

Biological nutrient removal using an alternating of anoxic and anaerobic membrane bioreactor (AAAM) process

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Abstract

An innovative alternating of anoxic and anaerobic membrane bioreactor (AAAM) process was developed to enhance nitrogen and phosphorus removal simultaneously. The process was composed of a continuous aerated MBR and an alternating anaerobic and anoxic zone containing two separated bioreactors (reactor A and reactor B). By switching on and off the control valves, the mixed liquor in the aerobic zone was recycled to the reactor A or reactor B alternately, thus, the anoxic conditions for denitrification and anaerobic conditions for phosphorus release were implemented in two single tanks alternately. Additionally, by this operation mode, the denitrifying bacteria and phosphate-accumulating organisms (PAOs) could make full use of the organic substrate present in the wastewater alternately. A laboratory-scale experiment treating synthetic wastewater was applied to investigate the performance of the AAAM process. The results showed that COD removal efficiency was high and stable (over 93%) throughout the experiment. Nitrogen and phosphorus removal efficiency also attained high treatment levels (67.4% and 94.1% respectively). Comparing to DO and pH, oxidation–reduction potential (ORP) could provide much better information about the processes going on in reactors A and B during the 120-min cycle. Analysis of membrane fouling showed that small particles seemed to play an important role on the membrane fouling.

Keywords: Nitrogen and phosphorus removal; AAAM process; Phosphate-accumulating organisms (PAOs); Denitrification; Oxidation–reduction potential (ORP); Membrane fouling

1. Introduction

Phosphorus and nitrogen removal from wastewater is now considered as a crucial and

effective procedure to prevent eutrophication problems in receiving waters. Biological nutrient removal (BNR) processes are generally the most efficient and cost-effective solution to achieve this [1].

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There is a growing interest in using membrane bioreactor (MBR) system for wastewater treatment and recent studies on wastewater treatment using MBR have focused on nutrient removal [2,3]. MBR process, in addition to its compactness, offers many advantages over the conventional activated sludge processes, such as the complete solid–liquid separation, high effluent quality, absolute control of solids and hydraulic retention times, preventing failure of biological systems due to biomass loss and/or bulking and maintenance of high mixed liquor suspended solids (MLSS) in the reactor [4].

Biological nitrogen removal has been achieved by two types of MBR systems: the single-reactor-type MBR and the modified Luzack-Ettinger (MLE)-type MBR. The single-reactor-type MBR introduced the alternating aerobic and anoxic conditions to a submerged MBR system by intermittent aeration in the aerobic tank for simultaneous removal of carbon and nitrogen [5]. However, to minimize the membrane fouling, filtration operation in this type of MBR is limited during only the aeration period. To make up for this drawback, the MLE-type MBR (a membrane-coupled anoxic/oxic process) [6], was developed for continuous filtration operation and removal of carbon and nitrogen simultaneously. The MLE-type MBR is composed of a continuous aerated MBR and a separated anoxic tank, in which the mixed liquor was recycled continuously from aerobic zone to anoxic zone. These MBR systems have shown an improvement on nitrogen removal, the removal of phosphorus, though, remains inadequate. To enhance biological phosphorus removal in MBRs, a sequencing anoxic/anaerobic membrane bioreactor (SAM) process was developed [7,8]. In this process, sequencing anaerobic and anoxic conditions were implemented in a single tank by switching on and off the recirculation from the following aerated reactor where also the membranes were installed. By supplying strict anaerobic conditions without an internal recycle, the phosphorus release was induced in a

significant amount, resulting in excellent uptake of phosphorus in the aerobic zone. So the phosphorous removal efficiency was much better. But nitrogen removal efficiency in the SAM process was lower than that of the MLE-type MBR process, so process optimization still has to be done.

On the basis of previous researches, the aim of current work, therefore, was to develop an innovative method to enhance nitrogen and phosphorus removal simultaneously. The process we adopted was composed of a continuous aerated MBR and two separated tanks. The alternating of anoxic and anaerobic membrane bioreactor (AAAM) process could be a more efficient process for biological nitrogen and phosphorus removal by providing favorable environments for different functional microorganisms namely nitrobacteria, denitrifying bacteria and phosphate-accumulating organisms (PAOs) and giving full play of the organic substrate present in the wastewater. Membrane fouling was controlled by intermittent suction and sub-critical flux operation, which were useful methods for fouling alleviation.

2. Materials and methods

2.1. AAAM system description

The experimental set-up was shown in Fig. 1. The laboratory-scale AAAM process was composed of a continuous aerated MBR (reactor C) and an alternating anaerobic and anoxic zone containing two separated bioreactors (reactor A and reactor B).

The working volume of the bioreactors was 24 L (reactor A), 24 L (reactor B) and 48 L (reactor C), respectively. The mixed liquor was recycled from reactor C (aerobic zone) to reactor A or reactor B alternately. A MF membrane module was immersed in the bioreactor C. The characteristics of the membrane module were showed in Table 1. An air diffuser was installed underneath the membrane module to provide air

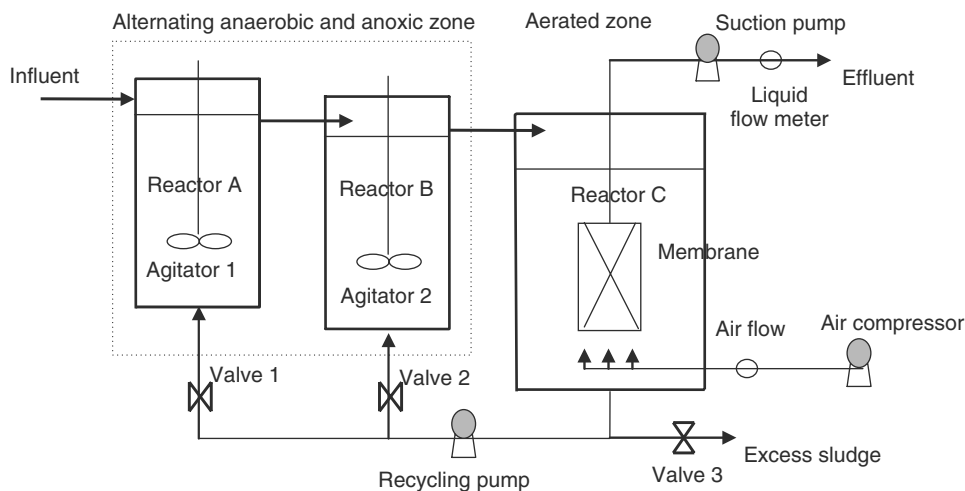


Fig. 1. Schematic diagram of the AAAM system.

at 10 L/min. The bubbling air, therefore, was used to serve for three purposes: providing oxygen for the microorganisms in aerobic tank, mixing the aerobic tank and removing of cake deposition on the membrane surface.

The excess sludge was discharged regularly and the mixed liquor suspended solid concentrations (MLSS) concentrations in the aerobic zone were maintained in a range of 5500–6500 mg/L during the operation. SRT was kept at 20 days, In order to prevent rapid fouling, the membrane flux (10 L/h) was maintained below the critical level. What is more, a peristaltic pump was used in intermittent mode (on/off = 10/2 min) to permeate the effluent through the membrane.

Table 1
Characteristics of the membrane module

Membrane material	PVDF
Membrane module type	Hollow fiber
Effective filtration area (m ²)	1.0
Inside diameter (mm)	0.65
Outside diameter (mm)	1.0
Nominal pore size (μm)	0.22
Producer	MOTIMO, China

2.2. Operation mode

Two agitators were kept working throughout the operation only to maintain the mixed liquor solids in suspension. Operation mode was controlled using timers which were connected to the suction pump, valve 1 and valve 2. The alternating anoxic and anaerobic conditions were produced by the alternation of the recycling mixed liquor directly from the aerobic zone (reactor C) to reactor A or to reactor B. When the mixed liquor was recycled to reactor A, the valve 2 was turn off. The anoxic conditions could be induced in reactor A because of the introduction of dissolved oxygen and nitrate in the recycled liquor. Meanwhile, no recycling would induce strict anaerobic conditions in reactor B, in which the contents were gently agitated so as to maintain the mixed liquor solids in suspension but retain anaerobicity. One hour later, the valve 1 was switched off and the mixed liquor from the aerobic zone was recycled to reactor B. As a result, the anaerobic conditions could be induced in reactor A and the anoxic conditions in reactor B. Both of the anaerobic conditions phosphorus release for and the anoxic conditions for denitrification were

maintained for 1 h periodically with the mixed liquor recycle at the rate of 2.

2.3. Influent quality and sludge seeding

Synthetic wastewater, comprising sodium acetate, NH_4Cl , KH_2PO_4 and mineral solution (Mg, Ca and Fe), was fed to the lab-scale reactor continuously throughout the operation. The initial influent contained 300 mg COD/L, 30 mg NH_4^+ -N/L and 6 mg PO_4^{3-} -P/L. The pH value was about 7.3.

The seeding sludge was supplied from the aeration tank at a municipal wastewater treatment plant in Xuzhou city (China). After seeding, the sludge was cultivated with the synthetic wastewater over 2 months for acclimation of the microorganisms.

2.4. Analytical methods

All the results we presented were obtained from the AAAM system at steady state. The permeate from one entire cycle was collected in a container, and the mixed liquor was used to assay the intraday effluent quality. The samples taken from bioreactors were filtered using 1.2 μm filter

paper. Dissolved oxygen concentration (DO) was measured using the DO meter (WTW Oxi 340, Germany). pH and oxidation–reduction potential (ORP) value using the pH meter (PHSJ-4A, China). Particle size distribution was determined using the laser granulometer (Microsizer, Malvern). Measurement of COD, MLSS, total nitrogen (TN), oxidized nitrogen (NO_3^- -N and NO_2^- -N), ammonium nitrogen (NH_4^+ -N), orthophosphate concentration (ortho-P) and total phosphorus (TP) followed standard methods [9].

3. Results and discussion

3.1. Mechanism

In order to understand the metabolic characteristics of the AAAM process in alternating anaerobic and anoxic zone, an intense sampling campaign over an entire cycle was conducted when the AAAM system was at a global steady state. The experiment was performed when the mixed liquor began to be recycled from reactor C to reactor A. The effluent samples from bioreactor A and bioreactor B were taken every 15 min and assayed for quality. Fig. 2 showed how the nitrate and orthophosphate (ortho-P) concentrations

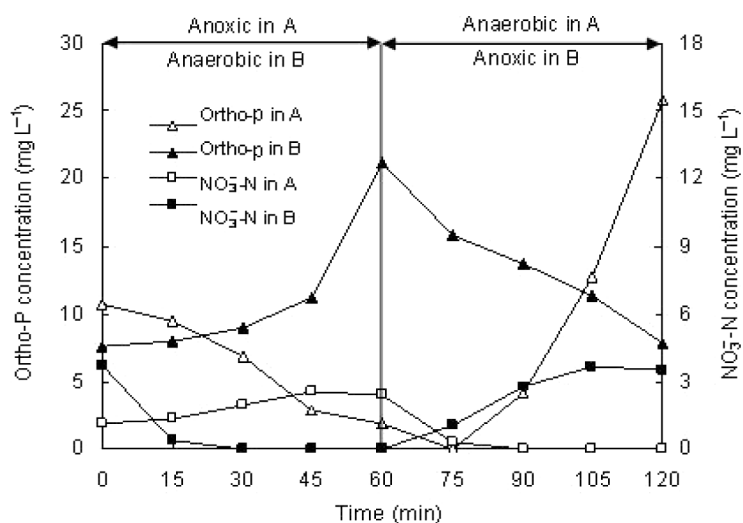


Fig. 2. Profiles of orthophosphate (ortho-P) and nitrate (NO_3^- -N) in a typical cycle of AAAM operation.

changed in the alternating anaerobic and anoxic zone (reactor A and reactor B) during a 120-min cycle.

As shown in Fig. 2, during the former semi-period, the mixed liquor was recycled to reactor A. As a result, the anoxic condition was induced in reactor A because of the introducing of a mass of nitrate and dissolved oxygen from the aerobic zone. In this operational mode, the heterotrophic denitrifying bacteria were given priority of utilizing the organic substances in the influent. The denitrification, therefore, was enhanced which could be confirmed by the low level (<3 mg/L) and slow increase of nitrate concentrations in reactor A although a mass of nitrate was introduced into it. Meanwhile, no recycling induced anaerobic condition in reactor B. Under anaerobic conditions, microbes deplete the carbon source from the wastewater, accumulate storage biopolymers and release soluble ortho-P. Presence of nitrate in anaerobic zone could prohibit the release of polyphosphate because of the carbon source competition between denitrifying bacteria and PAOs. Therefore, in reactor B, the slow increase rate of ortho-P concentrations during the initial 15 min might be due to the presence of nitrogen (3.72 mg/L). After 15 min, the nitrate was almost consumed and the strict anaerobic condition, favorable for PAOs, was induced. Thus the ortho-P began to be released rapidly, achieving 21.18 mg/L at the end of this period.

At 60 min, the mixed liquor was recycled to reactor B (the later semi-period started), which induced the anaerobic condition in reactor A and anoxic condition in reactor B. The presence of a small amount of nitrate at the start of the anaerobic phase may stimulate the growth of denitrifying phosphate-accumulating bacteria (DPB). DPB, a fraction of PAOs, can also take up phosphorus under anoxic conditions using nitrate as the electron acceptor instead of oxygen [10]. Therefore, the decrease of ortho-P concentrations in reactor A during the initial 15 min (from 60 to 75 min) in the later semi-period might attribute to the

simultaneous denitrification and phosphorus uptake by DPB. At 75 min, no enough nitrates were available for DPB and the PAOs could take full advantage of organic substances in the influent. As a result, the ortho-P was released to a large amount, achieving 25.75 mg/L at the end of the anaerobic phase. The more the phosphorus was released in the anaerobic phase, the more it was up-taken in the aerobic phase. Therefore, the phosphorus removal was improved in this period.

As presented in Fig. 2, the ultimate ortho-P concentration in reactor A was 4.57 mg/L higher than that in reactor B under the anaerobic conditions and the increase rate of nitrate concentrations in reactor A was a little lower than that in reactor B under the anoxic conditions. These results indicated that better performance of phosphorus release and denitrification occurred in reactor A. Since the same operation mode was adopted by the reactors A and B, the difference in performances might attribute to the arrangement of the two bioreactors. Functional microorganisms in reactor A were given the priority to utilize the organic substances present in the influent, indicating that reactor A played an important role in nitrogen and phosphorus removal. Nevertheless, reactor B was also important for it provided favorable environments for microorganisms in reactor A as well as achieving nitrification although the nitrogen removal efficiency was a little lower than that in reactor A.

It is reported that nutrient concentrations are in a way related to profiles of pH, DO and ORP. They can be used to identify specific points. In the 120-min cycle, pH, DO and ORP in reactor A and reactor B were measured every 15 min to determine a correlation between P release in the anaerobic phase and P uptake in the anoxic phase and denitrification processes. Although DO increased as the mixed liquor was introduced to reactor A or B, the DO values were still keep at a low level (<0.5 mg/L) throughout the cycle (data not shown). That was probably due to the high MLSS concentration in the recycling mixed liquor

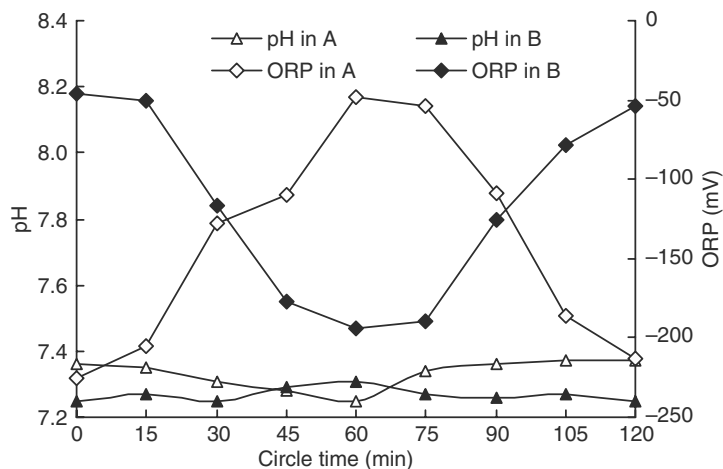


Fig. 3. Profiles of pH and ORP in a typical cycle.

which could consume the DO quickly. When the mixed liquor was halted in a reactor (A or B), DO decreased to zero in a short time. Therefore, DO could not be used as an efficient parameter to identify specific points in the AAAM system.

Unlike DO, pH values decreased in the recycling phase and increased in the non-recycling phase. The change in pH profile within a cycle was given in Fig. 3. Take reactor A for analysis, the decrease in pH, at the beginning of the recycling, was mainly due to the inflow of H^+ in the mixed liquor, which was released during nitrification taking place in the aerobic zone (reactor C). The pH continuously decreased and eventually reached a “valley” coinciding with the end of recycling. On the one hand, the H^+ from mixed liquor could lead to the decrease in pH, on the other hand, denitrification taking place in the reactor could cause pH increase resulting in the increase of alkalinity. Therefore, the pH decrease in this period was not markedly. At 60 min, the mixed liquor began to be recycled to reactor B. No nitrate flowing into reactor A, the pH began to increase corresponding to the simultaneous denitrification and phosphorus uptake by DPB (Fig. 2). After 75 min, pH reached a constant value (about 7.37).

As shown in Fig. 3, although pH decreased when the recycling began and increased when the recycling halted, the variation was not significant. pH values in reactors A and B were kept at a level of about 7.28 throughout the operation. Therefore, pH also couldn't identify specific points effectively in the AAAM system.

Comparing to DO and pH, ORP could provide much better information about the process going on in the 120-min cycle. Under anaerobic and anoxic conditions, ORP were negative values in the two reactors. The ORP level in the cycle was between -225 to -48 mV and -194 to -50 mV in reactor A and reactor B, respectively. Changes of the ORP values in different phases of the cycle were also given in Fig. 3. The denitrification process as well as substrate consumption, therefore, are reduction reactions. Under these conditions, ORP begins to decrease [11]. In this study, however, a significant increase in ORP level was observed in the anoxic phase, which might attribute to the inflow of DO and nitrate contained in the recycling mixed liquor. The ORP eventually reached a maximum value (about -48 mV in reactor A) coinciding with the end point of recycling. After this point, no nitrate and DO were introduced, ORP began to

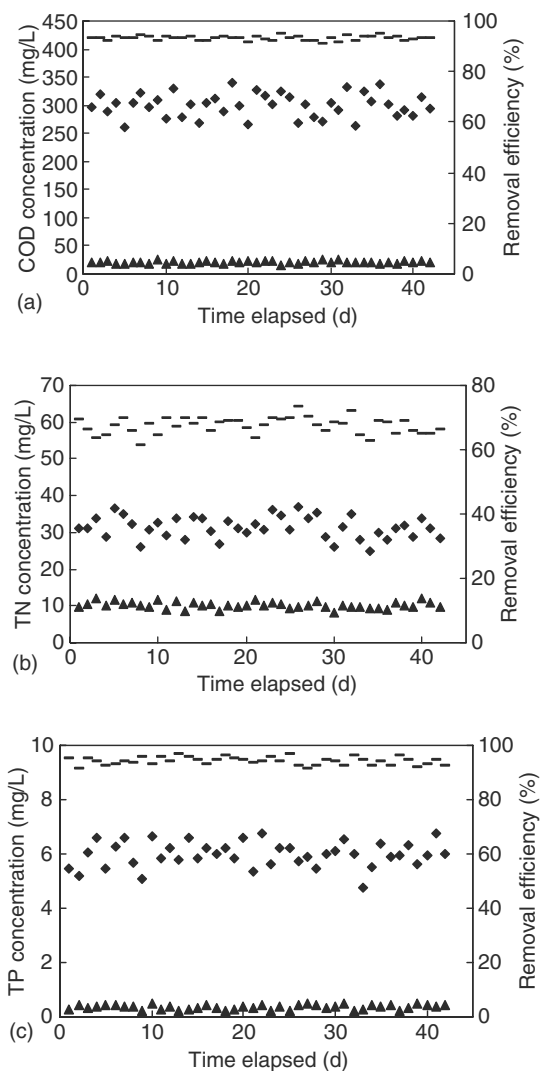


Fig. 4. Influent and effluent qualities and removal efficiency: (a) COD, (b) TN and (c) TP: Symbols: (◆) influent; (▲) effluent; (□) removal efficiency.

decrease as a result of denitrification. At 75 min, nitrate was completely consumed and strict anaerobic conditions prevailed in reactor A. Then phosphate release as well as substrate consumption resulted in a sharp decrease in the ORP level. This indicated the end of the anoxic period (for denitrification) and the start of strict anaerobic period (for phosphorus release). So

ORP level at this point (-54 mV in reactor A and -50 mV in reactor B) could be used to identify the shift from anoxic to anaerobic conditions.

In AAAM system, nitrogen and phosphorus removal could be improved by adjusting the duration of the recycling cycle and the recycling rate according to the influent quality to meet the different requirement of effluent quality. ORP could be used as an effective parameter to determine the optimum operation mode for the best removal efficiency of both nitrogen and phosphorus.

3.2. Nutrient removal

3.2.1. COD removal

Daily variation of the influent and effluent qualities and removal efficiency was studied during the operation. As illustrated in Fig. 4 (a), despite the fluctuations in influent quality, the COD removal efficiency was high and stable, and was more than 93% on average throughout the operation. The average effluent COD concentration was less than 21 mg/L correspondingly.

Since sodium acetate is easily degradable substrate, most of the organic compounds were consumed before entering the aerobic zone. As a result, microorganisms in the aerobic zone attributed less to the COD removal. The high COD removal efficiency in the experiment was due to the efficient utilization of organic compounds in the alternating anaerobic and anoxic zone (reactor A and reactor B) for denitrification and phosphorus release. In addition, the high removal rate could also be attributed to complete particulate retention of suspended COD and BOD_5 , high molecular weight organics, and biomass [12].

3.2.2. Nitrogen removal

As presented in Fig. 4(b), the average TN removal efficiency was accomplished at about 67.4%, corresponding to 10.20 mg N/L (SD = 0.96 mg N/L) in the effluent.

NH_4Cl was used as the sole nitrogen source in the synthetic wastewater, no ammonium nitrogen, however, was detected in the effluent, which implied that complete nitrification was achieved in the AAAM process. The high ability for nitrification could be attributed to the excellent retainability of the membrane to biomass which caused the nitrobacteria flourish in the aerobic zone. Furthermore, most of the biodegradable COD was utilized in the alternating anaerobic and anoxic zone, low COD level in the aerobic zone also ensured the predominance of autotrophic bacteria.

The high nitrogen removal efficiency of the AAAM system was also attributed to the excellent denitrification performance in the alternating anaerobic and anoxic zone. Denitrifying bacteria, the microorganisms capable of reducing nitrogen oxides to molecular nitrogen, are mostly heterotrophic and use organic substrates as electron donors for respiration and growth. When the mixed liquor from aerobic zone was recycled to reactor A, the denitrifying bacteria could make full use of the readily bioassimilable COD in the influent. As a consequence, the denitrification was improved thus the enhanced nitrogen removal was achieved. It should be noted that denitrification in the AAAM system was accomplished not only by normal denitrifying bacteria but also by denitrifying phosphorus removing bacteria, resulting in the increase of denitrification capacity.

In addition, since microorganisms need nitrogen for growth, N was also removed with the excess sludge.

3.2.3. Phosphorus removal

Enhanced biological phosphorus removal (EBPR) was achieved in the AAAM system. As shown in Fig. 4(c). The AAAM system had showed high performance on phosphorus removal. The TP removal efficiency was approximate 94.1% averagely, corresponding to the average 0.35 mg P/L in the effluent.

The high phosphorus removal efficiency indicated that the operational mode of AAAM process induced enhanced phosphorus release in the alternating anaerobic and anoxic zone as well as excess phosphorus uptake in the aerobic zone (as discussed in Section 3.1). In addition, a part of the phosphorus was removed biologically because P is one of the essentials needed for bacterial growth [1.5–2.5% (w/w) based on dry weight].

3.3. Filtration characteristic

3.3.1. Transmembrane pressure (TMP)

In our experiment, membrane flux was kept at an imposed value of $10 \text{ L}/(\text{m}^2 \text{ h})$, which was substantially lower than the measured critical flux. Sustainable operation (up to 42 days) was maintained without any cleaning or additional fouling control measures other than continuous aeration and intermittent filtration. Fig. 5 showed the transmembrane pressure profile during the experiment. For the first 26-day of filtration, TMP values kept at a low level (less than 5.0 kPa) and the rate of TMP rise (dP/dt) was very small ($<0.09 \text{ kPa/d}$). However, TMP began to increase rapidly after 26 days' operation and reached 15.9 kPa at the day of 42. The rate of TMP rise was about 0.94 kPa/d during this period. The

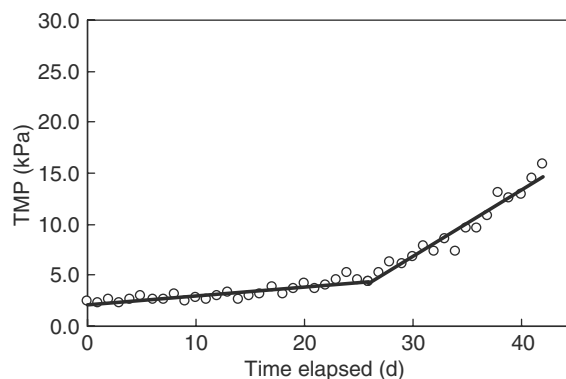


Fig. 5. Variation of TMP values during the operation.

sharp TMP increase was probably due to the development of irreversible membrane fouling.

Many investigations have showed that the accumulations of extracellular polymers (EPS) [13,14] in MBR system contributed a lot to membrane fouling. In our study, the mixed liquor in aerated zone was continuously recycled to the former bioreactors, which theoretically might decrease the accumulation of EPS in membrane zone, and thus alleviated membrane fouling. In this study, all the TMP values were at a relatively low level (less than 16.0 kPa) during the operation, so the membrane still could keep operating for a long time (until TMP reached 35 kPa) without any washing. Further study was needed.

3.3.2. Particle size distribution (PSD)

Foulant sizes may strongly affect fouling mechanisms in membrane filtration systems [15]. To evaluate the causes of membrane fouling in the AAAM process, particle size distributions (PSD), including activated sludge suspension in aerobic zone, exterior cake layer and interior cake layer, were analyzed in the range of 0.1–560 μm at the day of 42. As shown in Fig. 6, the PSDs of the different fractions were quite different. The

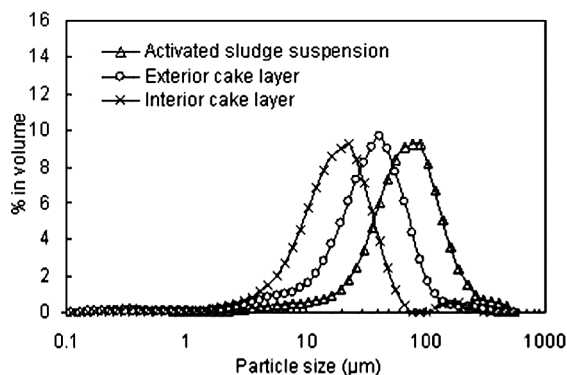


Fig. 6. Particle size distributions (PSD) related to membrane fouling

mean particle size of activated sludge suspension, exterior cake layer and interior cake layer was 75.01, 37.91 and 19.23 μm , respectively. The mean particle size of suspension was larger than that of the particles deposited on the membrane surface, and mean particle size decreased from exterior to interior in cake layer, indicating that small particles were easily to deposit or adsorb on the surface of membrane. From this point of view, it seemed that small particles in suspension liquor had precedence over large ones to adsorb in the pores of membrane or deposit on membrane surface which decreased effective filtration area and caused membrane fouling in AAAM system.

4. Conclusions

An innovative alternating of anoxic and anaerobic membrane bioreactor (AAAM) process was developed to enhance nitrogen and phosphorus removal simultaneously and its performance was investigated on a laboratory-scale system. Major findings from this study are summarized as follows:

- (1) The AAAM process exhibited high performance in removing carbon, nitrogen and phosphorus simultaneously. Under the operational conditions imposed, the removal efficiencies of COD, total nitrogen and total phosphorus were achieved at 93%, 67.4% and 94.1%, respectively.
- (2) The AAAM system could be improved by adjusting the duration of the recycling cycle and the recycling rate according to the influent quality to meet the different requirement of nitrogen or phosphorus removal. ORP could be used as an effective parameter to determine the optimum operation mode for the best removal efficiency of both nitrogen and phosphorus.
- (3) The operation mode of AMMM system might reduce the accumulation of EPS in membrane zone, and thus alleviate membrane fouling.

Therefore, membrane could keep operating for a long time without any washing.

- (4) Analysis of particle size distribution showed that small particles in suspension liquor seemed to be the main fraction responsible for the membrane fouling in AAAM system.
- (5) Further investigations should be focused on the optimization of operation mode according to different influent quality and on the effect of EPS on membrane fouling in the AAAM system.

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