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Nitrogen removal performances of a polyvinylidene fluoride membrane-aerated biofilm reactor

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A B S T R A C T

A membrane-aerated biofilm reactor (MABR) with polyvinylidene fluoride (PVDF) hollow fiber membranes was developed, and the nitrogen removal performances with the increase of influent NH_4^+-N concentration from 30 mg/L to 120 mg/L were investigated. The results indicated that simultaneous nitrification and denitrification (SND) process was realized in this MABR system, and the nitrogen removal performances were affected by the influent NH_4^+-N concentration. The limitation of influent NH_4^+-N concentration for this MABR system was 70 mg/L with a maximum specific NH_4^+-N conversion rate and specific total nitrogen removal rate of 55.67 and 52.87 kg/m^3 d, showing better performances compared with other studies. The nitrogen removal was realized through nitrification, denitrification and possible anaerobic ammonium oxidation (ANAMMOX) with partial nitrification. The polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE) was used to analyze the microbial community of biofilm in this MABR system, revealing the bacteria community diversity of biofilm. Besides various ammonia-oxidizing bacteria and nitrite-oxidizing bacteria, *Nitrosomonas* sp. and *Planctomycetes* sp. were found to conduct ANAMMOX with partial nitrification under oxygen limited condition.

Introduction

Wastewater contains a large quantity of organic matters, nitrogen and phosphorus. Up to now, researchers have mainly focused on bioprocesses for pollutant removal to protect water environment. Conventional biological nitrogen removal processes are usually conducted at least in two separate reactors or two separate phases for aerobic nitrification and anoxic denitrification. However, biofilm systems do demonstrate some advantages on nitrogen removal in a single bioreactor (Fu et al., 2010), which is known as simultaneous nitrification and denitrification (SND) process.

Recently, membrane-aerated biofilm reactor (MABR) has been proposed as a promising alternative to conventional biological wastewater treatment, and shows the advantages of lower operating cost and lower emission of volatile pollutants, compared with conventional bubbled diffuser systems (Rishell et al., 2004). In a MABR system, the role of membranes is twofold, as carrier to immobilize microorganisms, and as an oxygen supplier to provide bubbleless aeration for high oxygen transfer efficiency (LaPara et al., 2006; Lackner et al., 2010). The membrane used in MABR can be classified into microporous hydrophobic membrane, dense membrane and composite membrane (Hou et al., 2013). In this paper, polyvinylidene fluoride (PVDF) hollow-fiber membranes were used in MABR system, which is less applied in MABR system currently.

MABR creates a new symbiotic environment for aerobic and anaerobic microorganisms for nitrogen removal. In MABRs, SND can be achieved in a single biofilm system through providing oxygen and nutrients from opposite sides of the biofilm (Ni and Yuan, 2013). The air flows through membranes from membrane lumen and the oxygen is utilized by biofilm attached on membrane surface, meanwhile, the pollutants penetrate biofilm from wastewater side. Compared with conventional (co-diffusion) biofilm, the opposing substrate diffusion leads to a larger active thickness in biofilm, which provided the environment for SND process (Potvin et al., 2012; Gilmore et al., 2013). It is generally assumed that the nitrifying bacteria, including ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB), are dominant in the oxygen-rich region of membrane–biofilm interface, while denitrifying bacteria are dominant in the anoxic region of biofilm–liquid interface. The

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nitrogen removal can also be realized through combining the anaerobic ammonium oxidation (ANAMMOX) with partial nitrification in one reactor which named completely autotrophic nitrogen removal over nitrite (CANON) or oxygen-limited autotrophic nitrification-denitrification (OLAND) (Gong et al., 2008).

Currently, MABR has been mainly studied on municipal wastewater treatment, to remove carbon and nitrogen (Hu et al., 2008). The suitable influent ammonium concentration mostly ranged from 20 to 50 mg/L. There are few literature to reveal the MABR performances when the influent $NH_4^+ - N$ concentration of municipal wastewater or some kinds of industrial wastewater (such as livestock wastewater etc.) is higher. In this study, the nitrogen removal performances with the increase of influent $NH_4^+ - N$ concentration were investigated to find the limitation of nitrogen removal in the MABR system. Besides, the diversity of microbial communities was analyzed by polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE) to explore the nitrogen removal bioprocess.

Materials and methods

Materials

The artificial wastewater was composed of $C_6H_{12}O_6$, $CH_3COONa \cdot 3H_2O$ and NH_4Cl as the main sources of COD and $NH_4^+ - N$, and the concentration of COD and $NH_4^+ - N$ changed according to the requirement of experiments. The other components of artificial wastewater included $NaHCO_3$ (200 mg/L), KH_2PO_4 (11 mg/L), K_2HPO_4 (14 mg/L), $MgSO_4 \cdot 7H_2O$ (10 mg/L), $FeSO_4 \cdot 7H_2O$ (10 mg/L), $CaCl_2 \cdot 2H_2O$ (10 mg/L), $MnSO_4$ (0.1 mg/L) and trace element solution (1 ml/L). The trace element solution was prepared according to literature (Syron and Casey, 2008).

PVDF hollow-fiber membrane was used in the MABR system. The inner diameter of membrane was 1.0 mm, and the membrane thickness was 0.7 mm. The PVDF membrane belongs to microporous membrane with an average pore size of 0.2 μm, and the pores were evenly distributed in the surface of PVDF membrane. The membrane module included 163 fibers in parallel with a total surface area of 0.453 m^2, providing a specific surface area of 120.53 m^2/m^3.

Bacterial adhesion properties of PVDF membrane

Activated sludge with mixed liquid suspended solids of 6000 mg/L was centrifuged, the supernatant was discarded. The solids were washed with de-ionized water twice to remove residual substrates and extracellular polymer substance (EPS). The washed sludge was resuspended in a phosphate buffer saline ($pH = 7.2$) and 100 ml suspension was infused into the flask. A PVDF membrane with known surface area was immersed into the above sludge suspension (Liu et al., 2007). After shaking the flask at 100 r/min and 30 °C for 7 d, attached bacteria were sonicated for 30 min. The detached biomass was weighed and the total organic carbon (TOC) was analyzed with a TOC analyzer (TOC-V_{CPH}, Shimadzu).

MABR system start-up and operation

MABR system is schematically presented in Fig. 1. The reactor was constructed with plexiglas and had a working volume of 3.8 L with 250 mm length, 50 mm breadth and 360 mm height. The wastewater was supplied with a peristaltic pump at a flow velocity of 2.64 ml/min, and the effluent was discharged by overflow. Hydraulic retention time (HRT) of wastewater was maintained 24 h, system temperature was maintained at 20±2 °C, and influent pH was kept between 7.0 and 8.0. Compressed air was supplied to the membrane lumen by an air pump. Gas pressure and flow rate were controlled with a valve and adjusted by gas flow gages. The intramembrane pressure was kept at 0.025 MPa.

In biofilm culturing phase, the MABR was seeded with activated sludge of 1000 ml. After culturing for 15 d, the stable biofilm was formed and the sludge was discharged from the reactor. When the MABR system stably operated, the influent pollutant concentration was periodically increased to find out the limitation of nitrogen removal in the MABR system. The influent $NH_4^+ - N$ concentration ranged from 30 to 120 mg/L. The C/N ratio was maintained at 10:1 for successful nitrogen removal according to Roy et al. (2010). The performances of MABR system were evaluated with the COD, $NH_4^+ - N$ and total nitrogen (TN) removal.

To investigate the nitrogen conversion performance, the reactor was changed into sequencing batch reactor. After discharging all

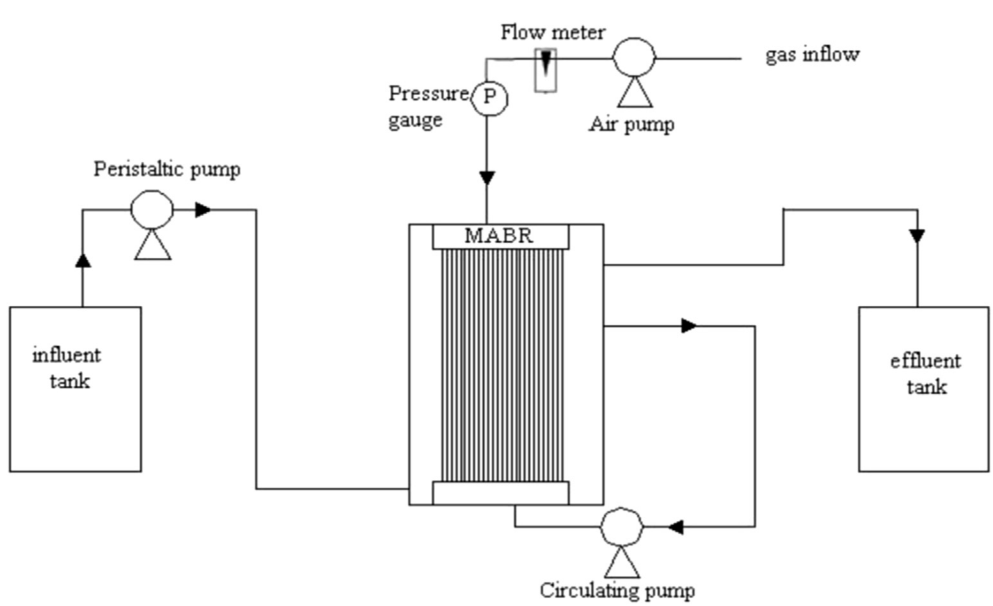

Fig. 1. Schematic diagram of PVDF membrane-aerated biofilm reactor.