



Performance of an innovative step-feed An-M(A/O)-MBR process for nutrients removal

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ABSTRACT

An innovative process step-feed Anaerobic-multiple Anoxic/Oxic-Membrane Bioreactor [An-M(A/O)-MBR] was developed for biological phosphorus and nitrogen removal from synthetic domestic wastewater. This process was composed of an anaerobic reactor, a multiple phases of aerobic and anoxic zones (multiple A/O zone) in sequence followed by a continuous aerated MBR. Performance of the laboratory-scale system was investigated at different organic compounds in the influent. The results showed that, under the conditions imposed, although the Chemical Oxygen Demand (COD) concentration fluctuated in the range of 120–1,200 mg/L, high performance on COD removal was achieved in the system and more than 95% removal efficiency was obtained throughout the operation, the COD concentrations in the effluent were lower than 50 mg/L throughout the operation. However, COD levels in the influent had great influence on nitrogen and phosphorus removal. When COD level was low (120–200 mg/L), poor performance of nitrogen and phosphorus removal were obtained because of carbon source deficiency. Good performance on total nitrogen (TN) and total phosphorus (TP) removal were achieved when COD level was in the range of 350–710 mg/L, and the average removal efficiency of TN and TP was above 85 and 84%, respectively. But when COD increased to a high level (1,110–1,200 mg/L), the performance of nitrogen and phosphorus removal also deteriorated possibly because of the shift of the composition of the microbial communities.

Keywords: Nitrogen and phosphorus removal; Step-feed; Organic compounds; Nitrification/denitrification; Membrane bioreactor

1. Introduction

To prevent eutrophication, effective control of nutrients is now required for all wastewater discharges to sensitive receiving waters [1]. Step-feed

anoxic/oxic activated sludge process (SAOASP) is one of the most practical methods for the upgrading of existing sewage treatment plants in terms of nitrogen removal. The step-feed process consists of two or more combinations of anoxic and oxic tanks in series, and receives influent at plural anoxic tanks. Recently, many research papers have focused on the parameters

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optimization, such as the dissolved oxygen (DO), the volumetric ratio of anoxic and oxic tanks, the distribution ratios of the influent, and so on. Good performance for nitrogen removal had been achieved through pilot-scale and full-scale experiments using step-feed processes [2–5].

Compared with other conventional methods, the SAOASP has many advantages [6]: (1) distributing organic substrate equally along with the reactor basin results in a similar F/M ratio for each pass, which, on one hand, decreases the difference between oxygen supplying rate and consuming rate, and on the other hand, stimulates the degradation ability of activated sludge micro-organism; (2) wastewater feeding into reactor gradually heightens the adaptability to shock load of flow rate and concentration of influent; (3) mixed liquor suspended solids (MLSS) decreases along with the length of reactor and the concentration of effluent stream is low, which reduces load of secondary clarifier and improves separation of clarifier; and (4) Grads of MLSS solid and liquid in secondary can come into being because recycled sludge feeds at the beginning of the reactor and influent feeds gradually, which lessens the flushing out of suspended solids at some certain point.

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 phosphorous removal (Bio-P) and/or phosphorous co-precipitation (Co-P), high and stable phosphorous removal can be expected [7].

On the basis of above knowledge, the aim of the current work, therefore, was to develop a step-feed Anaerobic-multiple Anoxic/Oxic-Membrane Bioreactor [An-M(A/O)-MBR] 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. Good performance for nutrients removal had been achieved in previous studies [8,9]. In this study, different organic compounds (120–1,200 mg COD/L) in the influent were investigated on a laboratory-scale system. Nutrient removal performance was studied at the different conditions imposed.

2. Materials and methods

2.1. An-M(A/O)-MBR system description

The experimental setup was shown in Fig. 1. The laboratory-scale An-M(A/O)-MBR system was

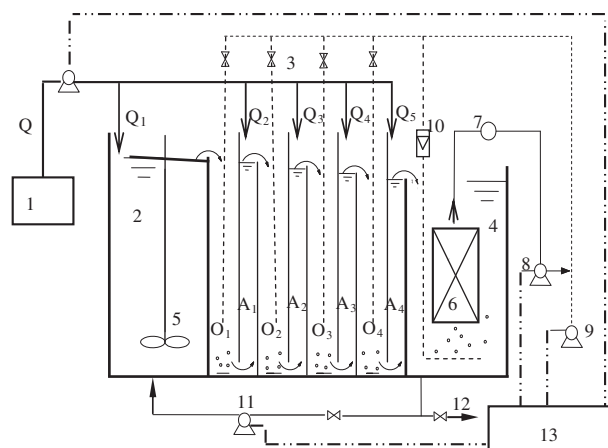


Fig. 1. Schematic diagram of the An-M(A/O)-MBR system. 1. Wastewater reservoir, 2. Anaerobic reactor, 3. Multiple phases of aerobic and anoxic zones, 4. MBR tank, 5. Agitator, 6. Membrane module, 7. Pressure gage, 8. Peristaltic pump, 9. Air blower, 10. Air flow meter, 11. Return sludge, 12. Excess sludge, and 13. PLC system (O— aerobic compartment; A— anoxic compartment).

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, 34.6, and 23.3 L, respectively. The multiple phases of aerobic and anoxic zones consisted of eight compartments, the aerobic and the anoxic tanks were arranged alternately, and the volume ratio of aerobic tank to anoxic tank was 2:1.

The first synthetic 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 fourth flow (Q_4), and the fifth flow (Q_5), was fed into the anoxic zone by stepwise feeding ($Q_1:Q_2:Q_3:Q_4:Q_5 = 6:1:1:1:1$). For the duration of the experimental period, the Chemical Oxygen Demand (COD) concentrations in the influent were controlled at different levels by adjusting the carbon source dosage. A MF hollow fiber membrane module (0.22 μm , 1.0 m^2 , MOTIMO, China) was immersed in the MBR tank. An air diffuser was installed underneath the membrane module to provide air at 0.5 m^3/h . The membrane flux was maintained at 12.5 $\text{L}/\text{m}^2\text{h}$. To alleviate membrane fouling, the membrane was operated in an intermittent mode (on/off = 10 min/2 min). The mixed liquor at the bottom of MBR tank was recycled to the anaerobic reactor continuously at a rate of 0.75.

2.2. Influent quality and analytical methods

The synthetic wastewater was composed of glucose, starch, NH_4Cl , KH_2PO_4 , NaHCO_3 , and mineral solution (Mg, Ca, and Fe). Different COD levels in the influent were implemented by adjusting the organic substances dosage. The initial influent contained COD 120–1,200 mg/L, TN 31.1–34.7 mg/L, $\text{NH}_4\text{-N}$ 27.8–32.9 mg/L, TP 3.8–5.2 mg/L, and the pH value was 6.7–7.3.

All the results presented were obtained from the An-M(A/O)-MBR system at its steady state. DO concentration was measured using the DO meter (WTW Oxi 340, Germany); pH using the pH meter (PHSJ-4A, China); the transmembrane pressure was measured using a pressure gage; and particle size distributions in mixed liquor were analyzed using a Laser Particle Size analyzer (WICS-50, ANKERMID, Dutch). Measurement of COD, MLSS, total nitrogen (TN), nitrate nitrogen ($\text{NO}_3\text{-N}$), ammonium nitrogen ($\text{NH}_4\text{-N}$), orthophosphate concentration (ortho-P) and total phosphorus (TP) followed standard methods [10].

2.3. Operation

On the basis of the previous studies, the initial influent was fed to the laboratory-scale system with a flow rate of $0.25 \text{ m}^3/\text{d}$ and the total HRT was 6.96 h. The MLSS concentration in the multiple A/O zone was kept at 3,800–4,200 mg/L and the excess sludge was withdrawn periodically to keep the sludge retention time (SRT) was about 26–27 d. The operation temperature was at 26–30°C during the operation. Table 1 shows the specifications of the experimental conditions.

3. Results and discussion

3.1. COD removal

Daily COD variation of the influent and effluent was studied during the operation. It seemed that COD

level in the influent had little influence on its removal in the An-M(A/O)-MBR system. Although the influent COD fluctuated from 120 to 1,200 mg/L, the COD removal efficiency was high and stable, and above 95% removal was achieved throughout the operation, indicating the system had relatively strong capacity in resisting shock organic loading. The COD concentrations were less than 50 mg/L in the effluent, which was sufficient to meet the standard of water reclamation in China.

To evaluate the COD removal mechanism in the An-M(A/O)-MBR system, typical variations of COD concentration in each tank were studied at different COD settings (data not shown). The results showed that COD level in the anaerobic zone, increased from 38.0 to 166.2 mg/L when it increased from 120–200 to 1,110–1,200 mg/L in the influent. Although 10% of influent was fed into the last anoxic tank in the multiple A/O zone, the COD concentrations, in the effluent of the multiple A/O reactor, were lower than 110 mg/L throughout the operation. The high COD removal might be attributed to the growth of high biomass concentration (3,800–4,200 mg/L) in the An-M(A/O)-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 [11].

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 nitrogen component in the effluent were presented in Figs. 2 and 3, respectively.

When COD concentrations were in the range of 350–410 mg/L in Run 1, as shown in Fig. 2, good performance of TN removal was achieved, the removal efficiency was 81.6–90.4%, and the average efficiency was 86.5%. As shown in Fig. 3, the TN concentrations

Table 1
Specifications of the experimental conditions

Items	Run 1	Run 2	Run 3	Run 4
Experimental period (d)	384–398	399–411	412–424	425–437
COD concentration in the influent (mg/L)	350–410	120–200	640–710	1,110–1,200
Influent distribution ratio	$Q_1:Q_2:Q_3:Q_4:Q_5 = 6:1:1:1:1$			
Membrane flux ($\text{L}/\text{m}^2 \text{ h}$)	12.5			
DO in the aerobic tanks in the multiple A/O zone (mg/L)	1.0–1.2			
DO in the MBR zone (mg/L)	2.0–3.0			
Sludge recycle rate (r)	0.75			
SRT (d)	26–27 d			

in the effluent were lower than 6.5 mg/L, little ammonia nitrogen was detected and it even became undetectable in the effluent, implying that nitrification was almost perfectly completed in the An-M(A/O)-MBR process. When COD in the influent decreased to 120–200 mg/L in Run 2, TN removal efficiency decreased from 81.7% at 399 d to 33.4% at 411 d. In Run 2, ammonia nitrogen in the effluent was lower than 0.3 mg/L, but the maximum value of nitrate ($\text{NO}_3\text{-N}$) was as high as 18.9 mg/L in the effluent, implying that the deterioration of TN removal was caused by poor denitrification performance in the system. Since the other operational conditions were kept at a relatively constant state, the poor denitrification performance might be attributed to the deficiency of organic substances in Run 2. When COD in the influent increased to 640–710 mg/L in Run 3, the TN removal efficiency began to increase and achieved to 85% after a few days. The ammonia nitrogen values in the effluent, like in Run 1 and Run 2, were also kept at a low level (<0.5 mg/L). When COD increased to 1,110–1,200 mg/L, the TN removal efficiency decreased to 18% some days later. Meanwhile, the inhibition of the nitrification process was observed and ammonia nitrogen concentration in the effluent increased to 22.4 mg/L. In Run 3, 60% of influent was fed into the anaerobic tank and the COD concentration reduced greatly, but a large quantity of the organic compounds was just adsorbed by the activated sludge and they could also be utilized by heterotrophic bacteria in the following multiple A/O zone. As a result, heterotrophic bacteria propagated rapidly and the growth of nitrobacteria, a kind of autotrophic bacterium, was prohibited [12]. Furthermore, additional 40% of influent was pumped into the multiple A/O zone to provide organic substrate for denitrification, which might further intensify the state of organic surplus in the system and stimulate the growth of heterotrophic bacteria. Ultimately, the ratio of nitrobacteria

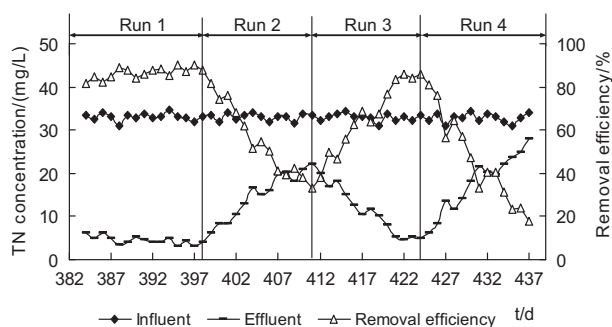


Fig. 2. Effect of influent COD concentration on TN removal.

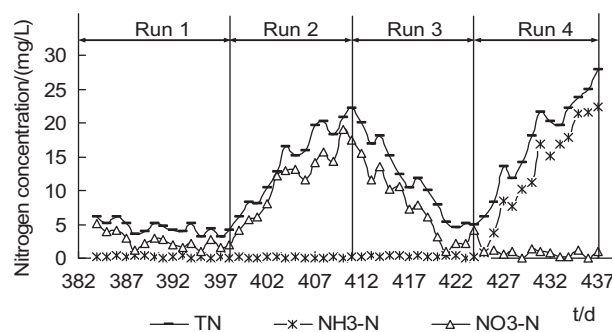


Fig. 3. Effect of influent COD concentration on nitrogen composition in the effluent.

in the activated sludge decreased, causing a negative effect on nitrification process and the nitrogen removal performance became worse accordingly.

3.3. Phosphorus removal

Effects of COD levels in the influent on the TP removal and on the TP concentrations in each tank were presented in Figs. 4 and 5, respectively.

As shown in Fig. 4, good performance of TP removal was achieved when COD concentration was in the range of 350–410 mg/L in Run 1, the removal efficiency was 84.5% on average and the effluent TP concentrations were lower than 1.0 mg/L, correspondingly. When COD in the influent was adjusted to 120–200 mg/L in Run 2, as shown in Fig. 5, TP concentration in the anaerobic tank decreased from 18.9 mg/L at 389 d (in Run 1) to 12 mg/L at 400 d (in Run 2), indicating that low COD level in the influent had negative effect on the phosphorus release. The TP removal efficiency of the system was kept at about 84% in the first 2 d (399 d and 400 d); however, an obvious decrease trend of TP removal was observed in the following days and the removal efficiency was

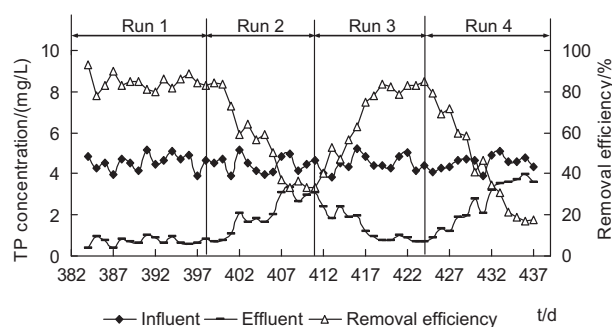


Fig. 4. Effect of influent COD concentration on TP removal.

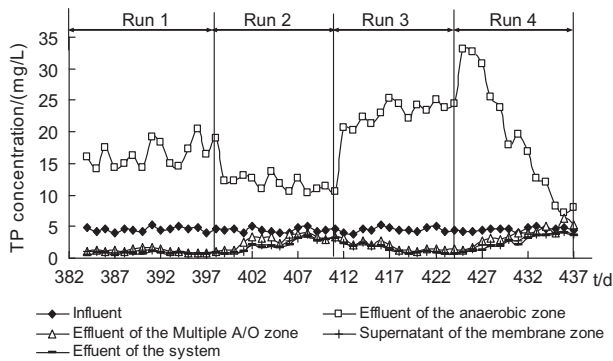


Fig. 5. Variation of TP concentration in each zone.

only 33% at the end of Run 2. When COD concentration was set at a low level (120–200 mg/L), the volatile fatty acid (VFA) in the anaerobic tank decreased accordingly, which had a timely negative effect on phosphorus release [13–15]. Meanwhile, low COD level could also have adverse effect on denitrification process, resulting in the increase of nitrate in the sludge circulation. Therefore, the carbon source competition between denitrification and phosphorus release was intensified [15,16], and the rate of phosphorus release was further reduced.

The effluent TP concentration began to increase at 401 d, and the removal efficiency decreased to 75%. This might contribute to the imbalance between phosphorus release and uptake rates. When COD in the influent decreased to a low level (120–200 mg/L), the phosphorus release decreased considerably in the anaerobic tank, but the phosphorus uptake had kept at the previous level in the first 2 d. When the quantity of polyphosphate particles in the PAOs reached saturation point, the P-uptake ability of PAOs began to decrease, leading to the increase of effluent TP concentration and only 33% TP removal was achieved at the end of Run 2.

To improve the performance of TP removal, COD concentration in the influent was increased to 640–710 mg/L in Run 3. After a 5–6-d operation, the TP concentration in the anaerobic tank was achieved to 19 mg/L and TP removal efficiency was restored to 80%. When COD was further increased to 1,110–1,200 mg/L in Run 4, phosphorus removal decreased accordingly. However, as shown in Fig. 5, the TP concentration increased from 24.5 to 33.0 mg/L in the anaerobic tank. It was reported that the rate of phosphorus release in anaerobic condition varied linearly with the VFA quantity in the influent and the storage compounds (poly- β -hydroxybutyrate [PHB]) in PAOs [17]; however, the rate of phosphorus uptake in aerobic condition did not vary linearly with the PHB strictly,

but had a saturation value [16,18]. Therefore, it could be inferred that the deterioration of TP removal at the beginning of Run 4 was the result of malfunction of phosphorus uptake after it is effectively released. The phosphorus release rate began to decrease at 427 d and the TP concentration reduced to 8.1 mg/L in the anaerobic tank, the total TP removal was lower than 20%, indicating the phosphorus removal was only caused by microbial assimilation.

High organic load rate (OLR) was beneficial to the growth of non-PAOs in the activated sludge, which might induce the change of microbial population and thus even made the system breakdown in an enhanced biological nutrients removal process [15,19,20]. In this study, after 437 d, the influent COD level was set back to 350–410 mg/L, but it took nearly one month to recover the performance of nitrogen and phosphorus removal in the An-M(A/O)-MBR system. Therefore, the deterioration of phosphorus removal performance might be the result of the shift of the composition of the microbial communities.

4. Conclusions

An innovative step-feed An-M(A/O)-MBR process was developed to treat synthetic domestic wastewater and its performance was investigated under different COD settings on a laboratory-scale system. Major findings from this study are summarized as follows:

- (1) Combining SAOASP with membrane separation, the step-feed An-M(A/O)-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. COD level in the influent was one of the most important parameters affecting the performance of the step-feed An-M(A/O)-MBR process.
- (2) COD level in the influent had little influence on its removal in the An-M(A/O)-MBR system. The removal efficiency was high and stable (95% on average) throughout the operation. The effluent COD level (<50 mg/L) was sufficient to meet the standard of water reclamation in China.
- (3) COD level in the influent caused significant differences in TN and TP removal efficiency in the An-M(A/O)-MBR system. Under the conditions imposed, when COD concentration was set at 350–710 mg/L, good performance of TN and TP removal was obtained and the

average removal efficiency was above 85 and 84%, respectively. But when COD was lower than 120–200 mg/L or higher than 1,110–1,200 mg/L, the performance of TN and TP removal deteriorated.

- (4) Low COD level (120–200 mg/L) could cause the deficiency of organic carbon source for denitrification and for phosphorus release. And high COD level (1,110–1,200 mg/L) could change the ecospecies in the system because of the excessive high OLR.

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