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Simultaneous nitrogen and organics removal using membrane aeration and effluent ultrafiltration in an anaerobic fluidized membrane bioreactor

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ABSTRACT

Dissolved methane and a lack of nutrient removal are two concerns for treatment of wastewater using anaerobic fluidized bed membrane bioreactors (AFMBRs). Membrane aerators were integrated into an AFMBR to form an aeration membrane fluidized bed membrane bioreactor (AeMFMBR) capable of simultaneous removal of organic matter and ammonia without production of dissolved methane. Good effluent quality was obtained with no detectable suspended solids, 93 ± 5% of chemical oxygen demand (COD) removal to 14 ± 11 mg/L, and 74 ± 8% of total ammonia (TA) removal to 12 ± 3 mg-N/L for domestic wastewater (COD of 193 ± 23 mg/L and TA of 49 ± 5 mg-N/L) treatment. Nitrate and nitrite concentrations were always low (< 1 mg-N/L) during continuous flow treatment. Membrane fouling was well controlled by fluidization of the granular activated carbon (GAC) particles (transmembrane pressures maintained < 3 kPa). Analysis of the microbial communities suggested that nitrogen removal was due to nitrification and denitrification based on the presence of microorganisms associated with these processes.

1. Introduction

An anaerobic fluidized bed membrane bioreactor (AFMBR) was first developed as a post-treatment method for an anaerobic fluidized bed bioreactor (AFBR), achieving 87% removal of chemical oxygen demand (COD), 82% of soluble COD (SCOD), and ~100% of total suspended solid (TSS) (Kim et al., 2011). In addition, a low energy demand of 0.028 kWh/m^3 was estimated for the process, which is 10 times lower than that needed for treatment using anaerobic membrane bioreactor (0.25–1 kWh/m^3) (Liao et al., 2006). Membrane fouling is well controlled in an AFMBR by mechanical scouring due to fluidization of granular activated carbon (GAC) particles. Effective treatment has also been obtained using AFMBRs as a second process that followed treatment by other types of bioreactors. For example, the effluent of a microbial fuel cell (MFC) treating domestic wastewater was reduced to a COD of 16 ± 3 mg/L and TSS of < 1 mg/L, at an AFMBR hydraulic retention time (HRT) of 1 h (Ren et al., 2014). Low effluent COD (11 mg/L) and negligible TSS were also achieved at an HRT of ~1 h for effluent from an anaerobic baffled bioreactor (ARB) (Lee et al., 2015). The combined AFBR and AFMBR process was found to have an additional advantage of effective removal of pharmaceuticals from wastewater (86–100%) (Dutta et al., 2014). A disadvantage of AFMBR treatment, however, is that the effluent contains dissolved methane (16 mL CH_4/L) which would need to be removed prior to discharge (Yoo et al., 2012). In addition, total nitrogen has not been reported to be reduced during AFMBR treatment, since a combination of anoxic and anaerobic conditions are required to achieve nitrification and denitrification.

Membrane-aerated bioreactors (MABRs) were developed to obtain efficient nitrogen removal through the growth of a biofilm on the aeration membranes. Oxygen is added by bubbleless gas transport through the membrane to the biofilm. Nitrification can occur in the stratified biofilm on the membrane, and the nitrate produced can reduce organics concentration to a low levels by denitrification, which in return favors nitrification in the biofilm (Gilmore et al., 2013). Ammonia-oxidizing bacteria (AOB) have been identified in the deep biofilm layer near the membrane, while denitrifiers and heterotrophic bacteria grow on the outer layer (Terada et al., 2003). Stratified biofilm growth of nitrifiers and denitrifiers has also been confirmed using fluorescence in situ hybridization (Gilmore et al., 2013). Typically the biofilms on the membranes are 50–200 μm thick (Casey et al., 1999a), which is usually deep enough to prevent oxygen transfer into the bulk liquid, thus maintaining anaerobic conditions in the solution (Casey et al., 1999b). Membrane aerators immobilized with microorganisms

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were first tested using synthetic wastewater (total organic carbon, TOC, 1000 mg/L and total nitrogen, TN, 58.5 mg-N/L) in batch mode (24 h), achieving a removal efficiency of 97.9% for TOC and 98.3% for TN with a lumen pressure of 245 kPa (pure oxygen) (Hirasa et al., 1991). When treating organic-free synthetic wastewater (217 mg-N/L of ammonium) in continuous-flow mode, separate arrays of hollow fiber membranes (HFMs) that supplied pure bubbleless hydrogen and oxygen in a redox controlled membrane bioreactor obtained a high ammonia removal flux (AR) of 5.8 g-N/m^2 membrane-d, with a nitrate and nitrite removal flux of 4.4 g-N/m^2-d, at a pressure of 861 kPa (Smith et al., 2008). A total nitrogen removal flux (NRF) of 1.7 g-N/m^2-d was achieved using an MABR supplied with air to treat COD-free wastewater (47.1 mM NH_3-N), with 75% removal of the influent nitrogen (Gilmore et al., 2013). Nitrogen and carbonaceous compounds in synthetic wastewater (TOC of 100 mg/L and TN of 25 g-N/m^3) were simultaneously removed using an MABR supplied with air, showing a carbon removal flux (CRF) of 7.4 g-C/m^2-d and NRF of 2.8 g-N/m^2-d (Hibiya et al., 2003). One disadvantage of using MABRs is that they require relatively long hydraulic retention times (HRTs) compared to other processes. HRTs can be as long as several days using air, for example 1 day (Smith et al., 2008), 4–6 days (Gilmore et al., 2013), 1.2–12 days (Gilmore et al., 2009) and 15 days (Terada et al., 2003). However, HRTs can be reduced to only ~1 to several hours by using pure oxygen, for example 0.6 h (Pankhania et al., 1994), 6 h (Hibiya et al., 2003) and 1–10 h (Brindle et al., 1998).

In order to achieve effective ammonia removal in an AFMBR, it was hypothesized that adding a membrane aerator module into the AFMBR could enable simultaneous removal of both carbonaceous and nitrogen compounds in a single aeration membrane fluidized bed membrane bioreactor (AeMFMBR). By infusing oxygen into the system, nitrogen could be removed through nitrification on aeration membranes, and denitrification by microorganisms on the aeration membranes or on GAC and in the mixed liquor. In addition, it was hypothesized that production of methane could be avoided through introducing a membrane aerator, which allows the production of nitrate via nitrification, resulting in an anoxic environment. A bench-scale AeMFMBR was constructed by integrating two different modules, the membrane aerators and the membranes used for ultrafiltration of the effluent, into a single reactor containing fluidized GAC. The performance of the AeMFMBR was initially examined using synthetic influent in fed-batch mode, and then by using synthetic or diluted domestic wastewaters in continuous flow mode. The mechanism of nitrogen removal was investigated through a microbial community analysis of the suspended biomass and the biomass on membrane aerators, and GAC.

2. Material and methods

2.1. Reactor setup

The AeMFMBR made of polyvinyl chloride (PVC, McMaster Carr) contained two chambers, one for filtration (lower section) and the other for aeration (upper section), with a total volume of 4.5 L (Fig. 1). The aeration membrane module contained 135 polyvinylidene fluoride (PVDF) HFMs (pore size of 0.1 μm, Kolon Inc., South Korea) that were sealed at one end. The ultrafiltration membrane module used to filter the wastewater had 54 PVDF HFMs. The total surface area was estimated to be 0.08 m^2 for the aeration membrane module (18 m^2/m^3), and 0.03 m^2 for the filtration membranes (7 m^2/m^3). A magnetic water pump (50 px-x, 1100 GPH, Pan World, Japan) was used to keep the mixed liquor recirculated at a constant flowrate of 4.3 ± 0.9 L/min. Two peristaltic pumps (model no. 7523-90, Masterflex, Vernon Hills, IL) were used for influent and effluent pumping. A mass flow controller (0–10 LPM, Air/He/Ar, Cole-Parmer, US) was used to measure the air flowrate, and a pressure gauge (type1490, Ashcroft, Stratford, CT) was used to measure the air pressure. GAC particles (45 g/L; DARCO MRX, 10 × 30 mesh, Norit Activated Carbon, Cabot, GA) were added into the filtration chamber for biofilm growth and to control membrane fouling.

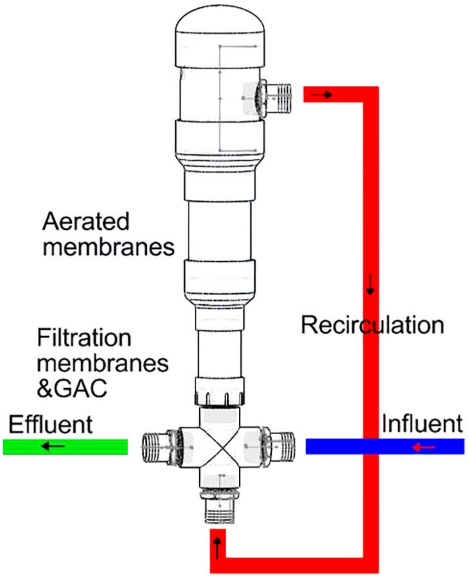

Fig. 1. Schematic of the AeMFMBR showing locations of the aeration and filtration membranes.

2.2. Operation

AeMFMBR operation was separated into six phases, with each phase used to sequentially examine the different aspects of the AeMFMBR components and test conditions, for example operation only with aeration membranes compared to operation with GAC and organic carbon in the feed, to identify the impact of the organic carbon on nitrogen removal. Each of these phases are identified with notation to indicate the specific aspects of operation, as follows (see Supplemental information): B for fed batch operation, or C for continuous flow operation; HN for high (~240 mg-N/L) and LN for low (50–80 mg-N/L) nitrogen concentrations; S for synthetic wastewater, and W for actual domestic wastewater; G for operation with GAC particles added to the reactor; U for operation with ultrafiltration of the effluent; and P for tests with a higher air pressure used in the aeration module compared to other tests (Table 1). For example, phase 3B-SG indicates phase 3 operation with fed batch conditions, a synthetic wastewater feed, and GAC fluidization (but no ultrafiltration of the effluent).

The membrane aerator (air flowrate of 1 mL/min) was inoculated with sludge from a nitrification tank (Pennsylvania State University Wastewater Treatment Plant) and feed solution (40 mM NH_4HCO_3, 14.3 mM NaCl, 3.7 mM $KHCO_3$, 0.8 mM $KHSO_4$, 1.25 mM KH_2PO_4, 0.83 mM $MgSO_4$, 1.23 mM $CaCl_2$, and 0.11 mM $FeCl_3$) (Gilmore et al., 2013) in a column with stirring for 50 days prior to phase 1B-HN. Each time the operational conditions were changed the reactor was operated for at least one week under the new conditions for reactor acclimation. The AeMFMBR was operated at a constant temperature room with 20 °C (minimum light source to avoid phototrophic growth).

In phase 1B-HN, the membrane aerator module alone was tested for ammonia removal with the reactor operated in batch mode (two repeated cycles), using a COD-free medium with a high concentration of ammonia (HN), same as the feed solution for acclimation of the biofilm for nitrification (236 ± 9 mg-N/L, Table 1). In all subsequent phases (2–6), lower nitrogen concentrations were used in the range of 49–79 mg-N/L, as indicated in Table 1. In phase 2B-LN, the AeMFMBR was therefore operated under the same conditions as phase 1 except the total ammonia concentration in the feed solution was reduced to 79 ± 11 mg-N/L.

In phases 3 through 6, GAC particles were added into the filtration