

# Treating both wastewater and excess sludge with an innovative process

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**Abstract:** The innovative process consists of biological unit for wastewater treatment and ozonation unit for excess sludge treatment. An aerobic membrane bioreactor (MBR) was used to remove organics and nitrogen, and an anaerobic reactor was added to the biological unit for the release of phosphorus contained at aerobic sludge to enhance the removal of phosphorus. For the excess sludge produced in the MBR, which was fed to ozone contact column and reacted with ozone, then the ozonated sludge was returned to the MBR for further biological treatment. Experimental results showed that this process could remove organics, nitrogen and phosphorus efficiently, and the removals for COD,  $\text{NH}_3\text{-N}$ , TN and TP were 93.17%, 97.57%, 82.77% and 79.5%, respectively. Batch test indicated that the specific nitrification rate and specific denitrification rate of the MBR were  $1.03 \text{ mg NH}_3\text{-N}/(\text{gMLSS}\cdot\text{h})$  and  $0.56 \text{ mg NOx-N}/(\text{gMLSS}\cdot\text{h})$ , and denitrification seems to be the rate-limiting step. Under the test conditions, the sludge concentration in the MBR was kept at 5000–6000 mg/L, and the wasted sludge was ozonated at an ozone dosage of  $0.10 \text{ kgO}_3/\text{kgSS}$ . During the experimental period of two months, no excess sludge was wasted, and a zero withdrawal of excess sludge was implemented. Through economic analysis, it was found that an additional ozonation operating cost for treatment of both wastewater and excess sludge was only 0.045 RMB Yuan (USD 0.0054)/ $\text{m}^3$  wastewater.

**Keywords:** wastewater treatment; excess sludge; aerobic membrane bioreactor (MBR); anaerobic reactor; ozonation; zero withdrawal of excess sludge

## Introduction

Urban wastewater is usually treated by conventional activated sludge process in China, which involves the biodegradation of organic pollutants by activated sludge in aerated bioreactors, and then the activated sludge could be separated by gravitational settling in clarifiers. The optimal sludge concentration is generally in the range of 2–3 g/L, which imposes large size of bioreactor (Defrance, 1998). In addition, a large amount of excess sludge is produced and the separation, dewatering, treatment and disposal of this excess sludge represent major investment and operating costs. With the rising costs and restrictions on sludge disposal, the minimization of sludge yield has become of increasing importance (Chen, 2000).

Up to now, numerous researches have been conducted to reduce the production of excess sludge in wastewater treatment (Wang, 1991; Yasui, 1996; Lee, 1996; Strand, 1999; Ghyyot, 2000). However, there are some drawbacks in their tests, i. e., minimization of excess sludge often results in sludge bulking, meanwhile, nitrogen and phosphorus can not be removed effectively and effluent often can not meet the requirement of National effluent standard. Therefore, it becomes necessary to modify conventional wastewater treatment process in order to reduce excess sludge production as well as to get high quality of effluent.

The aim of this study was to minimize the production of excess sludge as well as to get an excellent effluent. To achieve both the goals, combined aerobic MBR and anaerobic reactor process was chosen to treat wastewater, and excess sludge produced in MBR was reacted with ozone, then the ozonated sludge was returned to the MBR and mixed with influent for further biological treatment.

## 1 Materials and methods

### 1.1 Experimental equipment

A hollow fiber polyethylene ultrafiltration (UF) membrane module with a total surface area of  $2.0 \text{ m}^2$ , pore size of  $0.05 \mu\text{m}$  and a length of 0.35 m was used as the test membrane. The volumes of the aerobic and anaerobic reactors were 16.3 L and 5 L respectively. The temperature of both the reactors was



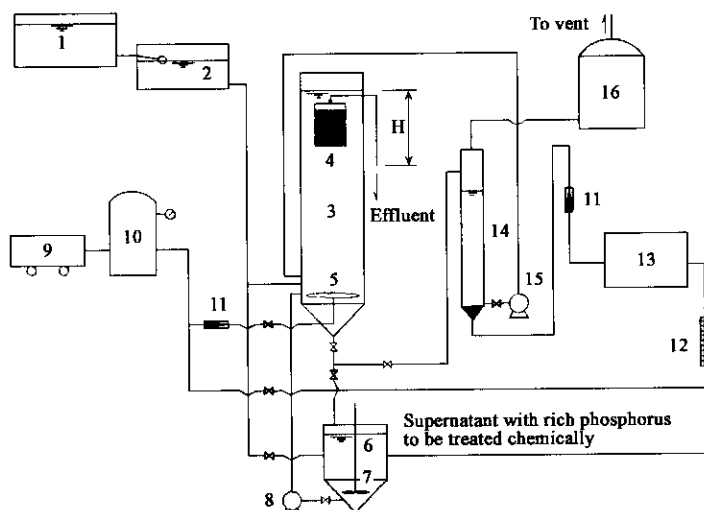


Fig.1 Schematic of the experimental system

1. storage tank; 2. constant high level tank; 3. aerobic reactor; 4. membrane module; 5. air diffuser; 6. anaerobic reactor; 7. stirrer; 8. return pump for sludge released phosphorus; 9. air compressor; 10. pressurized air storage vessel; 11. air flow-meter; 12. air drier; 13. ozone generator; 14. ozone contact reactor for sludge; 15. ozonated sludge return pump; 16. collecting device for ozone off-gas

maintained at 26°C with a thermostat. A water float cock was used to control water level of the aerobic reactor to keep the balance of influent and effluent (membrane permeate). The air was fed into the reactor with a micro-bubble air diffuser, and air flow was adjusted by an air flow-meter. The mixed liquid flowing into the anaerobic reactor was kept in suspension with a stirrer. The hydraulic retention times of wastewater in aerobic and anaerobic reactors were both 6 h. In addition, ozonation of the excess sludge was carried out in a contact column with an inner diameter of 5 cm and a height of 1.2 m. Ozone was introduced into the column through a diffuser located at the bottom of the column, and the ozonated sludge was returned to the aerobic reactor through a pump. A schematic diagram of the process configuration is presented in Fig.1.

## 1.2 Experimental raw water and analytical methods

A synthetic domestic wastewater was used in this test study, which was prepared by mixing tap water with certain quantities of starch, sugar,  $\text{NH}_4\text{Cl}$ ,  $\text{Na}_2\text{HPO}_4$ ,  $\text{NaH}_2\text{PO}_4$  and sodium bicarbonate, and its characteristics are summarized in Table 1.

Table 1 The characteristics of the test wastewater

Parameter	Range	Mean
pH	6.72—8.16	7.44
COD, mg/L	157.98—352.14	232.08
$\text{NH}_3\text{-N}$ , mg/L	28.54—39.85	32.95
TN, mg/L	32.25—42.56	36.89
TP, mg/L	2.5—5.0	3.72

## 1.3 Analytical procedures

COD,  $\text{NH}_3\text{-N}$ , TN, TP, MLSS and MLVSS were determined using the standard method issued by the Environmental Protection Agency(EPA) of China(1989). pH was measured using glass electrodes connected to a PHS-3C pH meter. DO was detected with an YSI(MODEL 50B) dissolved oxygen meter.

The biological treatment unit was run for 4 weeks under controlled conditions to reach steady state. Then the test continued for a total of two months, and the performance of whole treatment system was analyzed.

# 2 Results and discussion

## 2.1 Ozonation of excess sludge

Ozone is a very reactive oxidizing agent. It can react with sludge compounds in two different ways, i. e., the direct and the indirect reaction(Stahelin, 1985). Both reactions occur simultaneously. While the indirect reaction is based on the short-living hydroxyl radicals which do not react specifically, the direct reaction rate is lower and depends on the structure of the reactants. In this test, ozone was used as a cell lysis agent to disintegrate the sludge cells and improve their biodegradability, and then the ozonated sludge was returned to the aeration tank for following biological treatment.



2.1.1 Characteristics of excess sludge ozonation

In this process configuration, both sludge disintegration and wastewater treatment were done simultaneously in the same treatment system. The schematic diagram is shown in Fig.2.

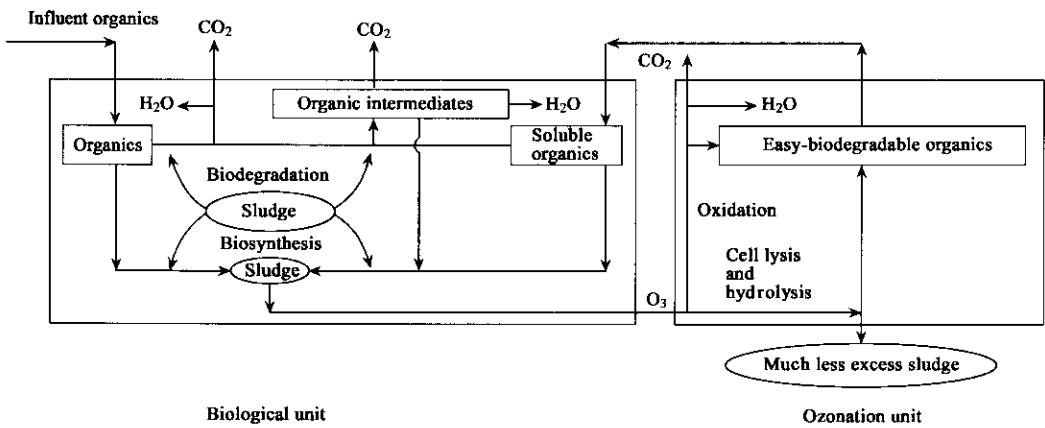


Fig.2 Schematic diagram of the treatment process

The process consists of ozonation and biodegradation stages. The ozonation enhances the biological degradability of the ozonated sludge, which is then decomposed into three parts: one is directly oxidized into CO<sub>2</sub> and H<sub>2</sub>O, and another part is converted into soluble intermediate organic products through cell lysis and hydrolysis, and the third part is the remained sludge, which accounts for only a small portion of the original excess sludge. With an assumption that the biomass mineralization at ozonation stage is negligible, then mass balances at biological treatment stage are formulated as follows(Yasui, 1994):

$$V \cdot \frac{dX_v}{dt} = YQ(L_i - L_o) - bVX_v - Q_wX_v - Q_RX_v + \gamma Q_R(X_v + X_i), \tag{1}$$

$$V \cdot \frac{dX_i}{dt} = - Q_wX_i - Q_RX_i + b'Q_R(X_v + X_i). \tag{2}$$

Under steady conditions, the left-hand sides of Equations (1) and (2) are equal to zero, and both the equations can be combined into the following form:

$$Q_w/V = YN_s - bX_v/X - [1 - (\gamma + b')]Q_R/V. \tag{3}$$

Where,  $V$  is the effective volume of biological reactor(L);  $X_v$  is the active biomass concentration in biological reactor(mg SS/L);  $X_i$  is the inactive biomass concentration in biological reactor(mg SS/L);  $Y$  is the sludge yield(g SS/g COD);  $\gamma$  is the yield of ozonated sludge(g SS/g SS);  $b$  is the specific decay rate ( $d^{-1}$ );  $b'$  is the residual ratio of ozonated sludge to original excess sludge(g SS/g SS);  $Q$  is the influent flow rate(L/d);  $Q_w$  is the flow rate of the wasted excess sludge(L/d);  $Q_R$  is the flow rate of ozonated sludge(L/d);  $N_s$  is the organic loading rate(kg COD/(kg MLVSS·d)).

It can be seen from the Equation (3) that the sludge wasting rate is related to the parameters of  $Y$ ,  $N_s$ ,  $b$ ,  $X_v/X$ ,  $Q_R$  and  $[1 - (\gamma + b')]$ . Once the biological treatment process and the quality of wastewater are determined,  $Y$ ,  $N_s$  and  $b$  become constant, and the sludge wasting rate is dependent on  $X_v/X$ ,  $Q_R$  and  $[1 - (\gamma + b')]$ .  $X_v/X$  represents the active component in sludge and  $[1 - (\gamma + b')]$  denotes the biological mineralization of the ozonated sludge. It can be concluded that these two items are both influenced by the conditions of sludge ozonation. The higher the degree of sludge ozonation, the more the biological mineralization of ozonated sludge takes place, and  $X_v/X$  also can be maintained at a high level. Therefore, it is thought that the reduction of ozonated sludge is only influenced by the conditions of



ozonation, i.e., ozone dosage and flow rate of ozonated sludge ( $Q_R$ ).

### 2.1.2 Determination of ozone dosage

Batch studies were conducted to evaluate the effect of ozone dosage on the degree of cell lysis and hydrolysis. The following procedures were carried out in order to determine the ozone dosage for treating excess sludge. Ozone gas was supplied at the bottom of the contact reactor, SCOD of the activated sludge were obtained at different reaction time. Then the plots of SCOD versus ozonation time were drawn. According to the ozonation, a suitable ozone dosage could reached the point at which cell could be liquefied and mineralized to a certain degree while the treatment cost for excess sludge should not be too expensive.

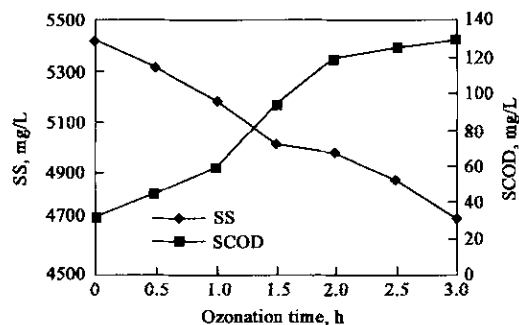


Fig.3 Relationship between ozonation time and SS, SCOD

It was found from Fig.3 that SCOD increased as ozonation dosage increased, after 2 h of ozonation, the increasing rate of SCOD became slower and stayed nearly constant thereafter, whereas SS kept decreasing during 3 h of ozonation time. Therefore, it was concluded that ozonation resulted in cell lysis and hydrolysis, and the increase in SCOD was mainly due to cell lysis, hydrolysis and solubilization. After 2 h of ozonation, the amount of COD solubilized by ozone is proportional to the amount of dissolved organics oxidized by ozone, thereby resulting in no net change in the SCOD concentration. The data were obtained with an

ozone gas concentration of 1.2 mg/L. The influence of higher ozone concentrations on the mineralization and solubilization of sludge should be further investigated.

Based on the above experimental findings, 2 h was considered to be an appropriate ozonation time. Ozone concentration multiplies by ozonation time and air flow, and divides by MLSS of the sludge, then the ozone dosage of 0.10 kg  $O_3$ /kg SS was obtained.

### 2.1.3 Determination of flow rate of sludge to be ozonated

To obtain an ideal performance of simultaneous nitrification and denitrification in MBR, a highly concentrated MLVSS and a long SRT are required. Because of the addition of an anaerobic tank, the removal of phosphorus cannot be affected by long SRT. On this base, MLSS of the reactor was kept at 5000—6000 mg/L, and the excess sludge was discharged from the MBR, and then was treated with ozone. In this test, the amount of ozonated excess sludge was about 0.5% of flow rate of the wastewater.

## 2.2 COD removal

It was found from Fig.4 that the MBR removed COD effectively, an average COD removal of 93.17% was achieved during the experiment. Sometimes, the filtrate COD of the reactor (the mixed liquid samples collected were filtered by qualitative filter papers) had a big fluctuation, whereas COD of the membrane permeate was often kept less than 20 mg/L. With the extension of sludge ozonation, it was noticed that the inorganics had a certain accumulation in aeration tank, VSS/SS varied from 0.92 to 0.74 in two months. Due to recirculation of ozonated sludge, some inert matters or refractory organics was returned to biological unit. Large molecular organics was retained in the aerobic reactor by ultra-membrane filtration and kept COD of permeate at a low level.

### 2.3 Nitrogen removal

During the test period, DO in MBR was kept at a low level of about 0.8 mg/L and pH of mixed liquid was varied between 7.2—7.8. It was found that the system had a mean removal of 97.57% for  $NH_3$ -N, which indicated that a DO concentration around 0.8 mg/L was suitable to achieve a complete nitrification.



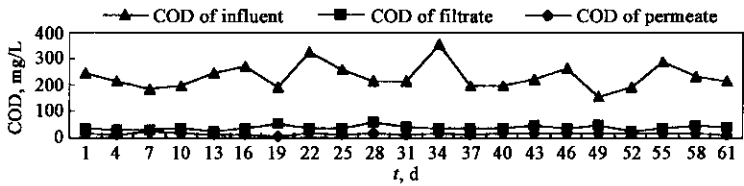


Fig.4 COD removal in MBR

It was observed that the  $\text{NH}_3\text{-N}$  of filtrate was varied between 0.62 mg/L and 2.21 mg/L while the  $\text{NH}_3\text{-N}$  of UF permeate was in the range of 0.38—1.57 mg/L, which means UF membrane has an average removal of 45.8% for  $\text{NH}_3\text{-N}$  of filtrate. Considering small molecular weight of  $\text{NH}_3\text{-N}$ , UF membrane could not retain it through size-exclusion, and the  $\text{NH}_3\text{-N}$  removal by UF membrane may be ascribed to the nitrifiers attached to the outer wall of the UF membrane. At the end of the test period, the UF membrane module was taken out from the reactor, and the biofilm on its surface was washed out with certain quantity of tap water, through the measurement and calculation, it was found that the amount of this biofilm accounted for about 5% of the total suspended activated sludge in MBR. Due to the coexistence of heterotrophs and autotrophs on the out wall, UF membrane showed a further biological removal for organics and nitrogen when the mixed liquid penetrated through the membrane. In aerobic MBR, due to highly concentrated MLVSS, there coexisted aerobic zone and anoxic zone, and simultaneous nitrification and denitrification could take place in the presence of carbon source and low DO, and a total 82.77% of TN removal was achieved in the system(Figs. 5, 6). As to the phenomenon of simultaneous nitrification and denitrification in MBR, which has been explained in detail in previous work(Zou, 2001; He, 2002).

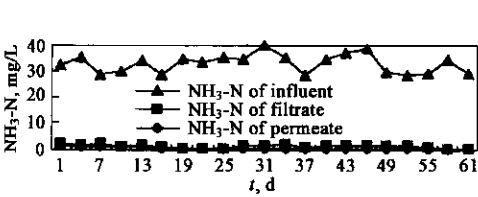


Fig.5  $\text{NH}_3\text{-N}$  removal in MBR

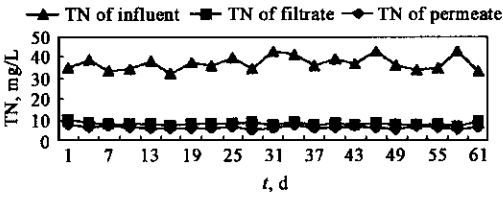


Fig.6 TN removal in MBR

2.4 Phosphorus removal

In general, the removal of phosphorus was realized by the withdrawal of excess sludge containing rich phosphorus. Therefore, to achieve a higher removal for phosphorus, a short SRT is required, whereas the short SRT is disadvantageous to the existence of nitrifiers. In this process configuration, an anaerobic tank was added for the release of phosphorus contained at the aerobic sludge, and then the sludge was returned to aerobic reactor for further uptake of phosphorus. Through batch tests, a sludge return ratio of 0.4 and a HRT of 6 h were chosen as operating parameters, and the removal of phosphorus is illustrated in Fig.7.

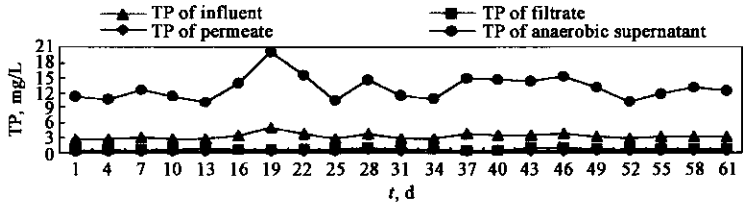


Fig.7 TP removal in MBR



According to Fig. 7, the TP of influent was in the range of 2.5—5.0 mg/L, and the effluent (UF permeate) always had a TP below 0.5 mg/L, and the average removal of TP was 79.5%. It was found that the phosphorus concentrations in anaerobic supernatant was very higher than those in influent, which means that the polyphosphate accumulating bacteria can take up a large amount of phosphorus in aerobic reactor, then release it into mixed liquid under the anaerobic condition, and this aerobic-anaerobic conditions is suitable for the growth of polyphosphate accumulating bacteria. The mass balance of phosphorus is formulated as follows:

$$Q_i \cdot P_i + Q_r \cdot P_r + Q_R \cdot P_R = Q_e \cdot P_e + Q_a \cdot P_a + Q_R \cdot P_{RO} \quad (4)$$

Where:  $Q_i$ ,  $Q_r$ ,  $Q_e$ ,  $Q_a$ ,  $Q_R$  are flow rate of influent, settled anaerobic sludge, effluent, anaerobic supernatant, and ozonated sludge respectively ( $\text{m}^3/\text{d}$ );  $P_i$ ,  $P_r$ ,  $P_e$ ,  $P_a$ ,  $P_R$ ,  $P_{RO}$  are phosphorus concentration of influent, settled anaerobic sludge, effluent, anaerobic supernatant, mixed liquid in MBR and ozonated sludge respectively ( $\text{mg/L}$ ); through calculation, it was found that the amount of phosphorus flowed out of the system accounted for 87.6% of that flowed into the treatment system. This small amount of phosphorus loss may be attributed to the experimental deviation.

## 2.5 Sludge yield in the process configuration

The ozonation was introduced into the biological treatment process to reduce the excess sludge production. For there was no sludge withdrawal from the system, it was assumed that sludge yield was the ratio of total generated sludge production to the organics removed.

$$Y = V(SS_i - SS_t) / \{QT(COD_{inf} - COD_{eff})\} \quad (5)$$

Where:  $Y$  is the sludge yield ( $\text{g SS/g COD}$ );  $V$  is the effective volume of aerobic reactor ( $\text{m}^3$ );  $SS_i$  is the sludge concentration in aerobic reactor at time  $t$  ( $\text{g/m}^3$ );  $SS_t$  is the sludge concentration in aerobic reactor at time  $i$  ( $\text{g/m}^3$ );  $Q$  is the influent flow rate ( $\text{m}^3/\text{d}$ );  $COD_{inf}$  is the COD of influent ( $\text{g/m}^3$ );  $COD_{eff}$  is the COD of effluent ( $\text{g/m}^3$ );  $T$  is the interval of sludge concentration measurement ( $\text{d}$ ).

The ozonation of the system continued for a total period of 61 d, and sludge concentration of 5500 mg/L was assumed to be the base value. During the experimental period, an average sludge yield of about zero was obtained, and the sludge yield of the system is plotted in Fig. 8.

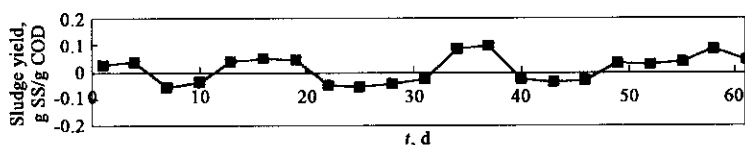


Fig. 8 Sludge yield during experimental period

## 3 Batch study results

Some batch studies for simultaneous nitrification and denitrification were conducted to investigate any limitation in nitrification or denitrification in MBR.

### 3.1 Nitrification

The mixed liquid with MLSS concentration of 5800 mg/L was prepared in a 2 L flask as a batch reactor, and the flask was spiked with a certain amount of ammonium chloride. DO was kept at 0.8 mg/L and pH was controlled around 7.5. Then the concentration of ammonium was monitored with time. The change of ammonium concentration is shown in Fig. 9.

From Fig. 9, it can be seen that ammonium decreased from 24.98 mg/L to 1.06 mg/L in 4 h. A linear reduction of ammonium was observed during this period. According to the test data, a specific nitrification rate of 1.03 mg  $\text{NH}_3\text{-N}/(\text{g MLSS} \cdot \text{h})$  was achieved.



3.2 Denitrification

To simulate the specific denitrification rate of MBR under the conditions of low organics concentration and low DO, two mixed liquids with MLSS 5800 mg/L, COD 60 mg/L and NO<sub>3</sub>-N 27.32 mg/L were utilized as test solutions, and DO was controlled at 0 mg/L and 0.8 mg/L respectively, then the variations of the concentrations of nitrate and nitrite with time are plotted in Figs. 10 and 11, respectively.

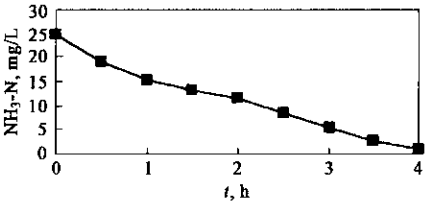


Fig. 9 Variation of NH<sub>3</sub>-N during batch nitrification experiment

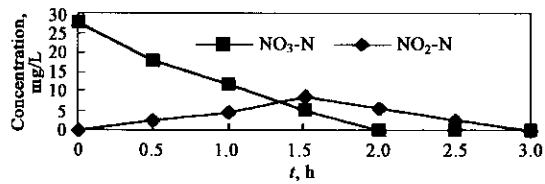


Fig. 10 Variations of nitrate and nitrite during batch denitrification experiment at DO = 0

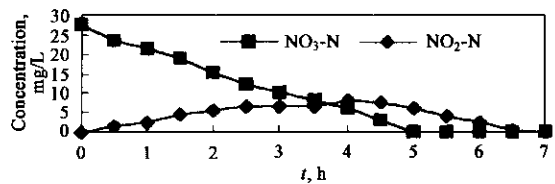


Fig.11 Variations of nitrate and nitrite during batch denitrification experiment at DO = 0.8 mg/L

At DO = 0, it took 3 h to consume all nitrate, while DO was at 0.8 mg/L, a complete denitrification lasted 7 h, which meant that under the conditions of highly concentrated MLSS and 0.8 mg/L of DO, denitrification can proceed, but with a lower rate compared to that at DO = 0. At DO 0 mg/L and DO 0.8 mg/L, the two specific denitrification rates were 1.30 mg NO<sub>x</sub>-N/(g MLSS·h) and 0.56 mg NO<sub>x</sub>-N/(g MLSS·h) respectively. In view of carbon source, the amount of SCOD in the solution is insufficient for denitrification, maybe endogenous denitrification played an important role in MBR.

Through batch test, it was found that the specific nitrification rate is slightly faster than the specific denitrification rate under the operating conditions of the MBR. For the HRT of 6 h is long enough for nitrification, but is slightly shorter for a complete denitrification, there was some nitrite and nitrate left in mixed liquor. If the concentration of ammonium in influent has a rise, to obtain a complete nitrification and denitrification at such a HRT, a higher concentration of MLSS is required.

4 Economic analyses

An energy consumption cost of about 15 RMB Yuan, equivalent to USD 1.8, is needed to produce 1 kg of ozone gas. Based on the experimental data, the operating cost of sludge ozonation could be calculated. A quantity of about 0.005 Q sludge with a 6000 mg MLSS/L of concentration was ozonated, and ozone dosage was 0.10 kg O<sub>3</sub>/kg SS, thus the sludge ozonation operating cost is equal to (15 RMB Yuan/kg O<sub>3</sub> × 6.0 kg SS/m<sup>3</sup> × 0.10 kg O<sub>3</sub>/kg SS) = 9.0 RMB Yuan(USD 1.08) /m<sup>3</sup> sludge. For the flow rate of ozonated sludge was about 0.005 Q, thus the additional ozonation cost for wastewater treatment is 9.0 RMB Yuan/m<sup>3</sup> × (0.005 Q/Q) = 0.045 RMB Yuan(USD 0.0054)/m<sup>3</sup> wastewater. Whereas, in a conventional wastewater treatment plant, the handling, treatment, and ultimate disposal of wasted biosolids accounts for from 50% to 60% of the operating costs of the plant(Brennan, 1999). Which means that the sludge ozonation process could reduce the operating cost of combined wastewater and excess sludge treatment.

5 Conclusions

The innovative process for both biological wastewater treatment and sludge ozonation treatment has been developed in the study. During the experimental period of two months, average removals of 93.17% , 97.57% , 82.77% and 79.5% was achieved for COD, NH<sub>3</sub>-N, TN and TP respectively, and a zero



sludge yield was obtained.

Through batch tests, nitrification rate and denitrification rate of the MBR were found to be  $1.03 \text{ mg NH}_3\text{-N}/(\text{g MLSS} \cdot \text{h})$  and  $0.56 \text{ mg NO}_x\text{-N}/(\text{g MLSS} \cdot \text{h})$  respectively. Under the test conditions, denitrification seemed to be the rate-limiting step.

Economic analysis indicated that this innovative process configuration could treat both the wastewater and excess sludge effectively and economically. On the base of obtaining high quality of effluent, an additional ozonation operating cost for treatment of both the wastewater and excess sludge is only 0.045 RMB Yuan(USD 0.0054)/ $\text{m}^3$  wastewater.

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