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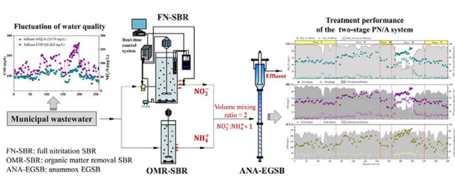
High-efficient nitrogen removal from municipal wastewater via two-stage nitritation/anammox process: Long-term stability assessment and mechanism analysis

Pengfei Jin, Baikun Li, Dongyang Mu, Xiyao Li, Yongzhen Peng*

National Engineering Laboratory for Advanced Municipal Wastewater Treatment and Reuse Technology and Key Laboratory of Beijing for Water Quality Science and Water Environment Recovery Engineering, Beijing University of Technology, Beijing 100124, PR China

GRAPHICAL ABSTRACT



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ABSTRACT

This study focused on the long-term stability of a novel two-stage partial-nitritation/anammox (PN/A) process treating municipal wastewater with fluctuated water quality. Specifically, two parallel sequencing batch reactors (SBRs) were used for removing organic matters and achieving complete nitritation, while the expanded granular sludge bed (ANA-EGSB) was used for anammox. With the influent ammonium concentration varying from 32 to 79 mg/L and the average hydraulic retention time of 3.39 h in this system, more than 93% of ammonium was removed and the effluent TIN was 4.8–11.8 mg/L. The partial denitrifying occurring in the anammox reactor could reduce nitrate to nitrite that was reused by anammox bacterium, enhancing the TIN removal efficiency. Further, the "overconsumption of ammonium" under anaerobic conditions was observed in ANA-EGSB. Microbial community analysis showed that *Nitrosomonas* (AOB) were the dominant nitrifying bacteria in the nitritation SBR and *Candidatus_Brocadia* with the relative abundance of 6–13% dominated in ANA-EGSB.

1. Introduction

Partial-nitritation/anammox (PN/A) process can save 60% aeration energy and nearly 100% organic carbon (Kartal et al., 2010), compared with conventional nitrification/denitrification nitrogen removal process. Until now, the PN/A process has been successfully implemented to treat high-strength ammonium wastewater such as dewatering liquor from anaerobic digesters in the side-stream of municipal wastewater treatment plants. More than 200 full-scale side-stream PN/A processes have been operated worldwide by the year of 2014 (Lackner et al., 2014), while only two mainstream PN/A processes are operated and require special conditions (Wett et al., 2013; Yeshi et al., 2016). Although the application of PN/A process to the mainstream wastewater processes has been proposed two decades ago (Kartal et al., 2010; Jetten et al., 1997), mainstream anammox was just studied extensively in recent years (Ma et al., 2011; Gilbert et al., 2014; Lotti et al., 2015; Miao et al., 2017). The implementation of mainstream PN/A processes faced two main barriers: first, the repression of nitrite-oxidizing bacteria (NOB) at low ammonium concentration; second, the impact of low water temperature and the frequent fluctuation of water quantity and quality on the stable operation (Kartal et al., 2010; De Clippeleir et al., 2013).

* Corresponding author.
E-mail address: pyz@bjut.edu.cn (Y. Peng).

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As we knew, there are two types PN/A processes: single-stage PN/A process with partial nitritation and anammox reaction combined in one reactor (e.g. CANON process), and two-stage PN/A process with partial nitritation and anammox in two separate reactors. Although the two-stage PN/A process needs more efforts for installations than single-stage PN/A process, its advantages are prominent, such as enriching AOB and anammox bacteria in different reactors, avoiding the competition for nitrite between anammox bacteria and NOB, and optimizing the control parameters for different functional organisms (Cao et al., 2017; Vlaeminck et al., 2012), which can facilitate the suppression to NOB. For the two-stage PN/A process, a reliable control strategy of the partial-nitritation stage is vital to provide suitable nitrite/ammonium ratio (around 1:1) for the anammox stage (Ma et al., 2011). Previous studies tried to achieve partial nitritation by controlling the reaction process in a single reactor (Ma et al., 2011; Isanta et al., 2015; Kouba et al., 2017), which might be difficult when the water quality fluctuated dramatically. In contrast, combining full nitritation reactor and organic matter removal reactor for mainstream partial nitritation was not common, and its long-term stability has not been tested.

Furthermore, the mainstream anammox process studied so far mainly treated the synthetic wastewater or the pretreated wastewater by adding nitrite and ammonium (Hendrickx et al., 2012; Ma et al., 2013; Lotti et al., 2014). However, the ingredients of real municipal wastewater was complicated but the studies on the mainstream two-stage PN/A systems treating real municipal wastewater were limited. The reported studies confirmed the feasibility of mainstream two-stage PN/A process and explored the microbial community response to influent shift and lowering temperature (Ma et al., 2011, Regmi et al., 2015; Liu et al., 2018). Until now, the long-stability assessment of two-stage PN/A processes treating mainstream municipal wastewater with fluctuated water quality has not been reported, and the nitrogen removal mechanism and the dynamics of microbial community structure under the condition have not been elucidated yet.

In this study, a novel two-stage PN/A process in which two parallel sequencing batch reactors (SBRs) were used for partial nitritation and the expanded granular sludge bed (ANA-EGSB) acted as the anammox stage was developed to treat municipal wastewater. The stability of the two-stage PN/A process was assessed over the long-term (260 days) treatment of municipal wastewater with fluctuated water quality for the first time. And the nitrogen conversion mechanism in the ANA-EGSB reactor were analyzed based on the stoichiometric correlation and batch tests. Furthermore, the microbial community structures of the nitritation reactor and the ANA-EGSB reactor were analyzed with high-throughput sequencing to elucidate the biological base of the two-stage PN/A process.

2. Materials and methods

2.1. Two-stage PN/A system set-up and operation strategy

The schematic of the two-stage PN/A system is shown in Fig. 1. The two parallel SBRs were used for organic matter removal and full nitritation and termed as OMR-SBR and FN-SBR, respectively. Municipal wastewater was pumped into the two SBRs individually and then the effluents of the two SBRs were mixed and further treated in the subsequent ANA-EGSB (the anammox reactor). Due to the oxidation of nitrite to nitrate in the pipe connecting (the red circle of Fig. 1) the middle tank 2 and ANA-EGSB, the mixture ratio between the effluents of FN-SBR and OMR-SBR was set as 1.37 in the first 115 days and then increased to 1.93 in the rest 155 days in order to provide sufficient nitrite to the ANA-EGSB.

The OMR-SBR (working volume: 5L) was operated with an anaerobic/aerobic mode in each circle and partial organic matters were removed from the municipal wastewater. To avoid the nitrification of ammonium and efficiently convert organic matters into biomass, short aeration time (30–60 min) and sludge retention time (SRT: 2–5 days) were used. The working volume of FN-SBR was 10 L and each circle consisted of feeding (18 min), anoxic reaction (30 min), aerobic reaction (changed based on the water quality), settling (50 min), idling and the pH, DO was continuously monitored in the course of circle. The pre-anoxic reaction made sure that the residual nitrite produced in the last cycle could be denitrified with the organic matters in the influent, which avoided the carbon source to be wasted in the aerobic reaction and alleviated the heterotrophic bacteria to compete with AOB for dissolved oxygen (DO). As reported by (Yang et al., 2007), the aeration duration of FN-SBR was controlled based on the real-time control system i.e. the aeration was stopped once the feature inflection point of pH value (also called the “ammonium valley”) appeared, in order to achieve complete nitritation. The exchange ratio of the FN-SBR was set as 0.7, in order to provide as much nitrite as possible for the subsequent anammox process. Furthermore, the DO and the effective SRT of the FN-SBR was controlled at 0.5–1.5 mg/L and 10–15 days, respectively.

The ANA-EGSB (working volume: 1L, diameter: 50 mm) was wrapped with black cloth to protect the anammox granular from light. The pretreated wastewater was continuously pumped into the ANA-EGSB at the bottom and the upflow rate was controlled as 150 ml/min by adjusting the recirculation ratio to maintain a good fluidization condition. The temperature of the ANA-EGSB and FN-SBR were controlled at 29–30 °C and 23–25 °C respectively, while the OMR-SBR operated at the ambient temperature since the removal of organic matter was less affected by temperature.

2.2. Characteristics of wastewater and inoculum biomass

The municipal wastewater was taken from a septic tank at the residential area of Beijing University of Technology (Beijing, China). The seed activated sludge of the FN-SBR and OMR-SBR was taken from a pilot-scale nitritation-denitrification SBR (volume: 8.8 m^3) at the Beijing University of Technology. Before the experiment, anammox granule sludge has been cultured in ANA-EGSB using the synthetic wastewater (NO_2^--N: 20 mg/L, NH_4^+-N: 30 mg/L) that simulated the low strength ammonium wastewater, and high nitrogen removal rate (5.17 kg N/(m^3·d)) was achieved.

2.3. Long-term stability tests of the two-stage PN/A system

The long-term stability of the two-stage PN/A system was tested in the 260-day experimental period, during which the influent and effluent samples of each reactor were taken three times per week. Considering that the substrate may be consumed by the microorganisms in the pipe, the influent sample of the ANA-EGSB was taken before the inlet point. The experiment was divided into five phases (I-V) throughout the 260-day period based on the fluctuation of wastewater quality and the performance of FN-SBR. In Phase I (day 1-day 70), the nitritation was well maintained and the influent of ANA-EGSB was replaced by the pretreated municipal wastewater. In Phase II (day 71-day 146), the COD and ammonium concentration of the wastewater increased abruptly. In Phase III (day 147-day 164), the substrate concentration in wastewater decreased suddenly and the control parameter wasn’t adjusted timely in the FN-SBR, leading to high dissolved oxygen (above 2 mg/L) and excessive aeration after the “ammonium valley”. In phase IV (day 165-day 221), the performance of nitritation in the FN-SBR deteriorated in the early phase due to the delay adjustment in phase III and then gradually recovered via shortening the aeration time and SRT. Moreover, as reported in literatures, nitrate could be reduced to nitrite or nitrogen gas by heterotrophic bacteria or anammox bacteria in the presence of organic matters and the nitrite produced in the reaction could be reused by anammox bacteria (Kartal et al., 2013; Du et al., 2014). Therefore, in phase IV, a portion of the effluent of the OMR-SBR was replaced with the raw wastewater and then mixed with the effluent of the FN-SBR as the ANA-EGSB’s influent, trying to improve the nitrogen removal efficiency in the ANA-EGSB. In phase V