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Enhanced nitrogen removal by membrane-aerated nitritation-anammox in a bioelectrochemical system

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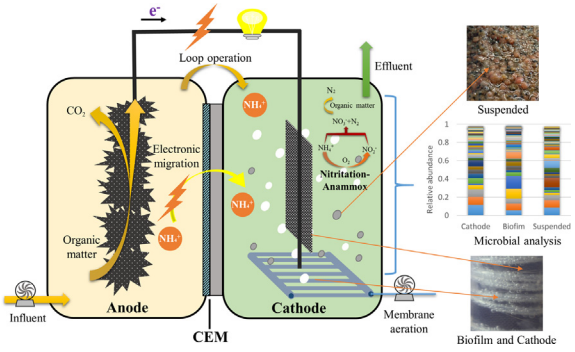
HIGHLIGHTS

- Membrane-aerated nitritation-anammox is conducted in a bioelectrochemical system.
- Biocathode with anammox can improve both organic and nitrogen removal.
- This BES is potentially energy efficient and has low specific energy consumption.
- AOB, anammox, denitrifier, and electrogenic bacteria are abundant in the cathode.
- The distribution for microorganisms is affected by aeration and location.

GRAPHICAL ABSTRACT



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ABSTRACT

A bioelectrochemical system (BES) containing membrane-aerated nitritation-anammox in its cathode has been developed for enhancing nitrogen removal. Long-term performance and microbial community structure were investigated. The BES using loop operation and external voltage achieved the highest total nitrogen removal efficiency of 94.8 ± 7.7%, and COD removal of 98.2 ± 3.3% at hydraulic retention time of 60 h and the lumen pressure of 10 psi. The energy consumption of the system was 0.90 kWh kg N^{-1} or 0.38 kWh kg COD^{-1}. Sequencing analyses revealed that ammonia oxidizing bacteria (0.2–7.4%), anammox bacteria (0.4–10.3%), denitrifying bacteria (5.8–13.1%), and electrogenic bacteria (4.6–12.8%) were in abundance of the microbial community in the cathode chamber, and their distributions were affected by the aeration and physical locations. These results encourage further investigation of membrane-aerated nitritation-anammox in BES for optimization and potential applications with actual wastewater.

1. Introduction

Ammonia is the most abundant inorganic nitrogen in various wastes and wastewaters such as municipal wastewater, landfill leachate and livestock waste. Excessive discharge of ammonia into water environment will create toxic effects to aquatic organisms and cause eutrophication. Ammonia can be removed by using physical, chemical and biological methods, and biological removal is of particular interest because of its cost effectiveness. Conventional biological ammonia removal such as nitrification and denitrification has been widely applied in full scale to treat industrial and municipal wastewater (Van Hulle et al., 2010). Discovery and application of anaerobic ammonium oxidation (anammox) coupled with nitritation has provided an innovative approach for ammonia removal with significant benefits of low energy consumption and chemical input (Kuenen, 2008).

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Precise control of oxygen supply is critically important to the successful operation of nitritation-anammox process, and can be accomplished using gas-permeable membrane (Gong et al., 2007) in so-called membrane-aerated biofilm reactor (MABR). In an MABR, gas-permeable membranes deliver oxygen to biological reactions and meanwhile serve as biofilm support for bacterial immobilization to favor growth of ammonia oxidizing bacteria (AOB) and/or anammox bacteria but repress nitrite oxidizing bacteria (NOB) activity (Downing and Nerenberg, 2008). MABRs have been studied for autotrophic nitrogen removal in the recent years (Gilmore et al., 2013; Gong et al., 2007; Li et al., 2016). However, the presence of organic compounds in the typical wastewater would favor the growth of heterotrophs over anammox bacteria due to very low specific growth rate of anammox bacteria, and thus the reduction of organic compounds becomes necessary prior to the implementation of anammox-based treatment.

As an emerging treatment concept, a bioelectrochemical system (BES) such as microbial fuel cell (MFC) or microbial electrolysis cell (MEC) has been employed to achieve simultaneous organic carbon removal and energy recovery from wastewater. BES takes advantage of microbial interaction with solid electron donors/acceptors to accomplish conversion of organic compounds to electricity (Rabaey et al., 2010). As a result of electron flow, nitrogen compounds may also be removed through ammonium transport or nitrate/nitrite reduction (Kelly and He, 2014). The integration of membrane aeration with BES has been performed to improve contaminant removal and electricity generation. For example, in a membrane-aerated MFCs, more than 99% of COD could be removed and nitrogen removal efficiency was doubled to 52% compared to that of an MFC with diffused aeration (Yu et al., 2011). Membrane aeration could also significantly decrease energy consumption due to enhanced oxygen diffusion efficiency in the MFC cathode chamber. To achieve similar electricity generation, a membrane-aerated MFC required energy input of only 0.05 kWh m^{-3}, much lower than 1.76 kWh m^{-3} of a diffused aeration MFC (Yang et al., 2016).

The advantages of both membrane aeration (e.g., precise control of oxygen supply for nitritation-anammox process) and BES (organic removal and conversion to bioenergy) intrigue the exploration of membrane-aerated nitritation-anammox in BES. Such integration may create mutual benefits like low energy consumption for system operation and removal of multiple contaminants simultaneously. In this study, a BES was designed to accommodate membrane-aerated nitritation-anammox in its cathode chamber and investigated for treating high-strength synthetic wastewater for organic and ammonia removal. The anode would function as a pretreatment step to remove a significant portion of organic compounds and generate electrons to drive ammonium ions transporting across ion exchange membrane into the cathode chamber, where nitritation-anammox would occur. An external voltage was applied to stimulate current generation and thus improve contaminant removal. For comparison, three control experiments were operated. The effects of operation mode and hydraulic retention time (HRT) were systmatically investigated for current density, treatment performance, and energy production/consumption. The microbial community at different locations in the cathode was analyzed and dominant species were identified.

2. Materials and methods

2.1. BES setup

The BES consisted of two compartments, physically separated by a piece of cation exchange membrane (CEM) (Membrane International Inc., Ringwood, NJ, USA). The anodic compartment (370 mL) was placed in a tubular container, creating a cathodic compartment of 1120 mL (Fig. 1). The anode electrode was non-wet proofed carbon brush, which was pretreated by being immersed in acetone overnight and then heated at 450 °C for 0.5 h, while the cathode electrode was wet-proofed carbon cloth (13.5 cm × 30 cm, Zoltek Corporation, St. Louis, MO, USA) containing 4 mg cm^{-2} activated carbon powder (Thermo Fisher Scientific, Bridgewater, NJ, USA) as a catalyst for oxygen reduction reaction (ORR). The activated carbon powder was coated to the carbon cloth by using a 15% polytetrafluoroethylene (PTFE) solution as a binder agent and heat-treated at 370 °C for 0.5 h. A gas-permeable silicone membrane module (5 m active length, outer diameter of 3.18 mm, and inner diameter of 1.98 mm, Silastic laboratory tubing #508-009, Dow Corning Corp., Midland, MI, USA) wrapped around the carbon cloth (Fig. 1), and was connected to a cylinder containing air (Airgas, Radnor, PA, USA). The oxygen supply rate was adjusted by a needle valve and a gas pressure meter. By adjusting the lumen pressure, the DO concentration in the cathode chamber could be controlled under an abiotic condition.

2.2. BES operation

The BES was operated in a continuous mode at room temperature (20 ± 1 °C) with an applied voltage of 0.80 V (3644 A, Circuit Specialists, Inc., Mesa, AZ, USA) across the external resistance of 1 ohm. The anode was fed with synthetic wastewater, composed of 2532 ± 27 mg L^{-1} sodium acetate (2000 ± 25 mg L^{-1} COD), 500 ± 10 mg L^{-1} NH_4^+-N, 15 mg L^{-1} $MgSO_4$, 20 mg L^{-1} $CaCl_2$, 500 mg L^{-1} NaCl, 100 mg L^{-1} $NaHCO_3$, 5.35 mg L^{-1} K_2HPO_4, 2.65 mg L^{-1} KH_2PO_4, and trace elements (Angenent and Sung, 2001). Such a synthetic solution was intended to mimic livestock wastewater or digester effluent. The anode HRT was set at 20, 40, 60, and 90 h by varying the influent flow rate of 0.3, 0.15, 0.1, and 0.07 mL min^{-1}. The anode was inoculated with 50 mL sludge from the anaerobic digester in a local wastewater treatment plant (Pepper's Ferry Wastewater Treatment Authority, Radford, VA, USA). In the non-loop operation, the cathode was continuously fed with a mineral solution containing 420 mg L^{-1} $NaHCO_3$, 27.2 mg L^{-1} KH_2PO_4, 59 mg L^{-1} $MgSO_4$, 180 mg L^{-1} $CaCl_2$, 1 mL L^{-1} trace elements solution I, and 1 mL L^{-1} trace elements solution II (Imajo et al., 2004). The cathode was seeded with 100 mL of inoculum containing both anammox granules and flocs from an upflow anaerobic sludge blanket (UASB) anammox reactor oper-

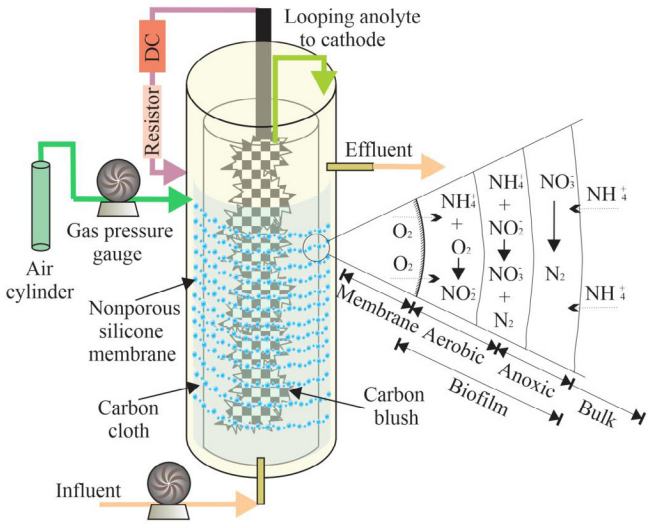

Fig. 1. The schematic diagram of membrane-aerated nitritation-anammox in a BES – loop operation of feeding anode effluent to cathodic chamber with an externally applied voltage. *Note:* "DC" means "direct current".