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Removal of nitrogen from wastewater for reusing to boiler feed-water by an anaerobic/aerobic/membrane bioreactor

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Abstract

The innovative process anaerobic/aerobic/membrane bioreactor (A/O/MBR) was developed to enhance pre-denitrification without the energy consumption of the recirculation pump for reusing wastewater to boiler feed-water. The performance of this bioreactor was investigated. Firstly, the septic tank wastewater with low ratio of COD/TN was disposed by a dynamic membrane bioreactor (DMBR). It was found that, although the high concentration of NO_2^-–N in the effluent implied the potential ability of DMBR to realize shortcut nitrification and denitrification, the effluent of single DMBR was difficult to reach the criteria of reusing to boiler feed-water. Then, the process A/O/DMBR in disposing the septic tank wastewater was studied. The results indicated that this process not only accomplished the removal of 91.5% COD, 90.3% NH_4^+–N and 60.2% TN, but also successfully realized pre-denitrification without additional recirculation pump. At last, based on the A/O/DMBR, a pilot plant A/O/MBR was built to dispose the municipal raw sewage. In the stable operation period, the average removal efficiencies for COD, NH_4^+–N, TP and turbidity reached 90%, 95%, 70% and 99%, respectively. During the tested HRT run of 9.0 h, the effluent of COD, NH_4^+–N, TP and turbidity was about 10 mg/L, 3 mg/L, below 1 mg/L and 1.2 NTU, respectively, which reached the criteria of the boiler feed-water in China.

Keywords: Pre-denitrification; Membrane bioreactor; Anaerobic/aerobic/membrane bioreactor; Boiler feed-water; Nitrogen

1. Introduction

Various conceptualized, studied and operating nitrogen removal processes can largely be classified into three major groups, namely post-denitrification, pre-denitrification and simultaneous nitrification and denitrification (SND) processes [1]. The requirement of external carbon addition and provision of anoxic basin in post-denitrification not only cease the possibility of use of influent organic carbon in denitrification, but also pose serious threat of exceeding final effluent BOD criteria in the case of overdosing and nitrogen loading variations. The attractive alternative of SND offers benefits such as no additional requirement of reaction space, energy savings, and recovery of alkalinity. The DO concentration gradient across large sludge flocs and intermittent aeration are two known basic mechanisms behind SND [2,3]. However, the essentiality of external carbon addition for complete denitrification, the evident complexity in the steep process control on floc size [4] and DO concentration [5,6] limit the acceptability of SND as a preferred treatment option for highly variable influent conditions experienced in many industrial applications. In conventional wastewater treatment plants, nitrogen removal is mostly achieved with pre-denitrification for two major reasons. Firstly, biodegradable organic matter available in the anoxic zone improves denitrification rate (DNR), hence reducing the required volume of biological reactor. Secondly, the oxidation capacity of nitrate degrades the part of the organic matter, hence reducing the oxygen demand and achieving savings in aeration requirement. Nevertheless, the nitrogen removal rate depends on the recirculation ratio that transfers the nitrate produced by nitrification in the aerated zone back to the anoxic zone and therefore consumes energy [7].

On the other hand, though conventional technologies of removing nitrogen have been playing an important role in disposing wastewater, there are many problems to be solved, such as the need of adding carbon source in treating the wastewater with low C/N rate, demanding more energy for oxidizing ammonia into nitrate than into nitrite, and occupying larger field and higher investment. So a series of new technologies of bio-denitrification

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for low C/N ratio wastewater have been developed. Firstly, as a highly effective technology, SND has been widely applied in disposing wastewater of low C/N rate. Andrade do Canto et al. using a sequencing batch biofilm reactor (SBBR) have produced the treated effluent only 0.2 mg/L NO_2^-–N, 4.6 mg/L NO_3^-–N and 1.0 mg/L NH_4^-–N remained by SND [8]. Chiu et al. have reported the influence of C/N rate on the removal rate of ammonia and COD in the SBR-SND system [9]. Secondly, as it demands less carbon for removing the same amount of nitrogen, the technology of shortcut nitrification and denitrification is usually the preferential choice in disposing wastewater of low C/N ratio. Ciudad et al. have realized stably shortcut nitrification and denitrification for 170 days by partial nitrification [10]. Yu et al. have also reported that a part of ammonia in the influent should be owed to shortcut nitrification and denitrification [11]. Thirdly, as a novel technology, anaerobic ammonium axidation (ANAMMOX) has much predominance, such as needing no carbon and demanding less oxygen, thus receiving more and more attentions. Waki et al. have examined the applicability of the ANAMMOX process to three kinds of low BOD/N ratio wastewaters from animal waste treatment processes in batch mode [12]. Pathak et al. have combined ANAMMOX and denitrification and realized 80% of nitrogen removal [13]. Finally, to solve the difficulty of disposing wastewater with low C/N rate, many studies on improving the nitrogen removing ability of microorganism have been performed, leading to the technology of full autotrophic biodenitrification. Rocca et al. have combined the heterotrophic and autotrophic denitrification for disposing drinking water [14]. Sun et al. have completely realized the autotrophic nitrogen-removal in the process of wetland [15]. However, the enhancement and optimization of process designs for denitrification, especially for employing pre-denitrification, are seldom reported.

Thus, in this work, we developed an integrative A/O/MBR system (Fig. 1), consisting of anaerobic zone, aerobic zone, sedimentation zone and MBR, which combined the technologies of A/O, biological contact oxidation process and membrane bioreactor. In this system, the nitrated water and activated sludge in the aerobic zone were circulated into the anaerobic zone by the aeration but without the help of recirculation pump, which could reduce the processing cost. Furthermore, the rate of circulation could be easily manipulated for the need of denitrification and biological phosphors removal. Thus, the pre-denitrification in the economical A/O/MBR plant and reusing the treated wastewater for boiler feed-water were realized in this work.

1.1. Special zones

Recirculation zone: As shown in Fig. 1, the nitrated water could be gingerly recirculated into the anoxic zone with the help of aerating in the aerobic zone, yielding the pre-denitrification without using recirculation pump, because the anaerobic zone and aerobic zone were equipped in the same reactor.

Sedimentation zone: In order to retain the sludge, a sedimentation zone was equipped between the aerobic zone and MBR. The volume of sedimentation zone could be adjusted according to the property of the sludge in this zone. At the prophase of the test, the cubage of sedimentation zone was necessarily increased to form a reticulate structure of flocculent sludge with lower settleability, which could strengthen the separation of sludge and water in this sedimentation zone. While the settleability of sludge was improved, especially appearing granulated sludge, the HRT in this sedimentation zone could be reduced. Moreover, the process of post-denitrification was also observed in the sedimentation zone, but its contributions to the nitrogen removal needed further studies.

1.2. Distinct features

The system of A/O/MBR not only has the common features of conventional MBR, but also has many distinct features.

(1) The integrative system was simple and occupied less area, which could reduce the plant investment.

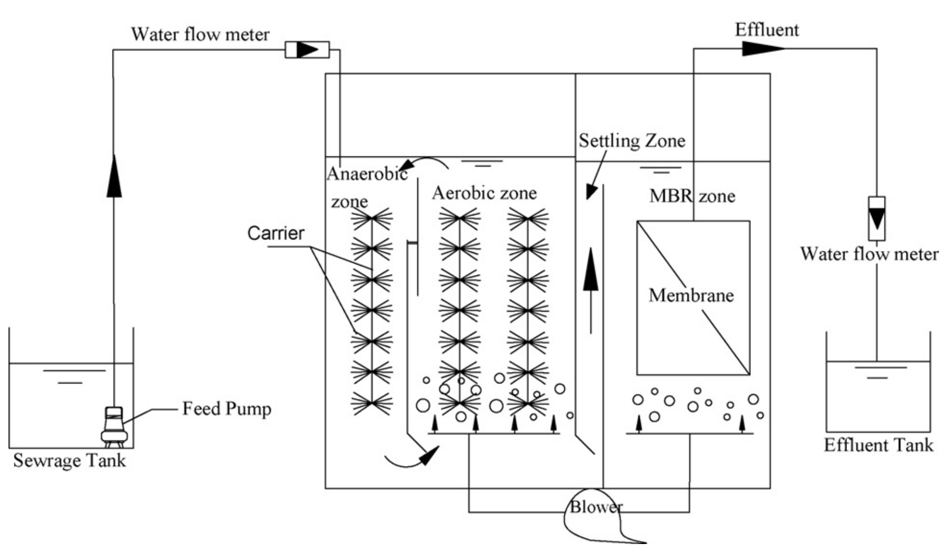

Fig. 1. Diagram of the integrative anaerobic/aerobic/MBR system.

Table 1
Quality of feed wastewater

Analysis items	COD (mg/L)	BOD (mg/L)	NH_4^+–N (mg/L)	TN (mg/L)	TP (mg/L)	Turbidity (NTU)	pH
Bench scale test	156–1286	–	150–230	199–86	21–40	240–600	7–8
Pilot test	78–126	38–67	9–19	–	1–3	56–84	6–8

(2) Though the zones of aerobic and anaerobic zones were separated by the clapboard, the bottoms and tops were joined, which provided the opportunity for realizing the pre-denitrification without special recirculation device.
(3) As shown in Fig. 1, both of the aerobic and anaerobic zones were installed with carriers, which not only improved the complexity of the microorganism and provided more microcosmic aerobic/anaerobic environments for SND, but also the SRT could be prolonged enough to enhance the nitrogen removal [16].
(4) Owing to the sedimentation zone and degrading of the matters in the A/O process, the MLSS in the MBR and the content of organic matters in the MBR influent were maintained in a low level, which could alleviate the pollution of membrane.
(5) The process of A/O/MBR realized the function of the process of anoxic/anaerobic/aerobic/MBR (A′/A/O/MBR or diversely A^2/O/MBR), putting the A′ zone ahead of the A zone, which could accelerate the removal of biologic phosphors.
(6) Richard [17] reported that the NH_4^+–N concentration over 10–15 mg/L and 0.1–1.0 mg/L had the adverse effects on the growth of nitrite-oxidizing bacteria and nitrate-oxidizing bacteria, respectively, by keeping the microbiology in the state of dormancy or low activity. However, in this system, the adverse effects of NH_4^+–N in the influent could be reduced by the nitrification at the top of the anaerobic zone and the dilution of the nitrated wastewater recirculated from the aerobic zone.

2. Materials and methods

2.1. Apparatus

Three plants were operated with different configurations on two different sites.

- One bench–scale plant (7.5 L) with MBR for simultaneous nitrification denitrication, here called DMBR and using dynamic membrane.
- One bench–scale plant (143 L) with A/O zone, sedimentation zone and MBR for pre-denitrification, here called A/O/DMBR and using DM.
- One pilot plant (2.5 m^3) with A/O zone, sedimentation zone and MBR for reusing the reclaimed water to the boiler feedwater, here called A/O/MBR and using hollow fiber microfiltration membrane.

In the system of A/O/MBR and A/O/DMBR, the wastewater was screened through 1 mm punch holes (rotatitive drum), and then flowed into the anoxic zone formed under the circulation bringing oxygen of aerobic zone into the anaerobic environment in the top of anaerobic zone. In the anoxic zone, NO_X–N could be translated into N_2 or N_2O and therefore nitrogen could be removed from the system. Then, the wastewater treated by the anoxic microorganism in the top of the anaerobic zone and anaerobic microorganism in the bottom of the anaerobic zone, flowed into the aerobic zone, sedimentation zone, MBR and passed through the membrane in sequence.

2.2. Source of wastewater

The bench–scale tests were fed with the septic tank wastewater, and the pilot test was fed with the municipal raw sewage, both of which were screened through 1 mm punch holes (rotative drum). The indexes of the influent quality were shown in Table 1.

2.3. Membranes

2.3.1. Membrane used in the pilot plant

The hollow fiber micro-filtration membrane of 0.6 mm inner diameter, 1.0 mm outer diameter and 0.2 μ pore-diameter was made of polyethersulphone by Tianjin Motimo Membrane Technology Ltd., China and used to construct a membrane module of the dead-end type in the pilot plant.

2.3.2. Dynamic membrane used in the bench–scale plant

The dynamic membrane subassemblies were similar to the plate-frame membrane and were made up of silk. The aperture of this kind of silk was 0.1 mm. The permeated water seeped through the surface of dynamic membrane and entered into the cavum inside of the membrane subassemblies, then effused from the reactor by the gather pipe and formed the effluent of the system at last.

2.4. Methods and process parameters

Conventional analysis items including pH, DO, COD, NH_4^+–N, TN, NO_X–N, turbidity and MLSS were carried out according to the standard methods issued by the China National Environmental Protection Agency [18]. The main process parameters were shown in Table 2.

3. Results and discussion

3.1. Performances of the DMBR

The detailed performances of the DMBR treating the septic tank wastewater were shown in Figs. 2 and 3. Due to the high con-

Table 2
Main process parameters in the three apparatuses

Apparatus	MLSS (mg/L)	HRT (h)	C/N	Temperature (°C)
DMBR	4000	5.0–10.0	0.8–5.7	15–25
A/O/DMBR	4000	9.0	0.8–5.7	15–25
A/O/MBR	6000	9.0	3.0–8.0	10–25

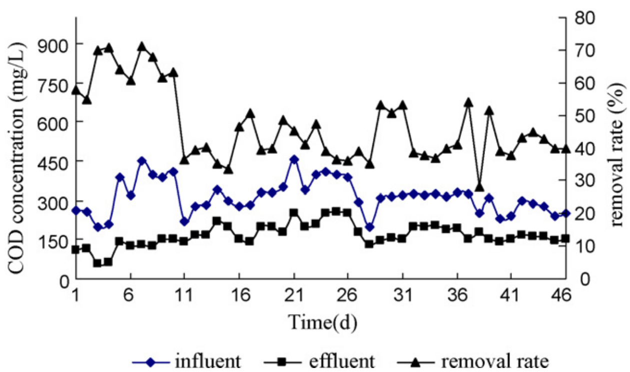

Fig. 2. Variation of the influent and effluent COD concentration and the removal rate of COD in DMBR.

centration of NH_4^+–N and low ratio of COD/TN in the influent, the DMBR, in the stable operation period, just reached the average COD and NH_4^+–N removals of 40% and 60%, respectively, and generated the effluent of COD and NH_4^+–N of more than 150 mg/L and 80 mg/L, respectively, during the experimented HRT run of 9.0 h and temperature of 20–26 °C. As shown in Fig. 4, neither the aims of SND in the DMBR were realized very well, although the nitrite of high concentration in the effluent implied the possibility of realizing shortcut nitrification and denitrication by improving the processing indexes.

3.2. Performances of the A/O/DMBR

3.2.1. Nitrification rate in the aerobic zone

Batch tests with washed sludge from the aerobic zone were implemented. As shown in Fig. 5, the correlation between the variation of the concentration of NH_4^+–N or NO_X–N and operating time implied that in a certain concentration range, the nitrification rate was free from the subtract concentration. The

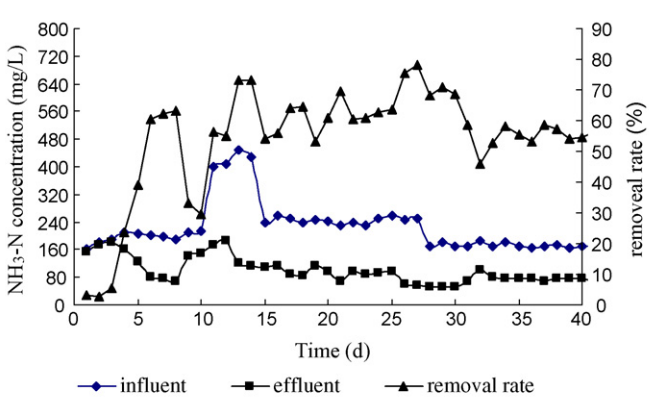

Fig. 3. Variation of the influent and effluent NH_4^+–N concentration and the removal rate in DMBR.

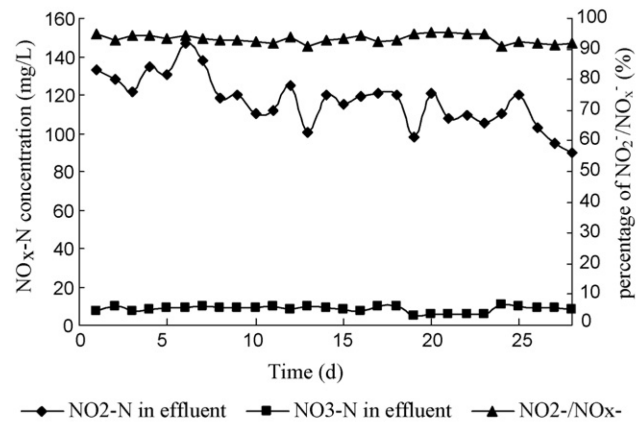

Fig. 4. Variation of the effluent NO_2^-–N and NO_3^-–N concentration and the rate of NO_2^-–N/NO_X–N in DMBR.

equation of the relationship between the nitrification rate and the concentration of subtract was in accordance with the zero order law according to some reports [19,20]. When the temperature was 20–27 °C, the nitrification rate was computed to be 0.236 d^{-1} according to Eq. (1).

$$V = \frac{r \times 60 \times 24}{M} \qquad (1)$$

V – nitrification rate, g NH_3–N/g MLSS d; r – the rate coefficient, M – MLSS, about 2107 mg/L.

3.2.2. Biomass specific denitrification rate in the aerobic and anoxic zones

The denitrification of the sludge recirculated from aerobic zone to the anaerobic zone not only realized the nitrogen removal, but also improved the nitrification rate in the aerobic zone by reducing the concentration of NO_X^-–N in the aerobic zone. Though the ratio of COD/TN in the influent was very low and unsuitable for denitrification, the reactor had an effective

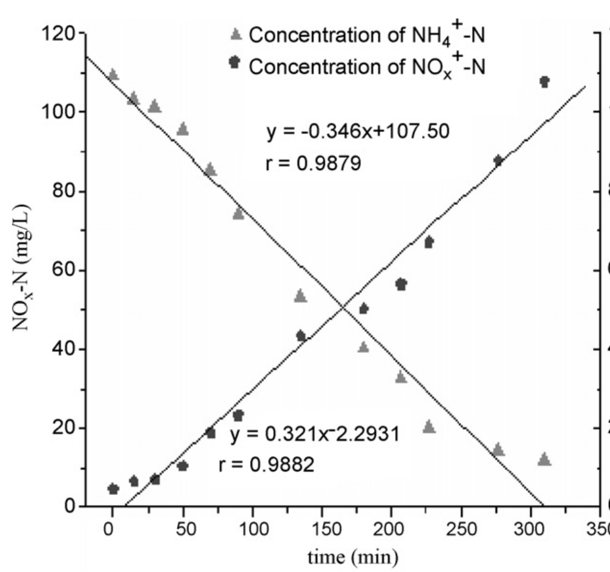

Fig. 5. Variation of NO_X–N and NH_4^+–N concentration with time in the aerobic zone of the A/O/DMBR system.

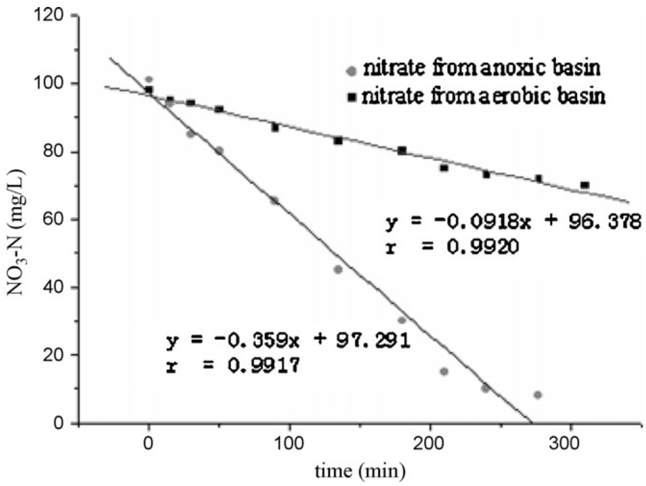

Fig. 6. Variation of NO_3–N concentration with time in the aerobic and anaerobic zones of the A/O/DMBR system.

nitrogen removal. The concentration of NO_3^-–N in the effluent could be reached as low as less than 25 mg/L when the influent NO_3^-–N concentration was 199–290 mg/L and the ratio of COD/TN was about 4.0. Furthermore, when the temperature was 20–30 °C, as shown in Fig. 6, the denitrification rates in the aerobic zone and anaerobic zone were 0.0627 d^{-1} and 0.231 d^{-1}, respectively, computed by Eq. (2).

$$V = \frac{r \times 60 \times 24}{M} \qquad (2)$$

V – denitrification rate, g NO_3^-–N/g MLSS d; r – the rate coefficient, M – MLSS, about 2107 mg/L in aerobic zone and 2235 mg/L in anaerobic zone.

Accordingly, in the system, the denitrification could be realized not only by recirculating the nitrated wastewater to the anaerobic zone for pre-denitrification, but also by the micro anaerobic–aerobic environments for the SND in the aerobic zone, although the rate of pre-denitrification was much quicker than that of SND. However, the system had great intrinsic ability for the nitrogen removal.

3.2.3. *Performance of the A/O/DMBR*

As shown in Figs. 7–9, though the fluctuating of the concentrations of COD, NH_4^+–N and TN in the influent were as high as 150–1300 mg/L, 150.4–230 mg/L and 198.5–285.5 mg/L, respectively, as the temperature was 20–30 °C, the system accomplished a COD removal efficiency of above 91.5%. However, the removal efficiency of TN and NH_4^+–N were influenced by the ratio of COD/TN in the influent, both of which could be improved by increasing the ratio of COD/TN, and could reach 60.2% and 90.3%, respectively, when the COD/TN was above 4.4. Interestingly, the increasing of COD/TN ratio not only improved the TN removal, but also accelerated the nitrification rate, which was impenetrable and on contrary to the theory of the nitrification. It is known that high ratio of COD/TN means the richness of organic substrates in the system, where heterotrophic bacteria can grow quickly and occupies most of oxygen and nutrition. Thus, as a kind of autotrophic bacteria, the

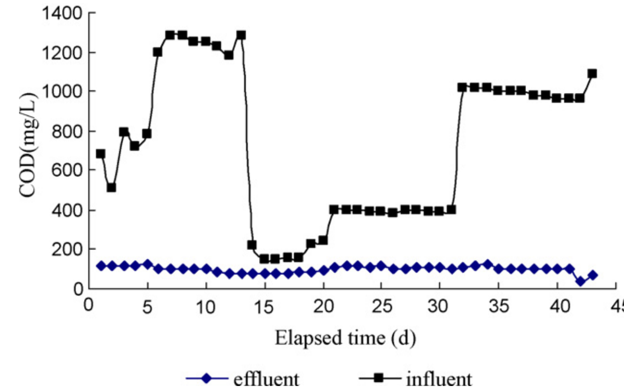

Fig. 7. Variation of the COD concentration in the influent and effluent of the system of A/O/DMBR.

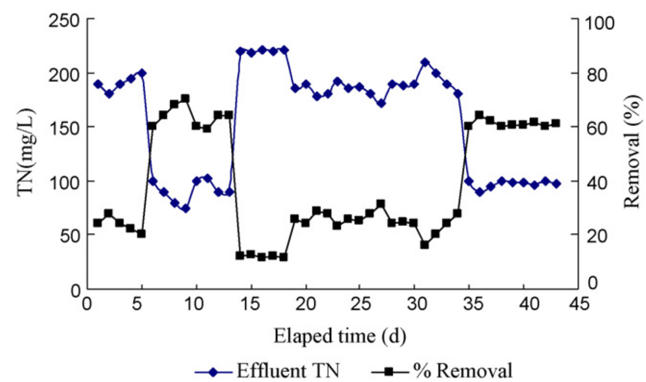

Fig. 8. Variation of the effluent TN and the removal rate of TN in the system of A/O/DMBR.

activity of nitrifiers is restrained, resulting in a low nitrification rate. However, in the test, the improvement of COD/TN ratio brought the increase of the removal rates for both of NH_4^+–N and TN. The reason might be as follows. In the system, the ratio of COD/TN was always very low, and the concentration of organic nutrition was not the main parameter influencing the growth of nitrifiers, but was the key index controlling the denitrification. The enhanced denitrification with higher ratio of COD/TN could remove the adverse effects of NO_X^-–N with high concentration on the growth of nitrifiers. However, if the environment was short of nutrition with high concentration of

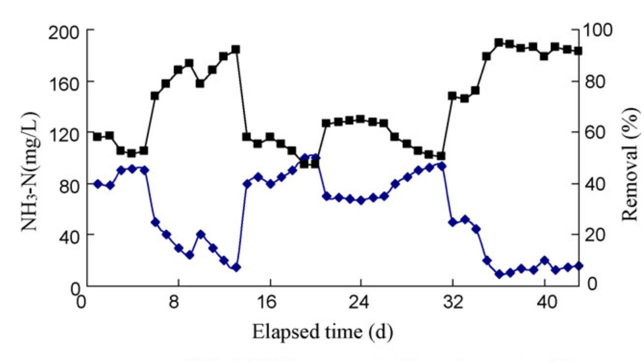

Fig. 9. Variation of the effluent NH_4^+–N and the removal rate of NH_4^+–N in the system of A/O/DMBR.

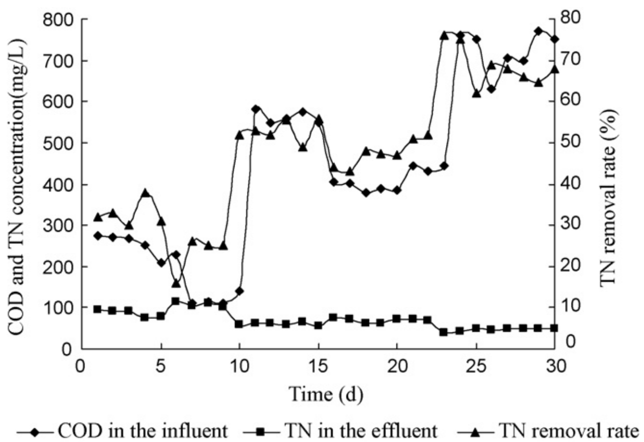

Fig. 10. Influence of the increase of the COD in the influent on the effluent TN concentration and the removal rate of TN in the system of A/O/DMBR.

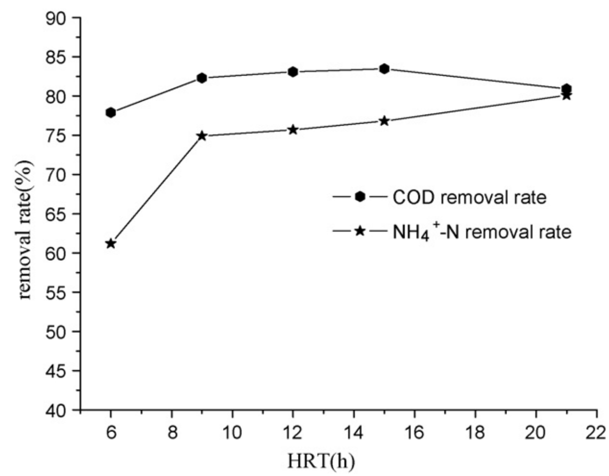

Fig. 11. Influence of HRT on the removal rates of NH_4^+-4 and COD in the system of A/O/DMBR.

NO_X^-–N, the concentration of NO_X^-–N would become higher and higher by accumulating and inhibiting both of denitrification and nitrification in the aerobic and anaerobic zone greatly at last.

3.2.4. The influence of C/N ratio and HRT on denitrification rate in A/O/DMBR

As shown in Fig. 10, when TN concentration was 136.5 mg/L, COD concentration was increased from 110.5 mg/L to 760.9 mg/L and the temperature was 20–30 °C, the denitrification rate was improved with the increase of COD/TN, and the removal rate of TN was improved from about 16% to 76% by increasing COD/TN ratio from 0.8 to 5.7. It had been reported that the denitrification rate could not be influenced by COD/TN ratio if it was more than 7.0. However, in this test, the largest COD/TN ratio was about 5.7, so the denitrification rate was restrained greatly by the low COD/TN ratio, implying that the system of A/O/DMBR with larger influent COD/TN ratio could reach much higher denitrification rate.

When temperature was 20–30 °C, the concentration of COD in the influent was 127.5–331.1 mg/L, and pH was 7.0–8.0, the average removal rates of COD and NH_4^+–N during the experimented HRT run of 6.0, 9.0, 12, 15 and 21 h were shown in Fig. 11. It was found that the influence of HRT on the removal rate of NH_4^+–N was greater than that on COD removal rate. And the optimal HRT should be about 9.0 h as considering both of the removal efficiency of the system and nitrogen removal.

3.3. Performance of the A/O/MBR

As shown in Table 3, the results inspected randomly by the Water Resources and Water Environment Monitoring Center of Wuhan City in China at the stable stage of system demonstrated that the effluent of the A/O/MBR was reliable enough for reusing to the boiler feed-water according to the standard shown in Table 4. Furthermore, the effluent of 6 m^3/h was stable in the overall period of more than 2 months, the removal rate of COD, NH_4^+–N, TP and turbidity were above 90%, 95%, 70% and 99%, respectively at the temperature of above 16 °C. The effluent of COD, NH_4^+–N, TP and turbidity were about 10 mg/L, about 3 mg/L, below 1 mg/L and 1.2 NTU, respectively. In addition, even though the concentration of coliform bacteria in the influent was as high as above 1,60,000 individuals/L, in the effluent it could reach at such a low level that the effluent could be used directly without disinfection.

Table 3
Quality of the influent and effluent monitored by the Water Resources and Water Environment Monitoring Center of Wuhan City of China

Indexes	Influent quality	Effluent quality	
		Concentration	Removal rate (%)
Temperature (°C)	15.8	17.0	
pH	7.1	7.0	
BOD_5 (mg/L)	48	<0.2	>99.6
COD (mg/L)	91.8	12.5	86.4
Fe (mg/L)	0.22	<0.01	>95
NH_4^+–N (mg/L)	18.84	0.32	98.3
TP (mg/L)	2.06	0.75	63.6
Coliform bacteria (individuals/L)	>160000	700	>99.6

3.4. The analysis of the fouling of membrane

During the 65 days, there was no chemical or physical washing for membrane cleaning. As mentioned above, the intermittent operation of the membrane (2 min rest for every 10 min

Table 4
Comparing of the quality between the standard of boiler feeding water in China and the effluent of the system of A/O/MBR

Indexes	The effluent quality of A/O/MBR	Standard of boiler feeding water
pH	7.1	≥7.0
SS (mg/L)	≤1.0	≤5.0
Total hardness (mmol/L)	≤0.01	≤0.03
Content of solid solute (mg/L)	≤100	<4000
PO_4^{3-} (mg/L)	<1.0	≤10
Fe (mg/L)	<0.01	≤0.3

of operation) and air bubbling were the only ways to control membrane fouling. As the volume ratio of gas/water, commonly 20:1 in conventional MBR, was 10:1, the trans-membrane pressure, effluent flux and the resistance of membrane were 0.015 ± 0.0025 MPa, 500–600 L/h and $4.3 \times 10^{11}\ m^{-1}$, respectively, at the first stage of the test. Then, as the volume ratio of gas/water was further reduced to 8:1, the trans-membrane pressure, effluent flux and the resistance of membrane were 0.020 ± 0.0025 MPa, 520 L/h and $5.5 \times 10^{11}\ m^{-1}$ respectively, demonstrating that the A/O/MBR could be operated at much lower ratio of gas/water. However, it was dangerous to reduce the ratio of gas/water. Furthermore, at the last stage of the test, the system was rest for one day resulting from accidental problems. It was found unexpectedly that the resistance of membrane was only $3.2 \times 10^{11}\ m^{-1}$ while the volume ratio of gas/water, trans-membrane pressure and effluent flux were 5:1, 0.0145 ± 0.0025 MPa, 650 L/h, respectively. This result means that there are gel layer and concentration polarization contributes to the membrane fouling although the resistance of filtration cake was the main part of the total resistance.

3.5. Analysis of expense of the A/O/MBR

The cost of MBR process included the investment of equipment and lands, and the operational cost of daily operation expanding and depreciation of equipment. If the scale was 20,000 m^3/d, in the conventional wastewater treatment, the investment and the cost of disposal were about 310.0 dollars/m^3 d and 0.264 dollars/m^3, respectively. However, in the system of A/O/MBR, the investment and cost of disposal were only 257.7 dollars/m^3 d and 0.214 dollars/m^3, respectively. The energy consumption of A/O/MBR system mainly included the consumptions of lift pump, sorption pump, air blower and excess sludge pump, total of which was about 0.8 kW h/m^3 wastewater. Thus, the development of integrative A/O/MBR system was suitable for the demanding of saving energy for the development of water treatment technology in the future.

4. Conclusions

(1) Though the pollutant removal ability of the DMBR in disposing the septic tank wastewater with low COD/TN is limited, the nitrite of high concentration in the reactor shows a possibility of realizing shortcut nitrification and denitrication in the DMBR. The high concentration of NH_4^+–N may accelerate the accumulation of nitrite, because NH_4^+–N of high concentration can restrain the action of nitrobacteria, but has few influences on nitrite-oxidizing bacteria.
(2) Though the ratio of COD/TN in the influent is much lower than the need for denitrification, the integrative A/O/MBR can realize the pre-denitrification without the energy consumption of the recirculation pump and thus reducing the consumption of energy greatly.
(3) The effluent of the innovative process A/O/MBR in disposing the municipal raw sewage is reliable for reusing to boiler feed-water. The application of this process not only meets the nowadays demanding of saving energy source and reducing investment in disposing wastewater, but also provides a successful attempt for reusing the treated wastewater in those fields with high quality water demanding.
(4) The membrane fouling is effectively alleviated by retaining the sludge in the sedimentation zone and degrading of organic matters in the zones before MBR. The volume ratio of gas/water for controlling of membrane fouling could be as low as 8:1.

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