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Simultaneous COD and nitrogen removal in a micro-aerobic granular sludge reactor for domestic wastewater treatment

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Abstract

The study on the integrated COD and nitrogen removal from domestic wastewater was carried out in a micro-aerobic granular sludge reactor. The influences of oxygen flux, hydraulic retention time (HRT), biomass concentration and sludge loading were investigated. The reactor showed stable and effective COD removal all through the experiment. Nevertheless, nitrogen removal much depended on the operation mode. Under the optimal condition, i.e. oxygen influx, HRT, MLSS and sludge loading rate (SLR) were 0.20g/L·d, 8h, 11~16g/L and below 0.10kgCOD/kgMLSS·d, respectively, total nitrogen (TN) removal efficiency achieved 82%, and the concentration of NH_4^+-N and TN in effluent were 5.5mg/L and 8.2mg/L, respectively. Besides, deteriorated sludge settling and filamentous granules were found as SLR increased to 0.18kgCOD/kgMLSS·d.

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Keywords:micro-aerobic granular sludge reactor; COD removal; simultaneous nitrification and denitrification; domestic wastewater;influence factors

1. Introduction

The recently reported micro-aerobic or oxygen-limited wastewater treatment has attracted more attentions from researchers in way of energy conservation and efficient operation [1, 2]. Compared with the separated aerobic and anoxic unit in the traditional processes, oxygen-limited system facilitates both micro-aerobic (with DO less than 1mg/L) and anaerobic micro-environment in single reactor due to the existence of DO concentration gradients within biomass matrix caused by mass transfer resistance and biological oxygen demand. And microbial populations including aerobe, anaerobe and facultative aerobe can flourish in different depth of biological floc to oxidize COD and remove nutrients simultaneously. Thus it is desirable to build an alternative configuration for organics and nitrogen removal different from the conventional biological nutrient removal (BNR) process such as intermittent aeration, two stage anoxic/oxic reactors and two stage oxic/anoxic reactors [3].

Biomass used for micro-aerobic process can be activated sludge [4], biofilm [5] and microbial granule [2, 6], among which granular sludge has the special nature that offers gradient descent of oxygen concentration from outer to intra

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granule, namely aerobic or facultative populations enrich in the outer periphery to consume oxygen while anaerobic ones flourish in the intra granule free from oxygen. This feature is particularly important for simultaneous nitrification and denitrification (SND). Study showed that both flocculent [7] and granular sludge [2,8-10] could be the inoculum for micro-aerobic granular sludge, moreover, nitrogen removal was much favored thanks to the increased SND [1, 2, 8, 9]. For example, Ren et al. [7] inoculated an expanded granular sludge bed (EGSB) reactor with the mixture of aerobic and anaerobic ammonium oxidation floc, and slightly aerated to build up nitrogen removal, gained 61% TN removal as DO was controlled about 0.5mg/L. Chu et al. [2] investigated the performance of micro-aerobic membrane bioreactor seeded with anaerobic granule for the treatment of synthetic domestic wastewater, and found that nitrogen was removed through SND process with 65~92% TN removed as HRT ranged in 16~24h.

Among the studies about micro-aerobic wastewater treatment, oxygen (generally represented by DO) was considered one of the most important factors that affect microbial reaction, especially for SND process [1, 3, 5, 11]. Other parameters such as HRT and sludge loading rate (SLR) were proposed to have influence more or less [11, 12]. It should be mentioned that the optimal DO concentration for micro-aerobic operation is related to various factors including influent concentration, floc size and structure, etc, and most of the researchers gave an approximate DO range. It is hard to determine the accurate DO value inside the high-concentration sludge blanket under anoxic condition, since the precision of experimental apparatus was limited at low DO concentration (< 0.5mg/L). Thus control of the oxygen influx into bioreactor may be a better choice for the micro-aerobic system, instead of the DO value of mixed liquid. And this is particularly suitable for the external oxygenated bioreactor as to associate nitrogen removal with oxygen utilization [13].

In this work, a micro-aerobic granular sludge reactor with external oxygenation was assembled from EGSB reactor and used to facilitate simultaneous organic and nitrogen removal from domestic wastewater. Factors that influenced treatment efficiency, such as oxygen, HRT and biomass concentration, were investigated to optimize the operation. Additionally, oxygen influx rate was engaged to evaluate the influence of oxygen.

2. Materials and methods

2.1. Reactor setup and operation

As shown in Figure 1, the experimental set-up included an 18L bioreactor and an 8L external aeration column (effective volume). The bioreactor was composed of reaction part (height: 1.8m, I.D.: 0.1m) and gas-solid-liquid separator (height: 0.5m, I.D.: 0.14m). A part of effluent was oxygenated and recirculated together with influent into the bottom of bioreactor reactor. In this way, a stable sludge bed was maintained to protect the granule from damaging by air bubbles. Besides, oxygen influx into micro-aerobic bioreactor could be controlled by manipulating the DO level in aeration column and recirculation rate. During the whole experiment, DO in aeration column changed gradually from 4.5mg/L to about 8mg/L with recirculation rate ranged 8:1~10:1, and DO near the outlet of the bioreactor was below 0.5mg/L.

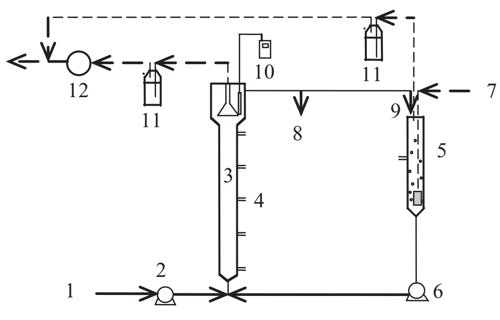


Fig. 1. Schematic diagram for micro-aerobic experiment: 1 influent; 2 inlet pump; 3 micro-aerobic reactor; 4 sample port; 5 aeration column; 6 recirculation pump; 7 air; 8 effluent; 9 recirculation; 10 DO meter; 11 water seal; 12 gas meter.

The reactor inoculum was the mixture of anaerobic granule (from a lab scale EGSB reactor for domestic wastewater treatment) and aerobic activated sludge (from a local municipal wastewater treatment plant) in volume ratio 4:1. Mixed liquor suspended solid (MLSS) seeded was about 16.0g/L. The influent was collected from the drainage system of a residential area nearby, with characteristics given in table1.

Table 1. Characteristics of domestic wastewater

Parameter	Range (*mg/L*)	Parameter	Range
COD	144~628	TP(*mg/L*)	2~14
NH_4^+-N	20~72	SS(*mg/L*)	75~240
NO_x^--N	0.2~2.5	pH	6.5~8.1
TN	26~90	Alkalinity(*mgCaCO$_3$/L*)	146~391

NO_x^--N is the total of NO_2^--N and NO_3^--N

After startup, the reactor was operated at room temperature. In order to seek the optimal running condition of simultaneous COD and nitrogen removal, three stages were engaged to investigate influences of oxygen influx rate, HRT and biomass concentration on the treatment effect (depicted in Table 2). During each period, series test were conducted with different value of the target parameter. For the first two periods, sludge concentration was maintained constant and oxygen influx or HRT was changed alternately; for the third period, oxygen influx and HRT were set at the optimal values respectively, and sludge concentration was altered through sludge discharge. Each value of the target parameter was kept about half month during the steady state. Oxygen influx rate into bioreactor was calculated as following:

$$O_{influx} = \frac{DO_{column} \times R \times Q}{V} \qquad (1)$$

Where O_{influx} is the oxygen influx rate, $gO_2/(L\cdot d)$; DO_{column} is the DO value in aeration column, mg/L; R is the reflux ratio; Q is the inflow rate, L/d; V is the effective volume of bioreactor, L.

Table 2. Operation conditions of the reactor for different periods

Period	O_{influx} (*g/L·d*)	HRT (*h*)	MLSS(*g/L*)
I	0.11/0.13/0.17/0.20/0.22/0.25	8	15~16
II	0.20	5/6/7/8/10	15~16
III	0.20	8	11/5.9

2.2. Analytical methods

COD, NH_4^+-N, NO_2^--N, NO_3^--N, TN, MLSS, and sludge volume index (SVI) were determined according to standard methods [14]. DO was measured by DO200 meter, pH value was monitored by pHS-3C meter. SEM pictures of the granules were taken with an environmental scanning electron microscope (FEI Quanta200). And samples were dried and precoated with gold before SEM analysis.

3. Result and discussion

3.1. COD and nitrogen removal at different oxygen influx

Table 3 shows the oxygen influx and reactor performance during period I. The system presented excellent COD removal (>90%) in despite of different oxygen influx value, which indicated less influence of oxygen on organics degradation. Nevertheless, nitrogen removal exhibited a close relationship with O_{influx}, i.e. less ammonium and TN were removed at low O_{influx}, but the removal rates rose to more than 80% as O_{influx} increase to above $0.20gO_2/L\cdot d$. It was considered that organics in domestic sewage could be used as the carbon source of both aerobic and anaerobic heterotrophs, so as to remain COD removal independent from oxygen influx. However, nitrogen conversion was more complicated in the SND system since a special environment was needed for the coexistence and cooperation of nitrifier and denitrifier, namely that the oxygen influx should be sufficient for nitrification in aerobic region but low enough not to inhibit denitrification in anoxic region. The low O_{influx} in series a~c might be insufficient for nitrification although denitrification went on well (proved by the little nitrite/nitrate accumulation in effluent, data not shown).

Table 3. Organic and nitrogen removal at different oxygen supply condition (HRT=8h)

Series	a	b	c	d	e	f
O_{influx} (g/L·d)	0.11	0.13	0.17	0.20	0.22	0.25
COD_{in} (mg/L)	343±73	440±100	408±119	340±107	303±106	447±80
COD removal (%)	93	94	94	93	93	95
NH_4^+-N_{in} (mg/L)	31.8±8.2	39.3±7.6	34.8±4.5	39.2±7.5	34.8±6.7	33.5±8.6
NH_4^+-N removal (%)	40	55	78	83	84	86
TN_{in} (mg/L)	41.7±10.1	48.5±8.6	45.9±7.7	47.7±9.0	46.5±9.5	45.5±10.2
TN removal (%)	51	61	79	82	82	82

Complete nitrification is the oxidation of ammonium to nitrate via nitrite, which can be inhibited at nitrite under oxygen-limited condition as the affinity for oxygen of ammonium oxidizing bacteria is better than that of nitrite oxidizing bacteria [1, 5, 11]. Thus it is desirable to achieve SND via nitrite by controlling oxygen influx. This partial nitrification-denitrification of single stage biodenitrification has many advantages such as decreasing aerating energy and saving carbon source. In Fig.2, volumetric TN removal increased remarkably from 0.06gN/L·d to 0.11gN/L·d as O_{influx} rose from 0.11 to 0.20gO_2/L·d, and then stagnated as O_{influx} continue to increase to 0.25gO_2/L·d. This indicated that complete nitrification occurred and there was no more TN removal for the additional oxygen influx. Therefore, it is important to regulate the oxygen influx for a cost-effective operation under micro-aerobic condition. Thus O_{influx} was set about 0.20gO_2/L·d for the later operation.

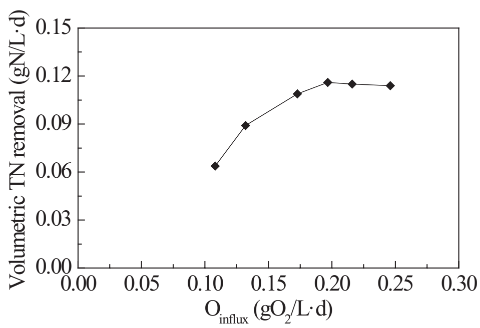


Fig. 2. Nitrogen removal rate at different oxygen influx rate

3.2. Influence of HRT

In the second period, influent COD, NH_4^+-N and TN ranged in 303~479mg/L, 34~47mg/L and 46~60mg/L, respectively. And the influence of HRT on COD and nitrogen removal was presented in Fig.3. Compared with the little change in COD removal (remained above 90% with effluent concentration less than 30mg/L) within HRT 5~10h, nitrogen removal rate presented an upward trend accompanied with the increase of HRT, e.g. only 54% NH_4^+-N and 60%TN were removed at HRT 5h, but these increased to 84% and 82% respectively with effluent concentration of 5.5mg/L and 8.2mg/L, as HRT was extended to 8h. This indicated that nitrogen removal in way of SND needs longer HRT than COD degradation in the micro-aerobic system. It should be mentioned that NH_4^+-N was the main component of TN in effluent during the whole experiment (meaning that nitrification was the rate-limiting step of SND), and NO_x^--N was negligible most of the time except for HRT 10h. As for the slightly accumulated NO_x^--N at HRT 10h, it is possible that too much hydraulic retention may result in the lack of carbon source for denitrification. This however, is contradictive with not only TN removal but also reactor effectiveness.

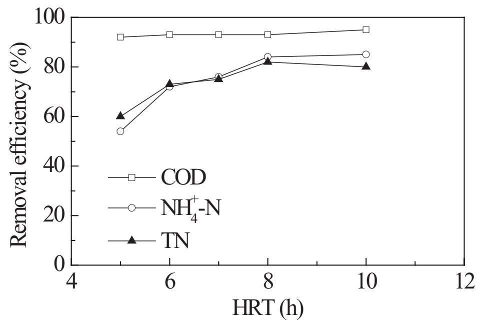


Fig. 3. Influence of HRT on COD and nitrogen removal

3.3. *Influence of biomass concentration and sludge loading*

High biomass concentration is benefit to nitrogen removal because the lower SLR and longer sludge retention in this condition can support the increase of slow-growing autotrophic nitrifiers. This is particularly true for this micro-aerobic SND system since the gain in the number of nitrifying organism could make up the loss of individual activity caused by low oxygen. In Fig.4, TN removal efficiency remained about 80% when MLSS was kept 11~16g/L, but sharply dropped down to 66% as MLSS decreased to 5.9g/L. It is likely that too much reduction of biomass cut down the number of nitrifiers, which played the key role during TN removal. Besides, this phenomenon can also be interpreted as the result of increasing SLR. As the reduction of MLSS to 5.9g/L, the average SLR increased from below 0.10kgCOD/kgMLSS·d to 0.18kgCOD/kgMLSS·d. It is possible that the higher SLR depressed nitrogen removal.

On the other hand, the excess sludge at high biomass concentration may serve as carbon source of denitrification through endogenous metabolism when the influent organic was not enough. This can be seen from the stable TN removal despite the influent COD fluctuated. Typically, the ratio of BOD:TN in influent is recommended more than 4 to sustain nitrogen removal in the conventional process, otherwise the addition of carbon source will become necessary. While in this work, the ratio of COD:TN in influent ranged in 2.6~13.8, and none decrease in TN removal was seen during the steady stage, even for the low ratio.

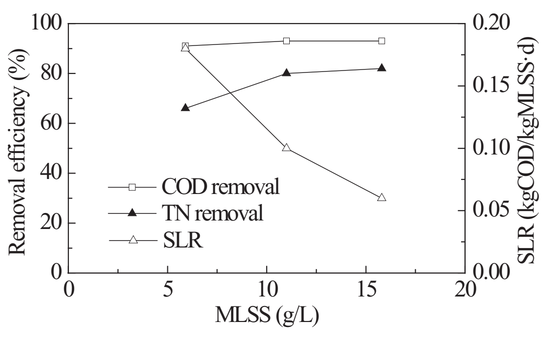


Fig. 4. Influence of sludge concentration on COD and nitrogen removal

3.4. *Sludge settleability and filamentous bacteria distribution*

As far as wastewater biotreatment is concerned, stable operation has close relationship to good sludge settling. And sludge settleability is an important feature to be considered for micro-aerobic process since insufficient oxygen may result in filamentous bulking and granules disintegration [15]. In this work, sludge settling and filamentous bacteria distribution were found much affected by the sludge loading. As SLR was less than 0.10kgCOD/kgMLSS·d, SVI of the low, middle and top of sludge blanket were 16~19ml/g, 23~27ml/g and 33~37ml/g, respectively (shown in Fig.5). And most of the biomass was composed of compact granules, within which filamentous bacteria was limited in both amount and growth region (Fig.6 A). However, sludge settleability much deteriorated as SLR rose to 0.18kgCOD/kgMLSS·d, some granules began to break or even broke into floc, which led to a great increase of SVI in the top sludge bed. On the other hand, filamentous bacteria grew predominantly in the outer layer of granule during this time (shown in Fig.6 B), which made the granular structure porous and loose. Obviously, the too much increased SLR accelerated the filamentous growth so as to deteriorate sludge settleability.

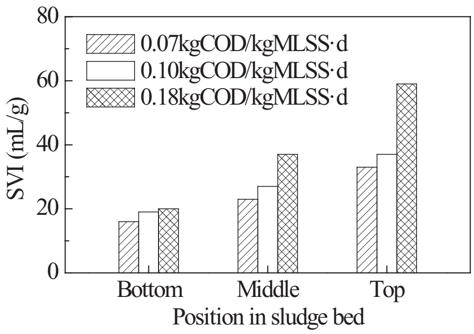
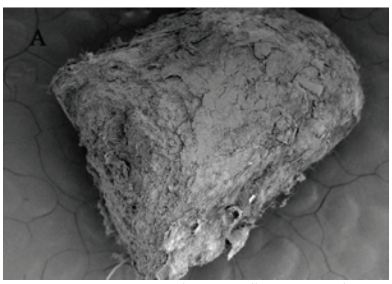
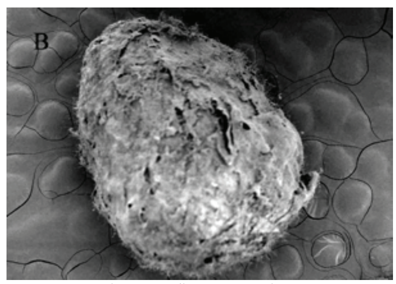


Fig.5. SVI at different sludge loading rate



A 0.10kgCOD/kgMLSS·d;



B 0.18kgCOD/kgMLSS·d

Fig. 6. Pictures of the granules at different SLR

4. Conclusions

The micro-aerobic granular sludge reactor had stable COD removal effect under different operation mode, whereas nitrogen removal much depended on oxygen influx, HRT, sludge concentration and sludge loading. Better TN removal was achieved when oxygen flux, HRT, MLSS and SLR were set at 0.20g/L·d, 8h, 11~16g/L and below 0.10kgCOD/kgMLSS·d, respectively. For sludge settleability, lower SVI and compact granular structure with restricted filamentous growth were observed at low SLR, but things deteriorated as SLR much increased.

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