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# Separation of competitive microorganisms using anaerobic membrane bioreactors as pretreatment to microbial electrochemical cells



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#### HIGHLIGHTS

- Anaerobic membrane bioreactors (AnMBRs) used as pretreatment to MECs.
- High current density (14.5 A/m<sup>2</sup>) achieved from the MEC fed with AnMBR permeate.
- Propionate fermentation rate limited current density in the MEC.
- No methane was produced in the MEC due to membrane separation in the AnMBR.
- Utilization of intracellular biopolymers resulted in CE above 100%.

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#### ABSTRACT

Anaerobic membrane bioreactors (AnMBRs) as pretreatment to microbial electrochemical cells (MECs) were first assessed for improving energy recovery. A dual-chamber MEC was operated at hydraulic retention time (HRT) ranging from 1 to 8 d, while operating conditions for an AnMBR were fixed. Current density was increased from  $7.5 \pm 0$  to  $14 \pm 1$  A/m² membrane with increasing HRT. MEC tests with AnMBR permeate (mainly propionate and acetate) and propionate medium confirmed that propionate was fermented to acetate and hydrogen gas, and anode-respiring bacteria (ARB) utilized these fermentation products as substrate. Membrane separation in the AnMBR excluded fermenters and methanogens from the MEC, and thus no methane production was found in the MEC. The lack of fermenters, however, slowed down propionate fermentation rate, which limited current density in the MEC. To symphonize fermenters,  $H_2$ -consumers, and ARB in biofilm anode is essential for improving current density, and COD removal.

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#### 1. Introduction

A need for energy-efficient wastewater treatment has been increased, as energy security and sustainability become issues to be addressed in current society. As a result, there has been growing interest on wastewater treatment technologies that are able to recover value-added products from organic wastes and wastewaters, such as anaerobic digestion, advanced anaerobic digestion (e.g., anaerobic membrane bioreactors), or microbial electrochemical cells (MECs). MECs are very attractive in that electrons from organic wastes and wastewaters are only captured using anode-respiring bacteria (ARB) as electrode catalysts. This feature allows manipulation of the recovered electrons into to a variety of value-added products on the cathode in MECs, such as electric power, hydrogen

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gas, methane gas, hydrogen peroxide, acetate, ethanol, and so on (Logan and Rabaey, 2012; Rabaey and Rozendal, 2010). MECs can recover more value-added products from high strength organic wastes and wastewaters than diluted wastewaters, due to high degree of carbon reductance in these wastes and wastewaters. However, MEC performance was relatively poor using high strength wastes and wastewaters as substrate, mainly due to ARB's poor hydrolysis of particulate matters (Zhang et al., 2009; Mohan et al., 2009). ARB need partners that are able to hydrolyze and ferment complex organic compounds into simple acids (e.g., hydrolyzing and acidophilic fermenters) (Parameswaran et al., 2009, 2010). In addition, inert particulate matters present in high strength organic wastes and wastewaters would attach on the anode, which can prevent ARB from forming biofilm on the anode and significantly decrease current density in MECs (Logan and Rabaey, 2012). Thus, a pretreatment step seems essential for MECs to utilize high strength organic wastes and wastewaters as feed. Several works used anaerobic digestion or modified anaerobic digestion processes as pre-treatment to MECs (Zhang et al., 2009; Clauwaert

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et al., 2008). Despite of hydrolysis and fermentation in upstream processes there is a significant challenge to be addressed in these combinations: microbial competitions for anode surface area or electron donor. A large number of fermenters from anaerobic digester flowing to an anode chamber would compete with ARB for anode surface area, which can decrease current density in subsequent MECs (Parameswaran et al., 2009; Chae et al., 2010). ARB would be outcompeted by methanogens for acetate or hydrogen gas (Parameswaran et al., 2009, 2010, 2011; Chae et al., 2010; Wang et al., 2009; Lee et al., 2008). Although ARB are survived in biofilm anode, high concentration of methanogens in digester effluent would easily divert substrate electrons into methane in subsequent MECs (Parameswaran et al., 2009; Chae et al., 2010; Wang et al., 2009). Methanogenesis accounted for 26-53% of substrate electrons as methane (Parameswaran et al., 2009; Chae et al., 2010). In addition to microbial competitions, inert particulates present in digester effluent would bother ARB biofilm formation on the anode. For MECs to maximize energy recovery and improve current density from high strength organic wastes and wastewaters, it is essential to utilize pre-treatment technologies that can remove particulate substances and competitive microorganisms, and provide soluble forms of simple acids for ARB. Anaerobic membrane bioreactors (AnMBRs) that combine anaerobic digestion with membrane separation have been used for treating organic wastes and wastewaters to improve effluent quality and methane production (Liao et al., 2006). Anaerobic biochemical reactions (hydrolysis, fermentation and methanogenesis) and membrane separation in AnMBRs can provide soluble, simple forms of organics (i.e., volatile fatty acids) for ARB from fermentable substrate, which allows ARB to proliferate on the anode without microbial competitions and anode fouling events by particulate matters. There are no studies that combine AnMBRs with MECs up to now, although the combined process of AnMBRs and MECs has high potential for energy recovery improvement.

In this study, the combined process of an AnMBR and MEC was tested using glucose medium as model substrate. First, AnMBR performance and permeates were characterized at a fixed operating condition. Second, current density, effluent quality, and coulombic efficiency were comprehensively tracked in an MEC operated

under different conditions. Finally, a hypothesis that ARB can directly oxidize propionate (a main volatile fatty acid in AnMBR permeates) for current generation was evaluated in the MEC fed with propionate medium.

#### 2. Methods

2.1. Configurations of anaerobic membrane bioreactor (AnMBR) and microbial electrochemical cell (MEC)

A laboratory scale anaerobic membrane bioreactor (AnMBR) built with polyvinylchloride was used in this study. A total volume of the AnMBR was 5.75 L with a working volume of 5.0  $\pm$  0.1 L. An ultrafiltration membrane module (ZeeWeed 500®, GE Water and Process Technologies, Canada) was immersed in the middle of the AnMBR. Fig. 1a shows schematic diagram of the AnMBR. The membrane surface area was 0.047  $\text{m}^2$  and its average pore size was 0.04  $\mu\text{m}$ .

To improve current density and attenuate applied voltage in MECs, a dual-chamber MEC was designed in which large specific surface area (anode surface areas to membrane surface area) can be provided for high ARB biofilm density, along with a small electrode distance (An and Lee, 2013). The MEC was fabricated with cylindrical plexiglass (Fig. 1b). The working volumes of the anode and cathode chamber were 280 mL and 120 mL, respectively. High density carbon fibers (2293-A, 24A Carbon Fiber, Fibre Glast Development Corp., Ohio, USA) that were connected with a stainless steel frame was used as the anode module. The total geometric surface area of anodes (carbon fibers) was 4480 cm<sup>2</sup>. The carbon fibers were pretreated for 3 days with nitric acid (1 N), acetone (1 N) and ethanol (1 N) for 1 day in series, and then washed them with MilliQ water (18.2 M $\Omega$ -cm). A stainless steel mesh was as the cathode (Type 304, McMaster Carr, OH, USA). An anion exchange membrane (AMI-7001, Membranes International Inc., USA) was placed between the anode and the cathode as separator, and the geometric surface area of the membrane was 28.1 cm<sup>2</sup>. Non-conductive polyethylene mat was used between electrodes and membrane to avoid possible short-circuiting and liquid leakage. The distance between electrodes was <1 cm.

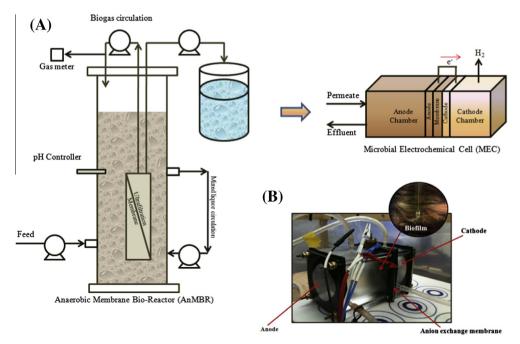


Fig. 1. (a) Anaerobic membrane bioreactor (AnMBR) and microbial electrochemical cell (MEC) integrated system, (b) photograph of sandwich-type MEC.

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