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Upflow anaerobic-microaerobic fixed biofilm reactor integrating methanogenesis with partial nitrification



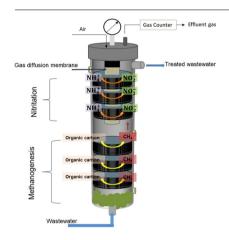
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HIGHLIGHTS

- Methanogenesis and partial nitrification were combined in a single bioreactor.
- High COD removal efficiency of 92–99% was achieved.
- Majority of the influent BOD₅ (80–91%) was anaerobically oxidized.
- Ammonium oxidation ranged between 69 and 86%, with nitrite accumulation of 15–24 mg/L.
- Known methanogens and nitrifiers were identified.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords: Wastewater treatment Anaerobic COD removal Partial nitrification Microaerobic Methane

ABSTRACT

An anaerobic-microaerobic fixed biofilm (AMFB) reactor, that integrates methanogenesis with partial nitrification within a single unit was investigated to achieve carbon removal simultaneously with ammonium oxidation in dilute wastewater. Membrane aeration was used for a controlled and efficient oxygen supply for partial nitrification and to prevent oxygen related inhibition of methanogens in the AMFB reactor. Removal of chemical oxygen demand (COD) and ammonium oxidation was first tested on synthetic wastewater, followed by domestic wastewater. The COD removal efficiency ranged between 92 and 99% on synthetic wastewater at hydraulic retention time (HRT) of 8–24 h. Nearly complete removal of biochemical oxygen demand (BOD $_5$) was obtained for domestic wastewater. Influent COD was mainly removed by fermentation and methanogenesis, resulting in high methane yields of up to 0.33 L_{CH4} gCOD $_{anaerobic}^{-1}$. Ammonium oxidation efficiency of 69–86% was obtained. Microbial community analysis showed proliferation of fermenters and methanogens exclusively in the anaerobic section of the reactor, while aerobic heterotrophs and nitrifiers were mainly identified in the membrane aerated section. This study first proves that the single-stage AMFB reactor can treat municipal wastewater economically to meet the wastewater standards, although further research for improving water quality (e.g., denitrification) would be required.

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1. Introduction

The development of compact wastewater treatment systems has garnered much attention in the past decade. This interest can be attributed to many reasons including: expansion/retrofitting of wastewater treatment plants (WWTPs) in urban and peri-urban areas with limited land availability; cold region plants placed inside heated buildings and small-scale decentralised WWTPs for rural and remote communities. Compact treatment systems currently in the marketplace typically utilize the activated sludge process and modifications thereof. Since this biological treatment depends on aerobic microbial metabolism, intensive oxygen supply is essential which features high energy consumption and operating costs. Methane-producing anaerobic biotechnologies such as anaerobic digestion (AD) provide an energy-efficient alternative to aerobic technologies. Contrary to aerobic counterparts, AD does not require oxygen for removal of organics and produces less sludge. Rapid improvements in bioreactor technology (e.g., immobilization, granulation, membranes, etc.) has resulted in advent of compact high-rate anaerobic bioreactors with treatment efficiency and process stability close to aerobic technologies. Multiple studies have reported organic removal efficiencies of 90-95% at a short hydraulic retention time (HRT) of 9-12 h in fixed-bed/film anaerobic digesters [1,2]. The immobilisation of the microbial community on inert media allows excellent biomass retention leading to enhanced degradation rate, reduced digestion time and a small footprint. Moderate effluent quality can also be achieved without the need for solid-separation. Despite the advantages, a major bottleneck in the widespread adoption of such compact AD systems for domestic wastewater treatment are limitations in removing nutrients, especially nitrogen.

Wastewaters typically contain considerable levels of nitrogen-based compounds such as ammonium that needs to be removed before discharge to water body for ensuring water quality and protecting aquatic ecosystems (e.g., algal bloom and toxicity to aquatic organisms). European (EN 12-566) and American standards (NS Standard 40) for small scale wastewater treatments, a niche market for anaerobic compacted systems, mandate effluent NH₄⁺-N concentrations of below 10 mg L⁻¹ [3]. Anaerobic systems transform a portion of nitrogen to soluble ammonia rather than removing it by nitrification. The requirement of oxygen for nitrification coupled with oxygen sensitivity of methanogens limits removal of nitrogenous pollutants in anaerobic reactors. Thus, additional removal steps are required after anaerobic digesters to achieve effluent quality suitable for discharge, which makes the entire process complex and expensive. A tangible solution to this challenge is a single compact reactor that combines AD with nitrification. This may be accomplished by exploiting the ability of methanogens to withstand low levels of dissolved oxygen (DO) combined with relatively low DO requirements for partial nitrification (i.e., nitritation), the oxidation of ammonia to nitrite. Transformation of nitrite to N₂ can be achieved in post-denitrification with less dose of electron donor, another potential benefit of partial nitrification.

Sustained methanogenic activity in mixed cultures under microaerobic conditions of 0.1-0.5 mg L⁻¹ of DO has been widely reported [4-6]. This tolerance in mixed cultures is attributed to oxygen consumption by facultative microorganisms which creates localised anaerobic environments where methanogens are protected. Similarly, low DO concentrations ($< 1 \text{ mg L}^{-1}$) are common in bioreactors catalysing partial nitrification [7-9]. Therefore, methanogenesis and partial nitrification could be combined in a single reactor provided low DO concentrations can be maintained by controllable and efficient oxygen supply; to maximize oxygen consumption by ammonia oxidizing bacteria (AOB), while minimizing oxygen related inhibition of methanogens. Advancement in membrane-based aeration allows for oxygen delivery at high rates and transfer efficiencies. Membrane systems have been applied where a conventional aeration system is unable to meet the oxygen requirements of a high rate system. With bubble-free aeration using membranes, oxygen transfer efficacy close to 100% is achievable with consequent energy savings [10,11]. In addition, the oxygen supply rate can be efficiently controlled by the intramembrane oxygen partial pressure and membrane surface area. Modulation of membrane into different geometries (e.g., tubular or flat membrane, etc.) allows for membrane integration into different reactor designs, increased membrane surface to volume ratio and high volumetric mass transfer [11,12].

This study presents the development and operation of an upflow bioreactor that can simultaneously accomplish carbon and ammonium removal in wastewater under microaerobic conditions. Organic carbon was removed by methanogenic biodegradation (anaerobic process), while ammonium removal was achieved by partial nitrification (aerobic process). To simplify workflow and attain a small land footprint, the aerobic and anaerobic processes were integrated in a single reactor by staging former on top of the latter without any physical separation. In both treatment steps, the active biomass was immobilized on a porous carbon felt media (fixed-biofilm). Micro-porous gas membrane was used for efficient oxygen delivery for partial nitrification, allowing simultaneous methanogenesis retained at the bottom of the reactor.

2. Material and methods

2.1. Bioreactor design

The schematic of the anaerobic-microaerobic fixed biofilm (AMFB) reactor developed in this study is depicted in Fig. 1. The reactor was fabricated using a plexiglass column with an inner diameter of 13 cm and a height of 105 cm. The empty bed volume of the reactor was ~14 L. The reactor was divided into two main sections: a lower anaerobic section occupying 8 L of the volume and an upper microaerobic section with a volume of 4 L. The headspace occupied $\sim 0.5 \, \text{L}$ of the reactor volume. The anaerobic zone was built for degradation of organic matter, while the microaerobic zone was designed primarily for partial nitrification (nitrite accumulation) using a gas membrane. The anaerobic and microaerobic zones were fluidically bridged without any physical barrier. A middle zone of 1.5 L in liquid volume bridged the two sections. Two separate recirculation loops were used for mixing the liquids in the anaerobic and microaerobic zones. A circulation rate of 8 L h⁻¹ was applied for both zones using peristaltic pumps (Masterflex L/S 7523-80, Cole-Parmer, Canada).

In both zones, the active biomass was immobilized on carbon-felt media (Speer Canada, Kitchener, ON, Canada). Carbon felt was selected due to low cost (\$0.85/kg), and high porosity (91%) and surface area $(0.5\,\mathrm{m}^2/\mathrm{g})$ which minimize mass transfer limitations and biofilm shearing, a major bottleneck in fixed-biofilm reactors [1,13]. The anaerobic zone was packed with three carbon felt modules: CF1, CF2 and CF3. The modules were prepared by packing cylindrical pieces of carbon felt on a stainless-steel mesh support with individual dimensions of 12.8 cm (diameter) \times 15 cm (height). Similarly, the microaerobic zone consisted of three carbon felt modules: CF4, CF5 and CF6. These hollow cylindrical modules had an inner and external diameter of 4 cm and 12.8 cm, respectively, with a height of 5.5 cm.

A woven carbon cloth membrane with hydrophobic micro-porous layer (Model# W1S1009, Fuel Cell store, TX, USA) was used for bubble-less aeration, creating the microaerobic zone. The custom-made membrane module consisted of a hollow steel tube with wide channels milled down its length and capped at the bottom end with a rubber stopper. The membrane was wrapped around the steel tube and sealed using a specialized adhesive (Fuel Cell store, TX, USA). Stainless-steel fittings connected the module to the removable reactor top (Fig. 1). Based on the assumption that most of the organics in wastewater would be stabilised to methane and carbon dioxide in the anaerobic zone, the air supply through the gas membrane was intended principally for partial nitrification in this study. The membrane had a thickness of 410 μ m, an effective surface area of $\sim 0.1 \, \text{m}^2$ and occupied $\sim 1.5 \, \text{L}$ of the microaerobic zone. To enhance gas transfer efficiency, the

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