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Methane production enhancement by an independent cathode in integrated anaerobic reactor with microbial electrolysis



Weiwei Cai^a, Tingting Han^a, Zechong Guo^a, Cristiano Varrone^c, Aijie Wang^{a,b,*}, Wenzong Liu^b

^a State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology (SKLUWRE, HIT), Harbin 150090, China
^b Key Laboratory of Environmental Biotechnology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
^c Technical University of Denmark, Department of Chemical and Biochemical Engineering, Lyngby, Denmark

HIGHLIGHTS

- Methane production rate was increased in cathodic anaerobic digestion process.
- Independent cathode mainly contributed to the increase of methane production.
- Fermentative liquid was efficiently utilized in integrated reactor.
- Economic revenue can self-cover the cost of input electricity.

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G R A P H I C A L A B S T R A C T

Integrating microbial electrolysis cell and anaerobic bioreactor to enhance methane production from organics.



ABSTRACT

Anaerobic digestion (AD) represents a potential way to achieve energy recovery from waste organics. In this study, a novel bioelectrochemically-assisted anaerobic reactor is assembled by two AD systems separated by anion exchange membrane, with the cathode placing in the inside cylinder (cathodic AD) and the anode on the outside cylinder (anodic AD). In cathodic AD, average methane production rate goes up to 0.070 mL CH₄/mL reactor/day, which is 2.59 times higher than AD control reactor (0.027 m³ CH₄/m³/d). And COD removal is increased ~15% over AD control. When changing to sludge fermentation liquid, methane production rate has been further increased to 0.247 mL CH₄/mL reactor/day (increased by 51.53% comparing with AD control). Energy recovery efficiency presents profitable gains, and economic revenue from increased methane totally self-cover the cost of input electricity. The study indicates that cathodic AD could cost-effectively enhance methane production rate and degradation of glucose and fermentative liquid.

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1. Introduction

The current demand in renewable energy has boosted the applications of anaerobic wastewater treatment, which could provide biogas and at the same time produce lower sludge amount from wastewater. The conversion of fermentation products, such as amino acids (AA) and volatility fatty acids (VFA) to methane



^{*} Corresponding author at: State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150090, China.

E-mail addresses: waj0578@hit.edu.cn, wzliu@rcees.ac.cn (A. Wang).

(by methanogens) is closely related to hydrolysis and fermentation of proteins and carbohydrates in AD (Chen et al., 2007; Eastman and Ferguson, 1981). Recently, a novel concept of microbial electrolysis cell (MEC) has been developed as a potential way to assist energy recycle from a wide range of organics, with a very high efficiency. Organic compounds are oxidized by anodic respiring bacteria and hydrogen is reduced on the Pt loading cathode with an small applied voltage ($E_{app} = 0.114$ V is theoretically needed, while $E_{app} > 0.130$ V is required due to overpotential (Call and Logan, 2008)). MEC have thus supplied a new pathway to effectively produce hydrogen from biomass, however cathode-induced methane production is still of major concern, due to the presence of methane-producing bacteria (Rozendal et al., 2006; Wagner et al., 2009).

Seemingly, microbial electrolysis cell has supplied a potential way to stimulate growth of hydrogenotrophs, which led to methane production by hydrogen consumption (or supplying electrons) at cathode (Villano et al., 2010). Recently, extra H₂ or electrons from cathode have been proved to increase methane production and enhance stability in conventional AD reactor coupled MEC (which are frequently associated with hydrogenotrophic methanogenesis). Bo et al. (2014), for instance, coupled anaerobic digestion in a singlechamber microbial electrolysis cell, using a barrel-shape stainless steel as cathode, and reported a COD of acetate removal efficiency that increased from initial 56.5% (for the AD process) to 100% (for the coupled process) in 72 h, while methane yield doubled. Zhang et al. (2015) operated a hybrid reactor with 0.3 V applied voltage and obtained 51 ± 2.2% COD removal, which was higher than AD reactor without voltage (44.3 \pm 1.2%), increasing at the same time hydrolysis of polysaccharides. Although hybrid reactors were tested for their ability to accelerate methane production and improve organics degradation (Zhang et al., 2015; Zhao et al., 2014), it is important to better understand the different roles played by anode and cathode in the AD system. Firstly, the most suitable substrates reported were different for anode respiring bacteria and cathodeinvolved bacteria (Liang et al., 2014). Secondly, reactions and/or products developed at the anode and cathode played different roles to methanogens, the presence of direct interspecies electron transfer between Geobacter and methanogens could primarily contribute to methane production (Zhao et al., 2015a), while cathode that was dominated by hydrogenotrophic methanogens take main responsible for the conversion from hydrogen/electrons to methane through biomass retention (De Vrieze et al., 2014; Siegert et al., 2015). However, single chamber integrated reactor consisted of anode and cathode was employed to enhance methane production in most studies, while the study concentrated on individual function of anode or cathode was still lack. Thus, a separated system between anode and cathode is expected to facilitate the evaluation of the relative contributions to AD improvement. The separated cathode chamber can avoid the effect of anode to methane production as the presence of direct interspecies electron transfer (DIET) in the anode biofilm (Rotaru et al., 2014; Zhao et al., 2015a). Thus the contribution of cathode could be studied independently. Moreover, in previous studies, up-flow reactor could exhibit better performance either for anaerobic digestion in methane production (Zhao et al., 2015b) or bioelectrochemistry system in current generation (Wang et al., 2012). Therefore, the integrated reactor was designed to couple up-flow construction and separated anode/cathode in order to achieve better performance and eliminate irrelevant effects.

The objective of this study was to investigate the contribution of cathode in methane production and organic removal efficiency. A novel reactor, two integrated ADs was constructed and connected by bioanode and biocathode independently to evaluate the effect on methane acceleration and COD removal. Glucose and synthetic fermentation liquid were used to evaluate the performance in new reactor, including enhancement and stability of methane production.

2. Methods

2.1. Bioreactor setup

A new integrated anaerobic reactor made of polymethylmethacrylate was tested, by integrated a bioelectrochemical system between two AD systems, which were separated by anion exchange membrane (AEM, Ultrex CMI-7000, Membranes Interna-tional, US.) into anodic AD and cathodic AD. The cathode was placed in the inner cylinder and the anode was in the outer cylinder. The total volume was 1.2 L, including 700 mL of inner and 500 mL of external. The cathode was made of stainless steel mesh, which was placed close to the AEM, in the inside cylinder. The anode was made of carbon brush, placed in the external tube. A 0.8 V applied voltage was added between anode and cathode, based on our previous study (Linji et al., 2013). A control reactor was operated synchronously but without external voltage.

Acetate (1.5 g/L) was used as the sole carbon source in the anode chamber. Glucose (2.5 g/L in 50 mM PBS) was used as carbon source in the cathodic AD for methane production and for reactor performance evaluation. Subsequently, sludge fermentation liquid (SFL) was used as complex carbons for application test. The main characteristics of sludge fermentation liquid was 750 mg/L acetate, 150 mg/L propionate, 200 mg/L n-butyrate, 300 mg/L polysaccharide and 650 mg/L protein. The mixture of SFL and 50 mM PBS (50:50, V:V) was used as influent to cathodic AD.

2.2. Bioreactor operation

Acetate was used as carbon source for anodic AD throughout the whole experiment. During startup, all reactors were inoculated by waste activity sludge taken from aeration tank of the Taiping Municipal Wastewater Treatment Plant (Harbin, Heilongjiang Province, China). Glucose was firstly used as simple carbon source for cathode AD. Two integrated reactors (0.8 V applied voltage) and one control reactor (without applied voltage) were operated in continuous mode at a hydraulic retention time of 24 h. All reactors were operated at room temperature of 25 °C for 30 days. To evaluate bioelectrochemical contribution to methane production in integrated reactors, the applied voltage of one reactor was cut for 7 days as operational control and recovered applied voltage back. Finally, sludge fermentation liquid was used as complex carbons for cathodic AD for organic removal and methane production for another 26 days.

2.3. Measurement and calculation

Agilent gas chromatography (GC) was utilized to analyze the composition of VFAs (Zhou et al., 2013b). The determination of SCOD, TCOD, carbohydrate and protein was performed as described previously (Zhou et al., 2013a). Voltage and current were recorded every 10 min, using a multimeter (model 2700; Keithley Instruments, the USA). Gases (hydrogen, carbon dioxide, methane) were analyzed by TCD GC (Fuli GC9790II, Zhejiang analytical instrument Inc., China).

The contribution of current was calculated as theoretical methane production.

$$\frac{lt}{nF} \cdot V_m = V_{CH_4} \tag{1}$$

where *I* is the current value, *t* the time, n = 8 represents 8 mol electrons that can be transfer into 1 mol of methane, F = 96,485 C/mol is

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