



Single-stage autotrophic nitrogen removal using Anammox and partial nitrification (SNAP) for treatment of high strength ammonia wastewater at low temperature

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ABSTRACT

In this study, long-term effect of low temperature on the SNAP (Single-stage Nitrogen removal using Anammox and Partial nitrification) process treating high strength ammonia wastewater was investigated. When the concentration of ammonium in wastewater was 2,000±20 mg/L, stable nitrogen removal was observed in the SNAP process with the ammonium and TN (total nitrogen) removal efficiencies of 96% and 85% at 10 °C, corresponding the nitrogen load of 0.1 kg N m⁻³ d⁻¹. This SNAP process may have applications for treating high strength ammonia and low C/N ratio wastewater such as mature landfill leachate at low temperature (≤20 °C).

Keywords: Anaerobic ammonium oxidation; Low temperature; Nitrogen removal; Partial nitrification; Sequencing batch biofilm reactor (SBBR)

INTRODUCTION

Ammonium pollution, which can cause eutrophication and be toxic to aquatic species, is becoming a serious environmental problem [1,2]. Traditional treatment of municipal waste water, ammonium (NH₄⁺-N) is converted into nitrogen gas via a two-step process starting with nitrification, which is the aerobic oxidation of ammonium (NH₄⁺-N) to nitrite (NO₂⁻-N) to nitrate (NO₃⁻-N), followed by heterotrophic denitrification under anaerobic condition. Furthermore, the process requires a large amount of energy (mainly for aeration) and the addition of an external carbon source for denitrification.[3]Which not only makes full-scale denitrification quite expensive but also causes secondary pollution, limiting the application in the treatment of low C/N ratio wastewater.

The anammox (anaerobic ammonium oxidation) had been recognized as a promising process to treat wastewater devoid of organic carbon [4]. Generally, major nitrogen compound in wastewater is ammonium, which must be nitrified partially to nitrite, but not to nitrate. Then the remaining ammonium together with the produced nitrite is converted to dinitrogen gas in anammox process. This partial nitrification and anammox process can be performed in two-stage reactors as the SHARON-ANAMMOX process [5] or in a single-stage reactor such as CANON (completely autotrophic nitrogen removal over nitrite) process [6,7] and SNAP (single-stage nitrogen removal using anammox and partial nitrification) process [8]. However, the nitrate is always inevitable in the stage of partial nitrification, since stable operation of partial nitrification process could not achieve 100% of the nitrite accumulation rate for long time even under the optimized conditions [9,10].

Nevertheless, application of anammox and partial nitrification for autotrophic nitrogen removal has mainly focused on

wastewater with high temperature ($\geq 25^{\circ}\text{C}$) [11]. The application of the autotrophic nitrogen removal process for the treatment of high strength and warm wastewater characterized by temperature exceeding 25°C is nowadays part of the state of the art [12]. Consequently, lower temperature of anammox reactor would result in an immediate large decrease in specific activity. De Clippeleir *et al.* [13] therefore investigated the feasibility of high rate ammonia removal at 25°C . They achieved nitrogen removal rates of $0.33\text{ kg N m}^{-3}\text{ d}^{-1}$. Despite the decrease in activity several laboratory studies have reported anammox and partial nitrification at lower temperature ($\leq 25^{\circ}\text{C}$) and reasonable conversion rates [14]. Enrichment of anammox and partial nitrification at low temperatures ($\leq 20^{\circ}\text{C}$) without inoculation with a large amount of biomass has not been shown previously. Low temperature application of this SNAP process becomes very interesting. And then the application of the lower temperature would extend the application potential of anammox-related processes.

Thus, this study was performed to evaluate the effect of low temperature ($\leq 20^{\circ}\text{C}$) on the SNAP process for treatment of high strength ammonia wastewater in a single SBBR (sequencing batch biofilm reactor). Some controlling strategies were optimized and the reactor performance was examined in the SNAP process.

EXPERIMENTAL SECTION

2.1 Reactor and operational strategy

The SBBR (sequencing batch biofilm reactor) was a plexiglas cylinder, the height of which was 500 mm and internal diameter 200 mm, with height to diameter ratio being 2.5. Semi-soft fibre fill was used as the biomass carrier and the packing rate was 50% (V/V). The reactor had a working volume of 10 L feeding with artificial wastewater. During the experiment period, all the reactors were placed in a thermostatic chamber, in order to maintain the temperature constant at low temperature.

The SBBRs were operated sequentially in 8 h-cycle, with intermittent aeration (aeration 4h / aeration stop 4h). Discharging and feeding were carried out during the last 10 min of each 3 cycles (24h) and the water filling ratio was 0.25. The aeration was controlled using air pumps to regulate the DO concentration of the reactor. The SBBR was run at limited aeration stage, with the concentration of DO strictly controlled around 2.5 mg L^{-1} by adjusting the air flow rate. At the aeration stop stage, the concentration of DO was at $0.1\text{--}0.2\text{ mg L}^{-1}$.

The strategy of limited aeration was adopted to inhibit (nitrite oxidizing bacteria) NOB activity and prompt (anaerobic ammonium oxidation bacteria) AnAOB proliferation, achieving autotrophic (ammonium oxidizing bacteria) AOB and AnAOB simultaneous growth.

2.2 Long term reactor operation and synthetic wastewater

The SNAP biomass was derived from an ongoing lab-scale SBBR using anammox and partial nitrification for treatment of high strength ammonium wastewater. The original SBBR was operated for 2 years with influent $\text{NH}_4^+\text{-N}$ concentration of $2,000 \pm 20\text{ mg/L}$. The experimental period described in this study was preceded by a long start-up in which the reactor was at room temperature ($20 \pm 1^{\circ}\text{C}$) and fed with ammonium as the only nitrogen source. After six months of operation at room temperature, the reactor was placed in a thermostatic chamber, in order to maintain the temperature constant at 20 ± 1 , 15 ± 1 and $10 \pm 1^{\circ}\text{C}$, respectively.

The composition of the synthetic inorganic media was as follows (L), NH_4HCO_3 : 11290 mg; KH_2PO_4 : 25 mg; EDTA: 25 mg; FeSO_4 : 6.25 mg; $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$: 200 mg; CaCl_2 : 300 mg; trace nutrient solution: 1.25 ml and the right amount of KHCO_3 to regulate pH to 8.0. The trace nutrient solution contained (g/L), $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$: 0.43; $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$: 0.24; $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$: 0.99; $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$: 0.25; $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$: 0.22; $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$: 0.19; H_3BO_3 : 0.014; $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$: 0.05.

2.3 Analysis methods

The influent and effluent samples were collected on a daily basis and were analyzed immediately. The concentrations of $\text{NH}_4^+\text{-N}$, $\text{NO}_2^-\text{-N}$, $\text{NO}_3^-\text{-N}$, TN and MLSS were measured according to standard methods for the examination of water and wastewater [15]. The system was equipped with suitable submerged probes, such as dissolved oxygen (DO) (Hach, HQ30d, USA), pH (Hach, sension2, USA) and oxidation reduction potential (ORP) (Hach, sension2, USA).

RESULTS AND DISCUSSION

3.1 Long term operation

The reactor was operated over a time period of 175 days. For a comprehensive view of the SNAP process performance, the profile which shows the effluent concentrations of $\text{NH}_4^+\text{-N}$, $\text{NO}_2^-\text{-N}$, $\text{NO}_3^-\text{-N}$, as well as nitrogen removal rate was presented in Figure 1. The concentration of $\text{NH}_4^+\text{-N}$ in influent was kept at $2,000 \pm 20$ mg/L, which simulated the ammonium concentration in mature landfill leachate [16,17]. It could be seen that a persistent, stable partial nitrification and anammox were achieved in the SBBR. AOB and NOB inhibition by free ammonia (NH_3) and free nitrous (HNO_2) [18] could be excluded throughout the whole experimental period, since their concentrations were calculated to be below 2.5 and 0.04 mg/L. A maximal total nitrogen (TN) removal rate of $0.34 \text{ kg N m}^{-3} \text{ d}^{-1}$ was achieved for the SNAP process with the $\text{NH}_4^+\text{-N}$ and TN removal efficiencies of 96.8% and 90.9%. It should be noted that the SBBR biofilm of the SNAP process was reddish.

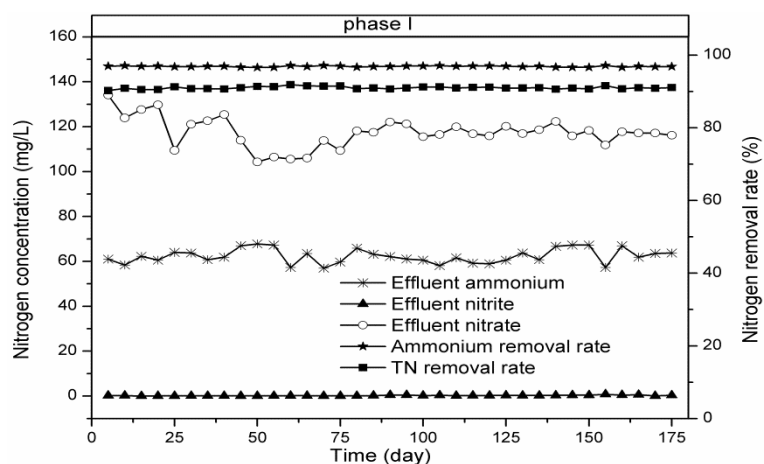


Figure 1 Profiles of nitrogenous compounds concentration and removal rate during the long term operation

3.2 Nitrogen removal performance of the SNAP process at different temperature

The reactor temperature was decreased gradually from 20 to 15 °C after the first 175 days of phase I. The short-term effect of the reactor caused by temperature decreased was a sharp decrease in anammox activity for the nitrite began to accumulate (day 182), which would also inhibited anammox activity gradually [19]. While the AOB activity was hardly affected. And then the concentration of ammonium and nitrite in effluent increased rapidly, corresponding with the ammonium removal rate from 96.8% to 85.1%. The applied load of reactor was then reduced to $0.30 \text{ kg N/m}^3/\text{d}$ after decreasing the temperature about one week.

Table 1 Ammonium conversion and TN removal rate as well as the temperature during the different phases of the reactor operation

Phase	Time/d	Temperature	Ammonium concentration in effluent (mg/L)	Applied load ($\text{kg N/m}^3/\text{d}$)	Ammonium conversion		TN removal	
		°C			$\text{kg N/m}^3/\text{d}$	%	$\text{kg N/m}^3/\text{d}$	%
I	1-175	20	64 ± 2	0.40	0.387	96.8	0.364	90.9
II	298-357	15	110 ± 2	0.30	0.284	94.5	0.265	88.3
III	452-505	10	188 ± 3	0.10	0.091	90.6	0.085	85.1

The reactor was then stable for about two months when the reactor was operated at 15 °C. The TN removal rate was $0.28 \text{ kg N m}^{-3} \text{ d}^{-1}$ with the $\text{NH}_4^+\text{-N}$ and TN removal efficiencies of 94.5% and 88.3%. The reactor temperature was decreased stepwise since day 358. The performance of the reactor worsened on the second day. The nitrogen removal rate continuously decreased in time accordingly. And then the nitrogen load decreased down to $0.1 \text{ kg N m}^{-3} \text{ d}^{-1}$ when the reactor was operated at 10 °C. As shown in the Table 1, the reactor was then successfully operated for two months (days 452-505) in which total nitrogen was removed at a rate of $0.085 \text{ kg N m}^{-3} \text{ d}^{-1}$ with an efficiency of 85%.

Overall, it could be concluded safely that the system had the capacity of resisting the shock of low temperature. Therefore, the simultaneous partial nitrification and anammox process could be applied for nitrogen removal in treating low C/N ratio wastewater such as mature landfill leachate at low temperature, even though the temperature changed frequently at some time. Nevertheless, some unexpected factors might affect the stability of the system at low temperature, which would be further investigated in the future.

CONCLUSION

In a long-term study, the SNAP process demonstrated that advanced autotrophic nitrogen removal could be achieved at low temperature in a single-stage SBBR. A maximal total nitrogen (TN) removal rates of 0.1 kg N m⁻³ d⁻¹ was achieved in the SNAP process with TN removal efficiencies of 85% at 10 °C. Thus, the SNAP process can be used to treat wastewater at low temperature.

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