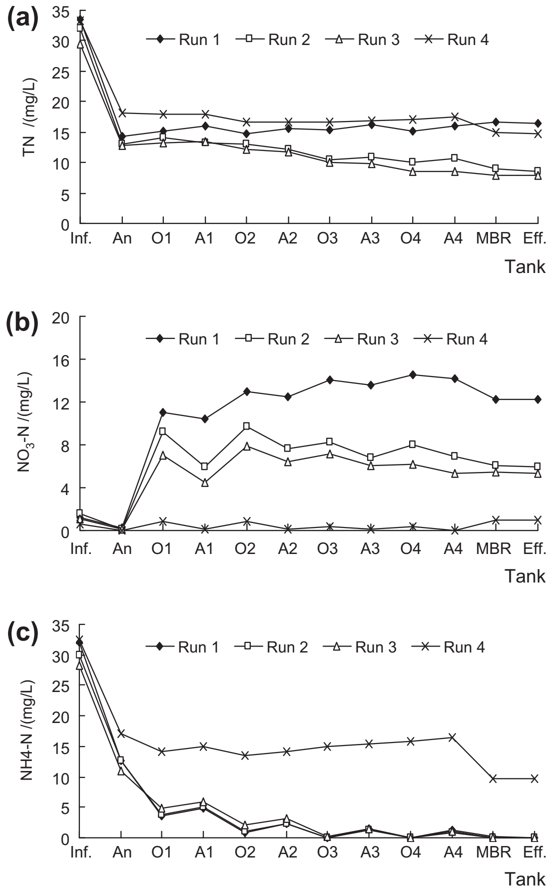

Fig. 5. Typical variation of nitrogen concentration in each tank (a) TN, (b) NH_4–N, and (c) NO_3–N.

nous respiration of micro-organisms could produce biodegradable organic nitrogen which could be transformed into ammonia nitrogen ultimately, resulting in the TN increase in the effluent [15]. This could be used to explain why the ammonia nitrogen increased in the last two anoxic tanks (A_3 and A_4) in this run.

Therefore, DO level in the aerobic tanks of the multiple A/O zone had great influence on the nitrogen removal in the An-(O/A)^n-MBR system. Studies showed that high performance on nitrogen removal could be achieved when DO level was at the range of 0.8–1.7 mg/L. Lower DO concentration in the aerobic tank could allow nitrification and denitrification to occur simultaneously, and could decrease oxygen quantity circulated from the aerobic tank to the anoxic tank, resulting in the improvement of TN removal performance and the decrease of energy consumption [16]. Therefore, as far as nitrogen removal concerned, DO concentration in the aerobic tank of the multiple A/O zone was kept at 0.8–1.2 mg/L was more preferable. Previous study showed that, in the An-(O/A)^n-MBR system, aeration in the MBR tank caused a turbulent flow and induced shear forces in the mixed suspension liquor that changed both the composition and the characteristics of the sludge floc [16]. In this study, PSDs of the mixed liquor in MBR tank were analyzed at the 6 d point and at the end of the operation, and a decrease, from 79.65 to 72.58 μm, of the mean particle size was found. Generally speaking, small particle sizes were beneficial for oxygen mass transfer in the mixed liquor, and thus lower DO level required for good performance in a biological wastewater treatment process. However, the optimal DO level (0.8–1.2 mg/L) in this study was much higher than that obtained by Wang et al. [17]. Therefore, the difference might be caused by some other floc properties and/or some factors affecting the properties in the hybrid MBR process, such as the composition of activated sludge, the impact of substrate, the factors affecting bioflocculation, and so on. To understand the mechanisms, further research is required.

3.3. Phosphorus removal

As shown in Fig. 6, under the conditions imposed in the An-(O/A)^n-MBR system, a similar trend in TP removal was observed as for TN reduction, but the difference of TP among four DO settings was less greatly found as compared to that of TN. When TP concentration was set at the range of 3.7–5.2 mg/L, the average TP removal efficiency was 51.3%, 68.5%, 71.4%, and 60.9% in Run 1, Run 2, Run 3, and Run 4, respectively. According to the typical variation of TP concentration in each tank (shown in Fig. 7), the TP concentration in the anaerobic tank was 7.1 mg/L, 11.6 mg/L, 13.3 mg/L, and 8.8 mg/L in Run 1, Run 2, Run 3, and Run 4, corresponding to 2.6 mg/L, 1.4 mg/L, 1.0 mg/L, and 2.1 mg/L in the effluent.

Studies showed that both too low (0.4–0.6 mg/L in Run 4) and too high (2.0–2.4 mg/L in Run 1) DO level had negative effects on phosphorus removal. In Run 4, DO concentration in the anoxic tank was very low (close to 0 mg/L) for the decrease of DO circulated from the aerobic tank to the anoxic tank, meanwhile, nitrate concentration in the anoxic tank was also very low due to the DO restraint in nitrification process in the aerobic tanks and the nitrate consumption in denitrification process. Therefore, strict anaerobic condition appeared in the anoxic tank and phosphate-accumulating organisms (PAOs) could assimilate the organic substances in the raw wastewater distributed into the anoxic tanks and thus phosphorus release was achieved [16]. This might explain why the phosphorus was released repeatedly in anoxic tanks of the multiple A/O zone in Run 4 (shown in Fig. 7). Simultaneously, PHB stored in PAOs could not be decomposed effectively because of DO deficiency in the aerobic tank and thus no enough ATP was produced. This could lead to the restraint of excessive phosphorus uptake and the continuous PHB accumulation in PAOs [18]. Consequently, the mechanism of PHB synthesis was affected adversely, resulting in the decrease of phosphorus release in the anaerobic tank. When DO level was as high as 2.0–2.4 mg/L (Run 1), massive nitrate in return sludge

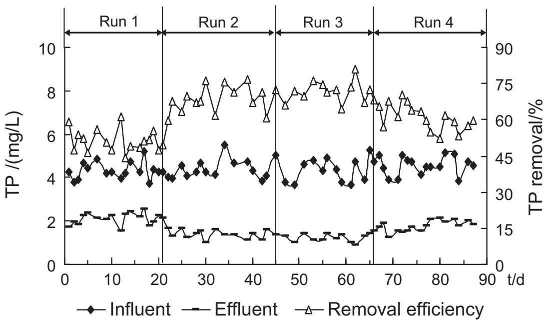

Fig. 6. Effect of DO concentration on TP removal.

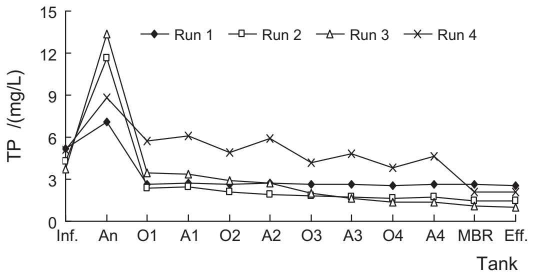

Fig. 7. Typical variation of TP concentration in each tank.

recycle was carried into the anaerobic tank, causing the organic substances competition between denitrification bacteria and PAOs [6,18,19], so the capacity of phosphorus release in the anaerobic tank decreased correspondingly. Furthermore, it was observed that phosphorus was uptaken in the anoxic compartments as well as in the aerobic compartments in this system. Studies show that the presence of a small amount of nitrate may stimulate the growth of denitrifying phosphate-accumulating bacteria (DPB). DBP, a fraction of PAOs, can also take up phosphorus under anoxic conditions using nitrate as the electron acceptor instead of oxygen [20]. Therefore, the decrease of phosphorus concentrations in the anoxic compartments might attribute to the simultaneous denitrification and phosphorus uptake by DPB. Since denitrification in the anoxic tanks was restrained under high DO level condition, the simultaneous denitrification and phosphorus uptake by DPB decreased accordingly [21], resulting in the increase of phosphorus concentration in the effluent.

Therefore, to achieve high performance on phosphorus removal in the system, the DO concentration in the aerobic tanks should be kept at the range of 0.8–1.2 mg/L.

4. Conclusions

An innovative step-feed An-$(O/A)^n$-MBR process was developed to treat synthetic domestic wastewater and its performance was investigated on a lab-scale system. Major findings from this study are summarized as follows:

(1) Combining SAOASP with membrane separation, the step-feed An-$(O/A)^n$-MBR process might be a promising process alternative for wastewater treatment because of its elimination of internal recycling and optimizing organic carbon utilization as well as its high effluent quality. DO level in the aerobic tank of multiple A/O zone was one of the most important parameters affecting the performance of the step-feed An-$(O/A)^n$-MBR process.

(2) DO level in the aerobic tanks of the multiple A/O zone had little influence on the organic pollutants removal in the An-$(O/A)^n$-MBR system. COD removal was high and stable (above 94%) throughout the operation. The effluent COD level (<22 mg/L) was sufficient to meet the standard of water reclamation in China.

(3) DO level in the aerobic tanks caused significant differences in TN and TP removal efficiency in the An-$(O/A)^n$-MBR system. When DO in the aerobic tanks was set at 2.0–2.4 mg/L, 1.3–1.7 mg/L, 0.8–1.2 mg/L, and 0.4–0.6 mg/L in Run1 to Run 4, under the experimental conditions imposed, the average TN and TP removal efficiency in each run was 54.2%, 71.2%, 74.8%, 57.1% and 51.3%, 68.5%, 71.4%, and 60.9%.

(4) Nitrification could be fully achieved when DO was at the range of 0.8–2.4 mg/L and the ammonia nitrogen removal efficiency was above 99% accordingly. High TN and TP removal performance can be obtained when DO was in the range of 0.8–1.2 mg/L. In this condition, DO can meet the demand of nitrification and phosphorus uptake simultaneously, and the SND occurred in the aerobic tanks under lower DO concentration. Meanwhile, the DO circulated from the aerobic tank to the anoxic tank decreased, denitrification was enhanced and the nitrate quantity in the sludge recycle system decreased, resulting in the decrease of carbon substrate competition between denitrfcation and phosphorus release in the anaerobic zone. Ultimately, the TN and TP removal efficiency were enhanced.

Further investigations should be conducted on the effect of selected synthetic wastewater containing different organic substances and real municipal wastewater, the optimization of influent distribution according to different influent as well as on the microbial kinetic analysis of the step-feed An-$(O/A)^n$-MBR process.

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