Chemosphere 200 (2018) 412–418

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Effect of Fe (II) in low-nitrogen sewage on the reactor performance and microbial community of an ANAMMOX biofilter

Xiaojing Zhang*, Yue Zhou, Siyu Zhao, Rongrong Zhang, Zhaoxue Peng, Hanfei Zhai, Hongzhong Zhang**

Henan Collaborative Innovation Center of Environmental Pollution Control and Ecological Restoration, School of Material and Chemical Engineering, Zhengzhou University of Light Industry, Zhengzhou 450001, China

HIGHLIGHTS

- Fe (II) in 1–5 mg L^{-1} effectively improved AAOB bioactivity and nitrogen removal.
- Fe (II) in 10–30 mg L^{-1} performed reversible inhibition on ANAMMOX process.
- The irreversible suppression threshold of Fe (II) on ANAMMOX was 50 mg L^{-1}.
- DHA was a pre-indicator for nitrogen removal performance of ANAMMOX system suffered Fe (II).
- Fe (II) feeding lowered the relative abundance of *Candidatus Kuenenia.*

ARTICLE INFO

Article history:
Received 11 December 2017
Received in revised form
1 February 2018
Accepted 21 February 2018
Available online 22 February 2018

Handling Editor: A Adalberto Noyola

Keywords:
ANAMMOX
Nitrogen removal
Fe (II)
AAOB
DHA
Microbial community

ABSTRACT

In this study, the effect of Fe (II) on Anaerobic Ammonium Oxidation (ANAMMOX) process was investigated by step-wise increasing the Fe (II) in influent from 1 to 50 mg L^{-1}. The nitrogen removal, biofilm property and the microbial community were analyzed in each phase. Results showed that, the anaerobic ammonia-oxidizing bacteria (AAOB) bioactivity and the nitrogen removal of ANAMMOX system were slightly improved to 0.58 from the initial 0.51 kg m^{-3} d^{-1} by Fe (II) in 1–5 mg L^{-1}. The nitrogen removal was suppressed and could recover to the initial level during the same period under 10–20 mg L^{-1} Fe (II), while it did not recover to the initial level under 30 mg L^{-1} Fe (II) and showed no recovery performance under 50 mg L^{-1} Fe (II). The irreversible suppression threshold of Fe (II) was calculated as 50 mg L^{-1}. The iron content in ANAMMOX biofilm presented linear correlation with the influent Fe (II) in 1–20 mg L^{-1}, which then tended to be stable when Fe (II) was higher. Dehydrogenase activity (DHA) showed similar and faster response to Fe (II) than the microbial activity, and it was an effective pre-indicator for the nitrogen removal performance in the ANAMMOX system suffered Fe (II). The Fe (II) feeding firstly led to the relative abundance of AAOB decreased to 11.04% from the initial 35.46%, and finally picked up to 19.39% after the long-term acclimatization.

1. Introduction

In the traditional nitrification-denitrification nitrogen removal process, the ammonia was sequentially oxidized to nitrate by aerobic ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB) (Guimaraes et al., 2017). Afterwards, denitrifying bacteria use organic substrate as electron donor to reduce nitrate to nitrogen gas (Zhang et al., 2017a). However, many wastewaters do not contain sufficient amounts of biodegradable carbon, making them less suitable for nitrogen removal via the nitrification-denitrification process (Zhang et al., 2016a). Not to mention, organic compounds in wastewaters are expected to be converted to biogas by anaerobic treatment process, which was feasible with the present state of the art (Kartal et al., 2010). The conversion leads to excessive amounts of nitrogen in the wastewater because of the lack of organic carbon for denitrification (Vega De Lille et al., 2015). The discovery of anaerobic ammonium oxidation (ANAMMOX) process revolutionized the understanding of microbial nitrogen cycling. The functional bacteria of ANAMMOX process was

* Corresponding author.
** Corresponding author.
E-mail address: zhangxiaojing@zzuli.edu.cn (X. Zhang).

https://doi.org/10.1016/j.chemosphere.2018.02.131

anaerobic ammonia-oxidizing bacteria (AAOB), which utilizes nitrite as oxidant to oxidize ammonia nitrogen directly to nitrogen gas (Francis et al., 2007). However, AAOB grows slowly and is sensitive to environmental conditions, such as metal ions (Yang et al., 2014), salinity (Puthiya Veettil et al., 2015), antibiotics (Chen et al., 2016) and substrate concentration (Ma et al., 2017), which then limits the development of ANAMMOX process. Recently, the application of ANAMMOX process in municipal wastewater treatment has attracted increasing attention. Therefore, it is important to study the possible effects of the materials existed in municipal wastewater on ANAMMOX, to facilitate the development of the process.

With the development of smelting, beneficiation, and machinery manufacturing, more and more metal ions have entered into the wastewater (Hou et al., 2016a). Metal ions were not biodegradable, eventually accumulated into the activated sludge or biofilm, and then destroyed the sewage biological treatment system (Zhang et al., 2015). Fe (II) was one of the most common metal ions existed in nitrogen-containing wastewater, which was also the necessary nutrients for microbial activities and coenzyme factor for substances such as microbial metal proteinases or certain functional enzymes (Ribeiro et al., 2017; Zhang et al., 2017b). When Fe (II) was insufficient, the intracellular transport system would be restricted and then limited the activity of microorganisms. When Fe (II) feeding was extensive, Fe (II) would enter into the microorganisms through microbial adsorption, transmembrane transport and intracellular accumulation (Vigneri et al., 2017). Some scholars have investigated the effect of Fe (II) or Fe (III) on the taxonomical and functional microbial community dynamics in an ANAMMOX-ASBR system (Wang et al., 2016), and autotrophic denitrification process (Liu and Horn, 2012). However, no research has been performed to evaluate the long-term effects of elevated Fe (II) on the nitrogen removal performance, as well as the microbial community of ANAMMOX process treating low-nitrogen sewage.

The main goal of this study was to (i) to investigate the effect of Fe (II) on the nitrogen removal performance and to detect the suppression threshold of Fe (II) on ANAMMOX process, (ii) to determine the effect of Fe (II) on the biofilm property of ANAMMOX process, and (iii) to clear the evolution of microbial community characteristics under Fe (II) effects.

2. Material and methods

2.1. Sludge and wastewater

A biofilter (Height: 20 cm; Diameter: 10 cm; Effective volume: 1 L) filled with volcanic rocks (particle size: 6–8 mm, specific surface area: 25.5 $m^2\,g^{-1}$) was adopted as the experimental reactor in present study, as shown in Fig.1. The reactor was seeded for 500 mL nitrification sludge (4.1 $g\,L^{-1}$) and 100 mL ANAMMOX sludge (5.7 $g\,L^{-1}$), to start-up ANAMMOX process. The synthetic wastewater contained 0.236–0.942 $g\,L^{-1}$ $(NH_4)_2SO_4$ as the ammonia resource, 0.246–0.986 $g\,L^{-1}$ $NaNO_2$ as the nitrite resource and 2.685 $g\,L^{-1}$ $NaHCO_3$ for adjusting pH as about 8.0, as well as 68 $mg\,L^{-1}$ KH_2PO_4, 150 $mg\,L^{-1}$ $MgSO_4{\cdot}7H_2O$, 68 $mg\,L^{-1}$ $CaCl_2$, and 1 $mL\,L^{-1}$ trace element solution I and II. The effect of Fe (II) was investigated by directly adding the $FeSO_4{\cdot}7H_2O$ into the influent, made the concentrations as 0, 1, 5, 10, 20, 30 and 50 $mg\,L^{-1}$, respectively in each phase.

2.2. Experimental set-up

The whole experiment was carried out as six phases. In P0 (day 1–39), ANAMMOX process was started-up, which was further conducted as P_{01}, P_{02} and P_{03}. In P_{01}, the influent NH_4^+-N and NO_2^--N were both 200 $mg\,L^{-1}$, then in P02 they were both decreased to 100 $mg\,L^{-1}$, and finally in P_{03} decreased to 50 $mg\,L^{-1}$. After the reactor got stable for treating wastewater with 50 $mg\,L^{-1}$ NH_4^+-N and 50 $mg\,L^{-1}$ NO_2^--N, the influence of Fe (II) was investigated in P1 to P6. In P1 to P6, Fe (II) with different concentrations was added to the influent, as shown in Table 1. Other detailed operational conditions including the pH, temperature and hydraulic retention time (HRT) for each phase were summarized as Table 1.

2.3. Analytical methods

Concentrations of NH_4^+-N and NO_2^--N were daily measured in different colorimetric methods and NO_3^--N was analyzed in ultraviolet spectrophotometric method. TN was calculated as the sum of NH_4^+-N, NO_2^--N and NO_3^--N. The pH and T were detected using portable instruments with specific probes (WTW, Germany). The

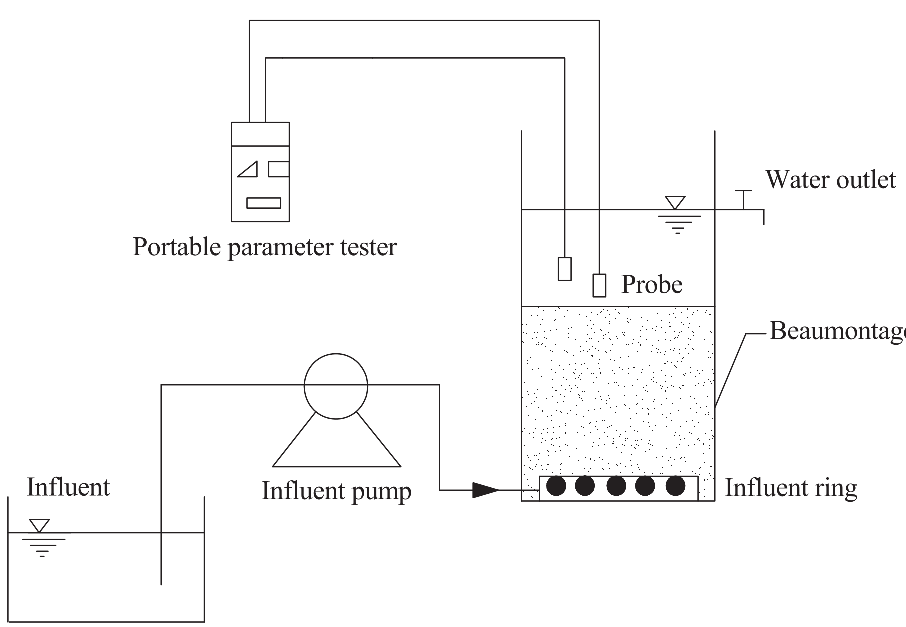

Fig. 1. Schematic diagram of the experimental system.