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Inhibition and recovery of nitrification in treating real coal gasification wastewater with moving bed biofilm reactor

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Abstract

Moving bed biofilm reactor (MBBR) was used to treat real coal gasification wastewater. Nitrification of the MBBR was inhibited almost completely during start-up period. Sudden increase of influent total NH_3 concentration was the main factor inducing nitrification inhibition. Increasing DO concentration in the bulk liquid (from 2 to 3 mg/L) had little effect on nitrification recovery. Nitrification of the MBBR recovered partially by the addition of nitrifying sludge into the reactor and almost ceased within 5 days. Nitrification ratio of the MBBR achieved 65% within 12 days by increasing dilute ratio of the influent wastewater with tap water. The ratio of nitrification decreased to 25% when influent COD concentration increased from 650 to 1000 mg/L after nitrification recovery and recovered 70% for another 4 days.

Key words: moving bed biofilm reactor; coal gasification wastewater; nitrification inhibition; nitrification recovery

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Introduction

Coal gasification wastewater is generated in the process of coal gas purification and contains large quantities of organic and inorganic pollutants (Ye et al., 2002). Main pollutants in the wastewater are phenolic compounds which can inhibit biotransformation (Hill and Robinson, 1975) and ammonia which is one of the worst contaminators for aquatic life in the form of free ammonia (Effler et al., 1990).

Biological treatment is widely used to treat the wastewater after being pretreated by the processes of ammonia stripping and phenols solvent extraction to reduce the concentration of toxic compounds (Zhang et al., 2006). Ammonia is usually removed by autotrophic nitrification which is sometimes affected severely in the presence of high organic content and inhibitory compounds containing in the wastewater (Gupta and Gupta, 2001; Okabe and Oozawa, 1996), because the activity and growth of autotrophic nitrifying bacteria are suppressed easily by fast-growing heterogeneous bacteria (Ling and Chen, 2005). Ammonia removal of the coal gasification wastewater avoiding environmental damage is a complicated process and all pollutants must be treated simultaneously and effectively.

Free ammonia is an important factor in the nitrification for the treatment of high strength ammonia wastewater (Yun and Kim, 2003; Kim et al., 2003; Peng et al., 2004). Nitrification requires two bacteria populations performing two series processes which are the oxidation of ammonia to nitrite by the ammonia oxidizing bacteria (AOB) and the oxidation of nitrite to nitrate by the nitrite oxidizing bacteria (NOB). NOB are more sensitive to free ammonia in the range of 0.1–1.0 mg/L while AOB are inhibited by free ammonia in the range of 10–150 mg/L. Nitrification can be incomplete or even ceased if free ammonia level is high (Ganigué et al., 2007).

Nitrification can be affected in treating wastewater containing organic pollutants for oxygen shortage (Figueroa and Silverstein, 1992). Moreover, heterogeneous bacteria compete with nitrifying bacteria for available oxygen and nourishment in the reactor. The oxygen saturation coefficients were found to be 0.3 g O_2/m^3 for AOB and 1.1 g O_2/m^3 for NOB (Wiesmann, 1994). Ruiz et al. (2003) reported that DO had no influence on nitrite accumulation at 5.7–2.7 mg/L, a temporal accumulation of nitrite was happened at DO of 1.7 mg/L, and both nitrite accumulation and ammonia consumption decreased at DO of 0.5 mg/L.

Many other factors, such as lack of inorganic carbon (Vázquez et al., 2006), temperature fluctuation (Fdz-Polanco et al., 1994), nitrifying bacteria being washed out in suspension systems (Blackburne et al., 2007), can also induce nitrification inhibition. Attached biofilm systems

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have high sludge retention time and less sensitivity to the change of environment which are suitable for treating wastewater containing high strength ammonia and inhibitory compounds. At the same time, a continuous stirred tank reactor (CSTR) is recommended to avoid abnormal increase of inhibitory compounds concentration in some parts of the reactor (Kim et al., 2006).

Nitrification can be affected either in adhered or suspended wastewater treatment systems and some adjustments of the system should be done to recover nitrification. Nitrification recovery is more difficult in the suspended system than in the adhered system because the nitrifying bacteria are more susceptible in suspended form. Kim et al. (2009) pointed out that nitrification was almost ceased in treating coke plant wastewater in a full-scale treatment process for shorting of alkalinity and nitrification was not recovered by adding $NaHCO_3$. Kim et al. (2007) reported that a full-scale suspended nitrification process was instable in summer due to washing out of nitrifiers by fast growth of competitive microorganisms at higher temperature with increasing phenol and thiocyanate concentrations. Adhered system is suitable for nitrification because the nitrifying bacteria grow adhering to carrier and can not be washed out easily. Lee et al. (2004) reported that nitrification of the nitrifying biofilm was affected when it had been exposed to organic carbon-containing wastewater for a prolonged period and was recovered for heterotrophic biofilm deterioration after the substrate shifting from acetate to ammonia.

In this study, a moving bed biofilm reactor (MBBR) was used to treat real coal gasification wastewater because of its high capacity on mixing and tolerance to the inhibitory compounds. Nitrification ratio of the MBBR increased gradually at the beginning of start-up period and almost ceased suddenly. Possible factors inhibiting nitrification of the MBBR were discussed. Some adjustments of the reactor operation conditions were also performed to investigate the feasible method to recover nitrification.

1 Materials and methods

1.1 Moving bed biofilm reactor

The MBBR used had a working volume of 4 L with cylindrical shade, with height 30 cm and radius 14 cm. The reactor did not use stirred device and movement of carriers depended on aeration solely. Compressed air was supplied at the bottom of the reactor using four separate diffusers with adjustable valve each. Such configuration would prevent anoxic zone in the reactor at low air flow rate. Carrier used in the reactor was made of polyethylene and the density was about 0.98 kg/m^3. The carrier had cylindrical shape with cross configuration inside and biofilm grew on the inner surface of the carrier.

1.2 Coal gasification wastewater

The coal gasification wastewater used in the experiment was collected from Harbin Coal Gasification Factory and had been pre-treated by ammonia stripping and phenols solvent extraction processes to facilitate subsequent biological treatment. Properties of the pre-treated wastewater are shown in Table 1. As can be seen, concentrations of pollutants in the wastewater changed greatly. One or several pollutants might increase abruptly beyond the given range because of abnormal operation of the pre-treatment process.

Table 1 Properties of experimental wastewater

Parameter	Range	Mean
pH	5–8	6.5
COD (mg/L)	1500–2500	2000
NH_4^+-N (mg/L)	100–250	175
Total phenol (mg/L)	300–600	450

1.3 Experimental setup

The MBBR was operated at (33 ± 1)°C. DO concentration in the bulk liquid of the reactor was controlled at (1.5 ± 0.2) mg/L by regulating air flow rate to keep partial nitrification to nitrite in the reactor. Inoculum sludge of the MBBR was collected from full-scale activated sludge process treating the coal gasification wastewater in the coal gasification factory. Sedimentation pond with 1 L volume was used to separate sludge from effluent wastewater by gravity. Part of sludge at the bottom of the sedimentation pond was recycled into the MBBR to maintain the suspended sludge retention time around 30 days during start-up period.

Carrier filling ratio was an important factor affecting MBBR performance. Increasing carrier filling ratio would elevate biomass in the reactor, and enhance the collision among carriers. Wang et al. (2005) studied carrier filling ratio varied from 10% to 75% using suspended carrier biofilm reactor, and the optimum carrier concentration was 50%. At the same time, high carrier filling ratio needed high air flow rate to assure carriers movement in the reactor. Low air flow rate was used to control DO concentration around 1.5 mg/L in the reactor, thus, carrier filling ratio of the MBBR was 40% in the experiment to maximize the biomass on the premise of carriers movement in the reactor.

Influent of the MBBR, supplied at a rate of 0.13 L/hr, was diluted with tap water based on COD concentration of the pre-treated coal gasification wastewater. Sodium bicarbonate (1 g/L) was added into the feed of the MBBR serving as inorganic carbon source to support nitrifying bacteria growth.

1.4 Analytical methods

The samples were taken from effluent of the MBBR every day and were analyzed immediately after filtered through 0.45 μm filter paper. Soluble COD, total phenol, total NH_3 (TNH_3), NO_3^--N and NO_2^--N were measured in accordance with standard methods. DO and pH in the bulk liquid were measured using oxygen meter (Model 58, YSI Instrument Company, USA), and pH meter (PHS-3D, Ltd., China), respectively.

2 Results and discussion

2.1 Nitrification inhibition

Nitrification ratio of the MBBR increased gradually during start-up period and reached approximately 35% after 25 days. Oxidation of total ammonia was almost NO_2^--N (Fig. 1a), and effluent NO_2^--N concentration achieved 25 mg/L on day 27.

The MBBR had good effects on COD and total phenol removal at the beginning of the experiment because the inoculum sludge had been used to treat the wastewater for more than one year (Fig. 1b). Effluent COD concentration was in the range of 200–250 mg/L when influent COD concentration was 1000 mg/L. Influent total phenol concentration was fluctuated from 150 to 250 mg/L and effluent was stable after 20 days.

Nitrification ratio decreased suddenly at day 29 and almost ceased within 4 days. Operation conditions of the reactor, such as DO concentration and temperature, were normal through out the period of nitrification failure. Thus, nitrification failure was not due to failure of aeration and heater. Wett and Rauch (2003) pointed out that inorganic carbon limitations may be the main reason for decrease of nitrifying bacteria activity. Scarcity of inorganic carbon could not be the factor affecting nitrification because $NaHCO_3$ was added into the influent of the reactor in the experiment.

Phenolic compounds were another factor inducing nitrification inhibition at certain concentration. Dyreborg and Arvin (1995) indicated that pseudo-critical concentration of phenol was 3.7 mg/L in pure culture of nitrifying bacteria; Kim et al. (2008) reported that threshold phenol concentration for inhibiting nitrification was 200 mg/L in treating cokes wastewater with batch activated sludge system. Tremendous difference of the phenol inhibition concentration would be due to different operation conditions. Extended aeration process was used in the experiment, and influent total phenol concentration was in the range of 150–250 mg/L. Nitrification ratio increased gradually (Fig. 1b), which indicated that nitrification of the MBBR could be achieved at such total phenol concentration. Influent and effluent total phenol concentration of the reactor had no obvious variety during nitrification inhibition period, thus, nitrification inhibition of the MBBR must not be caused by phenolic compounds.

TNH_3 in the wastewater has two forms which are ammonium (NH_4^+-N) and free ammonia (NH_3-N). NH_3-N concentration, relevant to total ammonia concentration, temperature and pH value, was an often mentioned factor affecting nitrification. NH_3-N concentration could be calculated by the following equation (Anthonisen et al., 1976).

$$[NH_3\text{-N}] = \frac{TNH_3 \cdot 10^{pH}}{\exp^{[6334/(273+T)]} + 10^{pH}}$$

Influent TNH_3 concentration of the MBBR did not control at the beginning of start-up period and increased to 200 mg/L abruptly (Fig. 1a) on day 30, although influent wastewater was diluted almost twice to control influent COD concentration.

NH_3-N concentration based on effluent TNH_3 concentration and ratio of NH_3-N/TNH_3 are shown in Fig. 2. NH_3-N concentration in the reactor was around 5 mg/L at the beginning of the experiment and increased to 40 mg/L sharply for sudden increase in influent TNH_3 concentra-

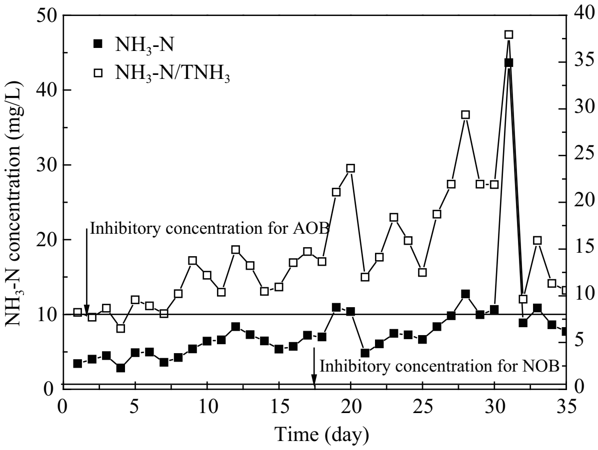

Fig. 2 NH_3-N concentration and ratio of NH_3-N/TNH_3 before and after nitrification inhibition.

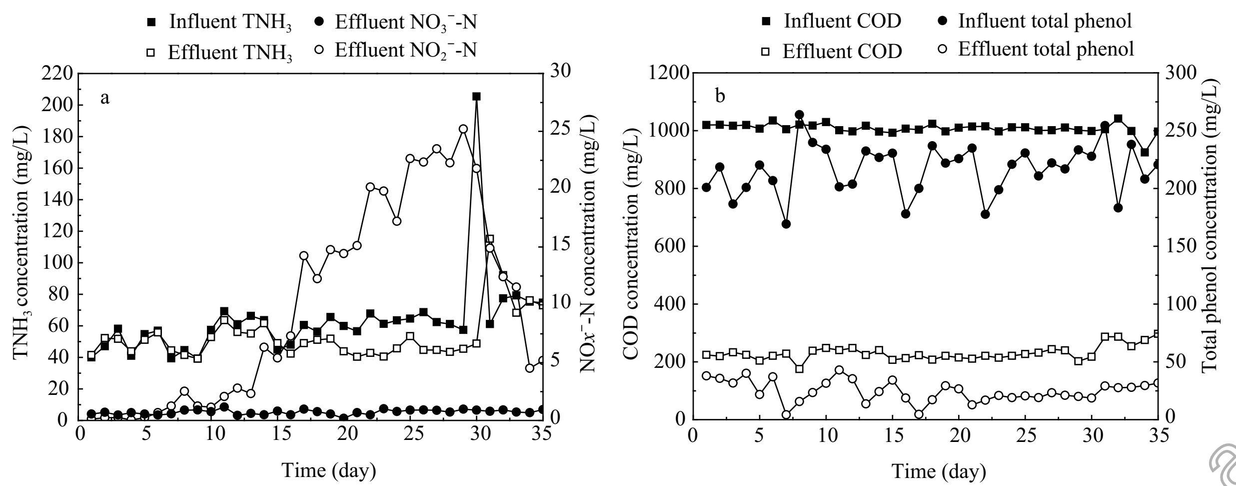

Fig. 1 Total ammonia removal (a) and COD and total phenol removal (b) of the MBBR before and after nitrification inhibition.

tion. Ratio of NH_3-N/TNH_3 achieved 38% when TNH_3 concentration increased sharply. Free ammonia inhibition effect on the activity of the AOB during nitrification was studied by many researchers and inhibition concentration was different greatly (10–150 mg/L) (Kim et al., 2006). In our experiment, NH_3-N concentration around 10 mg/L had no obvious inhibitory effect on nitrification (Fig. 2). The faster growing heterogeneous bacteria would be established at the outer biofilm layer while slower growing nitrifying bacteria would be developed near the attachment surface in treating high C/N ratio wastewater. This arrangement increased mass transfer resistance for ammonia to be diffused into the deeper lying nitrifying biofilm layer and the configuration would decrease NH_3-N inhibitory effect on the nitrifying bacteria. Nitrification of the MBBR was almost ceased while NH_3-N concentration increased to 40 mg/L suddenly which indicated that the activity of the AOB was inhibited almost completely. Ratio of NH_3-N/TNH_3 was highest as well. Thus, NH_3-N inhibition was the main reason inducing nitrification failure of the MBBR during start-up period.

2.2 Nitrification recovery

Although NH_3-N concentration decreased around 10 mg/L soon after the sharp increase by replacing influent wastewater of the MBBR, nitrification of the MBBR did not recovery for another 5 days (Fig. 1a). Some adjustments of the MBBR operation conditions were done to recover the activity of nitrifying bacteria so as to recover nitrification.

2.2.1 Dissolved oxygen (DO) concentration

DO is a key factor affecting nitrification. Sudden increase in NH_3-N concentration inhibited activity of the nitrifying bacteria and the balance between nitrifying bacteria and heterogeneous bacteria in the biofilm was broken. Increasing DO concentration in the bulk liquid would enhance oxygen penetration in the deeper layer of the biofilm which might recover activity of nitrifying bacteria.

Nitrification of the MBBR did not recover by increasing DO concentration from 2 to 3 mg/L (Fig. 3). Influent COD concentration was well controlled around 1000 mg/L. TNH_3 and total phenol concentrations were in the normal range compared with the level before nitrification inhibition. COD and total phenol removal of the MBBR was enhanced a little with the increase in DO concentration. Dissolved oxygen consumed by heterogeneous bacteria had no obvious increase for the limit of available organic pollutants in the influent. Thus, available oxygen in the deeper layer of the biofilm was increased for the nitrifying bacteria.

The increase of DO concentration in the MBBR was achieved by increasing air flow rate at the bottom of the reactor. Biofilm sloughing would be enhanced with the increase of air flow rate. In our study, we wanted to know whether the activity of nitrifying bacteria was recovered by increasing DO concentration based on the configuration of the biofilm, and DO concentration should be increased at certain range to prevent badly breakage of the biofilm. At the same time, the aim of the experiment was to achieve partial nitrification at low DO concentration. Being exposed at high DO concentration for long time would shift partial nitrification to complete nitrification in the reactor, thus, period of increase DO concentration should not be long.

2.2.2 Sludge addition

Biomass in the MBBR was divided into two parts that were adhered on the carrier and suspended in the bulk liquid, respectively. Nitrification of the MBBR was due to the co-effect of the adhered and suspended nitrifying bacteria, although adhered nitrifying bacteria played a major role. Adding nitrifying sludge into the reactor might have some effects on nitrification recovery of the MBBR. Nitrifying sludge of 0.4 L (about 6000 mg VSS/L) was added into the MBBR at the bottom of reactor. The nitrifying sludge was cultured with SBR model using the same influent wastewater as the MBBR used, therefore, the sludge adapted the wastewater well and had good nitrifying ability. Performance of the MBBR after adding sludge is shown in Fig. 4.

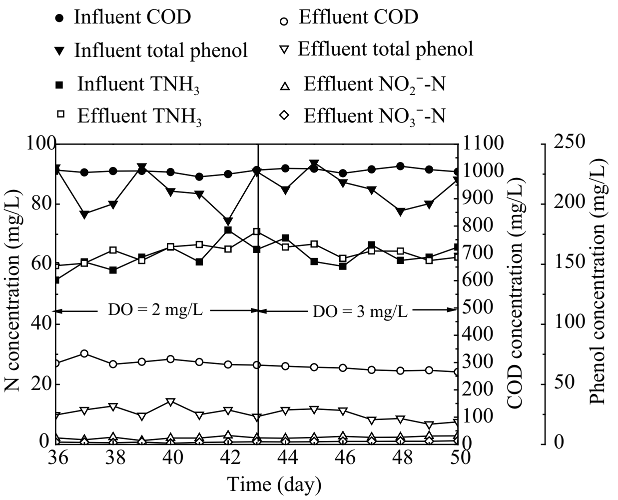

Fig. 3 Performance of the MBBR after increasing DO concentration in the bulk liquid.

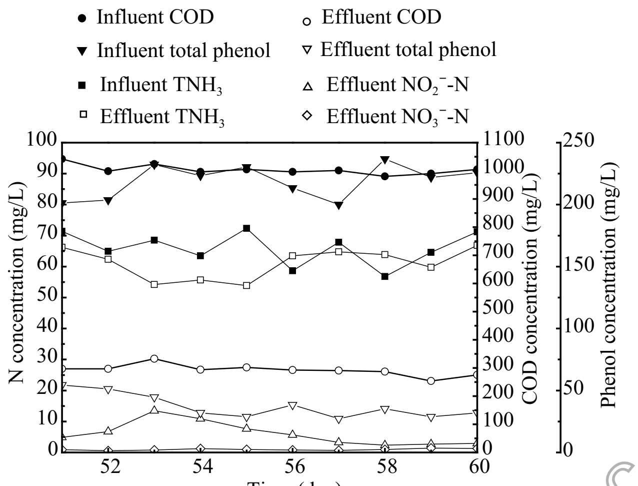

Fig. 4 Performance of the MBBR after adding sludge.

Nitrification of the MBBR was improved after adding nitrifying sludge and effluent NO_2^--N concentration reached 15 mg/L at day 53, but decreased gradually and almost ceased at day 58 (Fig. 4). The sludge adding into the MBBR was cultured in a batch manner which would promote nitrifying bacteria growing. That was the reason why nitrification of the system recovered partly after adding nitrifying sludge. COD and total phenol removal were improved little after adding the sludge, the results indicated that heterogeneous bacteria adhered to the carrier were enough to effectively degrade most pollutants in wastewater and organic pollutants in the effluent were difficult to be biodegraded using aerobic process (Lai et al., 2007).

Influent total phenol and TNH_3 concentration were in the normal range after adding sludge into the MBBR which indicated that nitrification of the reactor ceased gradually was not caused by the inhibition of NH_3-N or phenol. The nitrifying bacteria were difficult to adhere and grow on the biofilm that was already occupied by the active heterogeneous bacteria. Nitrification recovery would be due to the activity of the suspended nitrifying bacteria. Although the washed sludge was recycled once a day, the nitrifying bacteria would be suppressed in the deposition tank for lacking oxygen. Then, the recycled sludge would have less nitrifying ability than the sludge added into the MBBR at first. This was the reason why nitrification of the MBBR was weaker and weaker with the operation of MBBR.

2.2.3 Organic and TNH_3 load

Influent wastewater of the MBBR was diluted with tap water to decrease COD concentration to around 650 mg/L. COD and total phenol concentrations in influent and effluent are shown in Fig. 5a and TNH_3 removal of the MBBR is shown in Fig. 5b.

Influent COD concentration decreased to 650 mg/L and TNH_3 concentration varied between 42.1 and 58.9 mg/L from day 60 to day 72 (Fig. 5a, b). Effluent COD concentration was around 200 mg/L. Nitrification of the MBBR recovered gradually and the nitrification ratio reached 65% at day 72. Then, influent COD concentration increased to 1000 mg/L and effluent COD concentration increased a little to 250 mg/L. Nitrification ratio of the MBBR decreased to 25% at day 74 and recovered to 70% for another 4 days. Effluent NO_2^--N concentration reached 55 mg/L at day 89 and nitrification ratio of the MBBR achieved around 90%. NO_3^--N concentration was below 5 mg/L through out the start-up period of the MBBR.

Effluent COD concentration decreased only 50 mg/L when influent COD concentration decreased to 650 mg/L. Thus, available organic pollutants for heterogeneous bacteria in the reactor were decreased which approved indirectly that the activity of the heterogeneous bacteria was affected and the oxygen demanding could be reduced. Although nitrification inhibition of the MBBR was not caused by being short of oxygen (afore mentioned), shortage of available organic matter would make more heterogeneous bacteria shed from biofilm and nitrifying bacteria would gain more nourishment (such as inorganic carbon, phosphorus nutrition) from the bulk liquid because of biofilm attenuation.

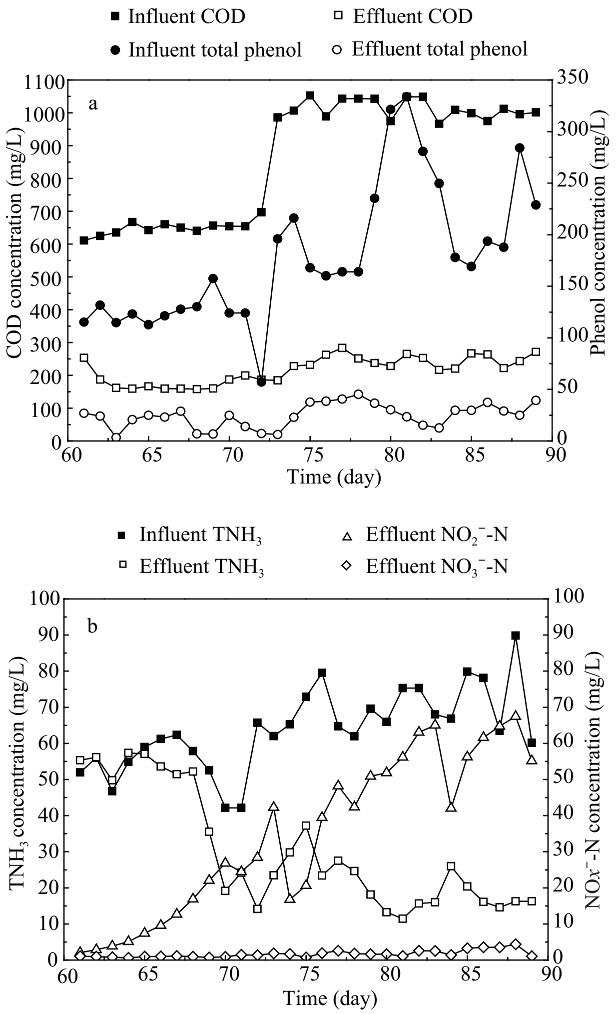

Fig. 5 COD and total phenol removal (a) and TNH_3 removal (b) of the MBBR after adding more dilute water into the influent.

At the same time, recovery of the nitrification would also be due to the decrease of influent total ammonia concentration. NH_3-N concentration and ratio of NH_3-N/TNH_3 during the experiment of nitrification recovery are shown in Fig. 6.

NH_3-N concentration was around 10 mg/L and occupied approximately 15% of TNH_3 during the period of DO concentration increase and sludge addition. Nitrification of the MBBR was not affected with FA concentration above 10 mg/L and ratio of NH_3-N/TNH_3 around 15% before nitrification inhibition and was not recovered although the NH_3-N concentration decreased to 10 mg/L after nitrification inhibition (Fig. 2). Effluent TNH_3 concentration decreased a little comparing with influent TNH_3 concentration because of nitrifying bacteria inhibition and NH_3-N concentration in the MBBR was around 10 mg/L which would inhibit activity of the nitrifying bacteria seriously. To add more dilute water into the influent of the MBBR would reduce TNH_3 concentration so as to reduce NH_3-N concentration in the MBBR and facilitate nitrification recovery. NH_3-N concentration decreased gradually and was below 1 mg/L after 20 days when nitrification of

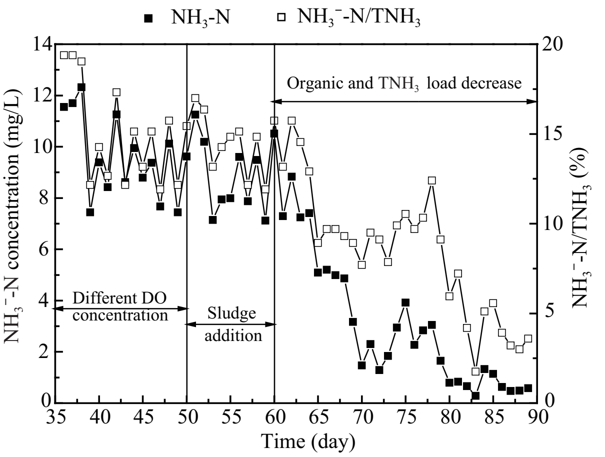

Fig. 6 NH_3-N concentration and ratio of NH_3-N/TNH_3 during the period of nitrification recovery.

the MBBR recovered. Ratio of NH_3-N/TNH_3 decreased to around 5%. Thus, NH_3-N concentration was an important control parameter in treating the coal gasification wastewater.

Nitrification of the MBBR was affected when influent COD concentration increased to 1000 mg/L and recovered within 4 days. Nitrification of the MBBR was recovered fast which indicated that the nitrifying bacteria were affected slightly by increasing organic and TNH_3 loading. Increase in organic load would enhance the activity of the heterogeneous bacteria and consume more oxygen which could reduce oxygen supply for nitrifying bacteria inside the biofilm, and increase of TNH_3 concentration might be another factor affecting nitrification. Whereas, shortage of oxygen supply for nitrifying bacteria was temporary because DO concentration in the bulk liquid was controlled at certain range and oxygen concentration in the inner part of the biofilm would turn to normal soon. NH_3-N concentration in the MBBR rose to 4 mg/L with the increase of TNH_3 concentration, but the negative impact on the activated nitrifying bacteria would be slight for the adaptive AOB.

3 Conclusions

Moving bed biofilm reactor was used to treat real coal gasification wastewater. Nitrification of the MBBR was almost inhibited because of acute increase of NH_3-N concentration during the start-up period. DO concentration increased from 2 to 3 mg/L had no effect on the recovery of the nitrification. The addition of nitrifying sludge into the MBBR had some effect on nitrification recovery, but the effect was temporary due to the nitrifying bacteria being washed out. To decrease influent COD and TNH_3 concentrations by adding more tap water was a feasible method for improving nitrification of the MBBR, and nitrification ratio could achieve 65% within 12 days. Nitrification of the MBBR was affected when influent COD concentration increased to 1000 mg/L after nitrification recovery, and recovered within 4 days.

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