A Novel Nearly Plug-Flow Membrane Bioreactor for Enhanced Biological Nutrient Removal

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The feasibility of enhanced biological nutrient removal by a new process called nearly plug-flow membrane bioreactor (NPFMBR) is studied. Results of long-term observations showed that average removal degrees of (1) chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) reached 95%, 85%, and 89%, respectively, at the steady operation period. Process success was further corroborated by batch experiments. Data of specific oxygen uptake rates demonstrated the abundance and/or high activities of ammonium oxidation bacteria and nitrite oxidation bacteria in the sludge. Observed specific rates of nitrification, denitrification, phosphorous release/uptake of the sludge were higher than those reported in previous research. Because of the unique flow pattern of sludge and alternant aerobic–anoxic operating conditions in the bioreactor, mass transfer and biotransformation of nutrients were expected to be improved. The NPFMBR could offer a new option for the wider application of MBRs. © 2012 American Institute of Chemical Engineers *AIChE J,* 59: 46–54, 2013
Keywords: wastewater treatment, membrane fouling, nitrification, denitrification, phosphorus removal

Introduction

Membrane bioreactors (MBRs) have been widely used for wastewater treatments because of their unique advantages, such as the good effluent quality and compact structure, over conventional activated sludge processes. MBRs can not only degrade organic matter but also are able to eliminate nitrogen and phosphorus more effectively. As such, a considerable number of MBR plants were constructed for biological nutrient removal (BNR).[1–4]

So far, most of the MBR plants for BNR are based on anaerobic/aerobic (A/O) and anaerobic/anoxic/aerobic (A^2/O) processes.[5] In these MBRs, the membranes are usually submerged in the aeration tank to make the best use of aeration energy and to alleviate membrane fouling. In addition, sludge recirculation is needed to realize nitrification and denitrification process. Thus, the nutrient removal efficiencies strongly depend on the recirculation ratio of the mixed liquor. An increase of recirculation ratio can improve the BNR performance, which however consumes more pumping energy. The A^2/O MBRs can be classified into two types: predenitrification (anaerobic/anoxic/aerobic process) and postdenitrification (anaerobic/aerobic/anoxic process).[6] The predenitrification-based MBRs are expected to achieve good denitrification performance even without the addition of an external carbon source. The postdenitrification, on the contrary, usually needs the addition of external carbon sources for denitrification. However, Vocks et al.[7] observed that enhanced biological phosphorus removal (EBPR) sludge could offer a carbon source for postdenitrification that is not present in non-EBPR sludge. Nitrogen removal can also be accomplished by simultaneous nitrification–denitrification (SND) processes.[8–10] But, the SND processes require either optimization of aeration modes/rates[8,11,12] or configuration modification.[13,14] In addition, some other novel MBRs, such as vertical submerged MBR[15,16] and double-deck aerated biofilm MBR,[17] have been developed, with aims to achieve good effluent quality and/or high membrane permeability. These previous findings reveal that the optimization of operating conditions or MBR configurations is an alternative to enhance the BNR performance. However, the current configurations of MBRs are mostly limited to the continuous stirred tank reactor (CSTR) and/or batch reactor. Another typical MBR configuration, based on the plug-flow reactor (PFR), has not yet been studied. These three kinds of reactors are different in either structures or operation modes. They also have their own limitations and benefits when being used for wastewater treatments or chemical reactions. The main advantages of PFR are the following: (1) no backmixing and (2) higher reaction efficiency as compared to the CSTR with the same volume. Consequently, the PFR-type MBRs are expected to achieve better performance than the CSTR-type MBRs in some occasions.

Therefore, the overall aim of this study is to propose a nearly plug-flow MBR (NPFMBR). In the NPFMBR, the bioreactor was divided into five long and narrow channels with different dissolved oxygen (DO) concentrations, which enable the sludge under alternant aerobic–anoxic conditions. The long-term performance of the NPFMBR on nutrient removals was investigated. Microorganism activities, such as specific oxygen uptake rates (SOURs), specific nitrification

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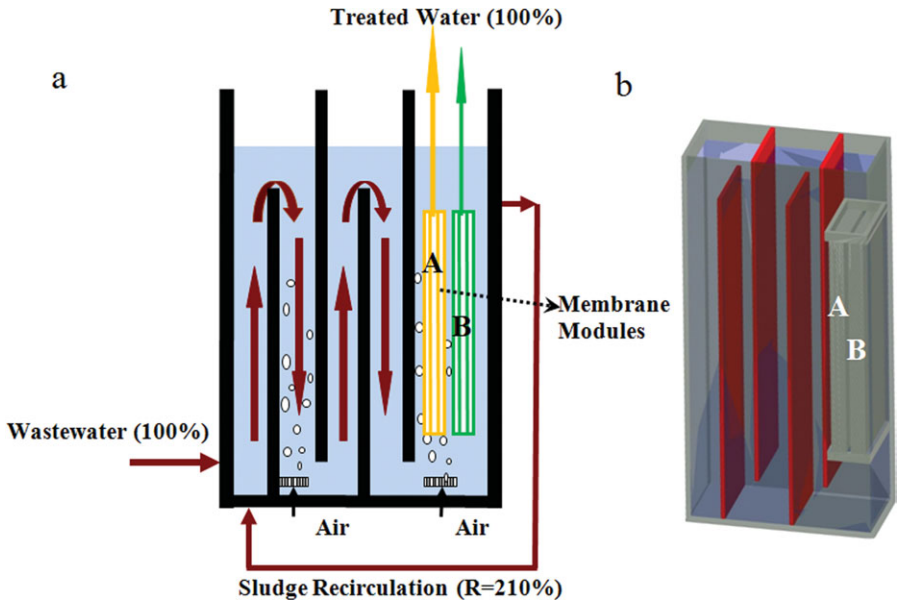

Figure 1. Process flow diagram (a) and three-dimensional structural drawing (b) of the NPFMBR.

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rates (SNRs), specific denitrification rates (SDNRs), specific phosphorus release rates (SPRRs), and specific phosphorus uptake rates (SPURs), were also measured using batch experiments. Fouling behaviors of the NPFMBR were also explored. Finally, optimization of two significant operating parameters (pH and DO) was discussed according to the long-term operation experience.

Experimental

Configuration and operation of the NPFMBR

Figure 1 shows the configuration of the NPFMBR with an effective working volume of 50 L. The use of baffles divided the bioreactor into five high and narrow tanks or channels, which were controlled at anoxic (DO = 0.1–0.5 mg/L), aerobic (DO > 0.5 mg/L), anoxic (DO = 0.1–0.5 mg/L), anaerobic (DO < 0.1 mg/L), and aerobic conditions (DO > 0.5 mg/L) on the basis of the DO concentrations, respectively. Height, width, and length of each channel were 800, 180, and 70 mm, respectively. The seed sludge in the bioreactor was derived from a full-scale municipal wastewater treatment plant (Li-Jiao plant, Guangzhou, China). As a result of the sludge recirculation and feed wastewater inflow, the mixed liquor in the bioreactor flowed through the channels in turn, with a nearly plug-flow pattern. Two identical flat-sheet membrane modules (i.e., Modules A and B) (polyvinylidene fluoride, 0.1 μm, Sinap Corp., Shanghai, China) with a total surface area of 0.23 m^2 (i.e., 0.115 m^2 for each module) were submerged in the second aerobic channel. Synthetic wastewater was fed as substrate for the microorganisms of the NPFMBR (see Table 1). Over the whole period of operation, hydraulic retention time (HRT) and solid retention time (SRT) were set at 12–14 h and 20 days, respectively. Aeration rates of the first and second aerobic zones were controlled at 60–80 and 120–150 L/h, respectively. In this study, the Modules A and B were operated with different membrane flux in different periods, respectively. The detailed operating conditions of the membrane flux are summarized in Table 2. In Period 1, the Modules A and B were connected together and sucked by one peristaltic pump, and also one vacuum pressure gauge was used. As such, their membrane flux was kept at 17.4 L/ (m^2 h) (LMH) on average. During the Period 2, the membrane flux of Modules A and B was set at 26.1 and 8.7 LMH, respectively, which were sucked by two peristaltic pumps. The transmembrane pressure (TMP) of the two modules was recorded, respectively. In Period 3, the membrane flux of Modules A and B was set at 34.8 and 0 LMH, respectively. Although the Module B did not work, it was still submerged

Table 1. Composition of Synthetic Wastewater

Substrates	Concen. (mg/L)	Trace Elements	Concen. (mg/L)
Starch	162	$CuSO_4$	0.06
Milk powder	200	$CaCl_2$	0.44
Sucrose	141	H_3BO_3	0.06
Peptone	32	$NaMoO_4{\cdot}2H_2O$	0.19
Yeast extract	60	$CoCl_2{\cdot}6H_2O$	0.13
Beef extract	60	$NiCl_2{\cdot}6H_2O$	0.04
Na-acetate	35	$MgCl_2$	0.19
Urea	50	$FeSO_4{\cdot}7H_2O$	2.50
NH_4Cl	80	$ZnCl_2$	0.06
KH_2PO_4	23	$MnCl_2{\cdot}4H_2O$	0.06
K_2HPO_4	28		
$NaHCO_3$	80		
Na_2CO_3	80		

Table 2. Imposed Flux of the Two Membrane Modules of the NPFMBR

	Membrane Flux (LMH)			
	Module A	Module B	Average	Cleaning Method
Period 1 (0–150 days)	17.4	17.4	17.4	Physical backwashing and Chemical cleaning
Period 2 (151–330 days)	26.1	8.70	17.4	Chemical cleaning
Period 3 (331–360 days)	34.8	0	17.4	Chemical cleaning

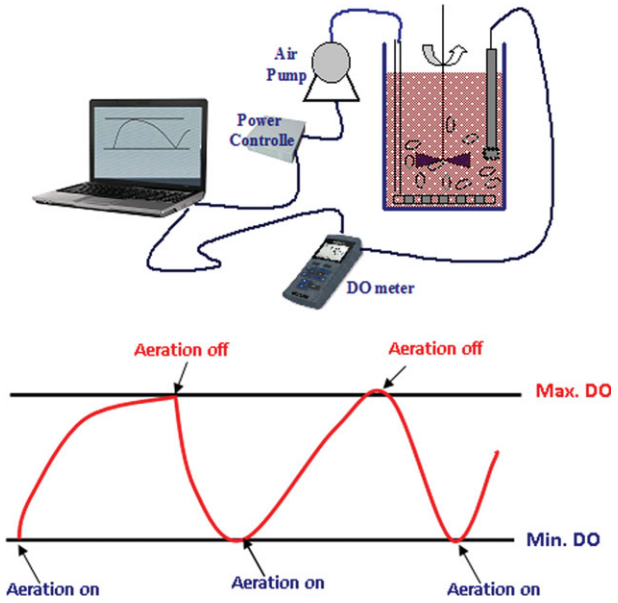

Figure 2. Scheme of the automatic apparatus designed for SOUR measurements.

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in the membrane channel to keep the same hydraulic conditions. Despite the variations of their flux in the different operation periods, the total effluent rate of the two modules was maintained at 4 L/h to keep the steady HRT. During the operation period of 0–150 days, both physical backwashing with 2 L pure water and chemical cleaning via soaking the fouled membrane module in 0.3% NaClO solution for about 12 h were conducted. Physical backwashing was conducted once TMP was over 15 kPa. When physical backwashing was not enough to recover the flux to an acceptable level, chemical cleaning was performed. During the operation period of 151–360 days, however, only the chemical cleaning was performed. As such, the irreversible fouling and reversible fouling could be investigated.

Measurements of SOURs

As shown in Figure 2a, the setup used for SOURs measurements was developed according to Iversen et al. and Rosenberg.[18,19] To achieve automatic control, an oxygen probe (CellOX 3310i, WTW, Germany) was connected with a computer, and hence the DO data were collected every 5 s. The DO concentration of the mixed liquor was set in a range of 4–6 mg/L. When the DO concentration was below 4 mg/L, fine bubble aeration (i.e., approx. 80 L/h) will be provided by an air pump. Once it reached the maximal value (i.e., 6 mg/L), the aeration was stopped automatically (see Figure 2b). Subsequently, the DO concentration will decline due to the respiration of microorganisms. According to the decrease of the DO concentration as a function of operating time, SOURs can be calculated. To keep the mixed liquor in suspension, it was gently stirred in the whole process (approx. 150 rpm).

About 500 mL of mixed liquor was taken out from the membrane tank (the second aerobic channel), and then it was filtered through a filter paper (about 10 μm) to remove the remaining organics in the bulk. Afterward, the sludge was resuspended with pure water, with concentrations of mixed liquor volatile suspended solids (MLVSS) in a range of 2000–3000 mg/L. Finally, the SOURs of the sludge were measured in the apparatus mentioned above. At least three measurements were performed for each sample, and average data were given in this article. Moreover, oxygen uptake rates of ammonium oxidation bacteria (AOB) and nitrite oxidation bacteria (NOB) were also monitored by spiking 10 mg/L nitrite and 30 mg/L ammonium into the sludge suspension. Detailed methods regarding this experiment can be found elsewhere.[20,21]

Measurements of specific rates of nutrient removal

SNRs of the sludge were measured as follows: (1) approx. 500 mL of sludge suspension from the second aerobic channel of the bioreactor was filtered through filter papers, (2) the filtered sludge was then resuspended to 1000 mL with pure water, (3) ammonium solution was added into the sludge suspension (with a final ammonium concentration of about 25 mg/L) and aerated it with the apparatus in Figure 2 (DO = 4–6 mg/L), and (4) samples were taken out and analyzed regularly. The SNRs were determined by the decrease of the ammonium concentration as a function of operating time. With respect to the measurements of SDNRs, the sludge from the anoxic channel of the bioreactor was also treated as that in the SNRs measurements. Afterward, nitrate (approx. 50 mg/L) and acetate (300 mg-COD/L) solutions were dosed into the sludge suspension at anaerobic conditions (aerated with nitrogen gas). According to literature,[22,23] the SDNRs were calculated by the decrease of values of "nitrate+0.6*nitrite" as a function of operating time.

Phosphorus removal potential of the sludge was characterized by SPRRs and SPURs. In this set of batch experiments, the treated sludge from the second aerobic channel was fed with acetate solution (300 mg-COD/L) and stirred at anaerobic conditions. Samples were taken out regularly for total phosphorus (TP) analysis. According to the increase of the TP concentration as a function of operating time, the SPRRs can be obtained. After a complete P release, the sludge was then aerated with DO concentrations in a range of 4–6 mg/L (P uptake will occur). The SPURs were determined according to the decrease of the TP concentration in aeration period.

The measurements of SNRs, SDNRs, SPRRs, and SPURs were performed in four replicates (Oct., Nov., Dec. 2010, and March 2011) to obtain solid data. In addition, the SNRs and SPURs under different DO ranges (1–2 mg/L, 2–4 mg/L, and 4–6 mg/L) were measured.

Chemical analysis

pH values of the sludge suspension were monitored by a pH meter (PHS-3D, Shanghai Precision & Scientific Instrument Corp.). Concentrations of ammonium, nitrite, nitrate, TN, TP, COD, MLSS, and mixed liquor suspended solids (MLSS) were measured according to standard methods.[24]

Results

Development of MLSS and MLVSS

As plotted in Figure 3a, concentrations of MLSS and MLVSS in the membrane tank increased from 2500 and 1500 mg/L to about 6000 and 5000 mg/L after 40 days of operation. Afterward, they kept in a range of 5500–7000 mg/L and 3500–5500 mg/L (see Figure 3a), respectively, although they always varied due to the decrease of the membrane permeate as a result of membrane fouling. The sudden decrease of MLSS and MLVSS on day 190 was due to the long-term maintenance of the membrane modules (e.g.,

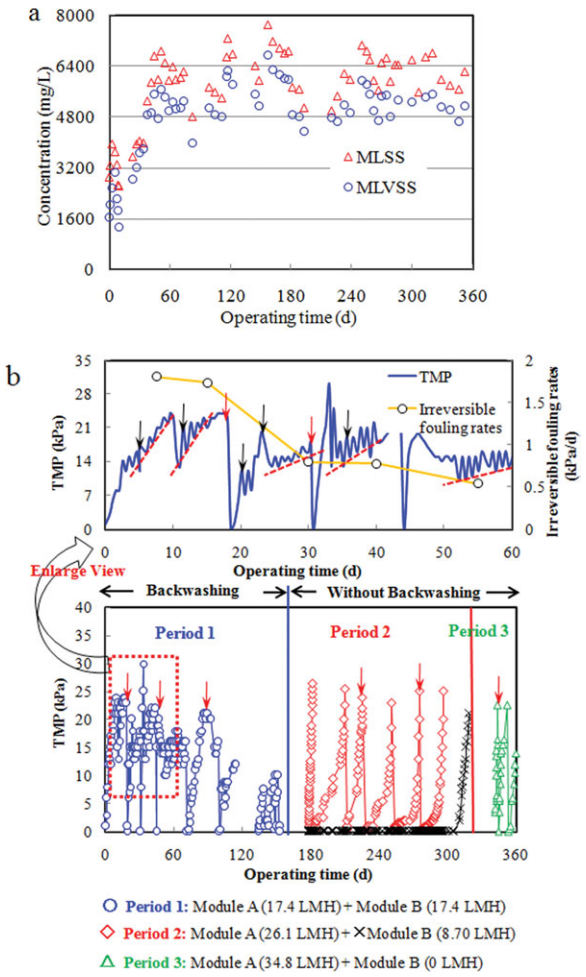

Figure 3. Evolution of MLSS/MLVSS concentrations (a) in the 606 membrane tank and 607 variations of TMP (b) over the entire operation period (where the red and black arrows represent chemical cleaning and physical backwashing, respectively; and the red broken lines represent the evolution of irreversible fouling).

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repeated physical backwashing and chemical cleaning). It should be mentioned that because of membrane retention, the MLSS and MLVSS in the membrane tank were much higher than those in other tanks (see Table 3). Note that keeping higher biomass concentrations is helpful for the accomplishment of BNRs.

Membrane performance of the NPFMBR

Over the whole period of operation, the membrane performance was indicated by the development of TMP as a function of filtration time. As showed in Figure 3b, both physical backwashing and chemical cleaning were performed during the Period 1 (0–150 days). It can be seen that physical backwashing could to some extent recover the membrane permeability, but the irreversible fouling was found to accumulate gradually after repeated physical backwashing due to the accumulation of the substances remaining on/in the membranes. This phenomenon is in good agreement with previous study.[25] The evolution rates of both irreversible fouling and total membrane fouling were much higher during the startup period of the NPFMBR (0–60 days). The chemical cleaning intervals could reach up to 8–12 days during this operating period (0–60 days). Interestingly, it was found that the development rates of irreversible fouling gradually declined from 1.8 to 0.54 kPa per day. In fact, the sludge had a lower MLVSS/MLSS ratio of 0.6–0.7 in the startup period (data not shown), and thus the presence of inorganic matter could cause severe irreversible fouling as a result of the formation of precipitate on/in the membranes. Nonetheless, the chemical cleaning intervals increased to 26–30 days when the MLVSS/MLSS concentrations increased to 0.8–0.9, demonstrating that the fouling rates became slower in the steady operation period.

In the Period 2 (151–330 days), only chemical cleaning was performed. Obviously, the increase rates of TMP of membrane Module A (26.1 LMH) in the Period 2 were lower than those in the startup period (0–60 days) but higher than those of the steady operation period (61–150 days) of the Period 1. This phenomenon implies the significance of both the imposed flux and physical backwashing. In comparison, the Module B (8.7 LMH) was slowly fouled and the TMP reached 21 kPa after being operated for 140 days. In the Period 3 (331–360 days), we noted that the TMP of the Module A with a much higher membrane flux (34.8 LMH) showed a dramatic increase, thus much more frequent chemical cleaning was required (every 5–9 days). Similar to a previous study,[26] a higher membrane flux can cause a faster increase rate of the TMP. Based on the long-term operation of the NPFMBR with different flux, the critical flux can be set at about 17.4 LMH, which is similar to the commercially used flat-sheet membranes, for example, the critical flux of Kubota membrane was reported as 16–24 LMH.[27] Generally, the recovery of membrane permeability and membrane fouling rates strongly depend on membrane materials, operating conditions (e.g., imposed membrane flux, aeration condition, and temperature), and sludge characteristics. Membrane fouling mechanisms of this NPFMBR will be further investigated by focusing on the deposition and biotransformation of biomolecules on/in the membranes.

Nutrients removal performance of the NPFMBR

Because of the combined effect of biodegradation and membrane rejection, the NPFMBR could obtain satisfying removal of organics, for example, 95% of COD was

Table 3. Average Values of pH, DO, MLSS, and MLVSS (9) in the Five Tanks of the NPFMBR*

	Anoxic Tank (First One)	Aerobic Tank	Anoxic Tank (Second One)	Anaerobic Tank	Membrane Tank (Aerobic Tank, Too)
DO (mg/L)	0.11	0.87	0.12	0.07	2.76
pH	6.95	6.85	7.17	7.23	7.25
MLSS (mg/L)	4733	4947	4904	5046	6273
MLVSS (mg/L)	4043	4163	4153	4267	5328

*Data were given as average values based on repeated measurements ($n = 10$).

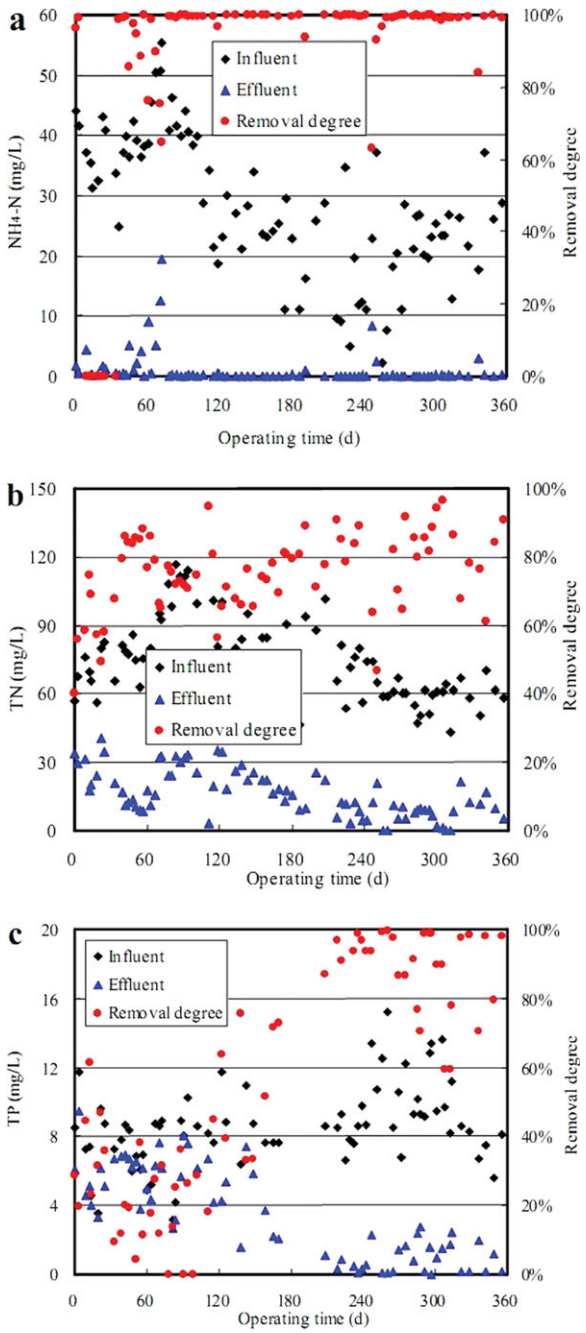

Figure 4. Removal performance of ammonium, TN, and TP in the NPFMBR.

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mineralized in this MBR (data not shown). Figure 4 depicts the removal of ammonium, TN, and TP. A complete nitrification of the ammonium was also found (see Figure 4a), indicating that the NPFMBR can achieve a good nitrification process. It can be seen that the average removal degree of TN reached 85% after the startup period of the NPFMBR. It demonstrates that most of the nitrogen present in the feed wastewater was eliminated by nitrification and denitrification. Nevertheless, the remaining nitrogen in the membrane permeate was present in the form of nitrate rather than nitrite (data not shown), indicating that transformation of nitrite to nitrate was very complete. This evidence also suggests that the denitrification needs to be further improved to eliminate the remaining nitrate in the bioreactor. We should keep in mind that the feed wastewater had low COD/TN ratios in a range of 5–8. Theoretically, to denitrify 1 g nitrate, it needs at least 8.67 g COD.[23] In addition to this, the microorganisms proliferate rapidly once they have access to COD, resulting in a low available carbon source for denitrification. Thus, we can conclude that the NPFMBR can complete satisfying nitrogen removals for wastewater with such low COD/TN ratios.

As seen in Figure 4c, TP was also well removed in the steady operation period of the NPFMBR (from day 200 to 360); the removal degree of TP was in a range of 70–100% (89% on average). The sporadic low removal degrees of TP on day 200–360 were due to the decrease and unsteady of DO concentrations in the aerobic tanks. As the P uptake by polyphosphate-accumulating organisms (PAOs) strongly depends on DO concentrations, we can assume that the decrease of DO concentrations can cause lower removal efficiencies of TP. In this study, we found that DO concentrations higher than 2 mg/L could be desired to achieve better TP removal. Details will be discussed in the following sections.

Specific activities of microorganisms responsible for BNR

In this study, the SOURs of ordinary heterotrophic organisms (OHO), AOB, and NOB were measured. As shown in Figure 5, the average SOURs were determined to be 3.1, 3.0, and 5.1 mg-O_2/(MLVSS h) for OHO, AOB, and NOB, respectively. It suggests that OHO and AOB had similar activities in terms of oxygen uptake; the NOB had much higher activities. This phenomenon also reveals that the NOB can transform the nitrite produced by AOB quickly. This is why the nitrate was the major nitrogen species in the NPFMBR effluent.

The nitrifying activities of sludge were investigated using batch experiments. As shown in Figure 6a, the added ammonium decreased linearly as a function of operating time, and the nitrate was also produced gradually. The nitrite was not found to accumulate in the entire experiments, which had a level lower than 0.5 mg/L. The SNRs were determined to be 2.9 mgNH_4^+-N/gMLVSS h on average. This value is much higher than the results of Han et al.[28] (1.09–1.29 mgNH_4^+-N/gMLVSS h varying with SRT). This shows that the sludge in the NPFMBR had high nitrifying activities, which should be attributed to the special configuration of the bioreactor and the alternating anoxic–aerobic–anaerobic conditions.[29] As plotted in Figure 6b, nitrate was also well removed in the batch experiments of SDNRs. A fast nitrate removal was found in the initial 2 h of the experiments. Accordingly, nitrite accumulation occurred in the initial 2 h. There could be two reasons for this accumulation:[30] extensive growth of microorganisms was able to transform nitrate to nitrite and/or the inhibition of nitrite reduction enzymes. In this study, we observed that the nitrite accumulation occurred only in the initial 2 h, and it disappeared in the following experimental period. This evidence suggests that the nitrite accumulation was a consequence of the extensive growth of nitrate reduction bacteria rather than the inhibition of nitrite reduction enzymes. According to previous literature,[22] the SDNRs was corrected by Δ(nitrate + 0.6*nitrite), because the reduction of 1 mg nitrite to nitrogen gas consumes the

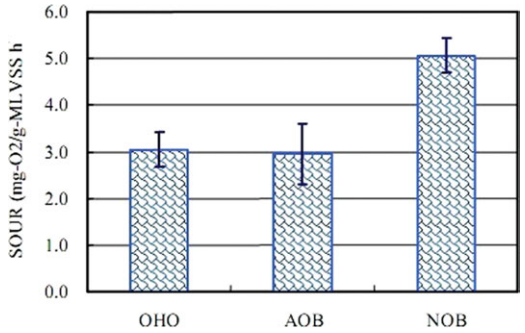

Figure 5. SOURs of OHO, AOB, and NOB (these data reveal the biological activities of aerobic bacteria in the sludge).

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

same amount of electrons as the reduction of 0.6 mg nitrate to nitrogen gas.[23] Thus, the corrected SDNRs were determined to be 8.1 ± 1.7 mg-NO_3^--N/(MLVSS h) on the basis of four reduplicate measurements on different days. These data are higher than those reported by Vocks et al.,[7] who determined the SDNR to be 5.8 mg-NO_3^--N /(MLVSS h) in an MBR used for enhanced nutrient removal as well. In addition, Bracklow et al. also reported that the SDNRs of five MBRs for BNR were in a range of 0.2–1.5 mg-NO_3^--N/(MLVSS h) on average.[31] Therefore, compared with previous studies, the sludge in the NPFMBR had a high denitrification potential.

The results of batch experiments for P release and uptake under anaerobic and aerobic conditions were plotted in Figure 6c. As expected, considerable amount of P was released by the sludge within 1 h of the P release experiments, and part of the released P was assimilated in the following aeration period. The average SPRRs and SPURs were measured to be 10.7 and 6.8 mg-P/(g-MLVSS h), respectively. It is widely known that the P elimination in the activated sludge processes is completed by the alternate release and uptake of P. Thus, the data of SPRRs and SPURs can to some extent reveal the P removal potential of the PAOs. Previous research reported lower SPRRs and SPURs, for example, 3.5 and 0.15 mg-P/(g-MLVSS h) of an SPRR and SPUR were observed by Oehmen et al.[32]; and a much lower SPRR of 0.016 mg-P/(g-MLVSS h) was reported in a recent study.[33] One possible reason for the high SPRRs and SPURs in the NPFMBR could be high abundance of PAOs, but more research is needed to confirm it. Another explanation is that the BNR in the NPFMBR was a predenitrification process that has a higher rate of P release than that in the postdenitrification process.[33] In addition, the nearly plug-flow pattern of substrates and microorganisms can promote the growth of PAOs with high activities.

Optimization of DO and pH in the NPFMBR

As DO and pH are two significant parameters affecting the performance of NPFMBR, they were optimized in the startup period of the operation. The pH value can be considered as an indication of alkalinity in the bulk liquor. Most bacteria, particularly the nitrifying and denitrifying bacteria, are sensitive to alkalinity. In this study, a considerable amount of sodium carbonate and sodium bicarbonate was added to supply sufficient alkalinity for the nitrifying bacteria. As seen in Table 3, the pH in the first aerobic tank was lower, which is due to alkalinity consumption during nitrification process. On the contrary, the higher pH values in the anaerobic and anoxic tanks were a consequence of alkalinity generation during the denitrification process. In fact, the combined use of different tanks can help to make the best use of the alkalinity added in the feed wastewater or that produced by the denitrification process.

On the other hand, the optimization of DO concentrations cannot only influence the nutrient removals but also determine the aeration cost of the MBR. On the premise of keeping satisfactory nutrient removals (i.e., P uptake and nitrification) and good membrane permeability, the aeration rates in the two aerobic tanks were kept as low as possible (as shown in Experimental). From Table 3, we can see that the

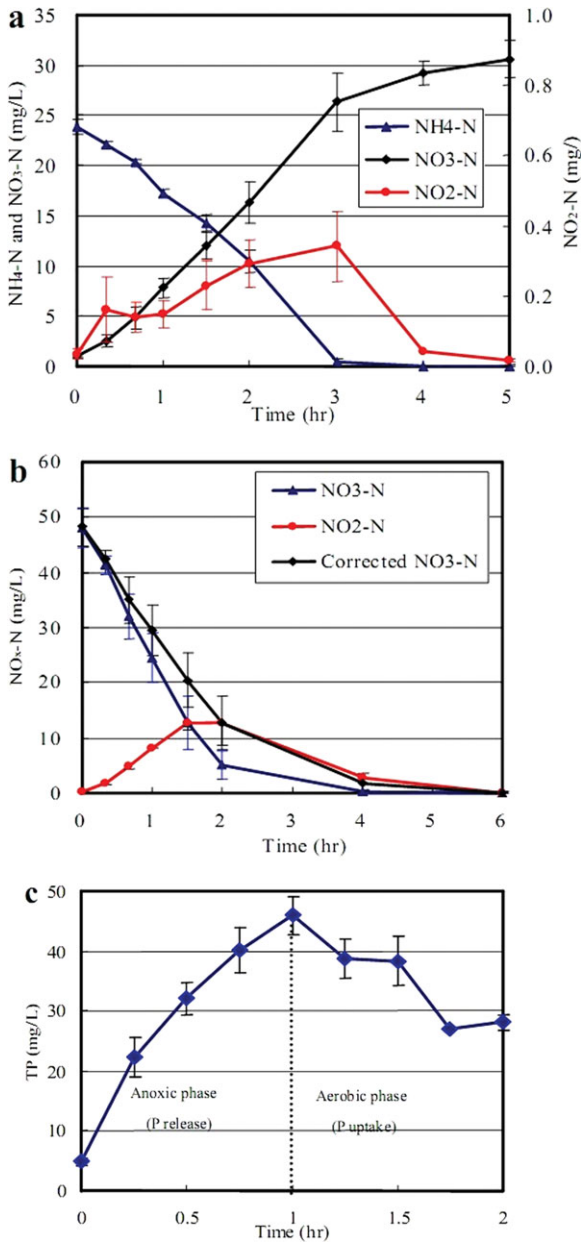

Figure 6. Specific utilization rates of ammonium, nitrate, and TP in the well-controlled batch trials.

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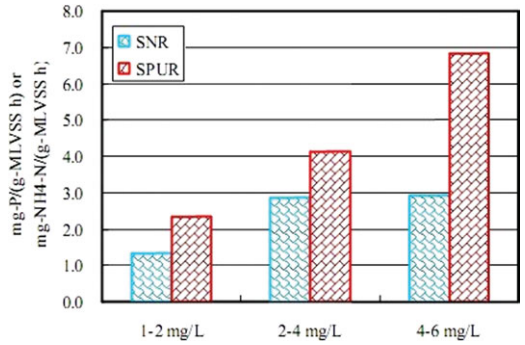

Figure 7. Influence of DO concentrations on SNR and SPUR.

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DO concentrations in the anoxic and anaerobic tanks were kept around or below 0.1 mg/L; the DO concentrations in the two aerobic tanks were around 0.9 and 2.8 mg/L, respectively. The lower DO levels in the first aerobic tank were due to the more extensive biotranformations of nutrients (especially COD degradation), because this tank should be subjected to higher organic loading rates than the second one (i.e., membrane tank). On the other hand, a too high DO level in this tank could lead to the consumption of more carbon source, which can limit the occurrence of denitrification in the following tanks. In this study, we found that DO concentrations in a range of 0.8–1.5 mg/L were preferred in the first aerobic tank. Nonetheless, the second aerobic tank or the membrane tank was expected to play important roles in the occurrence of nitrification and P uptake, which usually need sufficient oxygen. To confirm this hypothesis, more batch tests, focusing on the influence of DO concentrations on nitrification and P uptake, were conducted. As shown in Figure 7, the SNR was much higher as the DO concentrations were higher than 2 mg/L. Both, however, declined greatly at lower DO concentrations (in a range of 1–2 mg/L). It is well-known that the AOB and particularly NOB have a much lower oxygen affinity, and thus low DO concentrations can inhibit ammonium and nitrite oxidation, resulting in low nitrification rates and/or the nitrite accumulation.[34,35] This study also found nitrite accumulation at low DO concentrations (data no shown). Results of Figure 7 also show that the SPURs increased with increasing DO concentrations, suggesting that a higher DO level could benefit for P uptake. A previous study also showed that P release began at DO concentrations between 0.1 and 0.5 mg O_2/L.[36] Our current data, together with previous findings, reveal that keeping a higher DO concentration in the aerobic tank is desirable for P uptake.

Discussion

In this study, we reported on the performance of a new NPFMBR developed on the basis of PFR rather than CSTR. The NPFMBR may provide an alternative for wastewater treatment, especially for the treatment of high-strength wastewater. To date, the major problems limiting the BNR performance in MBRs could be (1) the low mass transfer of solid/liquid/gas in the mixed liquor, (2) the mixing of the treated and untreated pollutants, and (3) the difficulty in enrichment of specialized microorganisms responsible for BNR. In the NPFMBR, the bioreactor was divided into high and narrow channels, which allows the sludge and pollutants to transport in a nearly plug-flow pattern. In this way, the biological reactions can proceed when the pollutants go through the channels. We can expect that the reaction rates are high at the inlet, but they decline as the untreated pollutants decrease and the products increase in the channel. Therefore, PFR-type MBR can produce concentration gradients and reaction rate gradients along the axis of the channels, finally leading to high mass-transfer coefficients and thus high biotransformation rates.[37] Crucially, the PFR-type reactors have higher potential to control filamentous bulking, such as *Microthix parvicella*,[38] because the imposed substrates gradients can create favorable conditions for the growth of floc-forming bacteria and limit filaments growth.[38] At this point, the PFR-type MBRs can aid in mitigating filament-causing membrane fouling. Previously, Smith and Oerther[39] found that that *Nitrosomonas europea* was the major AOB in the PFR-type reactor, while *Nitrosomonas marina* and *Nitrosomonas europea* were enriched in the CSTR-type reactor. Therefore, the mass gradients, such as DO concentrations and pollutants concentrations, can enhance the growth and cultivation of specialized microorganisms. It can be expected that the MBR configurations should have substantial influence on the bacterial community of the sludge. This needs further investigation. More interestingly, the plug flow can avoid backmixing between treated and untreated pollutants and, therefore, aid in the separation of treated water from the mixed liquor. Thus, the decrease of backmixing can to some extent improve the biological treatment efficiency. As a result of the above-mentioned features, the NPFMBR is expected to remove nutrients more efficiently. In comparison to the reported CSTR in series MBRs by Bracklow et al.,[31] the NPFMBR obtained good performance on nutrient removals at much lower COD/TN ratios of the feed wastewater and at lower sludge recirculation rates. Because of the different feed wastewater compositions as well as the different operating conditions (e.g., SRT and HRT), it is difficult to conclude which type of MBRs is better. But, the long-term measurements and batch experiments in this study can corroborate the benefits of PFR-type MBRs.

Over a long-term operation of 1 year, results showed that the NPFMBR can accomplish high removal efficiencies of COD, ammonium, TN, and TP. Process success was corroborated by batch experiments. Nitrification and P uptake could be enhanced at elevated DO concentrations (>2 mg/L). The batch experiments not only suggest the presence of AOB and NOB but also demonstrate the high SNR and SDNR in comparison with previous research. These findings, together with the high rates of P release and uptake, suggest that the specialized microorganisms, including nitrifying bacteria (AOB and NOB), denitrifying bacteria, and PAOs, were enriched in this NPFMBR. Previously, the research group at TU Berlin has conducted considerable work on the BNR with MBRs composed of a series of CSTR-like tanks.[7,31] They observed that the lab-, pilot-, and full-scale MBRs can achieve satisfying BNR. It should be noted that most of these MBRs accomplished BNR through postdenitrification rather than predenitrification. In the postdenitrification process, part of the substrate (i.e., COD) derived from the feed wastewater was stored as the form of intracellular polymers or glycogen, which will act as the main energy or carbon source for the following denitrification and P removal.[7,33] In the predenitrification process, the organics in wastewater can be used as the carbon source for denitrification directly, and

the abundance of organics can help to the proliferation of PAOs in the first aerobic tank. Therefore, these two processes are of different mechanisms in BNR. Different to previous research, only one pump was set for sludge recirculation in the NPFMBR. It not only can save pump energy and investment but also is easy for operation.

In addition, the membrane tank in the NPFMBR was also very compact, which is expected to control membrane fouling with as low as possible aeration rates. As a new process, however, possible limitations cannot be avoided. The main problem is the much more complex structure of bioreactor configuration, leading to more work to design and fabricate it. Additionally, mixers should be set in the anaerobic and anoxic tanks to keep the sludge in suspension. To achieve high performance, the DO concentrations and alkalinity in the sludge of each tank should be optimized. These problems have to be considered in the real application of this process.

Conclusions

In this study, a nearly plug-flow MBR was designed, and its performance in BNR was investigated on the basis of long-term monitoring. The main findings can be summarized as follows:

- The NPFMBR can achieve higher removal efficiencies of TN and TP up to 85% and 89%, respectively, at lower COD/TN ratios of 5–8. The remaining TN was found in the form of nitrate, indicating a complete nitrification process. These findings indicate that enhanced BNR was achieved by the NPFMBR.
- The high oxygen uptake rates of AOB and NOB suggest the abundance and high activities of these nitrifying bacteria in the sludge. The higher SNRs, SDNRs, SPRRs, and SPURs also demonstrate that the sludge had a high potential to nitrification, denitrification, and phosphorus removal.
- DO concentrations and alkalinity of the sludge suspension should be optimized to enhance TN and TP removal in the NPFMBR. DO concentrations higher than 2 mg/L is needed to warrant the good performance of nitrification and P uptake.
- As a new configuration of MBR, the nearly plug-flow can to some extent enhance the mass transfer in the mixed liquor, and importantly it can mitigate the occurrence of backmixing between treated and untreated pollutants. However, the limitations of the NPFMBR should be addressed before real application being considered.

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