Nitritation and denitritation of ammonium-rich wastewater using fluidized-bed biofilm reactors

Sukru Aslan [a,*], Mohamed Dahab [b]

[a] *Cumhuriyet University, Department of Environmental Engineering, 58140 Sivas, Turkey*
[b] *Department of Civil Engineering, University of Nebraska-Lincoln, W348 Nebraska Hall, Lincoln, NE 68588, USA*

Abstract

Fluidized-bed biofilm nitritation and denitritation reactors (FBBNR and FBBDR) were operated to eliminate the high concentrations of nitrogen by nitritation and denitritation process. The dissolved oxygen (DO) concentration was varied from 1.5 to $2.5 \, g/m^3$ at the top of the reactor throughout the experiment. NH_4-N conversion and NO_2-N accumulation in the nitritation reactor effluent was over 90 and 65%, respectively. The average NH_4-N removal efficiency was 99.2 and 90.1% at the NLR of 0.9 and $1.2 \, kg \, NH_4$-N/m^3 day, respectively. Increasing the NLR from 1.1 to $1.2 \, kg \, NH_4$-N/m^3 day decreased the NH_4-N elimination approximately two-fold while NH_4-N conversion to NO_2-N differences were negligible. The NO_2-N/NO_x-N ratios corresponded to 0.74, 0.73, 0.72, and 0.69, respectively, indicating the occurrence of partial nitrification. An average free ammonia concentration in the FBBNR was high enough to inhibit nitrite oxidizers selectively, and it seems to be a determining factor for NO_2-N accumulation in the process.

In the FBBDR, the NO_x-N (NO_2-N + NO_3-N) concentrations supplied were between 227 and $330 \, mg \, N/l$ (NLR was between 0.08 and $0.4 \, kg/m^3$ day) and the influent flow was increased as long as the total nitrogen removal was close to 90%. The NO_2-N and NO_3-N concentrations in the effluent were 3.0 and $0.9 \, mg/l$ at $0.08 \, kg/m^3$ day loading rate. About 98% removal of NO_x-N was achieved at the lowest NLR in the FBBDR. The FBBDR exhibited high nitrogen removal up to the NLR of $0.25 \, kg/m^3$ day. The NO_x-N effluent concentration never exceeded $15 \, mg/l$. The total nitrogen removal efficiency in the FBBRs was higher than 93% at $21 \pm 1 \, °C$.

1. Introduction

Biological systems for nitrogen removal can be improved by separate treatment of highly concentrated waters, such as supernatant produced during dewatering of digested sludge, effluents from the fertilizer industry and fish canning industry, manure systems and landfill leachates. In wastewater treatment plants (WWTP) with anaerobic sludge digestion, a recirculated supernatant contributes to 15–20% of the influent nitrogen load. Therefore, it is proposed to separately treat the supernatant rather than return it to the WWTP inlet for treatment as a part of the main flow [1–3].

Biological nitrification and denitrification is the most studied process for nitrogen removal from wastewaters. Nitrification is an autotrophic aerobic process that converts ammonium into nitrate. Ammonium is oxidized into nitrite during the first step by ammonium-oxidizing bacteria (AOB), and nitrite is oxidized into nitrate during the second step by nitrite oxidizing bacteria (NOB). In the denitrification process, nitrate is anoxically converted into nitrite, then into nitrogen gas [4,5].

Operational costs of the biological nitrogen removal (BNR) process are, to a great extent, related to the oxygen and organic matter requirements for nitrification and denitrification, respectively. Several studies have been preformed in order to optimize BNR and new methods have been developed such as a partial nitrification/complete denitrification (nitritation/denitritation) [6–13]. This process is based on the facts that, since nitrite and nitrate are intermediary compounds in nitrification and denitrification, the partial nitrification to nitrite

* Corresponding author. Tel.: +90 346 219 10 10x1296;
fax: +90 346 219 11 77.
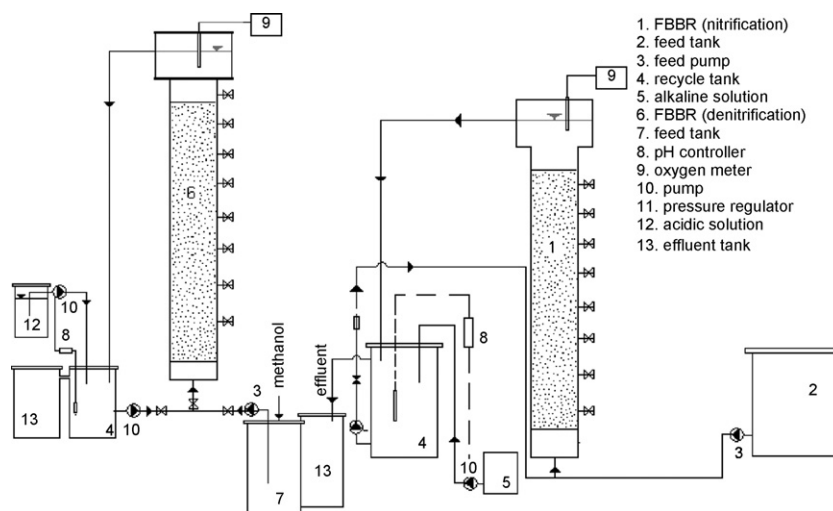*E-mail address:* saslan@cumhuriyet.edu.tr (S. Aslan).

Fig. 1. Schematic diagram of FBBRs.

and the denitrification from accumulated nitrite, instead from nitrate, would be feasible [7]. In contrast to the traditional separate BNR process, the nitritation/denitritation process results in savings amounting to 25% of the oxygen supply for nitritation while carbon requirements for denitritation are approximately 40% lower than for denitrification [10,14–16] and denitrification rates with nitrite are 1.5–2 times greater than with nitrate [17].

In order to perform nitritation and denitritation process, nitrite oxidation should be controlled without affecting the AOB and NOB must be adapted to high concentrations of nitrite. Anthonisen et al. [18] reported that unionized ammonia inhibited the nitrification reaction. Recent studies also suggested that free hydroxylamine, an intermediate of ammonia oxidation, might be a key factor that caused inhibition to nitrite oxidation [19]. Other factors that could be used for control of nitrite oxidation were DO [16,20,21], pH [14,20,22], temperature [20,23–25], sludge retention time [26] and so on.

Fluidized-bed biofilm reactor (FBBR) system was operated for elimination of nitrogen in the wastewater [27–33]. However, there are few studies reported in the literature on the nitritation and denitritation of wastewater containing high levels of nitrogen compounds using the FBBR. The results from laboratory and field pilot scale studies have consistently illustrated the technical advantage of the fluidized bed over most other suspended and attached growth biological systems. Typically, the efficiency of the FBBR can be as much as 10 times that of the activated sludge system and it typically occupies as low as about 10% of the space required by stirred tank reactors of similar capacities [34].

This study aimed at investigating the feasibility of nitrogen removal from high strength wastewater by nitritation and denitritation in the FBB nitritation and denitritation reactors (FBBNR and FBBDR). In this study the operational parameter of nitrogen loading rate (NLR) was studied for each process to determine the maximum NO_2-N accumulation and adaptation in the nitritation step and the removal of NO_x-N (NO_2-N + NO_3-N) in the denitritation step.

2. Materials and methods

The reactor system, shown in Fig. 1, consisted of an up flow reactor (76 mm diameter and 1.5 m height). The DO probes were placed at the top of reactors and connected to the DO meters. The reactors were loaded with 1.0 kg uniform sand which served as a biofilm carrier. Table 1 gives the sand properties. The DO concentration varied from 1.5 to 2.5 mg/l at the top of the FBBNR and 4 to 6 mg/l in the recycling tank of the nitritation column. The DO was provided during recycling of the mixed liquor and external air was supplied two times for 1 h when the DO concentration dropped lower than 0.5 mg/l to maintain the DO level of about 1.8 mg/l in the FBBNR.

The DO concentration at the top of the denitritation reactor was maintained lower than 0.5 mg/l by adding Na_2SO_3 at the beginning of the operation. The recycle tank of the FBBDR was completely closed to keep the DO concentration lower than 0.5 mg/l. Since biological denitritation relies on an available carbon source (typically measured as a COD/N) is one of the most important parameter in the process. The FBBDR was operated at a high COD/N ratio of 5 by considering effluent total NO_x-N concentration of the FBBNR using following equation;

$$\frac{COD}{N} = \frac{COD\,(mg/l)}{effluent\,(NO_2\text{-}N + NO_3\text{-}N)\,(mg/l)} \quad (1)$$

2.1. Synthetic wastewaters

Synthetic wastewater was prepared using demineralized water, a nutrient mix and other chemicals for pH buffering and to provide the trace metals and vitamins necessary to maintain

Table 1
Sand properties

Mean diameter (mm)	0.83
Specific density (g/cm^3)	2.65
Void ratio	0.42
Dry density (g/cm^3)	1.54
Specific area (m^2/m^3)	4200

Table 2
Composition of synthetic wastewaters

Chemical	Concentration (mg/l)
Nitritation	
Na_2EDTA	4.83
$CuSO_4$	0.0046
Thiamine	0.1
$ZnSO_4 \cdot 7H_2O$	0.023
$CoCl_2 \cdot 6H_2O$	0.0119
$Na_2MoO_4 \cdot 2H_2O$	0.066
D-Biotin	0.0005
Vitamin B12	0.0005
$MgSO_4 \cdot 7H_2O$	36.97
$NaHCO_3$	226
$CaCl_2 \cdot 2H_2O$	36.74
H_3BO_3	1.0
$FeCl_3 \cdot 6H_2O$	0.316
K_2HPO_4	1960
KH_2PO_4	1920
Denitritation	
KH_2PO_4	4000
K_2HPO_4	5300
$NaHCO_3$	32.5
$FeSO_4 \cdot 7H_2O$	0.816
$NaMO_4$	0.237
$MnSO_4 \cdot 7H_2O$	0.157
$CoCl_2 \cdot 6H_2O$	0.526
Na_2SO_3	20

the bacterial growth. The synthetic wastewater composition for the FBBNR and FBBDR are presented in Table 2.

2.2. Reactor operation

The reactors were seeded with 2 l biomass inoculums from the nitrification-denitrification basin at the Theresa Street Wastewater Treatment Plant in Lincoln, NE, USA. The initial concentrations of the suspended and volatile suspended solids (SS and VSS) in the reactors after seeding were about 1.95 and 1.7 kg/m^3, respectively. The inoculations lasted approximately 2 months for microbial growth with daily replenishment of NH_4-N and NO_2-N for the FBBNR and FBBDR, respectively.

The influent wastewater was pumped continuously to the reactors using peristaltic pumps, and discharged from the top of the reactors to the recycling tanks. Effluent was allowed to overflow from the top of the recycling tanks. The nitrified effluent was combined with methanol, phosphate and other nutrients to eliminate the possibility of nutrient deficiency and fed to the FBBDR.

The reactors were operated at room temperature ($21 \pm 1\,^\circ$C). The pH was measured continuously using pH probes connected to pH controllers. Automatic pumping of 10% NaOH and HCl solutions into the recycling tanks of the FBBNR and FBBDR, respectively, maintained the pH at 7.5 ± 0.1.

Variation of the NLR was controlled by changing the influent NH_4-N concentration of the FBBNR. The experimental increase of the influent nitrogen load into the FBBNR was planned with the goal not to overload the system and damage the denitritation process. The decision to increase the influent nitrogen load was

based on precise analyses of the concentrations of NH_4-N and NO_x-N in the effluent of the FBBNR. During the start-up period, the reactor was operated at a low NLR of about 0.5 kg NH_4-N/m^3 day. The DO concentrations in the feeding tanks of the reactors were kept about zero by adding Na_2SO_3.

The total NO_x-N load was raised by increasing the influent flow rate step-by-step to obtain optimum nitrogen load to the FBBDR.

2.3. Analytical methods

All analyses were performed on grab samples taken from the reactors influents, effluents, and completed in accordance with Standard Methods (1998) [35]. NH_4-N concentration was determined by an ammonia-specific electrode, which was calibrated with known concentrations before each use. Samples were withdrawn daily from the reactors and filtered using 0.45 μm, white, 47 mm radius filters. All samples were tested for NO_3-N and NO_2-N concentrations using a Hach DR 2800 spectrophotometer with Hach Chemicals Nitriver5 and Nitriver2, respectively, and a Dionex DX-500 LC 20 Ion Chromatography (IC) with a CD 20 suppressed conductivity detector and a column model Dionex Ionpac AS9-SC 4 mm × 250 mm. The eluent used for this IC contained 1.7 mM $NaHCO_3$ and 1.8 mN Na_2CO_3. The pumping flow rate of the eluent was 2 ml/min. The DO was measured by the DO meters (YSI 5100). Biomass concentrations of samples were determined as SS and volatile solids (VSS) [35].

3. Results and discussion

Stable nitritation in the first FBBR can be combined with denitritation in the second FBBR to ensure total nitrogen removal throughout the biological process. The NO_x-N produced and the remaining NH_4-N, including the nitritation biomass, from the FBBNR (about 0.65 kg/m^3) was used as an influent in the subsequent FBBDR after adding methanol and other nutrients.

3.1. Start-up period

The FBBRs were start-up at a low NLR. In about 2 months of operations, the biomass concentration in the FBBNR and FBBDR reached to about 5.5 ± 1.7 and 5.2 ± 1.0 kg VSS/m^3, respectively, and the bed of the reactors expanded to a height of 1.0 m. Biomass measurement was carried out on the bioparticles which were withdrawn in the FBBRs and in the effluent water. The withdrawn bioparticles were mixed vigorously in a rapid mixer to detach the biofilm and the clean sand was returned to the reactors [31].

3.2. NH_4-N loading affects on nitritation

The nitritation process realization was observed by NH_4-N, NO_2-N and NO_3-N measurements in the effluent of the FBBNR. Stable operation was achieved throughout most of the start-up period with complete NH_4-N removal by regulating recycled water flow, sand fluidization, and biomass development onto the sand.

During the first 2 weeks of the start-up period, NH_4-N removal efficiency was lower than 60%, and then increased to about 80% at the NLR of 0.5 kg/m^3 day. Afterwards NH_4-N removal increased reaching a stable level above 90% in approximately 1 month.

At first the decrease of NH_4-N was almost completed by a significant accumulation of NO_3-N and a small fraction of NO_2-N. A higher concentration of NO_2-N than NO_3-N was kept up for the duration of the experimental study.

After nitritation was established in the FBBNR, the NLR was increased slowly by changing the influent NH_4-N concentration in the feeding wastewater. The feed flow rate was kept at 18 l/day and the NH_4-N concentration was slowly increased from 250 to 350 mg/l, corresponding to NLR increase from 0.9 to 1.2 kg/m^3 day. This change resulted in incomplete oxidation of ammonium and the effluent NH_4-N and NO_2-N concentration increased. The purpose of this exercise was to determine the maximum NLR, the resultant NH_4-N removal efficiency, and the maximum NO_2-N accumulation potential.

The average NH_4-N removal efficiency was 99.2 and 90.1% (standard deviation = 1.6) at the NLR of 0.9 and 1.2 kg NH_4-N/m^3 day, respectively (Fig. 2). Nearly complete NH_4-N removal was obtained at 0.9 kg/m^3 day and about 94% NH_4-N removal was achieved at 1.1 kg/m^3 day of NLR with an average effluent NH_4-N concentration of lower than 20 mg/l. Increasing the NLR from 1.1 to 1.2 kg NH_4-N/m^3 day, increased the effluent concentration of NH_4-N approximately by two-fold while the differences in NO_2-N accumulation were negligible. As shown in Fig. 3, it was evident that the system was unable to provide an ammonium removal rate of more than 1.1 kg NH_4-N/m^3 day. Although nitritation remained very efficient at the NLR up to 1.2 kg/m^3 day, NH_4-N effluent concentration exceeded 20 mg/l which represented 90% removal efficiency.

The average NH_4-N removal at various NLRs is presented in Fig. 4. At the low NLR of 0.9 kg N/m^3 day, the removal rate was about 0.68 kg/m^3 day and increased to 0.86 kg/m^3 day when the NLR was increased to 1.2 kg/m^3 day.

During the start-up period, NO_2-N accumulation was (57 mg/l) when the NLR was lower than 0.9 kg/m^3 day, because the NH_4-N loading affects the NO_2-N accumulation.

Fig. 5 represents the NO_2-N and NO_3-N concentrations in the effluent and NH_4-N removal efficiency at various NLR. NH_4-N conversion and NO_2-N accumulation in the FBBNR effluent
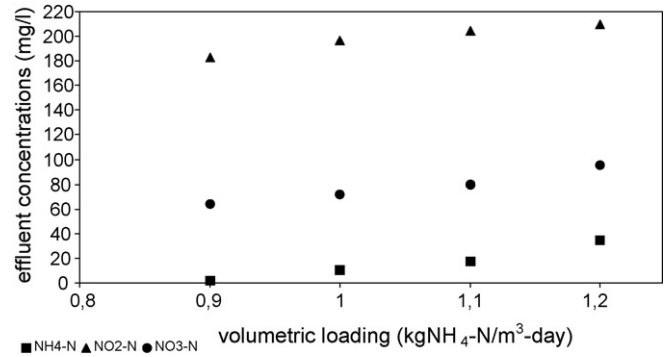

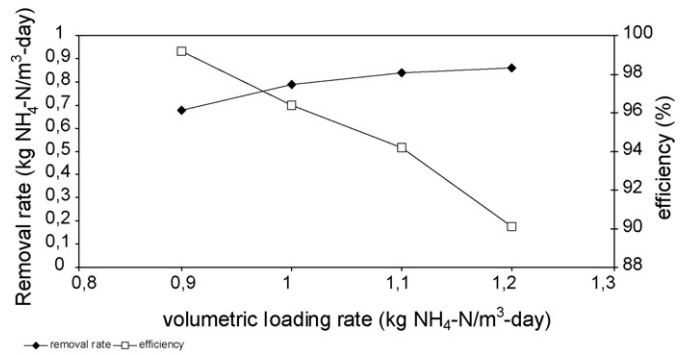Fig. 3. Effect of applied NLR on the NO_2-N and NO_3-N accumulations in the nitritation FBBR effluents.

Fig. 4. NH_4-N removal rate at various NLR in the FBB nitritation reactor.

was over 90 and 66%, respectively. The formation of NO_2-N was suppressed and more than 65% of the oxidized compounds of nitrogen at various NLR were represented by NO_2-N. The remarkable performance of NO_2-N accumulation was achieved at 0.9 kg/m^3 day of NLR while NH_4-N removal efficiency was about 99% and NO_2-N and NO_3-N concentrations in the effluent were 183 and 64 mg/l, respectively.

NH_4-N could be oxidized into the NO_2-N and NO_3-N under aerobic conditions. NO_2-N was detected as the main nitrification product in the effluent of the FBBNR through the experimental study. The effluent NO_2-N concentration averaged at 183, 197, 204, and 210 mg/l at the NLR of 0.9, 1.0, 1.1, and 1.2 kg/m^3 day,
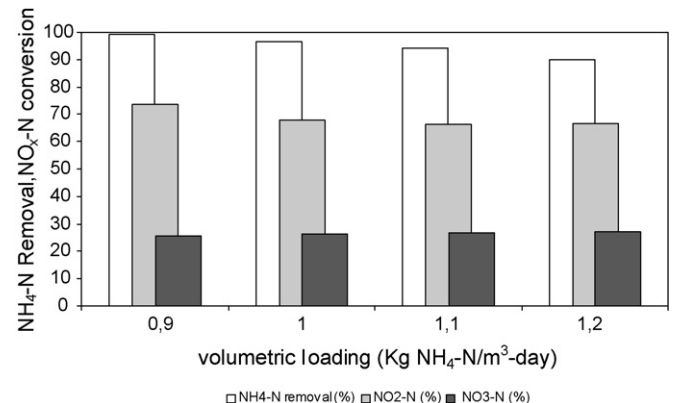

Fig. 2. Effect of applied NLR on the effluent NH_4-N concentrations.

Fig. 5. NH_4-N removal efficiency and NO_x-N accumulations at various NLR in the FBB nitritation reactor.

respectively. The NO_2-N/NO_x-N ratios corresponded to 0.74, 0.73, 0.72, and 0.69, respectively, indicating the occurrence of partial nitrification. The NO_3-N concentration was below 100 mg/l throughout the study which indicates the conversion of NO_2-N to NO_3-N was inhibited.

Although significant NO_2-N accumulation (an average of 200 mg NO_2-N/l) was achieved at various NLR, the difference of the NO_2-N concentration in the effluent water at the lowest and the highest NLR was about 27 mg/l, while the effluent NH_4-N concentration increased about from 2.1 to 34.5 mg/l.

Anthonisen et al. [18] reported that the AOB and NOB were inhibited 10–150 and 0.1–1.0 mg/l of free ammonia (FA) and all nitrifying bacteria were inhibited above 0.2 mg/l of free nitric acid. FA and free nitric acid concentrations can be calculated according to Eqs. (2) and (3).

$$FA\ (mg/l) = \frac{17}{14} \frac{\sum NH_4\text{-}N\ (mg/l) \times 10^3}{(k_b/k_w) + 10^{pH}} \tag{2}$$

where $k_b/k_w = e^{6344/(273+T)}$

$$HNO_2\ (mg/l) = \frac{47}{14} \frac{\sum NO_2\text{-}N\ (mg/l)}{\exp^{-(2300/273+T)} \times 10^{pH}} \tag{3}$$

Suthersan and Ganczarczyk [36] found that the NO_2-N accumulation was achieved at high pH, suggesting that it is possible to accumulate NO_2-N controlling the pH. The FBBNR was operated $21 \pm 1\,^{\circ}C$ temperature and the reactor pH was kept constant at 7.5 ± 0.1 during the experimental study. In the present study, the calculated average concentration of FA and nitric acid was 4.9 mg/l (4.0–5.7 mg/l) and 0.05 mg/l for the NLR of between 0.9 and 1.2 kg NH_4-N/m^3 day, respectively. An average FA concentration in the FBBNR was high enough to inhibit nitrite oxidizers selectively, and it seems to be a determining factor for NO_2-N accumulation in the process. As a result the NOB, but not the AOB was inactivated in the FBBNR.

The results indicated that the NH_4-N removal efficiency was decreased as the NLR load was increased even tough the DO concentration at the top of the FBBNR was over 2.0 mg/l which was higher than the optimal DO [37]. An et al. [38] observed also 80% NO_2-N ratio at the high DO concentration in a membrane bioreactor. The NO_2-N production efficiency above 66% was achieved throughout in this study. This indicates that the DO was not the limiting factor for nitritation. The reason for it was probably high concentration of FA.

Controls of the DO concentration in the reactor, various level of NO_2-N accumulation have been reported. In a fluidised-bed reactor, 34% of NO_2-N accumulation and 40% of NH_4-N oxidation was achieved [39]. Garrido et al. [37] achieved high NO_2-N accumulation (50%) at the DO concentration between 1 and 2 mg/l. Bernet et al. [40] observed about complete conversion of NH_4-N at the DO concentration of 0.5 mg/l with above 90% of NO_2-N/NO_x-N ratio. The FBBNR was operated at the high DO concentration and about 72% NO_2-N/NO_x-N ratio was achieved in this study.

Yang and Alleman [19] concluded that the DO level alone did not appear to be the dominant factor behind NO_2-N buildup and its correlation with FA concentration alone was also

erratic. High NO_2-N accumulation was observed when the ratio of DO/FA was below 5 [22]. The calculated minimum FA concentration in the FBBNR was 4 mg/l which corresponded to 4.9 mg NH_3/l. For the DO to FA ratio to be above 5, the DO level in the FBBNR must be higher than 20 mg/l (impossible). The calculated FA concentration demonstrated that DO/FA ratio was below 5 throughout the experimental study.

Ammonium oxidizers grow faster than the nitrite oxidizers at elevated temperatures (>15 $^{\circ}C$). At the operational temperature of 35 $^{\circ}C$, the maximum specific growth rate of nitrite oxidizers is approximately half of that for the ammonium oxidizers [41]. Only at temperatures above 25 $^{\circ}C$ is it possible for the ammonium oxidizers to effectively out compete the nitrite oxidizers [8,42]. In this study, the temperature in the laboratory was kept constant at $21 \pm 1\,^{\circ}C$ which was lower than the reported optimum value of 25–30 $^{\circ}C$ for nitrifiers. When the activated sludge process was operated at 25 and 30 $^{\circ}C$ temperatures, 75 and 65% NO_2-N accumulation was observed and the NH_4-N removal efficiency was over 90% [7,43]. The nitrification rates reported in the literature for various system treating high strength wastewater at 30 $^{\circ}C$ are 0.6–1.3 kg NH_4-N/m^3 day for nitrifying sludge, 0.8–1.0 kg NH_4-N/m^3 day for nitrifying biofilm SBR [25] and 0.47–1.6 kg NH_4-N/m^3 day for partial nitrification using downflow hanging sponge reactor [44]. The nitrification rate in this study was around 0.9 kg NH_4-N/m^3 day at 21 $^{\circ}C$ and about the same accumulation of NO_2-N and NH_4-N removal efficiency was observed in this study with previous study.

3.3. NO_x-N removal efficiency of the FBB denitritation reactor

Excess methanol was used as the external carbon source in the FBBDR. The reactor was operated in a start-up mode by feeding NO_2-N and NO_3-N to acclimate microorganisms to the synthetic wastewater for 2 months. The reactor was then fed with effluent water from the FBBNR after adding methanol and nutrients at a low flow rate. The NH_4-N concentration was measured to check the removal of NO_2-N caused by the denitritation or ammonification process. However, NH_4-N conversion was not observed in the effluent during the experiment. After 1 month of operation at the low NO_x-N load, the influent flow rate was gradually increased to examine the effects of the NO_x-N loading rate on the denitritation. The influent NO_2-N concentrations of the FBBDR was between 170 and 290 mg/l at which range more than 80% NO_x-N removal was achieved at various influent flow rates.

The NO_x-N supplied was between 220 and 330 mg N/l (the NLR was 0.08–0.4 kg/m^3 day) and the influent flow was increased as long as the total nitrogen removal was close to 90%. NO_2-N and NO_3-N concentrations in the effluent were 3.0 and 0.9 mg/l at 0.08 kg/m^3 day loading rate. About 98% of NO_2-N, NO_3-N, and NO_x-N removals were achieved at the lowest NLR.

Fig. 6 presents the inlet and outlet NO_2-N and NO_3-N concentrations and the total NO_x-N removal efficiency at various NO_x-N loading rates of FBBDR. The reactor exhibited high nitrogen removal up to a loading rate of 0.25 kg/m^3 day. During this period, NO_x-N effluent concentration never exceeded 15 mg/l.

Table 3
Performances of various nitritation and denitritation processes

| Reactor	NLR or inlet NH$_4$-N (mg/l)	NH$_4$-N removal efficiency (%)	NO$_2$-N/NO$_x$-N	Factors of control			References
				pH	T (°C)	DO (mg/l)	
UASB/MBR	24.00–43.8 mg/l	98.2	79.6	7.8–8.1	30–32/28–30	3.0–4.0	[38]
SBR	0.32 kg NH$_4$-N/m^3 day	63.5	60	7.18–8.42	21	1.5–4.0	[12]
Biofilm and Anammox	Average 568 mg/l	62.4 ± 13.5		7.61–8.30	17–31	0.5–5.97	[46]
Activated sludge	5.87 ± 0.06 kg NH$_4$-N/m^3 day	93.5 ± 0.9	73.8 ± 6.2	7.85 ± 0.5	30	1.1	[7]
FBBRs	1.1 kg NH$_4$-N/m^3 day	94.2	72	7.5	21 ± 1	≥2.5	This study
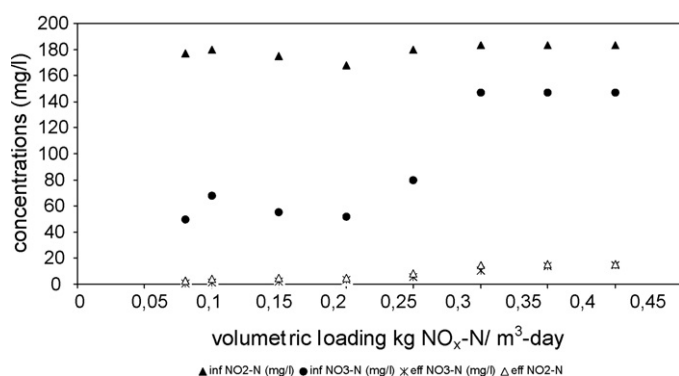
Fig. 6. Influent and effluent concentrations of NO$_2$-N and NO$_3$-N at various volumetric loading rate in the FBB denitritation reactor.

As shown in Fig. 6, the NO$_x$-N concentration in the effluent increased and the removal efficiency decreased due to the increase in the NLR by increasing influent flow rate. The average NO$_2$-N and NO$_3$-N concentration in the effluent, up to the NO$_x$-N loading rate of 0.25 kg/m^3 day was lower than 9 and 6 mg/l and the removal efficiencies were higher than 94 and 92%, respectively. The average nitrogen concentration in the effluent continued to increase and was 25, 29, and 30.5 mg NO$_x$-N/l, at NLR of 0.3, 0.35, and, 0.4 kg NO$_x$-N/m^3 day, respectively.

The NO$_2$-N, NO$_3$-N, and the total NO$_x$-N removal efficiencies are depicted in Fig. 7. Effluent concentrations of NO$_2$-N and NO$_3$-N were increased by increasing the loading rate of nitrogen. Although the removal efficiency was higher than 90% at the highest loading rate, NO$_2$-N and NO$_3$-N concentrations in the effluent were shown to be above 15 mg/l when the NO$_x$-N loading was higher than 0.25 kg N/m^3 day.
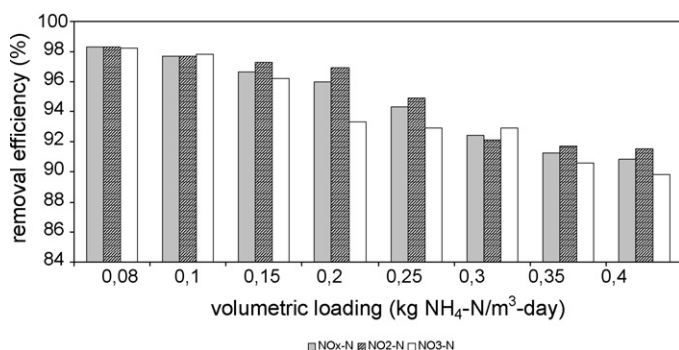

Fig. 7. Nitrogen removal efficiency at various volumetric loading rates in the FBB denitritation reactor.

The denitrification rates reported in the literature for various systems are 0.36 kg NO$_x$-N/m^3 day in a submerged biofilter, 0.2–0.38 kg NO$_2$-N/m^3 day in a packed film reactor and 3.23–18.70 kg NO$_2$-N/m^3 day in a two phase FBBR at 30 °C [45]. The denitritation rate in this study was around 0.24 kg NO$_2$-N/m^3 day at 21 °C. The denitritation rate could be increased by operating the reactor at a high temperature or modifying the reactor operation.

3.4. System performance of the combined FBBNR and FBBDR

Biological nitrogen removal can be carried out by various processes. The results and operational parameters of several processes for the nitritation and denitritation are presented in Table 3.

Operational parameters were examined for each process for the maximum NO$_2$-N accumulation in the FBBNR and NO$_x$-N removal in the FBBDR. In order to elucidate the nitritation and denitritation of the FBBRs for wastewaters with high concentrations of NH$_4$-N and NO$_2$-N accumulation and the control factors were compared with previous studies.

NO$_2$-N accumulations occurred by the inhibition of FA and/or nitric acid for the last three experimental studies and by the DO control at the first study. Because the inlet NH$_4$-N and NO$_x$-N concentration in the nitritation and denitritation reactors were lower and the reactors were operated at higher temperatures than in this study, high removal efficiency were obtained by using a UASB/MBR. Considering operating conditions of the presented literature in Table 3 high nitrogen removal efficiency was observed in this study by using FBBRs. The total nitrogen removal efficiency was higher than 93% and high NO$_2$-N/NO$_x$-N ratio was obtained at 21 °C by applying FBBRs.

4. Conclusion

During this study, FBBRs were successfully operated and maintained as a nitritation and denitritation system for the treatment of nitrogen-rich wastewater with high removal efficiency under low DO. Stable oxidation of NH$_4$-N to NO$_2$-N with lower formation of NO$_3$-N than NO$_2$-N at various nitrogen loading rates was obtained in the nitritation reactor. During the operation, an average NH$_4$-N removal efficiency of 90% was achieved at the highest nitrogen loading and 65% of the NH$_4$-N was converted to NO$_2$-N. In the denitritation reactor more than 90% NO$_3$-N than NO$_2$-N elimination was achieved at various NO$_x$-

N loading rates. The denitritation process reached an average NO_x-N elimination between 98.3 and 90.8% at the lowest and the highest loading rates, respectively. Although the process was operated at 21 °C, which is lower than the reported optimum temperature, comparable NO_2-N accumulation and the nitrogen elimination was achieved in the FBBRs.

Acknowledgements

References

[1] L. Gut, E. Płaza, J. Trela, B. Hultman, J. Bosander, Combined partial nitritation/Anammox system for treatment of digester supernatant, Water Sci. Technol. 53 (12) (2006) 149–159.

[2] J.J. Beun, J.J. Heijnen, M.C.M. van Loosdrecht, N-removal in a granular sludge sequencing batch airlift reactor, Biotechnol. Bioeng. 75 (1) (2001) 82–92.

[3] B. Wett, R. Rostek, W. Rauch, K. Ingerle, pH-controlled reject-water-treatment, Water Sci. Technol. 37 (12) (1998) 165–172.

[4] E. Bock, H.P. Koops, H. Harns, Cell biology of nitrifying bacteria, in: J.I. Prosser (Ed.), Nitrification, IRL Press, Oxford, 1986, pp. 17–38.

[5] E. Bock, H.P. Koops, B. Ahlers, H. Harns, Oxidation of inorganic nitrogen compounds as energy source, in: A. Balows, H.G. Trüper, M. Dworkin, W. Harder, K.H. Schleifer (Eds.), The Prokaryotes: A Handbook on Biology of Bacteria: Ecophysiology, Isolation, Identification, Applications, Springer, Berlin, Heidelberg, New York, 1992, pp. 414–430.

[6] G. Ruiz, D. Jeison, R. Chamy, Nitrification with high nitrite accumulation for the treatment of wastewater with high ammonia concentration, Water Res. 37 (2003) 1371–1377.

[7] G. Ruiz, D. Jeison, O. Rubilar, G. Ciudad, R. Chamy, Nitrification–denitrification via nitrite accumulation for nitrogen removal from wastewaters, Bioresour. Technol. 97 (2006) 330–335.

[8] Y.Z. Peng, Y. Chen, C.Y. Peng, M. Liu, S.Y. Wang, X.Q. Song, Y.W. Cui, Nitrite accumulation by aeration controlled in batch reactors treating domestic wastewater, Water Sci. Technol. 50 (10) (2004) 35–43.

[9] J. Gorska, A. Cichon, K. Miksch, Nitrogen removal from wastewater with high ammonia nitrogen concentration via shorter nitrification and denitrification, Water Sci. Technol. 36 (10) (1997) 73–78.

[10] C. Fux, S. Velten, V. Carozzi, D. Solley, J. Keller, Efficient and stable nitritation and denitritation of ammonium-rich sludge dewatering liquor using an SBR with continuous loading, Water Res. 40 (2006) 2765–2775.

[11] H. Yoo, K. Ahn, H. Lee, K. Lee, Y. Kwak, K. Song, Nitrogen removal from synthetic wastewater by simultaneous nitrification and denitrification (SND) via nitrite in an intermittently aerated reactor, Water Res. 33 (1) (1999) 146–154.

[12] P. Jenicek, P. Svehla, J. Zabranska, M. Dohanyos, Factors affecting nitrogen removal by nitritation/denitritation, Water Sci. Technol. 49 (5–6) (2004) 73–79.

[13] C.-C. Tseng, T.G. Potter, B. Koopman, Effect of influent chemical oxygen demand to nitrogen ratio on a partial nitrification/complete denitrification process, Water Res. 32 (1998) 165–173.

[14] W. Jianlong, Y. Ning, Partial nitrification under limited dissolved oxygen conditions, Process Biochem. 39 (2004) 1223–1229.

[15] O. Turk, D.S. Mavinic, Benefits of using selective inhibition to remove nitrogen from highly nitrogenous wastes, Environ. Technol. Lett. 8 (1987) 419–426.

[16] O. Turk, D.S. Mavinic, Maintaining nitrite build-up in a system acclimated to free ammonia, Water Res. 23 (1989) 1383–1388.

[17] U. Abeling, C.F. Seyfried, Anaerobic–aerobic treatment of high strength ammonium wastewater—nitrogen removal via nitrite, Water Sci. Technol. 26 (1992) 1007–1015.

[18] A.C. Anthonisen, R.C. Loehr, T.B.S. Prakasam, E.G. Srinath, Inhibition of nitrification by ammonia and nitrous acid, J. Water Pollut. Control Fed. 48 (1976) 835.

[19] L. Yang, J.E. Alleman, Investigation of batch-wise nitrite build-up by an enriched nitrification culture, Water. Sci. Technol. 26 (5–6) (1992) 997–1005.

[20] W. Bae, S. Baek, J. Chung, Y. Lee, Optimal operational factors for nitrite accumulation in batch reactors, Biodegradation 12 (2002) 359–366.

[21] K. Hanaki, C. Wantawin, S. Ohgaki, Nitrification at low levels of dissolved oxygen with and without organic loading in a suspended-growth reactor, Water Res. 24 (1990) 297–302.

[22] F. Cecen, I.E. Gonenc, Nitrogen removal characteristics of nitrification and denitrification fillters, Water. Sci. Technol. 29 (10–11) (1994) 409–416.

[23] C. Hellinga, A.A.J.C. Schellen, J.W. Mulder, M.C.M. van Loosdrecht, J.J. Heijnen, The SHARON process: an innovative method for nitrogen removal from ammonium-rich wastewater, Water Sci. Technol. 37 (1998) 135.

[24] T. Yamamoto, K. Takaki, T. Koyama, K. Furukawa, Novel partial nitritation treatment for anaerobic digestion liquor of swine wastewater using Swim-Bed technology, J. Biosci. Bioeng. 102 (6) (2006) 497–503 (The Society for Biotechnology, Japan).

[25] D. Bougard, N. Bernet, D. Che'neby, J.-P. Delgene's, Nitrification of a high-strength wastewater in an inverse turbulent bed reactor: effect of temperature on nitrite accumulation, Process Biochem. 41 (2006) 106–113.

[26] A. Pollice, V. Tandoi, C. Lestingi, Influence of aeration and sludge retention time on ammonium oxidation to nitrite and nitrate, Water Res. 36 (10) (2002) 2541–2546.

[27] J.S. Jeris, R.W. Owen, R. Hickey, F. Flood, Biological fluidized-bed treatment for BOD and nitrogen removal, J. Water Pollut. Control Fed. 49 (5) (1977) 816–831.

[28] L.T. Mulcahy, E.J. LaMotta, Mathematical model of the fluidized bed biofilm reator, Report No. 58-78-2, Department of Civil Engineering, University of Massachusetts at Amherst, 1978.

[29] S.W. Hermanowicz, M.W. Cheng, Biological fluidized bed reactor: hydro-dynamics, biofilm distribution and performance, Water Sci. Technol. 22 (1–2) (1990) 193–202.

[30] C.Y. Chen, C.D. Chen, Biofilm characteristics in biological denitrification biofilm reactors, Water Sci. Technol. 41 (4) (2000) 147–154.

[31] F.K.J. Rabah, D.F. Mohamed, Biofilm and biomass characteristics in high-performance fluidized-bed biofilm reactors, Water Res. 38 (2004) 4262–4270.

[32] F.K.J. Rabah, D.F. Mohamed, Nitrate removal characteristics of high performance fluidized-bed biofilm reactors, Water Res. 38 (2004) 3719–3728.

[33] A.E.F. Botrous, M.F. Dahab, High-Performance Side-Stream Nitrification and Denitrification of Municipal Biosolids Treatment Decants, vol. I. Side-Stream Nitrification Studies Final Report to USEPA, 1 October 2004, 189 pp.

[34] R.F. Gasser, R.W. Owens, J.S. Jeris, Nitrate removal from wastewater using fluid bed technology, Purdue Ind. Waste Conf. (1975) 202–1207.

[35] APHA, Standard Methods for the Examination of Water and Wastewater, 20th ed., American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC, USA, 1998.

[36] S. Suthersan, J.J. Ganczarczyk, Inhibition of nitrite oxidation during nitrification, some observations, Water Pollut. Res. J. Can. 21 (2) (1986) 257–266.

[37] J.M. Garrido, W.A.J. van Benthum, M.C.M. van Loosdrecht, J.J. Heijnen, Influence of dissolved oxygenconcentration on nitrite accumulation in a biofilm airlift suspension reactor, Biotechnol. Bioeng. 53 (2) (1997) 168–178.

[38] Y. An, F. Yang, H.C. Chua, F.S. Wong, B. Wu, The integration of methanogenesis with shortcut nitrification and denitrification in a combined UASB with MBR, Bioresour. Technol. 99 (2008) 3714–3720.

[39] M. Denac, S. Uzman, H. Tanaka, I.J. Dunn, Modeling of experiments on biofilm penetration effects in a fluidized bed nitrification reactor, Biotechnol. Bioeng. 25 (1983) 1841.

[40] N. Bernet, O. Sanchez, P. Dabert, A. Olaizola, J.J. Godon, J.P. Delgene's, Effect of solid hold-up on nitrite accumulation in a biofilm reactor molecular characterization of nitrifying communities, Water Sci. Technol. 11–12 (49) (2004) 123.

[41] J.H. Hunik, Engineering aspects of nitrification with immobilized cells, Ph.D. Thesis, Wageningen Agricultural University, The Netherlands, 1993.

[42] M. Brouwer, M.C.M. van Loosdrecht, J.J. Heijnen, One reactor system for ammonium removal via nitrite, STOWA Report, 96-01, STOWA, Utrecht, The Netherlands, 1996, ISBN 90 74476 55 4.

[43] G. Ciudad, O. Rubilar, P. Munoz, G. Ruiz, R. Chamy, C. Vergara, D. Jeison, Partial nitrification of high ammonia concentration wastewater as a part of a shortcut biological nitrogen removal process, Process Biochem. 40 (2005) 1715–1719.

[44] H.P. Chuang, A. Ohashi, H. Imachi, M. Tandukar, H. Harad, Effective partial nitrification to nitrite by down-flow hanging sponge reactor under limited oxygen condition, Water Res. 41 (2007) 295–302.

[45] A. Hirata, A.A. Meutia, Denitrification of nitrite in a two-phase fluidized bed bioreactor, Water Sci. Technol. 34 (1–2) (1996) 339–346.

[46] B. Szatkowska, G. Cema, E. Plaza, J. Trela, B. Hultman, A one-stage system with partial nitritation and Anammox processes in the moving-bed biofilm reactor, Water Sci. Technol. 55 (8–9) (2007) 19–26.