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Bioelectrochemically assisted anaerobic digestion system for biogas upgrading and enhanced methane production

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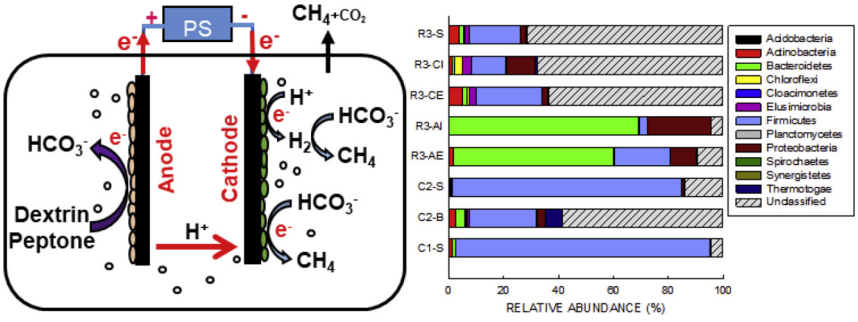
HIGHLIGHTS

- 2.0 V resulted in water electrolysis and H_2 production in an AD-BEC reactor.
- High CH_4 production and biogas CH_4 content (88.5%) in the AD-BEC reactor at 2.0 V
- Enrichment of exoelectrogens (anode) and hydrogenotrophic methanogens (cathode)
- AD-BEC more resilient to shock organic loadings than suspended biomass AD systems

GRAPHICAL ABSTRACT



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ABSTRACT

The objective of this study was to evaluate the effect of biofilm and external voltage on the performance and microbial community composition of batch-fed, combined anaerobic digestion-bioelectrochemical cell (AD-BEC) systems under different operational conditions. A dextrin/peptone mixture was fed at a range of organic loading rates (0.34 to 1.37 g COD/L-d). The hybrid system with both suspended biomass and biofilm without any external potential application achieved a substantially higher initial soluble COD consumption (53.7 ± 2.3% vs. 39.7 ± 3.7) and methane (CH_4) production (331 vs. 225 mL) within one day of feeding than the conventional AD system (suspended biomass only). Compared to the conventional AD system, the hybrid systems had higher resilience to shock organic loadings. A range of external potential (0.5 to 2.0 V vs. Ag/AgCl) was applied to AD-BEC reactors, developed with two different start-up procedures. A potential of 2.0 V resulted in water electrolysis leading to a higher CH_4 production rate (105 vs. 84 mL/L-d) and biogas CH_4 content (88.5 ± 1.4 vs. 64.5 ± 1.9%) in the AD-BEC reactor (closed vs. open circuit condition, respectively). Application of external potential enriched putative exoelectrogens at the anode biofilm and hydrogenotrophic methanogens at the cathode biofilm, which may have contributed to the observed enhanced CH_4 production in the AD-BEC system. A phylotype related to *Methanobacterium formicicum*, a hydrogenotrophic methanogen, dominated the archaeal community in the AD-BEC cathode biofilm.

1. Introduction

The increasing demand for energy and a world-wide shift away from fossil fuels have accelerated the development of renewable, net zero energy technologies. Anaerobic digestion, as a well-established process of converting organic waste into biogas (Angenent et al., 2018; Appels

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et al., 2008; Pavlostathis, 2011; Tezel et al., 2011), is an excellent choice for high-strength waste treatment, especially in the pursuit of sustainable, carbon neutral, net zero energy water resource recovery facilities (WRRFs) (WEF, 2014). With its advantages, including a lower volume of digested sludge requiring final disposal, low energy requirement, and biogas energy recovery, anaerobic digestion is considered a major and essential part of a modern WRRF (WEF, 2014). Despite its benefits, the potential for further optimization of anaerobic digestion, in terms of energy recovery, is still high. The typical range of biogas methane (CH_4) and carbon dioxide (CO_2) content is 55–75% and 25–45% (v/v), respectively (Rittmann and McCarty, 2001). The biogas CH_4 and CO_2 content is primarily determined by the mean oxidation state of the carbon in the organic matter fed to the digester (Gujer and Zehnder, 1983; Pavlostathis, 2011), which is related to the chemical composition of the feed, in particular its carbohydrate, protein, and lipid content.

The relatively high biogas CO_2 content constrains its commercial use, and biogas upgrading (i.e., increasing the CH_4 content) is required prior to use in standard equipment and applications. Multiple methods, including chemical scrubbing, physical separation, and biological consumption have been developed to decrease the biogas CO_2 content while increasing its CH_4 content and thus its calorific value. However, expensive material, high energy and chemical input, as well as capital cost have been the major concerns for these types of biogas upgrading processes (Muñoz et al., 2015). Meanwhile, bioconversion of CO_2 in ADs through addition of ex situ generated hydrogen (H_2) followed by hydrogenotrophic methanogenesis and/or acetogenesis, a process known as in situ biomethanation, has been studied (Al-mashhadani et al., 2016; Francioso et al., 2010; Luo and Angelidaki, 2012; Salomoni et al., 2011). Due to a number of challenges described by Angenent et al. (2018), industrial application of the in situ biomethanation process using exogenous H_2 is no longer considered. AD has also been considered for the bioconversion of exogenous CO_2 generated by other processes for carbon capture on-site and CH_4 production enhancement. However, further investigation of the mechanism of CO_2 utilization, the technologies promoting mass transfer, and the CO_2 injection conditions is needed before full-scale application of such processes becomes viable (Angenent et al., 2018; Bajón Fernández et al., 2017).

Bioelectrochemical systems (BESs) have been applied for wastewater treatment, nutrient, metal and energy recovery, as well as production of biofuels (Bajracharya et al., 2016; Li et al., 2014; Modin and Aulenta, 2017; Ren, 2013; Rozendal et al., 2008; Zhang and Angelidaki, 2014; Angelidaki et al., 2018). Bioelectrochemically assisted AD (AD-BEC) is a process in which the metabolic processes taking place in a conventional anaerobic digester are assisted by the application of a relatively low, external potential. As a result of the externally applied potential, microbial metabolism and interspecies interactions could be substantially altered (Moscoviz et al., 2016), resulting in enhanced CH_4 production. Electromethanogenesis, i.e., CH_4 production by hydrogenotrophic methanogens directly utilizing CO_2 and electrons from the cathode electrode, can be achieved in a BEC, even at a relatively low temperature (i.e., 10 °C) (Cheng et al., 2009; Liu et al., 2016). In an integrated AD-BEC system, biogas can be produced and upgraded simultaneously, which makes in situ biogas upgrading possible. The benefits of in situ biogas upgrading include a small process footprint, low capital cost and low energy demand, compared to ex situ biogas upgrading through physical and chemical processes (Muñoz et al., 2015). Although several studies have shown enhanced CH_4 production in AD-BEC systems (Liu et al., 2016; Moreno et al., 2016; Zhang et al., 2013; Zhao et al., 2015), limited research has been performed on such systems under different start-up conditions and a range of applied potentials. It is not yet well understood how the external potential affects the development and activities of bacterial and archaeal species in both the suspended and electrode-attached biofilm biomass. Hydrogenotrophic methanogenesis and electromethanogenesis are believed to be the two main processes through which enhanced CH_4 production and biogas upgrading can be achieved in AD-BEC systems (Bajón Fernández et al., 2017; Liu et al., 2016; Moreno et al., 2016; Zhang et al., 2013). However, the relative contribution of these two processes to reported enhanced CH_4 production in integrated AD-BEC systems maintained at a range of potentials is not fully investigated.

The objective of this study was to assess: (i) the effect of biofilm on the performance of hybrid AD systems; (ii) the performance of an integrated AD-BEC system at a range of external potential applied at different stages of electrode biofilm development; and (iii) the change in microbial community composition due to the application of external potential to an AD-BEC system.

2. Materials and methods

2.1. AD reactors

To investigate the contribution of the biofilm on CH_4 production without an external potential, two AD control reactors were developed and maintained throughout this study: a conventional AD with only suspended biomass (R1 reactor) and a hybrid (i.e., suspended biomass and biofilm), open circuit AD with two carbon felt electrodes attached to stainless steel (SS) collectors (R2 reactor). The two AD control reactors were developed using 2.8 L Spinner cell flasks (Bellco Glass, Inc., Vineland, NJ) with a liquid volume of 2.1 L. Each anode and cathode electrode in R2 had four carbon felt strips (Alfa Aesar, Ward Hill, MA; 6 × 1 × 1 in.), attached to a 6-mm diameter SS rod (Alfa Aesar, Ward Hill, MA) used as the collector. The reactor's headspace was connected to a graduated cylinder, which contained an acid brine solution (10% NaCl w/v and 2% H_2SO_4 v/v), used for gas collection and measurement by liquid displacement. An aliquot of 0.6 L stock mixed methanogenic culture and 1.5 L pre-reduced medium were anaerobically transferred to the reactor, after flushing the empty reactor and gas lines with N_2. The stock mixed methanogenic culture used in this study was developed initially with inoculum from a mesophilic, municipal anaerobic digester, batch-fed with a mixture of dextrin and peptone, and maintained at 35 °C for several years (Misiti et al., 2013). The medium contained (in g/L): K_2HPO_4, 0.9; KH_2PO_4, 0.5; NH_4Cl, 0.5; $CaCl_2 \cdot 2H_2O$, 0.1; $MgCl_2 \cdot 6H_2O$, 0.2; $FeCl_2 \cdot 4H_2O$, 0.1; $NaHCO_3$, 6.7; $Na_2S \cdot 9H_2O$; 8 mL of a vitamin stock solution; and 8 mL of a trace metal stock solution (Beydilli and Pavlostathis, 2005). The reactors' contents were continuously mixed using a magnetic bar-bearing impeller assembly (Bellco Glass, Inc., Vineland, NJ) magnetically driven by an Isotemp stirring plate (Fisher Scientific, Waltham; MA). The reactors were batch-fed every 3 and 4 days with a mixture of dextrin/peptone stock solution, resulting in an initial dextrin/peptone concentration of 1030/1370 mg COD/L upon feeding; the mean organic loading rate was 343 mg COD/L-day. The reactors were maintained at room temperature (22 ± 2 °C) with a hydraulic retention time (HRT) of 21 days. pH, gas production and composition, soluble COD, volatile fatty acids (VFAs) and volatile solids (VS) were periodically measured for both control reactors. Samples for VS measurements were taken from the liquid phase of the reactors.

To determine the ultimate anaerobic biodegradability and kinetics of the D/P mixture in a batch system at room temperature, a biodegradability test was conducted following the protocol from the previous work of Tandukar and Pavlostathis (2015). Two 500-mL glass bottles were used, with one serving as a seed blank control, and the other as a D/P-amended culture. An aliquot of 40 mL deionized water and 40 mL D/P stock solution (12 g/L dextrin, 6 g/L peptone) were transferred to the seed blank bottle and the D/P bottle (total initial COD 1.8 g/L), respectively. After flushing with N_2 gas, an aliquot of 160 mL of medium (4.1 ± 0.07 g TS/L; 0.5 g VS/L) and 200 mL seed culture (5.4 ± 0.08 g TS/L; 1.8 ± 0.12 g VS/L) obtained from R1 was transferred anaerobically to each bottle. The initial pH, total COD (tCOD), soluble COD (sCOD), and VFAs were measured for both the seed blank control and the D/P amended culture. Incubation was carried out at 22 °C and the bottles were shaken manually once a day. Throughout the incubation period,