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Nitrogen and phosphorus removal in an airlift intermittent circulation membrane bioreactor

Haiyan Guo[1,2,*], Jiandong Chen[1,2], Yun Li[1,2], Tengteng Feng[1,2], Shoutong Zhang[1,2]

1. School of Environmental and Chemical Engineering, Dalian Jiaotong University, Dalian 116028, China
2. Department of Environmental Science and Technology, Dalian University of Technology, Dalian 116024, China

Abstract

A new airlift intermittent circulation integrated bioreactor was developed for simultaneous nitrogen and phosphorus removal of wastewater, in which, circulation of mixed liquid between mixing zone and aeration zone was realized by aeration power, alternately anaerobic/anoxic bio-environment in mixing zone was realized by intermittent circulation and simultaneous nitrogen and phosphorus removal was obtained through strengthened denitrifying phosphorus removal process. Removal performance of the reactor was investigated and pollutant removal and transfer mechanism in one operation circle was analyzed. The experiment results indicated that under the influent condition of chemical oxygen demand (COD) concentration of 642.1 mg/L, total nitrogen (TN) of 87.4 mg/L and PO_4^{3-}-P of 12.1 mg/L, average removal efficiencies of COD, TN and PO_4^{3-}-P reached 96.4%, 83.2% and 90.5%, respectively, with the hydraulic residence time of 22 hr and operation circle time of 185 min. Track studies indicated that the separation of aeration and mixing zones and intermittent circulation of mixed liquid between the two zones provided distinct biological environments spatially and temporally, which ensured the occurrence of multifunctional microbial reactions.

Key words: nitrogen and phosphorus removal; membrane bioreactor; intermittent circulation

Introduction

Membrane bioreactor (MBR) is considered a promising technology for future wastewater treatment due to its higher efficient performance and smaller footprint than those of conventional activated sludge process. In MBR, complete separation of biomass by membrane filtration makes it possible to maintain high concentration of mixed liquor suspended solids (MLSS) in the reactor. Usually, intensive aeration is carried out in MBR for supplementation of oxygen to microorganisms and cleaning of the membrane, one obvious drawback of the intensive aeration is poor removal of nitrogen. One possible approach to overcome this drawback is to install an additional anoxic reactor in which denitrification occurs followed by the aerobic MBR (Ahn et al., 2003; Patel et al., 2005; Song et al., 2009). However, with such configuration, many important advantages of MBR such as compactness or ease to operation will be impaired. On the other hand, MBR with novel configuration or operating method for nitrogen removal in a single reaction tank has been examined by several researchers, e.g., through the approach of intermittent aeration (Hasar et al., 2001; Choi et al., 2008), through the sequencing batch reactor (SBR) operation mode (Yang et al., 2010), by inserting baffles or internal-loop into the membrane chamber (Kimura et al., 2008; Li et al., 2008), and appropriated method of wastewater addition. In the MBRs mentioned above, aerobic and anoxic conditions were induced spatially or temporally in a single reactor, however, anaerobic and anoxic conditions were not compartmentalized and simultaneous nitrogen and phosphorus removal was not effective due to organic substrate competition between phosphorus accumulating organism (PAOs) and denitrifying bacteria, especially under the condition of treating the domestic wastewater with lower concentration of organic matter.

Denitrifying phosphorus accumulation phenomenon in wastewater treatment plants and laboratory-scale experiments has been widely reported (Kuba et al., 1993; Ahn et al., 2002; Guo et al., 2012). It is suggested that some of the PAOs are capable of utilizing nitrate instead of oxygen as an electron acceptor for phosphorus uptake and nitrogen removal and biological phosphorus removal can be achieved simultaneously, therefore, the application of denitrifying phosphate-accumulating organisms (DNPAOs) has been highlighted to promote the nitrogen and phosphorus removal efficiency of wastewater with a

* Corresponding author. E-mail: ghy2721@sina.com

low organic carbon. To address the problems stated above, an airlift intermittent circulation membrane bioreactor was developed, in which, anaerobic, anoxic and aerobic conditions could be created both spatially and temporally, and denitrifying phosphorus removal process was supposed to be strengthened during nitrogen and phosphorus removal processes. In this article, the performance of the proposed reactor was evaluated and track study of parameters in a single operating circle was conducted to find the detailed path-way of pollutant removal.

1 Materials and methods

1.1 Airlift intermittent circulation membrane bioreactor

Figure 1 shows the concept of the airlift intermittent circulation membrane bioreactor. A guide baffle discriminated the reactor into two compartments, namely, mixing zone and aeration zone, both zones were filled with support carriers. Membrane module was set in the aeration zone, to prevent moving carrier impairing it, a pored baffle was inserted and separated it into two zones, called aerobic zone and membrane zone. Raw wastewater was pumped into the mixing zone intermittently and effluent was drawn out by membrane filtration constantly, variation of water level ranging from the set highest level to the lowest level resulted in the intermittent liquor recirculation between the aerobic and mixing zones. The aerobic zone was supposed to be aerobic and nitrification and P-taken up take place all the time, well, the mixing zone was supposed to be alternately anaerobic and anoxic and P-release, denitrification and denitrifying phosphorus removal processes could take place sequently. The experimental-scale reactor was made of transparent polyvinyl chloride plastic, total effective working volume was 26 L with mixing zone of 7.7 L, aerobic zone of 7.7 L and membrane zone of 10.4 L, plastic support material (diameter 10 mm, height 10 mm) were added to the mixing zone and aerobic zone with the filling ratio (volumetric filling in empty reactor) of 40%. Membrane module set in the aerobic zone was made of hollow fiber membranes with pore size of 0.1–0.2 μm and filtration area of 0.2 m^2.

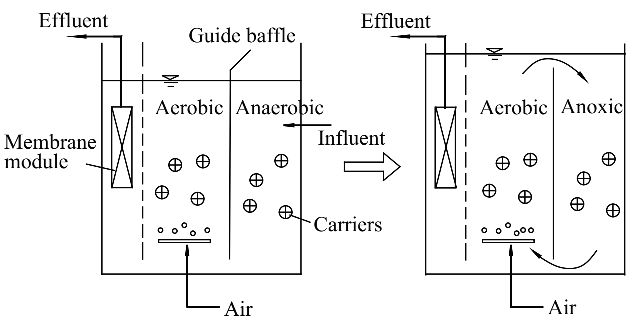

Fig. 1 Concept of the airlift intermittent circulation membrane bioreactor.

1.2 Feed wastewater and operation condition

Before regular monitoring in this study, the reactor was inoculated with activated sludge and biofilm on the carriers, which were cultivated in the way of intermittent aeration for about one month. The feed was synthetic wastewater which was produced by mixing glucose, NH_4Cl, KH_2PO_4 and $NaHCO_3$ with other required nutrients in tap water. COD, NH_4^+-N and PO_4^{3-}-P concentrations of the feed were in the range of 450–710 mg/L, 45–89 mg/L and 10.6–18.6 mg/L, respectively. Hydrogen retention time (HRT) of the reactor was 22 hr and intermittent circulation circle (operation circle) was 185 min. Aeration rate of the reactor was 0.2 m^3/hr and temperature of wastewater in the reactor was kept at 20–25°C during the whole operational period.

1.3 Analytical methods

A routine monitoring determined the overall performance of the new reactor, while a track study provided data on the concentration of specific compounds over each operational condition in a cycle. NH_4^+-N, NO_3^--N, NO_2^--N, COD and PO_4^{3-}-P were measured according to Standard Methods (APHA, 1999). TN was measured as the sum of NH_4^+-N, NO_3^--N and NO_2^--N. Dissolved oxygen (DO), pH and oxidation-reduction potential (ORP) were measured by a DO meter (Hanna HI9147, Italy) and a pH/mV meter (Hanna HI8424NEW, Italy).

2 Results and discussion

2.1 Removal performance of the reactor

Figure 2 shows the removal performance of the reactor. During the operation day of 1 to 15, the influent COD, NH_4^+-N and PO_4^{3-}-P concentration were 454, 46.9 and 16.7 mg/L respectively, and the effluent COD concentrations were less than 30 mg/L and TN concentrations less than 5.6 mg/L. But the averaged PO_4^{3-}-P concentration in the effluent was about 10 mg/L and PO_4^{3-}-P removal efficiency was only about 38.7%. Poor phosphorus removal performance in this period indicated that accumulation of PAOs was more sensitive than that of denitrifiers under the same cultivation condition. It was reported that the influent ratio of BOD_5/TP should be kept at 20–30 to provide enough organic substrate for PAOs to synthesized PHB (Shen and Wang, 1999) and PHB could only be decomposed under the condition of enough oxygen and limited organic carbon (Lv and Hu, 2004) . In this study, the influent COD/PO_4^{3-}-P ratio was about 27.3 and there was sufficient organic carbon for P-release, but considering that the operation circle time of the integrated reactor was not optimized and quite amount of influent organic carbon could be circulated to the aerobic zone and oxidized by the aerobic heterotrophic bacteria, during the operation day of 16 to 30, the influent COD concentration was increased for the better cultivation of PAOs, also, NH_4^+-N

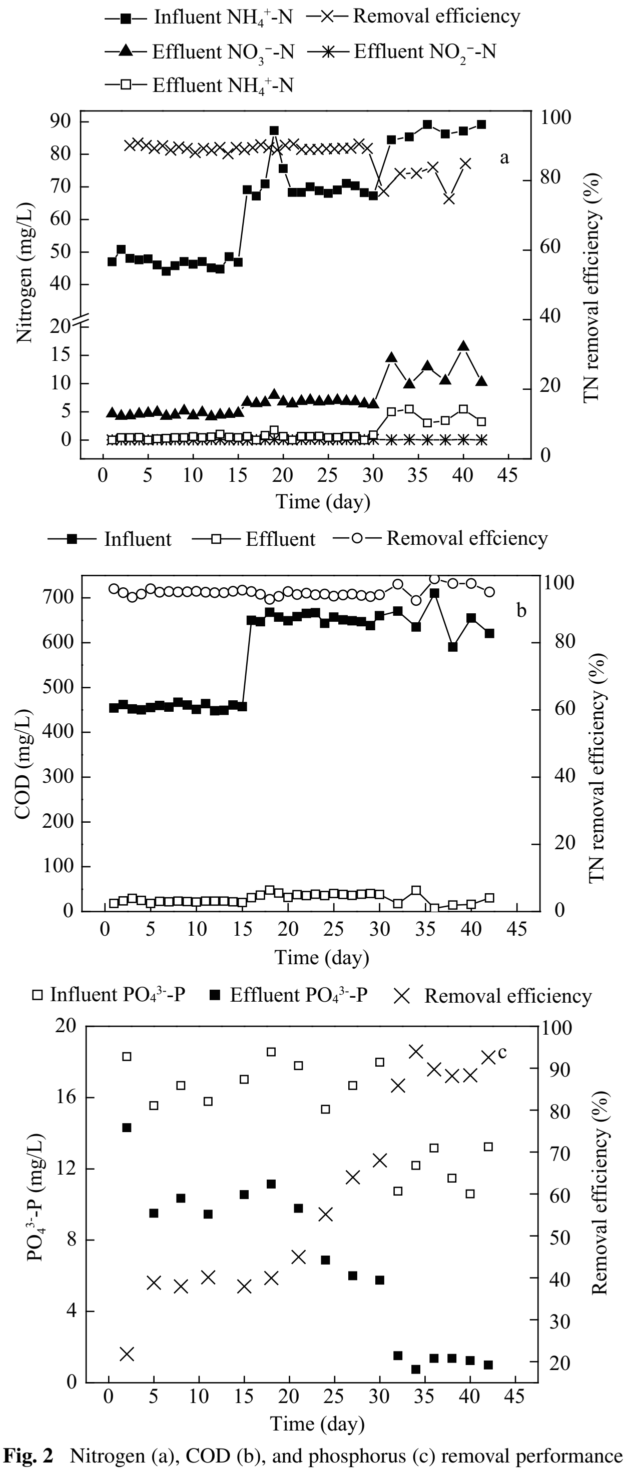

Fig. 2 Nitrogen (a), COD (b), and phosphorus (c) removal performance of the reactor.

concentrations in the influent was increased to investigate the nitrogen removal capability of the reactor.

As shown in **Fig. 2a** and **b**, nitrogen and COD concentrations in the effluent were steady and TN and COD removal efficiencies reached 89.5% and 94.2% respectively. PO_4^{3-}-P in the effluent dropped gradually to 5.7 mg/L during day 1 to 30, the improved phosphorus removal performance was resulted from the increased COD concentration in the influent, which provided more organic carbon for PAOs to conduct P-release reaction.

During the operation day 31 to 42, when the influent COD concentration was kept constant, NH_4^+-N concentration in the influent was further increased to 87.4 mg/L and PO_4^{3-}-P concentration in the influent was decreased to 12.1 mg/L to probe the nitrogen removal capability and obtain better phosphorus removal efficiency. **Figure 2a** shows NH_4^+-N concentration in the effluent rose slightly to 4.3 mg/L and NO_3^--N concentration increased to 12.4 mg/L, denitrification was the limiting step in the nitrogen removal process, however, averaged TN removal efficiency of 83.2% could still be abstained. **Figure 2c** shows PO_4^{3-}-P concentration in the effluent dropped significantly to 1.2 mg/L and PO_4^{3-}-P removal efficiency of 90.5% was obtained.

2.2 Removal characteristics in an operating cycle

Figure 3 shows the typical profiles of DO, COD, PO_4^{3-}-P and nitrogen in one operation circle of the reactor at a pseudo-steady state when simultaneous nitrogen and phosphorus removal was obtained. As shown in **Fig. 3**, one operation circle composed 4 stage. In T_1 stage, when the water level reached the set lowest level, addition of wastewater was initiated and liquid level was allowed to rise, the supply of organic matter in the feed and absence of nitrate made it possible for the occurrence of P-release in the mixing zone. In T_2 stage, when the water level was higher than the top of the baffle, mixed liquor was circulated from aeration zone to mixing zone, the continuous supply of organic matter in the feed and nitrate from aeration zone made it possible for the occurrence of denitrification in the mixing zone. In T_3 stage, addition of wastewater was stopped when the water level reached the set highest level, then the water level went down to the top of baffle due to membrane filtration, denitrifying phosphorus removal was supposed to happen due to the existence of nitrate from aeration zone and the absence of organic matter. In T_4 stage, water level went down continuously until to the set lowest level, the remained pollutants was further removed and microorganisms in both the aeration and mixing zones were ready for the next operating circle.

As indicated in **Fig. 3a**, DO concentration in the mixing zone was always below 0.2 mg/L during the whole operating circle, which provided the precondition for the occurrence of denitrification, P-release and denitrifying phosphorus removal processes. DO concentration in the aerobic zone and membrane zone had the similar variation trend, decreasing during T_1 and T_2 stages and recovered during T_3 and T_4 stages. The decrease of DO concentration during T_1 and T_2 stages in the aerobic zone was resulted from the oxidization of organic carbon and NH_4^+-N coming from mixing zone under the effect of liquid diffusion and circulation. The lowest DO concentrations in the aero-

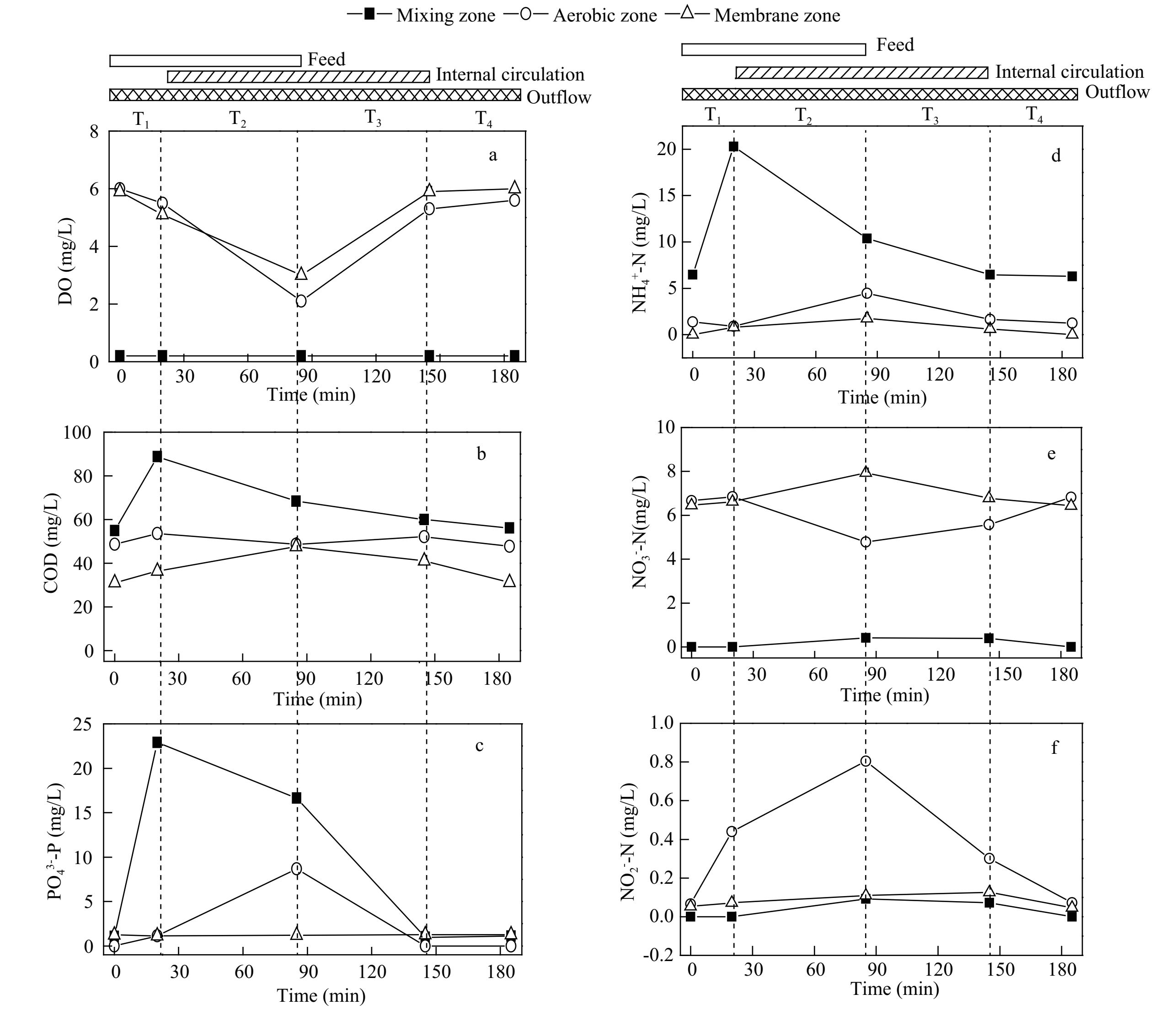

Fig. 3 Parameters DO (a), COD (b), PO_4^{3-}-P (c), NH_4^+-N (d), NO_3^--N (e) and NO_2^--N (f) variation in different zones of the reactor during one operating circle.

bic and membrane zone were 2.1 and 3 mg/L respectively, and nitrification process would not be interrupted under such DO condition. When the inflow feed stopped, DO concentration in the aerobic increased to 5.3 mg/L at the end of internal circulation stage.

Figure 3b shows, in spite of the induce of wastewater during the T_2 stage, in mixing zone COD decrease rate at T_2 phase was almost the same as that at T_3 stage, indicating that influent organic carbon was mainly used for denitrification process in mixing zone. COD concentrations in both the aerobic zone and membrane zone fluctuated in the range of 31–54 mg/L, and the COD concentration in the membrane zone was always lower than that in the aerobic zone.

Figure 3c shows PO_4^{3-}-P in the mixing zone demonstrated regular increase and decrease variation trend, while, PO_4^{3-}-P in the membrane zone was always less than 1.3 mg/L. For mixing zone, PO_4^{3-}-P concentration increased during T_1 stage and decreased continuously during T_2 and T_3 stages, and the maximum PO_4^{3-}-P concentration value attained to 22.9 mg/L the at the end of T_1 stage. Considering that the influent PO_4^{3-}-P concentration was only 12 mg/L and the influent volume of wastewater was only 1/4 of the volume of the mixing zone, the increase of PO_4^{3-}-P concentration during T_1 stage was mainly caused by P-release. During T_2 stage, in spite of continuous wastewater inflow, PO_4^{3-}-P concentration in mixing zone declined to 16.6 mg/L under the effect of mixed liquor circulation. It is important to note that PO_4^{3-}-P concentration dropped sharply to 1.1 mg/L during T_3 stage. This is due to the mixed liquid circulation to aerobic zone and the occurrence of denitrifying phosphorus removal in the mixing zone. DNPAOs in the activated sludge, especially those in the biofilm on support material in the mixing zone utilized NO_3^--N as electron acceptor for phosphorus uptake. Batch studies also indicated that biofilm on support material in the mixing zone showed higher denitrifying phosphorus removal rate than that from the aerobic zone (data not shown in this study).

Figure 3d, e, and **f** show the changes of nitrogen in one operation circle time. NH_4^+-N concentration in mixing zone was higher than that in aerobic and membrane zones, while, NO_3^--N concentration in mixing zone was far lower than that in aerobic and membrane zones, indicating that nitrification took place predominantly in aerobic and membrane zones and denitrification occurred mainly in mixing zones. NH_4^+-N in mixing zone and NO_2^--N in aerobic zone demonstrated regular variation with time in one operation circle, the accumulation of NO_2^--N in aerobic zone during T_1 and T_2 stages was probably resulted from the inhibition of the second step of nitrification due to the oxidation of organic matter.

3 Conclusions

In this study, a new airlift intermittent circulation membrane bioreactor was developed to treat synthetic wastewater, nitrogen and phosphorus removal performance and characteristics of the reactor was investigated. The reactor demonstrated satisfied pollutant removal performance, under the influent condition of COD concentration of 642.1 mg/L, TN of 87.4 mg/L and PO_4^{3-}-P of 12.1 mg/L, average removal efficiencies of COD, TN and PO_4^{3-}-P reached 96.4%, 83.2% and 90.5%, respectively, with the hydraulic residence time of 22 hr and operation circle time of 185 min. Track studies indicated that separation of aeration zone and mixing zone along with intermittent circulation of mixed liquid between the two zones provided distinct biological environments specially and temporally, which ensured the occurrence of nitrification, P-taken up, P-release, denitrification and denitrifying phosphorus removal processes.

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References

Ahn J, Daidou T, Tsuneda S, Hirata A, 2002. Characterization of denitrifying phosphate-accumulating organisms cultivated under different electron acceptor conditions using polymerase chain reaction-denaturing gradient gel electrophoresis assay. *Water Research*, 36(2): 403–412.

Ahn K H, Song K G, Cho E, Cho J, Yun H, Lee S et al., 2003. Enhanced biological phosphorus and nitrogen removal using a sequencing anoxic/anaerobic membrane bioreactor (SAM) process. *Desalination*, 157: 345–352.

APHA (American Public Health Association), 1999. Standard Methods for the Examination of Water and Wastewater (20th ed.). Washington DC, USA.

Choi C, Lee J, Lee K, Kim M, 2008. The effects on operation conditions of sludge retention time and carbon/nitrogen ratio in an intermittently aerated membrane bioreactor (IAMBR). *Bioresource Technology*, 99(13): 5397–5401.

Guo H Y, Guo Z, Feng T T, Zhang S T, Zhou J T, 2012. Influence factors and characteristics of nitrogen and phosphorus removal in a sequencing batch moving bed biofilm reactor. *The Official Journal of the Chemical Industry and Engineering Society of China*, 63(1): 251–257.

Hasar H, Kinaci C, Unlu A, Ipek U, 2001. Role of intermittent aeration in domestic wastewater treatment by submerged membrane activated sludge system. *Desalination*, 142: 287–293.

Li Y Z, He Y L, Ohandja D G, Ji J, Li J F, Zhou T, 2008. Simultaneous nitrification-denitrification achieved by an innovative internal-loop airlift MBR: Comparative study. *Bioresource Technology*, 99(13): 5867–5872.

Lv X H, Hu L X, 2004. Nitrogen and phosphorus removal technology of moving bed biofilm reactor (MBBR). *Journal of Chemical Engineer*, 9: 20–22.

Kimura K, Nishisako R, Miyoshi T, Shimada R, Watanabe Y, 2008. Baffled membrane bioreactor (BMBR) for efficient nutrient removal from municipal wastewater. *Water Research*, 42(3): 625–632.

Kuba T, Smolders G, Vanloosdrecht M C M, Heijnen J J, 1993. Biological phosphorus removal from waste-water by anaerobic-anoxic sequencing batch reactor. *Water Science and Technology*, 27(5-6): 241–252.

Patel J, Nakhla G, Margaritis A, 2005. Optimization of biological nutrient removal in a membrane bioreactor system. *Journal of Environmental Engineering*, 131(7): 1021–1029.

Song K G, Cho J, Ahn K H, 2009. Effects of internal recycling time mode and hydraulic retention time on biological nitrogen and phosphorus removal in a sequencing anoxic/anaerobic membrane bioreactor process. *Bioprocess Biosystem Engineering*, 32: 135–142.

Shen Y L, Wang B Z, 1999. New Technologies for Biological Wastewater Treatment, China. Environmental Science Press, Beijing.

Yang S, Yang F L, Fu Z M, Wang T, Lei R B, 2010. Simultaneous nitrogen and phosphorus removal by a novel sequencing batch moving bed membrane bioreactor for wastewater treatment. *Journal of Hazardous Materials*, 175: 551–557.

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