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Effect of nitrate concentration on performance of pre-denitrification moving bed biofilm reactor system in treating coal gasification wastewater

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ABSTRACT

An anoxic moving bed biofilm reactor (ANMBBR) followed by an aerobic moving bed biofilm reactor (AEMBBR) operated as pre-denitrification system was used to investigate the effect of different influent NO_3^--N concentrations on performance of the pre-denitrification system in treating real coal gasification wastewater. Influent NO_3^--N concentration was controlled at 50, 100, 200, and 400 mg/L during the experiments. Evolution of COD and phenols in the effluent of the ANMBBR and AEMBBR, and performance of nitrification in the AEMBBR with different influent NO_3^--N concentrations were studied. Almost complete denitrification was achieved when influent NO_3^--N concentration was 100 mg/L. NO_3^--N and NO_2^--N accumulated in the effluent of the ANMBBR with influent NO_3^--N concentrations of 200 and 400 mg/L. NO_3^--N concentration of 400 mg/L facilitated high NH_4^+-N removal efficiency in the AEMBBR when influent NH_4^+-N concentration was around 165 mg/L. The ratio of $COD_{consumed}/NO_3^-N_{consumed}$ along with phenols removal of the ANMBBR under different influent NO_3^--N concentrations was discussed.

Keywords: Moving bed biofilm reactor; Coal gasification wastewater; Nitrate concentration; COD; Phenols

1. Introduction

Coal gasification wastewater is generated in the process of coal gas purification and coal chemical production [1]. Chemical composition of coal gasification wastewater is very complex and varies from one factory to another. Phenolic compounds are the main organic constituents, and other organics include polynuclear aromatic hydrocarbons and heterocyclic compounds [2,3]. The wastewater also contains high concentration of nitrogenous compounds, mainly in the form of ammonium and thiocyanate [4]. Pollutants in the wastewater must be well treated to prevent environmental destruction.

Autotrophic nitrification can convert ammonium into nitrite (NO_2^--N) or nitrate (NO_3^--N) by nitrifying bacteria. Then, heterotrophic denitrification process is recommended to convert NO_2^--N and NO_3^--N nitrogen to nitrogen gas using organic matter as

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electron donor. Coal gasification wastewater contains high concentration of organic pollutants which affect the activity of nitrifying bacteria because of heterotrophic bacteria proliferation [5]. The wastewater also contains plenty of toxic matters which can restrain nitrification [6]. Alkalinity is consumed in nitrification inducing decrease of pH value [7], and nitrification will be affected seriously if pH value decreases below 6 without supplying alkalinity. To decrease alkalinity consumption during nitrogen removal process, some new methods were introduced. Partial nitrification–anammox process uses ammonium as electron donor and NO_2^--N as electron acceptor, and the process can save around 50% alkalinity without adding organic carbon [8]. However, it is difficult to maintain stable ratio of ammonium and NO_2^--N for anammox process, and start-up of anammox process is difficult and consumes long time in full-scale wastewater treatment process [9].

A pre-denitrification system, combining process for carbon and nitrogen removal, is suitable for treating coal gasification wastewater because of its efficiency and cost-effectiveness [3,10]. Activated sludge process is often used in the pre-denitrification system, and sludge recycle must be used to maintain the concentration of suspended solids at certain range. Moreover, nitrification is affected easily in suspended system because nitrifying bacteria are washed out easily, especially under abnormal conditions [11]. The process of moving bed biofilm reactor (MBBR) was introduced about 30 years ago and it has since become popular in Europe [12]. Basic idea of the MBBR is to have a continued operation biofilm reactor with a high density of biomass and without backwashing or sludge return. Most biomass in the MBBR is in the form of biofilm adhering to carriers, thus, the MBBR can tolerate impact of toxic pollutants and has good effect on nitrification in treating various wastewater [13,14].

Generally, effluent of the aerobic reactor containing relative high NO_3^--N concentration was recycled into the anoxic reactor in the pre-denitrification system to achieve nitrogen removal, and characteristics of organic carbon in the wastewater for heterotrophic nitrate reduction has a major effect on denitrification process [15]. Although COD concentration of the coal gasification wastewater was relative high, most of organic pollutants containing in the coal gasification wastewater have complex configurations which might be difficult to be used as organic carbon source for denitrification process. Therefore, two lab-scale MBBR, one in anoxic condition (defined as ANMBBR) and the other in aerobic condition (defined as AEMBBR), were operated in pre-denitrification mode to treat real coal gasification wastewater. And KNO_3 was added into the influent of the two-stage MBBR to investigate denitrification potential of the organic pollutants in the coal gasification wastewater, and effect of different influent nitrate concentrations on performance of the two-stage MBBR was studied as well to give some advices in treating coal gasification wastewater using the full-scale pre-denitrification MBBR system.

2. Experimental

2.1. Description of the pre-denitrification MBBR system

Two plexiglass MBBR with effective volume of 4 L each were used in the experiments. The reactor had an internal diameter of 14 cm, a height of 30 cm, and effective depth was 26 cm. Polyethylene carriers used in the MBBR were cylindrical shape (cross inside) with 10 mm diameter. Density of the carrier was about 0.98 kg/m^3 and specific surface area was around 1,200 m^2/m^3. Carriers filling ratio was 50% in ANMBBR and 40% in AEMBBR. Movement of carriers in the ANMBBR was provided by a blender located at lower part of the reactor. Carriers at top of the ANMBBR did not move, which could facilitate N_2 escaping from the reactor and prevent oxygen penetrating into the reactor. Carriers in the AEMBBR were moved by pressurized air through diffusers at the bottom of the reactor. Screen was placed below the effluent ports of both reactors to prevent carriers from being washed, and top of the reactors was not covered.

2.2. Wastewater

The coal gasification wastewater used in the experiment was collected from a coal gasification factory located in the northeast of China. The wastewater had been pretreated by ammonia stripping and phenols solvent extraction processes to facilitate subsequent biological treatment. Basic compositions of the pretreated wastewater using in the experiment is shown in Table 1.

2.3. Experimental setup

Temperature of the ANMBBR was controlled at 30 ± 2 ℃ and the AEMBBR was operated at 20 ± 2 ℃ by temperature regulator. Dissolved oxygen (DO) concentration in the AEMBBR was controlled around 5 mg/L. Inoculum sludge of both MBBR was collected from the full-scale activated sludge process treating coal gasification wastewater in the coal gasification factory. Batch culture with the wastewater was done in both

Table 1
Basic compositions of the experimental wastewater

Parameter	Range	Mean
COD (mg/L)	1,712–2,340	2026
Phenols (mg/L)	412–542	477
NH_4^+-N (mg/L)	182–279	230.5
SCN^- (mg/L)	99–176	137.5
NO_3^--N (mg/L)	<1	-
NO_2^--N (mg/L)	<1	-

of the MBBR before the experiments to facilitate biofilm formation on the carriers. The reactors were operated for approximately 160 days, and the concentration of NO_3^--N added into the influent was 50, 100, 200, and 400 mg/L, respectively. HRT of the ANMBBR and AEMBBR was controlled at around 28 h throughout the experiments.

2.4. Analysis methods

The samples were taken from effluent of the ANMBBR and AEMBBR every two days and were analyzed immediately after filtered through 0.45 μm filter paper. Soluble COD, phenols, NH_4^+-N, NO_3^--N, and NO_2^--N were measured in accordance with standard methods [16]. SCN^- was measured by ferric thiocyanate colorimetric method. DO concentration was measured using YSI O_2-electrode.

3. Results and discussion

3.1. COD and phenols removal

Influent COD concentration of the ANMBBR was controlled around 1,500 mg/L and phenols concentration varied from 300 to 342 mg/L during the experiments. COD and phenols concentrations in the influent of the ANMBBR and effluent of ANMBBR and AEMBBR at different NO_3^--N concentrations are illustrated in Fig. 1 and Fig. 2.

Both of the reactors were operated continuously after batch culture without adding KNO_3 within the first 15 days, and effluent COD and phenols concentrations of the AEMBBR were 324.6 and 36.1 mg/L on average, respectively. The ANMBBR had some effect on organic pollutants removal, and decrease of COD and phenols concentrations were limited to 248.15 and 24.08 mg/L on average. Configuration of the ANMBBR would prevent oxygen from penetrating deeper into the reactor, DO concentration decreased stepwise from surface of the reactor. Denitrifying bacteria were capable of using oxygen as an electron

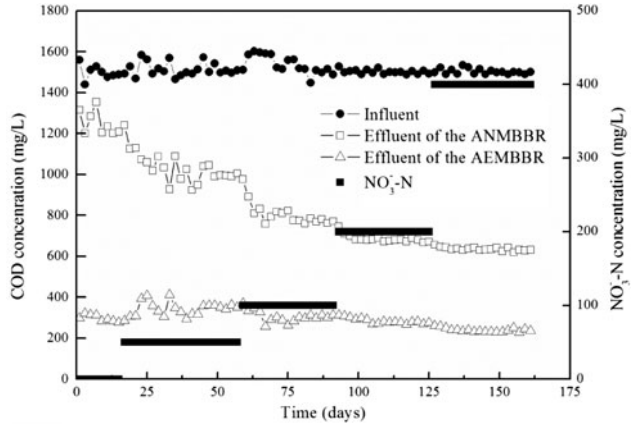

Fig. 1. Evolution of COD in the influent of the ANMBBR and effluent of the ANMBBR and AEMBBR at different NO_3^--N concentrations.

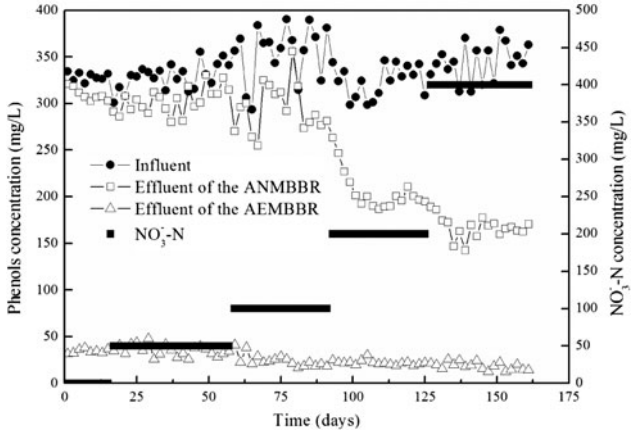

Fig. 2. Evolution of phenols in the influent of ANMBBR and effluent of ANMBBR and AEMBBR at different NO_3^--N concentrations.

acceptor for organic matter removal and oxygen had a negative effect on denitrification [17]. Biological oxidization might happen in the upper reactor surface where most of carriers were floating, and denitrification in the deeper reactor sector would not be affected by oxygen.

Then, KNO_3 was added into the influent of the ANMBBR to make NO_3^--N concentration to be 50, 100, 200, and 400 mg/L, and corresponding influent COD/NO_3^--N ratio was 30:1, 15:1, 7.5:1, and 3.75:1, respectively. Effluent COD concentration of the ANMBBR decreased with the enhancement of influent NO_3^--N concentration. Downtrend of COD concentration in the effluent of the ANMBBR was obvious when NO_3^--N concentration increased from 50 to 100 mg/L, however, COD concentration decreased

around 40 mg/L merely when influent NO_3^--N concentration increased from 200 to 400 mg/L. Effluent COD concentrations of the ANMBBR and AEMBBR were 681.9 and 282.8 mg/L on average when influent NO_3^--N concentration was controlled at around 400 mg/L. Phenols concentration in the effluent of the ANMBBR decreased with the increase of influent NO_3^--N concentration, and effluent phenols concentration of the AEMBBR was around 20 mg/L when influent NO_3^--N concentration was controlled at around 400 mg/L. The relationship between COD and phenols removal and influent NO_3^--N concentration would be stated in section 3.3.

3.2. SCN^- and NH_4^+-N removal

Influent SCN^- concentration changed from 74.5 to 114.5 mg/L and NH_4^+-N concentration varied from 147.7 to 197.6 mg/L during the experiments. SCN^- and NH_4^+-N concentration in the influent of the ANMBBR and effluent of the ANMBBR and AEMBBR at different NO_3^--N concentrations are illustrated in Fig. 3 and Fig. 4.

For the first 15 days, NH_4^+-N and SCN^- were not removed in the ANMBBR, which might be due to negative effect of high organic matter concentration on the activity of autotrophic bacteria that biodegraded NH_4^+-N and SCN^- [18,19]. Influent NH_4^+-N concentration of the system was around 165 mg/L and NH_4^+-N removal efficiency was poor in the AEMBBR.

Then, 50 mg/L of NO_3^--N was adding into influent of the ANMBBR and 1 g $NaHCO_3$ per liter wastewater was added into effluent of the ANMBBR as additional alkalinity so as to promote nitrification performance in the AEMBBR. High NH_4^+-N removal

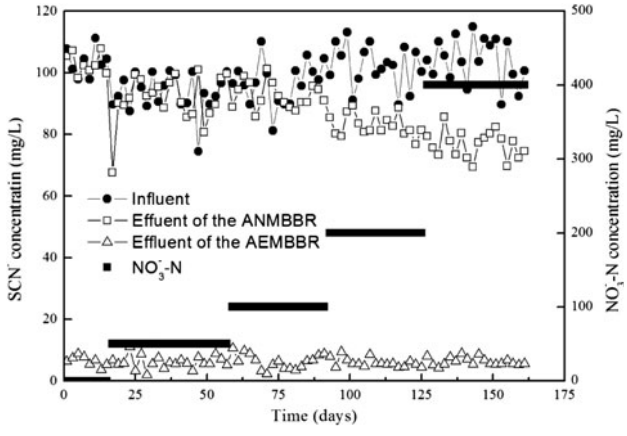

Fig. 3. Evolution of SCN^- in the influent of the ANMBBR and effluent of the ANMBBR and AEMBBR at different NO_3^--N concentrations.

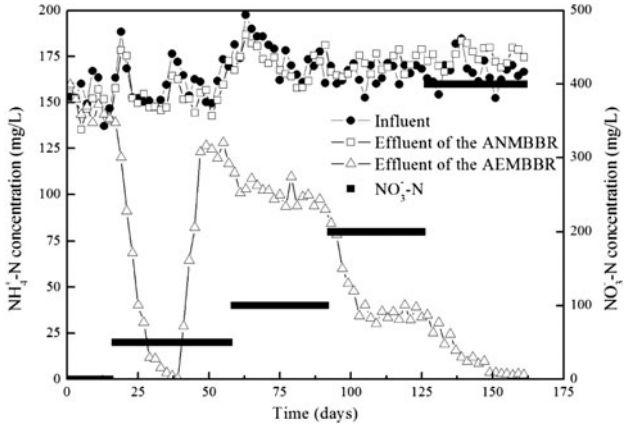

Fig. 4. Evolution of NH_4^+-N in the influent of the ANMBBR and effluent of the ANMBBR and AEMBBR at different NO_3^--N concentrations.

efficiency could be achieved in the AEMBBR within 10 days after addition of $NaHCO_3$. After good performance of nitrification was obtained in the AEMBBR, the reactor was operated without adding $NaHCO_3$, and influent NO_3^--N concentration was still 50 mg/L. NH_4^+-N concentration in effluent of the AEMBBR increased to around 125 mg/L within several days without adding $NaHCO_3$. The ANMBBR had little effect on SCN^- removal after addition of 50 mg/L NO_3^--N and effluent SCN^- concentration of the AEMBBR was below 10 mg/L.

Poor performance of the AEMBBR on NH_4^+-N removal for the first 15 days might be due to two reasons mainly, one was the activity of nitrifying bacteria being inhibited by toxic compounds in the wastewater in the beginning, and the other was due to low alkalinity concentration in the influent coal gasification wastewater (around 150 mg CaCO3/L) which could be supported by NH_4^+-N removal improvement with the addition of $NaHCO_3$ and decrease without adding $NaHCO_3$. Effluent NH_4^+-N concentration of the AEMBBR decreased gradually with the enhancement of influent NO_3^--N concentration, and NH_4^+-N concentration in the effluent of the AEMBBR was below 10 mg/L when influent NO_3^--N concentration was 400 mg/L without adding $NaHCO_3$. The results indicated that denitrification in the ANMBBR would promote nitrification of the AEMBBR, and the exact relation between NO_3^--N consumption and nitrification in the AEMBBR would be shown in section 3.3.

SCN^- decreased in the ANMBBR from the 90th day, and effluent SCN^- concentration of the AEMBBR was always below 10 mg/L. Literature did not support SCN^- biodegradation under anoxic environment [20]. Degradation of SCN^- was observed in the

ANMBBR when influent NO_3^--N concentration increased from 100 to 200 mg/L, which caused a little increase of effluent NH_4^+-N concentration. SCN^- removal would be due to effect of biological oxidation in the upper part of the ANMBBR by thiocyanate utilizing autotrophic micro-organisms. Effluent SCN^- concentration did not decrease when influent NO_3^--N concentration was below 100 mg/L, which might be due to the inhibiting effect of high concentration organic matters on autotrophic micro-organisms.

3.3. Evolution of NO_2^--N and NO_3^--N

NO_2^--N and NO_3^--N concentrations in the effluent of the ANMBBR and AEMBBR had direct relationship with the influent NO_3^--N concentration, and effluent NO_3^--N and NO_2^--N concentrations of the ANMBBR and AEMBBR are shown in Fig. 5.

NO_3^--N concentration in the effluent of the AEMBBR increased to around 190 mg/L gradually after 15 days with the addition of $NaHCO_3$, and decreased to around 60 mg/L obviously after the cease of $NaHCO_3$ addition. Then, effluent NO_3^--N concentration of the AEMBBR increased higher than 100 mg/L with influent NO_3^--N concentration of the ANMBBR being around 100 mg/L, and the concentrations of NO_3^--N and NO_2^--N in the effluent of the ANMBBR was below 10 mg/L which indicated that almost denitrification was achieved in the ANMBBR. After that, NO_2^--N accumulation could be observed in the effluent of the ANMBBR when the influent NO_3^--N concentration of the ANMBBR was higher than 200 mg/L, which indicated that the influent NO_3^--N concentration was excess when influent COD concentration was controlled at around 1,500 mg/L.

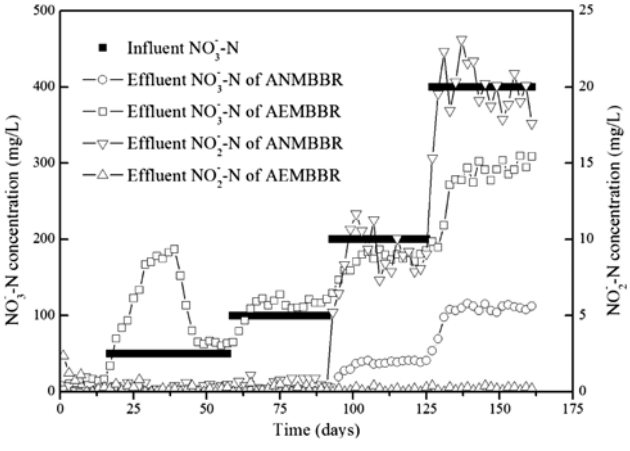

Fig. 5. Evolution of NO_2^--N and NO_3^--N in the effluent of the ANMBBR and AEMBBR at different NO_3^--N concentrations.

Almost complete denitrification was achieved in the ANMBBR when the influent NO_3^--N concentration was below 100 mg/L, and the corresponding ratio of $COD_{influent}/NO_3^-N_{influent}$ was 15. Incomplete denitrification appeared when influent NO_3^--N concentration increased to 200 mg/L, and $COD_{influent}/NO_3^-N_{influent}$ decreased to 7.5. It appeared that ratio of $COD_{influent}/NO_3^-N_{influent}$ should be higher than 7.5 to ensure complete denitrification in treating the wastewater. However, effluent COD concentration of the ANMBBR was around 680 mg/L with influent NO_3^--N concentration of 200 mg/L, which indicated that incomplete denitrification was not induced by organic matters exhaustion. Total COD in the wastewater could be separated into readily and slowly biodegradable substrate, and a lower denitrification rate was obtained in the presence of slowly biodegradable substrate [21]. Incomplete denitrification of the ANMBBR might be due to slow denitrification rate using slowly biodegradable substrate. Ratio of consumed COD and NO_3^--N concentration would show better utilization steps of organic matters in treating the complicated wastewater under denitrification process. Therefore, the ratio of $COD_{consumed}/NO_3^-N_{consumed}$ was varied under different influent NO_3^--N concentrations, which is shown in Fig. 6. At the same time, phenols reduction in the ANMBBR is illustrated in Fig. 6.

Consumed COD of the ANMBBR in Fig. 6 deducted the COD removed by oxidation. The value of deductible COD adopted 248.15 mg/L which was the average of consumed COD for the first 15 days. Operation conditions of the ANMBBR were similar during the experiments, which indicated that effect of biological oxidation on COD removal was almost consistent. Thus, we assumed that concentration of COD consumed by biological oxidation was around 250 mg/L with the addition of NO_3^--N. Many factors affect the utilization of organic matters in denitrification, such as, kinds of organic matter [22], configuration and operation conditions of the reactor. Operating conditions of the reactor did not change during the experiments, and complexity components in the coal gasification wastewater could be the main reason for different ratios of $COD_{consumed}/NO_3^-N_{consumed}$ with different influent NO_3^--N concentrations.

Denitrification rate was different with different types of organic carbon source. Xu [23] reported that the volatile fatty acid (VFA) could be an effective alternative organic carbon sources to methanol and acetic acid which can be directly inserted into the metabolic process of denitrification bacteria. Some VFA contained in the coal gasification wastewater is used

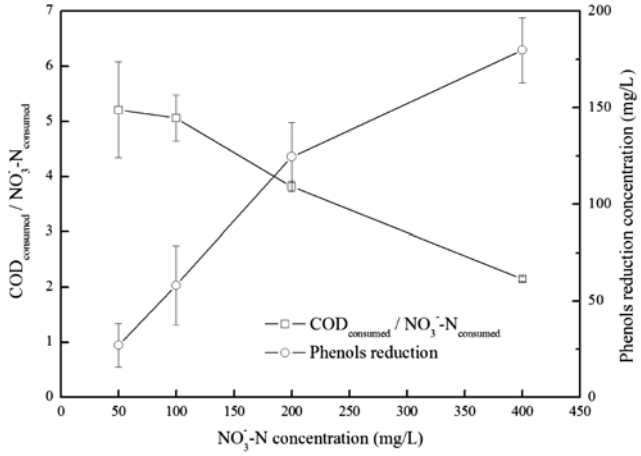

Fig. 6. Reduction of phenols and ratio of $COD_{consumed}$/NO_3^--$N_{consumed}$ under different influent NO_3^--N concentrations in the ANMBBR.

in the experiment (data were not shown). Phenolic compounds in the wastewater could be used as organic carbon source in denitrification as well, and denitrification rates were varied using different kinds of phenolic compounds [24]. The organic carbon used in denitrification would be VFA matters with influent NO_3^--N concentration of 50 mg/L, because phenols concentration decreased around 25 mg/L merely which was due to biological oxidation at the top of the ANMBBR (afore mentioned). With the increase of influent NO_3^--N concentration, the readily used VFA organic carbon was exhausted, and phenolic compounds turned to main organic carbon for denitrification. Phenols concentration decreased about 25 mg/L (deduct biological oxidation removal) with 100 mg/L NO_3^--N, which indicated that VFA and phenolic compounds were used for denitrification. Removal of phenols increased when influent NO_3^--N concentration increased to 200 mg/L. Sarfaraz [25] reported that 1 g NO_3^--N was consumed per 3.4 g phenol COD removal using sequencing batch reactor in anoxic condition. Average COD concentration decreased 108.2 mg/L when influent NO_3^--N concentration increased from 100 to 200 mg/L, and NO_3^--N reduction concentration increased 60.9 mg/L (9.3 mg/L NO_3^--N turned to NO_2^--N) on average. Thus, both of phenol and other complicated phenols were used for denitrification in this stage because of the low ratio of $COD_{consumed}/NO_3^-$-$N_{consumed}$, which would induce incomplete denitrification for the low denitrification rate using complicated phenols as organic carbon source. Moreover, decrease of effluent COD concentration in the AEMBBR with influent NO_3^--N concentration of 200 mg/L could also indicate that transformation of complex organic compound happened. Average phenols concentration decreased 55.2 mg/L when influent NO_3^--N concentration increased from 200 to 400 mg/L. The results indicated that transformation of complex compounds could be enhanced by increasing NO_3^--N concentration during denitrification, even in incomplete denitrification condition.

Effluent COD concentration of the ANMBBR decreased a little when influent NO_3^--N concentration increased from 200 to 400 mg/L, which indicated that the ANMBBR achieved its upper-limit capacity for denitrification at the operation conditions. Utilization of organic matters in the coal gasification wastewater using the ANMBBR could not be the real denitrification potential of the wastewater which was usually measured by nitrate utilization rate (NUR) method [26], because phenols removal of the ANMBBR increased when influent NO_3^--N concentration increased to 400 mg/L. Thus, denitrification capacity of the ANMBBR did not achieve maximum in treating the wastewater, and extension of the HRT would facilitate utilization of slowly biodegradable substrate to enhance denitrification capacity of the ANMBBR. However, it would be uneconomical to prolong the HRT for achieving maximum denitrification capacity of the denitrification process in full-scale wastewater treatment system. To ensure complete denitrification in the reactor would be an optimal choice for nitrogen and COD removal, and influent NO_3-N concentration of 100 mg/L was appropriate in the experiments.

Nitrification ratio of the AEMBBR increased with the improvement of influent NO_3^--N concentration, and complete nitrification achieved in the AEMBBR when addition of NO_3^--N concentration increased to 400 mg/L. About 7.14 g alkalinity was needed to convert 1 g NH_4^+-N to NO_3^--N through nitrification, and 3.57 g alkalinity was generated from denitrification of 1 g NO_3^--N. About 270.6 mg/L NO_3^--N was consumed completely and 20 mg/L NO_3^--N converted incompletely to NO_2^--N on average with influent NO_3^--N concentration of 400 mg/L. Alkalinity generated in denitrification was 970 mg/L theoretically. The generated alkalinity in denitrification combining with the containing alkalinity in the wastewater (147.5 mg $CaCO_3$/L on average) could ensure complete nitrification in the AEMBBR when influent NH_4^+-N concentration was around 165 mg/L.

4. Conclusion

Complete denitrification was achieved in the ANMBBR when influent NO_3^--N concentration was 100 mg/L, and NO_3^--N and NO_2^--N accumulated in

the effluent of the ANMBBR when the ratio of $COD_{influent}/NO_3^-\text{-}N_{influent}$ decreased to 7.5 by increasing influent NO_3^--N concentration. Increase of influent NO_3^--N concentration to 400 mg/L generated enough alkalinity in the effluent of the ANMBBR to achieve almost complete nitrification in the AEMBBR when influent NH_4^+-N concentration was around 165 mg/L. Increase of NO_3^--N concentration could facilitate biodegradation of refractory compounds in the wastewater, and incomplete denitrification of the ANMBBR was observed with influent NO_3^--N concentration of 200 mg/L and 400 mg/L. Influent NO_3-N concentration of 100 mg/L was appropriate in the experiments.

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