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Nitrogen removal via nitritation pathway for low-strength ammonium wastewater by adsorption, biological desorption and denitrification

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ABSTRACT

Stable nitritation for low-strength ammonium wastewater was the key obstacle for cost-effective and low-carbon biological nitrogen removal. A zeolite biological fixed bed (ZBFB) and an anoxic sequencing batch reactor (ASBR) were successfully applied for achieving nitritation-denitrification of low-strength ammonium wastewater by adsorption, biological desorption and denitrification. Based on free ammonia inhibition on biofilm, stable nitrite accumulation could be realized with suitable operational time and aeration in biological desorption. During cycle operation, adsorption effluent NH_4^+-N kept at 3.0–4.0 mg/L, biological desorption effluent NO_2^--N maintained at 226.8–243.2 mg/L with average nitrite accumulation ratio of 97.18%, and nitrite removal rate was about 0.628–0.672 kg NO_2^--N·m^{-3}·day^{-1}, revealing obvious feasibility of ZBFB and ASBR for low-strength ammonium wastewater treatment. High-throughput sequencing analysis results further presented significant microbial community variations happened after cycle operation, with ammonia oxidizing bacteria enrichment and nitrite oxidizing bacteria inhibition in ZBFB and dominance of denitrifiers in ASBR.

1. Introduction

As discharge of wastewater with ammonium can lead to eutrophication in receiving water (Huang et al., 2018), wastewater nitrogen emission standard in China has become increasingly strict, especially for environmental sensitive areas. For low-strength ammonium wastewater, in addition to meeting this strict discharge standard, it is still a big challenge to remove nitrogen compounds from low-strength ammonium wastewater economically and energy saving. Compared with complete nitrification-denitrification, nitritation-denitrification can save approximately 25% aeration demand and about 40% carbon source (Pollice et al, 2002; Peng and Zhu, 2006), while ANAMMOX can almost reduce 60% of aeration energy consumption and avoid addition of carbon source (Mariusz et al., 2017; Raudkivi et al., 2017; Rikmann et al., 2014), showing promising advantages in treating low-strength ammonium wastewater. Obviously, these technologies are definitely based on stable nitrite accumulation, which means that significant nitritation of low-strength ammonium should be realized preferentially.

The key for excellent nitritation lies in enrichment of ammonia oxidizing bacteria (AOB) and inhibition of nitrite oxidizing bacteria (NOB). Based on this, it was commonly reported that high temperature (van Dongen et al., 2001; Hellinga et al., 1998), low dissolved oxygen (DO) (Bernat, 2001; Wang et al., 2014), intermittent aeration (Jardin and Hennerkes, 2012), inhibition of free ammonia (FA) or free nitrous acid (FNA) (Blackburne et al., 2008; Ge et al., 2014) could be successfully applied for desired nitritation. However, most of these studies focused on high-strength ammonium wastewater, while few took account for low-strength ammonium because it is difficult to control NOB growth in treating low-strength ammonium wastewater (De Clippeleir et al., 2001).

Though the growth rate of AOB is higher than that of NOB at temperature higher than 25 °C (Hellinga et al., 1998), high temperature is not suitable for low-strength ammonium wastewater due to extra energy needed. Low DO was reported to achieve nitritation of low-strength ammonium wastewater (Xu et al., 2016). Nevertheless, strict operation should be considered for low DO and nitritation can be easily destroyed when DO is out of control. Besides, as oxygen is donor acceptor for ammonia oxidation, low DO may also result in a low ammonium removal rate as well as low biomass yield (Wang and Yang,

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2004). Intermittent aeration could successfully suppress NOB by turning off aeration before NOB activity restored for nitrite accumulation in treating low-strength ammonium wastewater (Ma et al., 2015). However, AOB could be affected by short aeration duration of intermittent aeration, which was adverse to stable nitritation (Miao et al., 2017). Anthonisen et al. (1976) had reported that FA inhibition limit for AOB and NOB was 10–150 mg/L and 0.1–1.0 mg/L, while FNA inhibition limit for these two bacteria was higher than 0.42 mg/L and 0.011–0.070 mg/L, respectively. That is to say, keeping an appropriate FA or FNA range may be available for nitritation for low-strength ammonium wastewater. Unfortunately, FA or FNA was hard to maintain in most cases because of gradual decrease of ammonium and poor nitrite accumulation in aeration phase (Wei et al., 2014).

As a kind of superior adsorbent for ammonium, zeolite was suitable for low-strength ammonium wastewater treatment with high standard effluent by adsorption. It was also reported that zeolite could be applicable biofilm carrier for enrichment of AOB (Li et al., 2013). During ammonium adsorption, adsorption equilibrium will eventually appear and portion of ammonium remains in liquid phase. Bases on this, the application of zeolite for biofilm carrier may be highly potential for keeping appropriate FA level in the reactor. This was verified by Yang et al. (2017) who found that zeolite biological aeration filter (ZBAF) could successfully realize efficient nitritation for high-strength ammonium wastewater.

For low-strength ammonium wastewater, ammonium concentration is typically low (usually less than 60 mg/L). In order to obtain high standard effluent, adsorption of low-strength ammonium wastewater by zeolite could be carried out firstly. Then adsorbed ammonium could be desorbed by biochemical reaction of nitrified biofilm on zeolite. Based on the enriched ammonium in zeolite, it might be possible to maintain an appropriate FA range in the liquid due to adsorption equilibrium, which would inhibited NOB for nitrite accumulation during the desorption process. If this was feasible, ammonium in the wastewater could be transformed to nitrite for the next nitrogen removal (such as nitritation-denitrification) and adsorption capacity of zeolite could recover for adsorption again, which means that high treatment standard, economical and energy saving treatment for low-strength ammonium wastewater is possible. However, there is scarcely any relevant report focusing on this issue in the literature and whether this hypothesis is feasible is still unclear.

In this study, a zeolite biological fixed bed reactor (ZBFB) was established for treatment of low-strength ammonium wastewater. Adsorption was firstly executed for high standard effluent. Then ZBFB was desorbed by nitrified biofilm for adsorption capacity recovery of zeolite (biological desorption). During biological desorption, nitrified products and effects of several factors (type of alkalinity, alkalinity dosage, reaction time, aeration) were investigated for estimating nitritation performance and determining optimal parameters. Subsequently, cycles of adsorption and biological desorption were carried out to verify feasibility of ZBFB for low-strength ammonium wastewater. In addition, an anoxic sequencing batch reactor (ASBR) was also started up for denitrification of biological desorption effluent, which further demonstrated possibility of stable nitritation-denitrification for low-strength ammonium wastewater. Moreover, nitrogen balance analysis, variations of microbial community structure of biomass and potential functional bacteria were also discussed.

2. Materials and methods

2.1. Experimental setup

The schematic diagram of adsorption-biological desorption-denitrification for low strength ammonium wastewater was shown in Fig. 1. The ZBFB with a diameter of 7.0 cm and height of 150 cm was filled with natural zeolite (V = 3.5 L, mean particle size: 1.80 mm, surface area: 16.20 m^2, adsorption capacity: 3–4 mg NH_4^+-N/g) as packings. In the adsorption step, low-strength ammonium wastewater was pumped into the bottom of ZBFB. After adsorption, aeration was supplied to ZBFB by an air compressor with rotor flow meter for controlling desired aeration volume and alkalinity was supplied with inner circulation for biological desorption. During this process, DO was controlled at 6.0–7.0 mg/L. The biological desorption effluent was discharged at the bottom of ZBFB. An ASBR with working volume of 7.5 L was started up and applied for denitrification of biological desorption effluent. Mechanical mixer (30 rpm) was installed in ASBR for stirring and temperature was controlled at about 27.0 ± 1.0 °C. The whole experiment was carried out in the key Lab of Pollution Control and Ecosystem Restoration in Industrial Cluster, South China University of Technology, Guangzhou, China.

2.2. Synthetic wastewater, chemical reagent and seed sludge

Low-strength ammonium wastewater with 50 mg/L NH_4^+-N was made from tap water with NH_4Cl as ammonium source. In the biological desorption step, $NaHCO_3$ or Na_2CO_3 was supplied for alkalinity. Glucose ($C_6H_{12}O_6{\cdot}H_2O$) chosen as carbon source for denitrification. $NaHPO_4$, 10.0 mg/L; $CaCl_2{\cdot}2H_2O$, 5.6 mg/L; $MgSO_4{\cdot}7H_2O$, 300 mg/L; $FeSO_4{\cdot}7H_2O$, 24.0 mg/L; and 1.0 mL/L of trace element solution (Yang et al., 2017; Aslan and Dahab, 2008) were added for biological desorption and denitrification, respectively. All reagents used in this study were AR grade (Purity ≥ 99.7%).

Activated sludge with mixed liquid suspended solid about 5000 mg/L was picked from an aeration unit of pilot AO process for biological nitrogen removal in our laboratory for inoculation of all reactors in this study.

2.3. Experimental procedure

2.3.1. Operation of ZBFB

ZBFB was carried out for 3 stages including start-up, single factor experiment and cycle operation.

Stage 1 (day 1–15): Synthetic wastewater was pumped into ZBFB with inflow rate of 2 V/h for ammonium adsorption, where V is the volume of zeolite packings. The adsorption was stopped after 30 h operation when effluent NH_4^+-N was about 3 mg/L (Supplementary data can be found in e-version of this paper online). Then 500 mL activated sludge, 3 g/L $NaHCO_3$ and other necessary elements were added into ZBFB for biofilm cultivation. During this process, aeration was controlled at 0.06 Nm^3/h with constant inner circulation (2 V/h) and 1 g/L $NaHCO_3$ was added every day. After almost 15 days, the start-up was finished with nitrite accumulation ratio (NAR) higher than 66.7%. Finally, liquid in ZBFB was discharged and backwash was run with 1 V of adsorption effluent.

Stage 2 (day 16–48): After the stage 1, adsorption was carried out again with inflow rate of 2 V/h in ZBFB. Adsorption effluent NH_4^+-N was detected and controlled lower than 5 mg/L with constant adsorption operation time. Then adsorption was stopped and biological desorption went on with constant inner circulation (2 V/h) and different kind of alkalinity, different dosage of Na_2CO_3, different aeration. After every biological desorption, ZBFB was switched for adsorption of low-strength ammonium wastewater and then again for biological desorption. This stage aimed to determine optimal dosage of alkalinity and aeration for biological desorption.

Stage 3 (day 49–78): In this stage, ZBFB was continuously run for repetitive adsorption and biological desorption with optimal biological desorption operational parameters obtained from stage 2. Every cycle included 3 h of adsorption, 16 h of biological desorption and 1 h of backwash. 30 cycles were carried out during this stage and all biological desorption effluent was stored for the next denitrification.

2.3.2. ASBR

ASBR was started up with inoculation of 7.5 L activated sludge and