



# Partial nitrification performance and mechanism of zeolite biological aerated filter for ammonium wastewater treatment



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## HIGHLIGHTS

- ZBAF succeeded in partial nitrification of influent  $\text{NH}_4^+\text{-N}$  from 250 to 550 mg/L.
- FA inhibition was proved to be responsible for excellent partial nitrification.
- Zeolite played an important role in maintaining appropriate FA range.
- Production of nitrite in ZBAF followed the zero-order kinetics model.
- Microbial analysis results showed the enrichment of AOB and inhibition of NOB.

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## ABSTRACT

A zeolite biological aerated filter (ZBAF) with continuous feeding was successfully applied for achieving stable partial nitrification. Excellent nitrite accumulation (higher than 98.0%) and high nitrite/nitrate production rate (NPR) (approximately  $0.760 \text{ kg/m}^3/\text{d}$ ) were obtained with increase influent ammonium concentration from 250 to 550 mg/L within a nitrogen loading rate (NLR) of  $0.854\text{--}1.200 \text{ kg/m}^3/\text{d}$ . Owing to the adsorption of zeolite to ammonium, free ammonia (FA) concentration could remain at an appropriate range for inhibition of nitrite oxidizing bacteria (NOB) and dominance of ammonia-oxidizing bacteria (AOB), which should be responsible for the excellent partial nitrification realized in ZBAF. Kinetic study showed that the production of nitrite in ZBAF followed the zero-order kinetics model and high-throughput sequencing analysis further presented the enrichment of AOB and inhibition of NOB in ZBAF. All the results demonstrated that ZBAF hold a great potential in the application of partial nitrification for ammonium wastewater treatment.

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## 1. Introduction

Traditional biological nitrogen removal (BNR) process consists of two main steps: ammonium biologically converted to nitrite (nitritation) and nitrate (nitratation) in nitrification and complete reduction of nitrate to  $\text{N}_2$  in denitrification. The operational costs of this traditional process are related to the oxygen requirement for nitrification and organic carbon source consumed in denitrification. In order to reduce operational costs, innovative biological processes are widely investigated in recent years, such as partial nitrification–denitrification and partial nitrification–ANAMMOX.

Compared to traditional BNR process, partial nitrification–denitrification, based on incomplete nitrification of ammonium and denitrification of nitrite, can save 25% of oxygen needed for nitrification and 40% of carbon source for denitrification, and also decrease reactor volume (Gabarró et al., 2012). Besides, BNR process consisting of partial nitrification and ANAMMOX is another environmental friendly and cost-effective technology for nitrogen removal. In this process, nitrite accumulation firstly happens in the former partial nitrification and then reduce to  $\text{N}_2$  with ammonium as electron donor under anaerobic condition without organic carbon needed (Tang et al., 2011). Basically, stable partial nitrification and nitrite accumulation are critical for these processes.

As nitritation and nitratation are catalyzed by two phylogenetically unrelated groups of autotrophic bacteria, the ammonia oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB) (Sinha and Annachatre, 2007), nitrite accumulation can be achieved by

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accumulation of AOB and inhibition of NOB. For achieving stable nitrification, several common strategies have been reported such as high temperature, low dissolved oxygen (DO) and free ammonia/ free nitrous acid (FA/FNA) inhibition (Ge et al., 2014). High temperature can cause AOB to out-compete NOB because of their different growth rate at temperatures higher than 25 °C (Hellinga et al., 1998) however, controlling high temperature will lead to extra energy consumption which is unfavorable in practical operation. Due to the differences between the Monod saturation constant of oxygen for AOB (0.3 mg/L) and NOB (1.1 mg/L) (Wiesmann, 1994), low DO concentration controlling is proved to be feasible for nitrite accumulation in lab (Bernat, 2001; Wang et al., 2014). However, the optimal DO range is narrow which makes it difficult to obtain reliable partial nitrification in a real-world wastewater treatment plant (Park et al., 2010), and low DO may result in a low ammonium removal rate and biomass yield (Jianlong and Ning, 2004). FA/FNA had been reported for nitrification due to different inhibition FA/FNA values on AOB and NOB. FA inhibition limit for NOB is 0.1–1.0 mg/L, while for AOB it is 10–150 mg/L. In addition, NOB is more sensitive to FNA compared to AOB. Inhibition on NOB occurs with FNA concentrations of 0.011–0.070 mg/L while on AOB FNA should be higher than 0.42 mg/L (Anthonisen et al., 1976). As reported in the literature (Anthonisen et al., 1976), FA/FNA is decided by ammonium/nitrite concentration, pH and temperature and can be calculated by the following equations.

$$\text{FA}(\text{mg/L}) = \frac{17}{14} \times \frac{\text{NH}_4^+ - \text{N} \times 10^{\text{pH}}}{\exp\left[\frac{6334}{273+T}\right] + 10^{\text{pH}}} \times 100\% \quad (1)$$

$$\text{FNA}(\text{mg/L}) = \frac{46}{14} \times \frac{\text{NO}_2^- - \text{N}}{\exp\left[\frac{-2300}{273+T}\right] + 10^{\text{pH}}} \times 100\% \quad (2)$$

According to Eqs. (1) and (2), FA can be the main inhibitor of nitrification at high pH, while FNA become the main inhibitor of nitrification when pH is low. Based on this, FA was commonly selected as a key parameter to realize nitrite accumulation, and compared to pH value and temperature, influent ammonia concentration had been proved more practical to acquire desirable FA for realizing inhibition on the NOB when treating high ammonium wastewater, such as landfill leachate, fertilizer wastewater and petrochemical effluent (Wei et al., 2014). Nevertheless, FA concentration is unstable and will decrease with the removal of ammonia nitrogen during aeration phase, in which could lose inhibition effect on NOB (Wei et al., 2014) and damage of nitrite accumulation. Thus it is worth to find a new way to main appropriate FA concentrations for nitrite accumulation.

Biological Aerated Filter (BAF) is a practical microbial immobilized process that has been successfully used in the traditional nitrification and denitrification process. The packing media in the BAF plays an important role in maintaining a high amount of activated biomass and a variety of microbial populations (Chang et al., 2009). As a kind of microporous aluminosilicate minerals, zeolite has been proved to a superior material for ammonium removal in biofilm process. It was reported that specific immobilization and enrichment of AOB could be achieved in the BAF with zeolite as fillings (ZBAF) due to the FA inhibition to NOB (Li et al., 2013), which suggested the possibility of zeolite as a potential BAF fillings to realize excellent nitrification. However, there are few reports about partial nitrification conducted in the ZBAF with continuous operation and information regarding the effects of zeolite on the ion-exchange and nitrification process is scarce.

In this study, ZBAF was established with different ammonium loading and influent ammonium concentrations to investigate whether excellent partial nitrification could be realized in ZBAF and the working mechanisms of this reactor. The partial nitrifica-

tion performance of ZBAF, and transport and transformation of nitrogen, nitrite accumulation mechanism and kinetics of partial nitrification were studied. In addition, shift in microbial community between seed sludge and biomass in zeolite was also analyzed using Illumina high-throughput sequencing technology.

## 2. Materials and method

### 2.1. The ZBAF set-up and operations

A ZBAF with a height of 1.2 m and internal diameter of 10 cm was used in this study (Fig. 1). The working volume of the reactor was 10 L, in which about 7.1 L was filled with natural zeolite as the biomass carriers. Table 1 gives the properties of natural zeolite. Synthetic wastewater was fed to the reactor using a peristaltic pump with continuous feeding, fully mixing with the air in the bottom. The reactor was covered with a heating jacket to control the temperature of the liquid flowed between  $25.0 \pm 1.0$  °C. The air was pumped to ZBAF by an air blower with gas flow was controlled at a rate of 0.4 mL/min. Corresponding to the air flow rate, DO concentration at the outlet of filter inside ZBAF was between 4.0 and 6.0 mg/L. The DO was monitored by a DO probe connected with display screen. Backwash operation was proceeded every seven days followed by 5 min of air scour with air flow rate of 4 L/min and then 10 min of combined air scour and water backwash.

### 2.2. Wastewater and seed sludge

The composition of synthetic wastewater was as follow:  $\text{NH}_4\text{-N}$  (as  $(\text{NH}_4)_2\text{SO}_4$ ), 150–550 mg/L;  $\text{NaHCO}_3$ , 2250–8250 mg/L;  $\text{NaH}_2\text{PO}_4$ , 10.0 mg/L;  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ , 5.6 mg/L;  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , 300 mg/L;  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ , 24.0 mg/L; and 1.0 ml/L of trace element solution. The composition of trace element solution was derived from the literature (Aslan and Dahab, 2008).

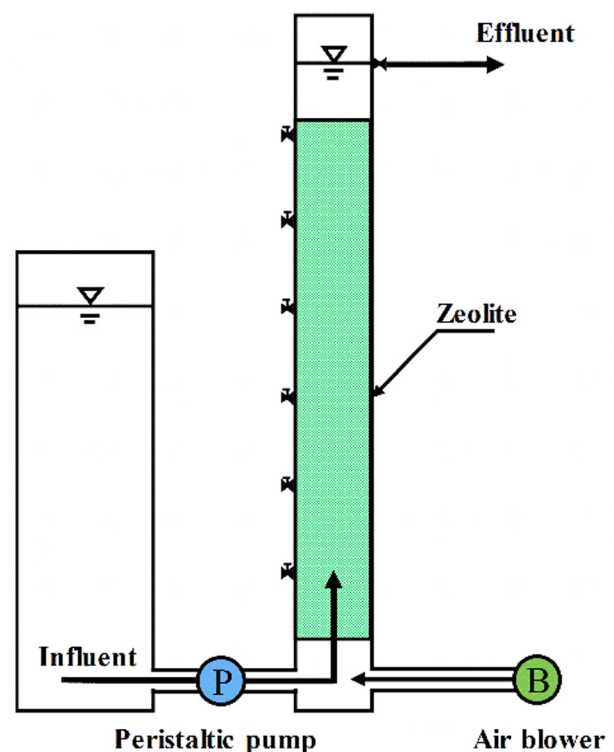


Fig. 1. Schematic diagram of the ZBAF.

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