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Nitrogen removal via nitrite in a partial nitrification sequencing batch biofilm reactor treating high strength ammonia wastewater and its greenhouse gas emission

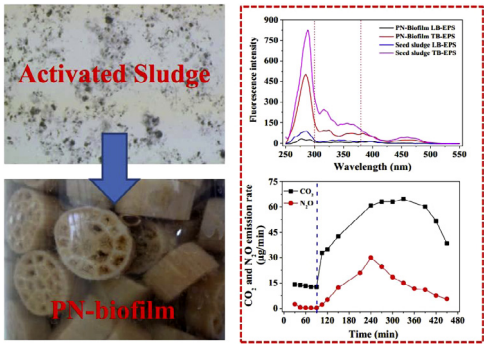
Dong Wei [a], Keyi Zhang [a], Huu Hao Ngo [b], Wenshan Guo [b], Siyu Wang [a], Jibin Li [a], Fei Han [a], Bin Du [a,*], Qin Wei [c]

[a] School of Resources and Environment, University of Jinan, Jinan 250022, PR China
[b] School of Civil and Environmental Engineering, University of Technology Sydney, Broadway, NSW 2007, Australia
[c] Key Laboratory of Chemical Sensing & Analysis in Universities of Shandong, School of Chemistry and Chemical Engineering, University of Jinan, Jinan 250022, PR China

HIGHLIGHTS

- Partial nitrification was successfully achieved and maintained in a SBBR.
- PN and PS contents were reduced during the achievement of PN process.
- 3D-EEM implied that LB-EPS and TB-EPS had similar chemical compositions.
- CO_2 and N_2O emission amounts per cycle were 67.7 mg and 16.5 mg.

GRAPHICAL ABSTRACT



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ABSTRACT

In present study, the feasibility of partial nitrification (PN) process achievement and its greenhouse gas emission were evaluated in a sequencing batch biofilm reactor (SBBR). After 90 days' operation, the average effluent NH_4^+-N removal efficiency and nitrite accumulation rate of PN-SBBR were high of 98.2% and 87.6%, respectively. Both polysaccharide and protein contents were reduced in loosely bound extracellular polymeric substances (LB-EPS) and tightly bound EPS (TB-EPS) during the achievement of PN-biofilm. Excitation-emission matrix spectra implied that aromatic protein-like, tryptophan protein-like and humic acid-like substances were the main compositions of both kinds of EPS in seed sludge and PN-biofilm. According to typical cycle, the emission rate of CO_2 had a much higher value than that of N_2O, and their total amounts per cycle were 67.7 and 16.5 mg, respectively. Free ammonia (FA) played a significant role on the inhibition activity of nitrite-oxidizing bacteria and the occurrence of nitrite accumulation.

1. Introduction

Biological nitrification-denitrification is typically utilized for nitrogen removal through two individual sequential processes: aerobic nitrification with the terminal conversion of NH_4^+-N to

* Corresponding author.
E-mail address: dubin61@gmail.com (B. Du).

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NO_3^--N and the subsequent anoxic denitrification with the conversion of NO_3^--N to molecular nitrogen (Adav et al., 2009). Recently, much attention has been paid to partial nitrification (PN) via nitrite process as a novel concept of BNR process. It theoretically saves approximately 25% of oxygen supply for nitrification, 40% of organic carbon as electron donor for denitrification and achieves a lower sludge production (Peng and Zhu, 2006). To date, PN process has been successfully applied for wastewater treatment containing high-nitrogen concentration or low carbon/nitrogen (C/N) ratio, such as municipal wastewater, landfill leachate and anaerobic sludge digester liquor etc (Ge et al., 2014; Qiao et al., 2008; Wang et al., 2010).

The main influencing factors to achieve and maintain PN process are including dissolved oxygen (DO), pH value, sludge retention time (SRT), free ammonia (FA) and free nitrous acid (FNA) etc (Wei et al., 2015a). Till now, most of partial nitrification processes are reported by using synthetic or real wastewater as substrate under well-controlled activated sludge reactors (Chen et al., 2016). However, little information is available for the achievement and maintenance of PN via nitrite through a biofilm system. Compared to suspended-growth activated sludge, biofilm system has the ability to provide different sub-zones for various types of bacteria, and therefore protect the slow-growing nitrifying bacteria from washout in the competition of heterotrophic bacteria (Yin et al., 2015). Therefore, various biofilm systems, such as sequencing batch biofilm reactor (SBBR), moving bed biofilm reactor (MBBR) and fixed bed biofilm reactor (FBBR) etc, are increasingly being applied for treating various municipal and industrial nitrogen-containing wastewaters (Gilbert et al., 2014; Jin et al., 2012; Zhang et al., 2016). Especially, it is evident that the degradation of pollutants largely depends on the amount and activity of microorganisms, which is attached onto the solid surface of carrier for biofilm formation in the presence of extracellular polymeric substances (EPS) (Czaczyk and Myszka, 2007; Li et al., 2016). Microbial EPS likely have a dynamic double-layered structure of loosely bound EPS (LB-EPS) diffused from the tightly bound EPS (TB-EPS) that surround the cells (Zhao et al., 2015). Moreover, the two kinds of EPS fractions play different roles in maintaining the sludge floc structure and functions (Liu et al., 2010). However, there is still a lack of destructive research to evaluate the changes of EPS in a PN-biofilm achievement, and thus, the major components of double-layered EPS have not been characterized.

Additionally, biological wastewater treatment is recognized as one of the major sources of greenhouse gas (GHG) emissions, particularly carbon dioxide (CO_2) and nitrous oxide (N_2O), causing a major challenge to global climate (Kong et al., 2016). It is well reported that CO_2 and N_2O can be produced from the processes of organic matter degradation and biological nitrogen removal, respectively. Especially, N_2O has about 300 times higher global warming potential than that of CO_2. As a by-product and intermediate product, N_2O emission from biological wastewater treatment is influenced by various operational parameters, including DO concentration, nitrite, COD/N, temperature and toxic compounds etc (Kampschreur et al., 2009). In particular, nitrite accumulation was considered to be a major parameter for affecting the emission of N_2O in both nitrification and denitrification stages, and therefore mitigate the environmental benefits of PN process (Wei et al., 2014a). Therefore, it is desirable to evaluate the GHG emissions in PN biofilm system in order to provide a better understanding on the basis of PN process.

In present study, the feasibility of PN process achievement in a SBBR was evaluated treating high strength ammonia wastewater. For more detailed insights, the changes of LB-EPS and TB-EPS were qualitatively and quantitatively analyzed by using chemical and fluorescence spectroscopic approaches during the PN-biofilm formation. After the PN-SBBR achieving stable operation, the productions of CO_2 and N_2O were carried out to evaluate the behavior of GHG emissions from wastewater treatment through PN process. The acquired results would help us to fully reveal PN-biofilm process by considering the point of GHG production.

2. Materials and methods

2.1. SBBR system and operational procedure

Fig. S1 shows the schematic diagram of the lab-scale SBBR used in present study, which was made of plexiglas with a working volume of 3.4 L (12 cm in diameter and 30 cm in height) at room temperature (25–27 °C). The SBBR was filled with 40% cylindrical biocarriers (K3, plastic media), and each carrier with 25 mm in diameter and 12 mm in height, respectively. The specific surface area and bulk density of each carrier were 500 m^2/m^3 and 110 kg/m^3, respectively. Influent wastewater was prepared in a storage tank (60 L) and introduced to the bottom of the reactor through a water pump. Air was supplied at the bottom of reactor by using an air pump. The reactor was automatically operated through a time controller.

The anoxic-aerobic SBBR was sequentially operated at a cycle of 480 min, consisting of 5 min influent, 85 min anoxic phase, 360 min aerobic reaction, 20 min settling and 10 min effluent and idle. Electromagnetic stirrer was used as the mixing method to keep the suspension of sludge during anoxic and aerobic phases. The reactor was operated with a volumetric exchange ratio of 50%, resulting in hydraulic retention time (HRT) of 16 h. Activated sludge was taken from a plant treating soy protein wastewater as the inoculation of biofilm system. The plant was located at Shandong province in China treating high strength ammonia wastewater. The sludge retention time (SRT) and nitrogen loading rate of plant were about 20 day and 0.15 kg NH_4^+-N/(m^3·day), respectively. The initial mixed liquor suspended solids (MLSS) concentration in the reactor was 3.0 g/L.

2.2. Synthetic wastewater

The compositions of synthetic wastewater were used in the experiment as follows: chemical oxygen demand (COD, as sodium acetate), 600 mg/L; NH_4^+-N (as ammonium chloride), 200 mg/L; K_2HPO_4, 112 mg/L; $CaCl_2$, 40 mg/L; $MgSO_4{\cdot}2H_2O$, 20 mg/L; $FeSO_4{\cdot}2H_2O$, 20 mg/L and trace element solution 1.0 ml/L. The composition of the trace mineral solution was as follows: H_3BO_3 0.05 g/L, $ZnCl_2$ 0.05 g/L, $CuCl_2$ 0.03 g/L, $MnSO_4{\cdot}H_2O$ 0.05 g/L, $(NH_4)_6MoO_24{\cdot}4H_2O$ 0.05 g/L, $AlCl_3$ 0.05 g/L, $CoCl_2{\cdot}6H_2O$ 0.05 g/L, $NiCl_2$ 0.05 g/L. The ratio of bicarbonate to NH_4^+-N was kept above 8.0 mg/mg to ensure the growth requirements of nitrifying bacteria, as similarly reported by Shi et al. (2009). As a reslut, the influent pH values of wastewater were controlled between 8.0 and 8.5.

2.3. EPS extraction and spectra analysis

A heating method was used to extract the double-layered EPS from seed sludge and biofilm, including loosely bound EPS (LB-EPS) and tightly bound EPS (TB-EPS), and the detailed procedure could be found in the previous literature (Li and Yang, 2007). 3D-EEM spectra were obtained by using a fluorescence luminescence spectrometer (LS-55, Perkin-Elmer Co., USA). EEM spectra were gathered with scanning emission (Em) spectra from 280 to 550 nm at 0.5 nm increments by varying the excitation (Ex) wavelength from 200 to 400 nm at 10 nm increments. Synchronous fluorescence spectra of EPS samples were measured by ranging the excitation wavelengths from 250–550 nm with a constant offset ($\Delta\lambda$) of 60 nm. The width of the Ex/Em slit and scanning speed