



Effects of COD/N ratio and DO concentration on simultaneous nitrification and denitrification in an airlift internal circulation membrane bioreactor

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Abstract

The effects of chemical oxygen demand and nitrogen (COD/N) ratio and dissolved oxygen concentration (DO) on simultaneous nitrification and denitrification (SND) were investigated using an airlift internal circulation membrane bioreactor (AIC-MBR) with synthetic wastewater. The results showed that the COD efficiencies were consistently greater than 90% regardless of changes in the COD/N ratio. At the COD/N ratio of 4.77 and 10.04, the system nitrogen removal efficiency became higher than 70%. However, the nitrogen removal efficiency decreased to less than 50%, as the COD/N ratio shifted to 15.11. When the operating DO concentration was maintained at 1.0 mg/L in AIC-MBR, a satisfying SND was achieved. Either low or high DO concentration could restrain SND.

Key words: simultaneous nitrification and denitrification (SND); chemical oxygen demand and nitrogen (COD/N) ratio; dissolved oxygen (DO) concentration

Introduction

In recent years, pollution and eutrophication of aquatic environment has become worse. As the discharge standard of nitrogen and phosphorus in the effluent has become stricter, a membrane bioreactor (MBR) has been proposed as an alternative for the conventional activated sludge process (Brindle and Stephendon, 1996). The MBRs have higher biomass concentrations and better retain slow growing microorganisms (such as nitrifiers), which enhance biological nutrient removal (de Silva *et al.*, 1998).

Wang *et al.* (2005) reported that a laboratory scale anoxic/aerobic MBR was capable of achieving 94% chemical oxygen demand (COD), 91% ammonia nitrogen ($\text{NH}_4^+\text{-N}$), and 74% total nitrogen (TN) removal, by continuous runs under appropriate operational conditions. Yeom *et al.* (1999), applied an intermittently aerated membrane bioreactor with a submerged fiber hollow membrane to treat household wastewater. With 8–15 h hydraulic retention time (HRT) and long solid retention time (SRT), COD and TN removal efficiency was 96% and 83%, respectively. Rosenberger *et al.* (2002) evaluated the performance of a bioreactor with submerged membranes for aerobic treatment of municipal wastewater. Although many MBR systems could efficiently eliminate nitrogen, the elimination of nitrogen was always time-consuming, requiring

complex configuration, higher energy, and operation cost.

On the basis of these considerations, simultaneous nitrification and denitrification (SND) has gained significant attention, because of its potential to eliminate the need for separate tanks required in conventional treatment plants, which consequently simplifies the plant design, saving space and time. Pochana and Keller (1999) have investigated the efficiency of nitrogen removal from wastewater by an SND-based sequencing batch reactor (SBR). They have observed that higher DO concentrations enhance the nitrification rates. Simultaneously, high DO concentrations inhibit the denitrification process, causing an accumulation of nitrite and nitrate in the reactor. On the other hand, at lower DO concentrations the nitrification process is inhibited and the denitrification process is enhanced. Therefore, the DO level is a factor critical to the SND process. It must be maintained at an appropriate level in the SND reactor. Zhao *et al.* (1999) has pointed out that DO must be available for the nitrifiers, but must not exceed a certain level at the same time for the denitrifiers and SND. Chiu *et al.* (2007) has studied how the C/N ratio controls the simultaneous nitrification and denitrification in a sequencing batch reactor. They have proposed that the SBR system can achieve nearly complete removal of both organic matter and $\text{NH}_4^+\text{-N}$, with no accumulation of intermediate $\text{NO}_2^-\text{-N}$ when the initial C/N ratio is controlled at 11.1.

Most of the SND studies have been conducted in

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sequential batch reactors (Pochana *et al.*, 1999), continuous flow extended aeration plants (Collivignarelli and Bertanza, 1999), and oxidation ditches (Rittmann and Langeland, 1985). There are limited studies on the achievability of SND in continuously aerated MBRs. A novel MBR, airlift internal circulation membrane bioreactor (AIC-MBR) has been established and applied for nitrogen removal through the SND process. Unlike the reactors mentioned earlier, this reactor has a simple configuration and is convenient to operate. On one hand, the configuration of internal circulation can use the energy that the aeration produces, for a nitrified liquid cycle. The energy consumption and operational cost will be reduced significantly. On the other hand, the special configuration generates the aeration and anoxic zone in the AIC-MBR. It overcomes the difficulty in DO control in conventional MBRs and provides an advantageous environment for the SND process.

The available literature shows that under suitable conditions, the SND process will occur in the wastewater treatment system. However, the conditions that lead to efficient SND in the AIC-MBR process are not yet well established. In this investigation, an SND in the AIC-MBR was studied, to resolve some of the conditions under which significant SND activities can occur.

1 Material and methods

1.1 Reactor setup and operation

Figure 1 shows the schematic diagram of the experimental setup, consisting of a Plexiglas reactor in which a membrane module is submerged in the upper part. The AIC-MBR, with a working volume of 18 L, is seeded with sludge from a local municipal wastewater treatment plant (Dalian, China). The membrane module used in this study is a bundle of hollow-fiber membranes of polyethylene (Daiki, Japan) with a pore size of 0.1 μm and a filtration area of 0.01 m^2 . An air diffuser, an 8-mm inner diameter tube with many 1 mm openings, is fitted under the membrane, for aeration. Two baffle plates are fixed on both sides of the membrane to form an upflow aerobic zone between the membrane and plates, and downflow

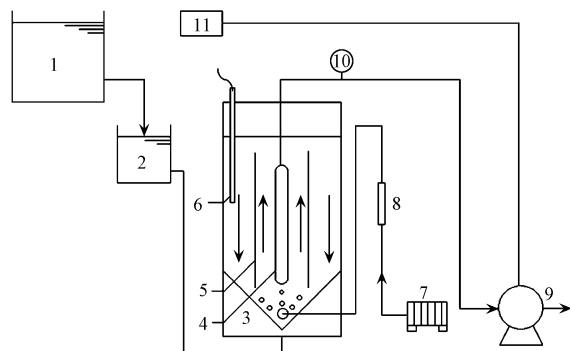


Fig. 1 Schematic diagram of the experimental setup. (1) feed tank; (2) balance tank; (3) airlift internal circulation membrane bioreactor (AIC-MBR); (4) membrane module; (5) baffle plates; (6) electric heater; (7) air compressor; (8) rotameter; (9) peristaltic pump; (10) effluent vacuum gauger; (11) time relay.

anoxic zone between the plates and reactor walls. The reactor temperature is maintained at about 25°C with an electric heater. To control membrane fouling, the suction mode is 6 min on, 2 min off by time relay. When the operating pressure increases to over 0.02 MPa, hydraulic cleaning is adopted. If hydraulic cleaning cannot improve the situation of membrane fouling, chemical cleaning is used with NaClO solution to recover the membrane flux. The operating parameters are summarized in Table 1.

Table 1 Operating parameters in AIC-MBR system

Parameter	Value
Mixed liquor suspended solids (MLSS) (mg/L)	5,000±300*
Mixed liquor volatile suspended solids (MLVSS) (mg/L)	4,000±300*
Hydraulic retention time (HRT) (h)	12
Solid retention time (SRT) (d)	40

* Mean ± SE.

1.2 Feed medium

Synthetic wastewater consisting of sugar, NH_4Cl , KH_2PO_4 , a mineral solution containing CaCl_2 , $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, and EDTA, and a trace element solution containing $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, H_3BO_3 , $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$, KI , $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$, ZnCl_2 , $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$, and $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, was used to simulate the composition of real domestic wastewater. The pH of the synthetic wastewater was controlled at 7.8 ± 0.2 by the addition of NaHCO_3 .

1.3 Analysis

The COD, $\text{NH}_4^+\text{-N}$, nitrate ($\text{NO}_3^-\text{-N}$), nitrite ($\text{NO}_2^-\text{-N}$), total suspended solids (TSS), and volatile suspended solid (VSS) contents were determined according to the standard methods (APHA/AWWA/WEF, 1995). TN concentration was evaluated by a TOC analyzer equipped with a total nitrogen-measuring unit (TOC-VCPH, Shimadzu, Japan). The DO concentration was measured using a DO meter (Model 55, YSI, USA).

1.4 Experimental procedure

The operating conditions of the AIC-MBR are shown in Table 2. Holding the influent COD concentration, the influent TN concentration was increased gradually, to change the COD/N ratio during run I. Then the influent COD/N ratio was kept constant, and a gradual increase of DO concentration was adopted in run II. DO concentration monitored during the whole experiment mainly aimed at the aerobic zone. Generally, the DO concentration in the anoxic zone was 40%–60% of the aerobic zone. The specific values of COD and nitrogen compounds for the influent, membrane effluent, and supernatant were monitored daily. The sludge concentration in the reactor was measured periodically.

1.5 Batch tests

The batch tests were conducted in a 400-ml beaker, which was placed in a water bath at 25°C. The batch reactor was equipped with compressed air installation.

Table 2 Operating conditions of the AIC-MBR

Run	Operational time (d)	Influent COD (mg/L)	Influent TN (mg/L)	COD/N ratio	DO (mg/L)
I	1–48	406.21 (10.86)	28.30 (1.83)	14.44 (1.06)	0.8 (0.1)
	49–96	406.21 (10.86)	42.47 (2.04)	9.59 (0.54)	0.8 (0.1)
	97–144	406.21 (10.86)	82.75 (2.77)	4.90 (0.12)	0.8 (0.1)
II	145–160	302.45 (7.37)	30.55 (1.67)	9.92 (0.42)	0.5 (0.1)
	161–175	302.45 (7.37)	30.55 (1.67)	9.92 (0.42)	1.0 (0.1)
	176–190	302.45 (7.37)	30.55 (1.67)	9.92 (0.42)	1.5 (0.2)
	191–205	302.45 (7.37)	30.55 (1.67)	9.92 (0.42)	3.0 (0.4)

The numbers in parentheses are standard deviation, $n = 150$.

Sludge samples of 200 ml were added to the beaker, which was fed with synthetic domestic wastewater.

Two experiment series were conducted. Series 1 evaluated the effect of COD/N ratio on organics and nitrogen removal, and series 2 was concerned with the effect of DO concentration on nitrogen removal. In series 1 the influent TN concentration fixed at about 40 mg/L, whereas, the influent COD was increased stepwise to obtain three different COD/N ratios (4.77, 10.04, and 15.11). The DO concentration of batch reactors was maintained at about 1.0 mg/L throughout the tests. In series 2 the same quality of synthetic domestic wastewater was added to the beaker to achieve initial COD and TN concentrations of 350 and 35 mg/L, respectively. Oxygen was supplied and the concentration was controlled at 0.5, 1.0, 1.5, and 3.0 mg/L, respectively. All the batch tests lasted for 6 h. During the period, samples were collected regularly and immediately analyzed for COD, TN, NH_4^+ -N, NO_3^- -N, and NO_2^- -N.

2 Results and discussion

2.1 Influence of nitrogen loading rate

Figure 2 presents the COD concentration in the effluent and COD removal efficiency during the whole operation period. The system showed excellent performance in organic carbon removal. An average organic removal of over 90% was achieved throughout the experiment. Although the supernatant COD fluctuated greatly from 30 to 120 mg/L, the membrane effluent COD stabilized at low levels. This was attributed to the complete retention of all particulate COD and macromolecular COD components

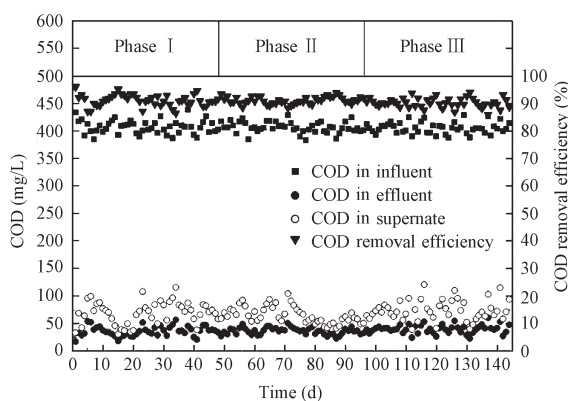


Fig. 2 Profiles of COD concentrations in the system when the nitrogen loading rate was adjusted at 0.013 (phase I), 0.020 (phase II), or 0.040 gN/(g VSS·d) (phase III).

by the membrane.

Figure 3 shows the removal performance of nitrogen in the three phases. During the whole operation period, with the given influent COD of 400 mg/L, the influent COD/N ratio was shifted, by varying the nitrogen loading rate supplied to the system.

During phase I, from day 1 to day 48, the nitrogen loading rate was maintained at 0.013 gN/(g VSS·d). NH_4^+ -N removal efficiency was about 69% and the effluent NH_4^+ -N was about 8 mg/L. NO_3^- -N and NO_2^- -N in the effluent were detected at low concentration level (0.59 and 0.08 mg/L, respectively). Good nitrification was not achieved because the heterotrophic microorganism had dominated the growth competition between autotrophic and heterotrophic microorganism populations and utilized the available DO. TN removal efficiency was only 68% as a result of the inhibition of nitrification.

In phase II, although the nitrogen loading rate was increased to 0.020 gN/(g VSS·d), NH_4^+ -N removal efficiency gradually increased to 73%. This was mainly the result of the growth and enrichment of the nitrifiers. The products of nitrification were reduced, and the effluent NO_3^- -N and NO_2^- -N was maintained at a low residual level. TN removal efficiency was increased to 73% because of the promotion of nitrification. Therefore, under the condition of phases I and II, nitrification was the limiting process and the efficiency of TN removal mainly depended on the degree of the nitrification in the system.

During phase III, from day 97 to day 144, the nitrogen loading rate increased to 0.040 gN/(g VSS·d). NH_4^+ -N removal efficiency increased rapidly to over 89% and the effluent NH_4^+ -N concentration remained at a low level. However, this condition was disadvantageous for aerobic denitrification. The effluent NO_3^- -N residual concentration was high, up to 37 mg/L. As the nitrogen loading rate increasing, the amount of COD could not supply enough carbon sources for denitrification. This situation resulted in the accumulation of nitrates and limited the SND process. The TN removal efficiency decreased to 45% because of the deterioration of denitrification. Zhang *et al.* (2007) pointed out that the key to achieving high total nitrogen removal lay in controlling the balance of nitrification and denitrification, when nitrification and denitrification occurred simultaneously in one reactor.

2.2 Influence of DO concentration

The TN removal was greatly influenced by the operating DO concentration. In phase I, when the DO

was maintained at 0.5 mg/L, nitrification was hampered because of oxygen limitation, with an average effluent $\text{NH}_4^+\text{-N}$ of 14 mg/L. TN removal efficiency was 55%. In phase II, to improve $\text{NH}_4^+\text{-N}$ removal, the operating DO was increased to 1.0 mg/L. Along with the improvement of nitrification, the efficiency of TN removal increased to 78%. The operating DO was further increased to 1.5 mg/L, and nitrification was enhanced, with an average effluent $\text{NH}_4^+\text{-N}$ of 3 mg/L. However, the average TN removal decreased to 55% (Fig.4).

SND in the system was realized from the analysis of the effluent nitrogen compounds. When the operating DO was maintained between 0.5 and 1.0 mg/L, no significant $\text{NO}_3^-\text{-N}$ and $\text{NO}_2^-\text{-N}$ were detected in the effluent. These results showed that the efficiency of aerobic denitrification was high when the DO was in this range. However, when the DO concentration was at 0.5 mg/L or lower, $\text{NH}_4^+\text{-N}$ became the main form of the effluent nitrogen compounds. Under this operating condition, the sludge floc tended to be anoxic, even anaerobic. This was capable of increasing the ability of aerobic denitrification of the whole system. Simultaneously, it also inhibited the nitrifiers existing in the sludge floc. Therefore, the removal of $\text{NH}_4^+\text{-N}$ and TN was affected significantly. When the operating DO was increased to 1.0 mg/L, the effluent nitrogen compounds

were all at a low level. The results indicated that the promotion of nitrification and maintaining of denitrification were the main factors that affected the SND of the system. Nevertheless, when the DO concentration was increased to 1.5 mg/L or higher, considerable $\text{NO}_3^-\text{-N}$ was measured in the effluent, and TN removal was greatly decreased as a result of the reduced denitrification rate. The higher DO concentration restrained the activity of the denitrifiers that were preferentially active in areas of very low DO concentration. The denitrification ability of the system was decreased greatly. In addition, the higher DO concentration enhanced the activity of heterotrophic aerobic bacteria and the organics could be biodegraded rapidly and thoroughly. Thus, although an anoxic zone could be formed in part of the sludge floc, the denitrification process was hampered because of lack of organics. Pochana and Keller (1999) explained that increased DO concentration in the reactor bulk liquid negatively affected SND.

These results reflected the relationship between the DO concentration and SND efficiency. The optimal DO concentrations for maximum efficiencies of SND presented a certain extent of difference among the previous studies. Katie *et al.* (2003) found that the optimal range of DO concentration was between 0.8–1.2 mg/L when they investigated SND that used reserved PHB as an electronic

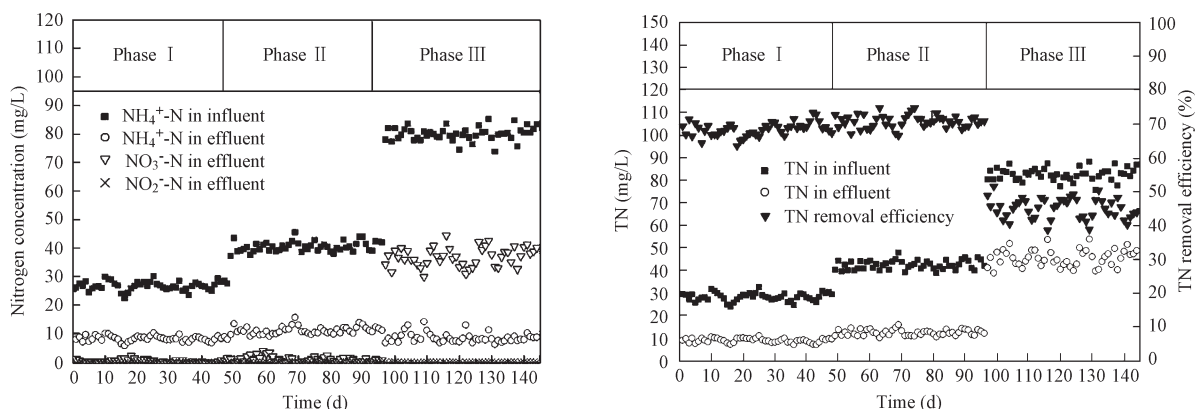


Fig. 3 Profiles of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, $\text{NO}_2^-\text{-N}$, and TN concentrations in the system when the nitrogen loading rate was adjusted at 0.013 (phase I), 0.020 (phase II) or 0.040 gN/(g VSS·d) (phase III).

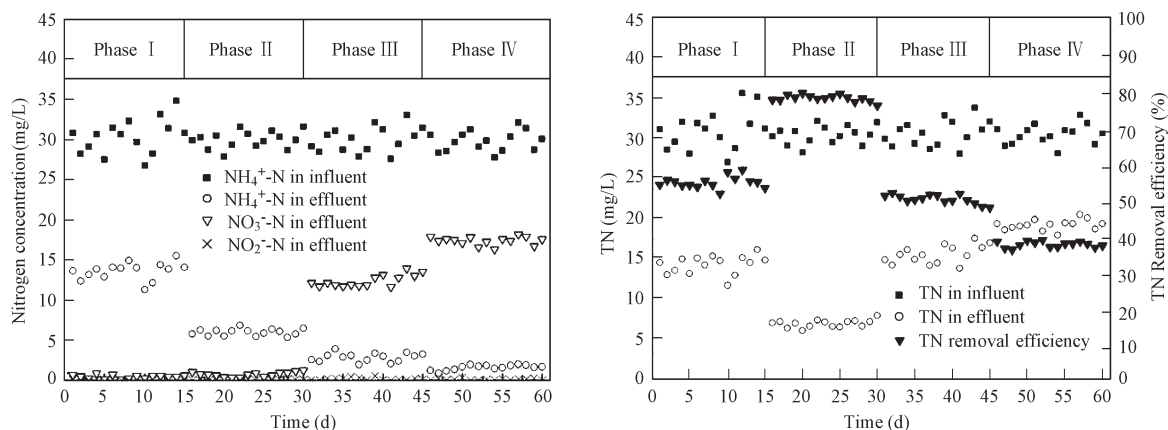


Fig. 4 Profiles of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, $\text{NO}_2^-\text{-N}$, and TN concentrations in the system when the DO concentration was controlled at 0.5 (phase I), 1.0 (phase II), 1.5 (phase III), and 3.0 mg/L (phase IV).

acceptor in an SBR. Pochana and Keller (1999) achieved more than 80% SND under DO conditions between 0.3 and 0.8 mg/L. Von Münch *et al.* (1996) required a DO concentration of 0.5 mg/L to achieve 100% SND. In general, the comparison of the studies suggested that an ideal SND process was achievable with low DO concentrations.

2.3 Batch tests

Under an initial COD/N ratio of 4.77, there was a noticeable decrease in the NH_4^+ -N removal. At the same time, peaking of NO_3^- -N was observed (Fig.5). This phenomenon indicated that nitrification by autotrophic bacteria was the major process occurring at low COD/N ratio, which is in agreement with Hooper *et al.* (1997). This also implied that the COD/N ratio was inadequate to provide enough degradable carbon sources as an electronic donor, for denitrification. Chiu *et al.* (2007) pointed out that the SND process required a sufficiently low degradable carbon substrate, to provide the power demanded by the denitrification reaction. Therefore this experimental condition was not fit for establishing an efficient SND process.

The efficiency of the SND process (E_{SND} ,%) was calcu-

lated by Eq.(1):

$$E_{\text{SND}} = \left(1 - \frac{C_{\text{NO}_x^- \text{-produced}}}{C_{\text{NH}_4^+ \text{-oxidized}}} \right) \times 100\% \quad (1)$$

where, $C_{\text{NO}_x^- \text{-produced}}$ (mg/L) was the sum of NO_3^- -N and NO_2^- -N present when the batch test ended and $C_{\text{NH}_4^+ \text{-oxidized}}$ (mg/L) was the amount of NH_4^+ -N oxidized during the batch test.

When the initial COD/N ratio was increased to a high level (10.04 and 15.11), there was no obvious NO_3^- -N and NO_2^- -N detected in the effluent (Fig.5). von Münch *et al.* (1996) proposed that an efficient SND process should not produce abundant NO_3^- -N or NO_2^- -N. The results of this experiment were consistent with the previous reports (von Münch *et al.*, 1996; Zeng *et al.*, 2003). Although the efficient SND process was attained and left trace amount of NO_3^- -N and NO_2^- -N, at a high COD/N ratio TN removal was not continuously enhanced along with the increase of COD/N ratio. When the COD/N ratio got to 15.11, TN removal showed a decrease to some extent, because of the limitation of nitrification. Denitrification was also affected by the lack of NO_x^- -N. The COD/N ratio

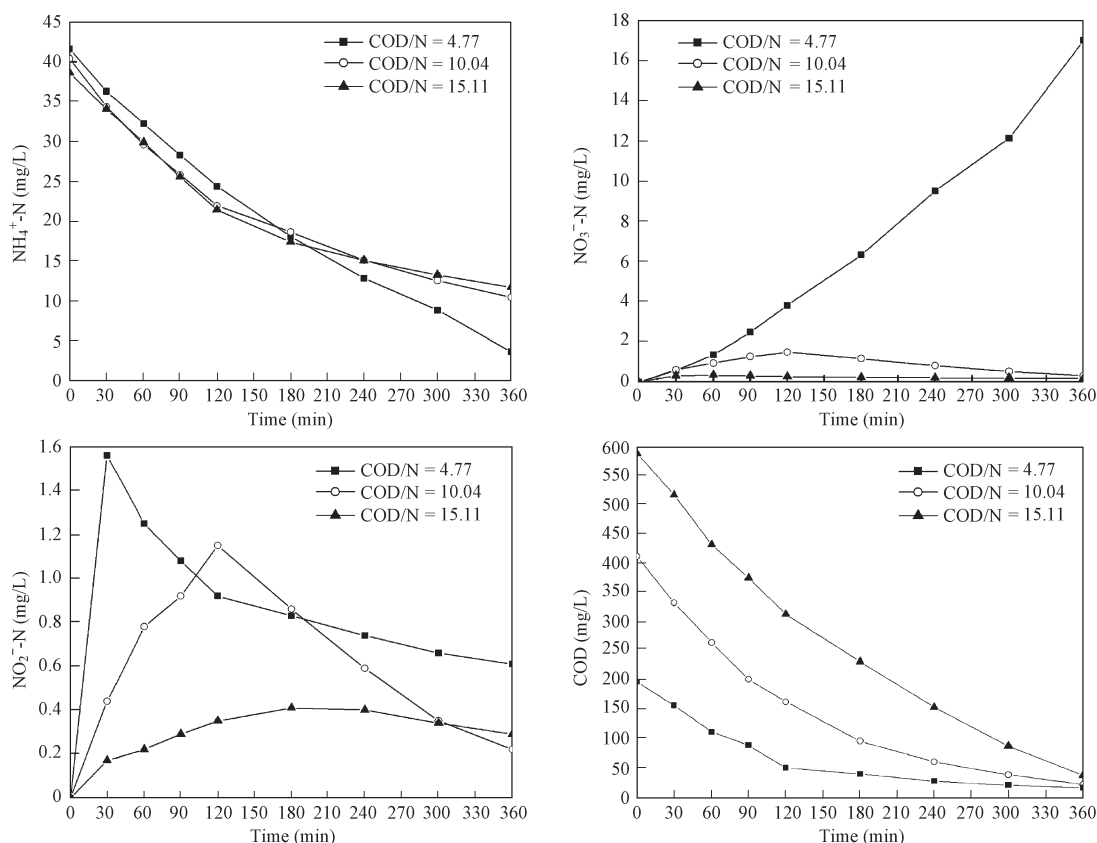


Fig. 5 Profiles of NH_4^+ -N, NO_3^- -N, and NO_2^- -N concentrations and COD during the batch tests.

Table 3 Performance of SND process under different initial COD/N ratios

COD/N ratio	Nitrification rate (mg NH_4^+ -N/(L·h))	Denitrification rate (mg NO_x^- -N/(L·h))	Efficiency of TN removal (%)	Efficiency of SND (%)
4.77	6.32	3.39	48.8	53.6
10.04	4.99	4.90	72.8	98.2
15.11	4.48	4.36	67.8	97.4

exerted some control over how heterotrophs and nitrifiers consumed common resources DO and space (Chu *et al.*, 2006). With the increase of COD/N ratio, heterotrophs gradually dominated and utilized the available DO and sludge floc space.

The nitrification and denitrification rates and efficiency of SND are summarized in Table 3. Chiu *et al.* (2007) proposed that an efficient SND process occurred when the nitrification and denitrification rates were in a balanced equilibrium. Therefore, 10.04 was considered as the optimum COD/N ratio value for SND in a single reactor in this study. With this COD/N ratio, the nitrification and denitrification rates were calculated as 4.99 mg NH₄⁺-N/(L·h) and 4.90 mg NO_x⁻-N/(L·h), respectively, basically reaching equilibrium. Furthermore, the nitrogen and organic carbon in the system were both optimally removed.

Figure 6 shows the impact of DO concentrations of 0.5, 1.0, 1.5, and 3.0 mg/L on SND. Running at 0.5 mg/L DO, almost all products from the nitrification process were converted to nitrogen gas. However, under this operating condition, the sludge floc tended to be anoxic even in an anaerobic environment that resulted in the limitation of nitrification. Moreover, the denitrification rate did not achieve a higher level as expected because of the lack of NO_x⁻-N. With the increase of DO concentration, a more significant increase in NH₄⁺-N removal was observed. Theoretically, the denitrification rate should be increasingly inhibited by high DO levels. Nevertheless, when the DO

concentration increased from 0.5 to 1.0 mg/L, the denitrification rate presented a certain extent of improvement. It could be explained that improved nitrification supplied a sufficient electronic acceptor for denitrification. At full aeration (DO concentration >1.5 mg/L), complete nitrification was noticed, but the percentage of SND decreased. This indicated that the oxygen supply was too high to allow for significant denitrification to occur simultaneously.

As shown in Fig.7, with the reduction in DO concentration, the percentage of SND could be improved. This result was in accordance with a previous research (Pochana *et al.*, 1999; von Münch *et al.*, 1996; Beun *et al.*, 2001). However, Katie *et al.* (2003) proposed that for satisfactory nutrient removal from wastewater not only the percentage of SND, but also the rate was important.

$$R_{SND} = \frac{C_{NH_4^+-oxidized} - C_{NO_x^--produced}}{T} \quad (2)$$

From the results, the maximum R_{SND} was achieved at a DO concentration of 1.0 mg/L, yet the percentage of SND at this DO concentration was not the optimal. The DO concentration was higher than 1.0 mg/L, yet both the rate and quantity of SND decreased; whereas, by lowering the oxygen supply (DO concentration < 1.0 mg/L), the percentage of SND greatly increased, but the rate decreased. Hence, there was a compromise between the rate and quantity at which SND occurred and total nitrogen removal. Katie *et al.* (2003) adopted the method where the rate of SND was multiplied by the fraction of the

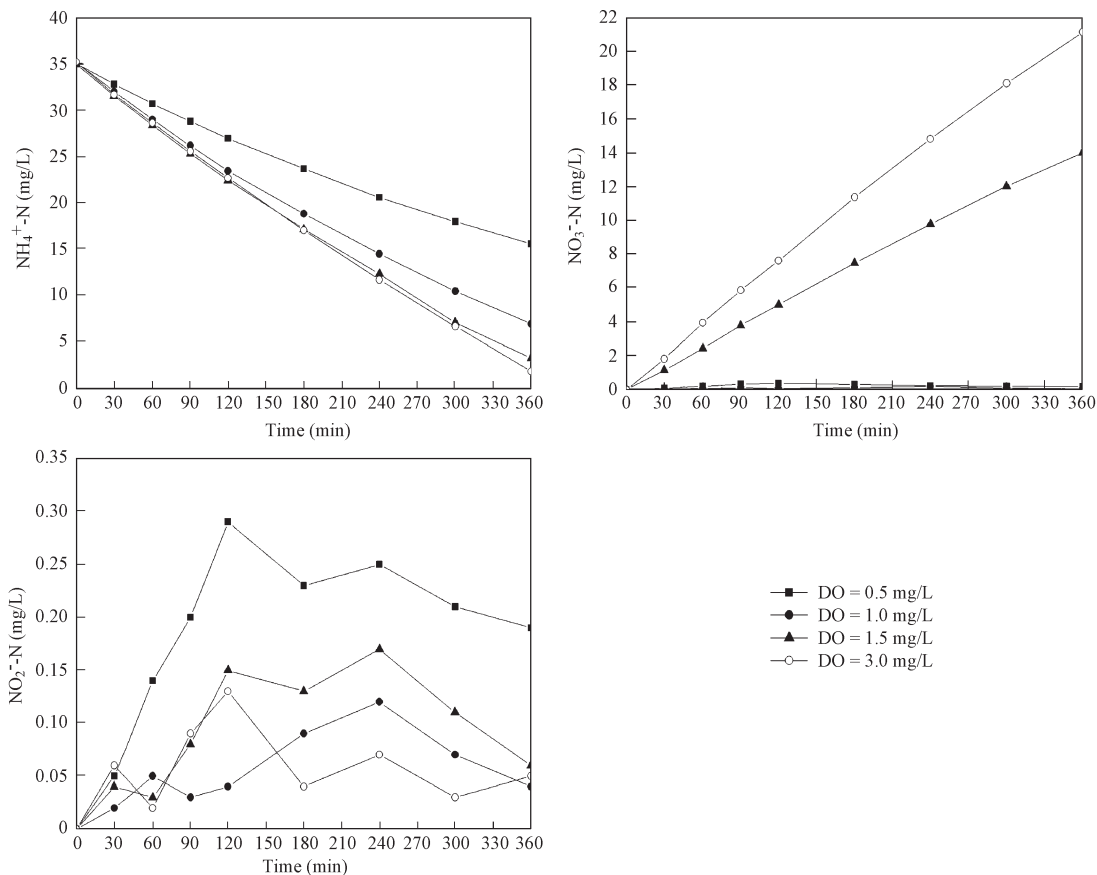


Fig. 6 Profiles of NH₄⁺-N, NO₃⁻-N, and NO₂⁻-N concentrations during the batch tests when the DO concentration was controlled.

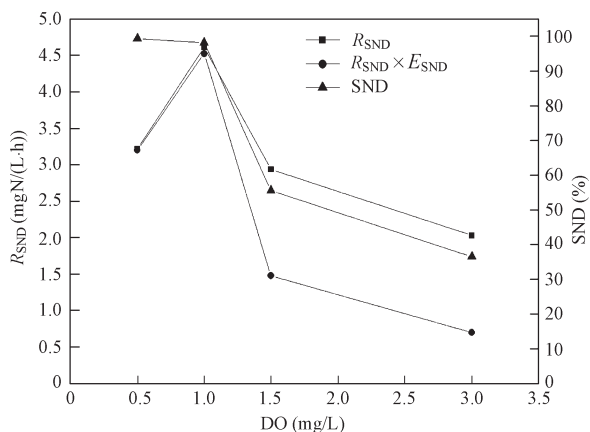


Fig. 7 Profiles of percentage of SND (E_{SND}) and the rate of SND (R_{SND}), and the SND rate multiplied by SND (%) when the DO concentration was controlled at 0.5, 1.0, 1.5 or 3.0 mg/L.

SND profile of DO concentration, to assess the area of DO concentration where the optimum SND rate and quantity occurred between 0.75–1.0 mg/L.

3 Conclusions

This study presents the manner in which the COD/N ratio and DO concentration affect the occurrence of the SND process in the AIC-MBR system.

Fixing the organic loading rate, the nitrogen loading rate was increased gradually, to investigate nitrogen removal. The study showed that it administered nitrification, but TN removal did not present an increasing trend along with the promotion of nitrification. The results indicated that there was an optimum COD/N ratio for nitrogen removal through the SND process.

At significant high or low DO concentration, the SND process was suppressed. It could be attributed to the change of aerobic or anoxic zones within the floc, by oxygen supply.

The results of the batch tests showed that the nitrification and denitrification rates reached equilibrium and resulted in nearly complete SND when the COD/N ratio was controlled at 10.04. With this COD/N ratio, nitrogen and organic carbon were both optimally removed. From the batch tests, 0.75–1.0 mg/L was taken as the optimum range of DO concentration for SND. This range of DO concentration decided the quantity and the rate of SND at which high levels of SND were achieved.

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