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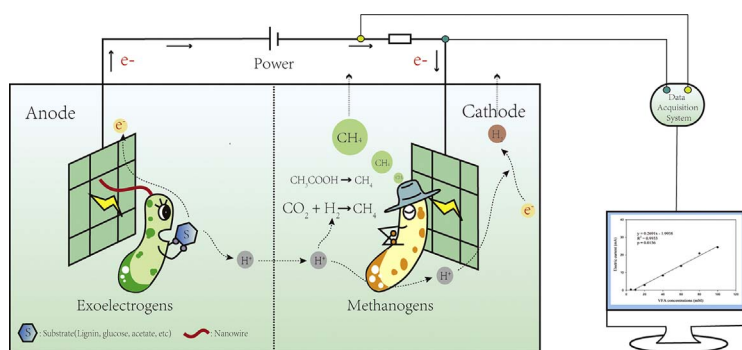
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## Review

## A review on the applications of microbial electrolysis cells in anaerobic digestion

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## GRAPHICAL ABSTRACT



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## ABSTRACT

Anaerobic digestion (AD) has been widely used for biogas or biofuel generation from waste treatment. Because a low production rate and instability of AD occur frequently, various technologies have been applied to improvement of AD. Microbial electrolysis cells (MECs), an emerging technology, can convert organic matter into hydrogen, methane, and other value-added products. Recent studies showed that application of MEC to AD (MEC-AD) can accelerate degradation of a substrate (including recalcitrant compounds) and alter AD microbial community by enriching exoelectrogens and methanogens thus increasing biogas production. With stable microbial communities established, improvement of MEC-AD for methane production was achieved. MEC-AD process can be monitored in real-time by detecting electric signals, which linearly correlate with substrate concentrations. This review attempts to evaluate interactions among the decomposition of substrates, MEC-AD system, and the microbial community. This analysis should provide useful insights into the improvement of methane production and the performance of MEC-AD.

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## 1. Introduction

Conventional anaerobic digestion (AD) is a technology developed for waste treatment and contains the following four major processes: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. By the action of different microorganisms, organic substrates are converted to renewable energy in the form of biogas containing methane and hydrogen (Guo et al., 2017). Various substrates, including almost all kinds of organic matter (such as cellulose in straw, sawdust, carbohydrate-rich food waste, vegetables, fruits, sugar, starch, and complex substrates like Fischer-Tropsch [FT] wastewater), can be subjected to AD via anaerobic fermentation (Guo et al., 2014). Even though application of AD to degrade organic pollutants and produce biogas has achieved great success, it still has limitations such as destabilization, weak decomposition of substrates and low biogas production. Although stable and highly efficient methane production (a 55% increase) is observed during treatment of pig slurry in a lab scale continuous stirred tank reactor for AD, destabilization occurs in an AD process with double organic-matter and nitrogen loads (Cerrillo et al., 2016; Park et al., 2018). Another example is that fermentation of lignocellulosic substrates inhibits the whole process owing to the high C/N ratio of cellulose (Xu et al., 2017). Another main source of substrates is food waste, which is rich in carbohydrates (Park et al., 2018). Because several easily biodegradable substrates are present in food waste, these substrates degrade rapidly and lead to the accumulation of volatile fatty acids (VFAs), which decrease pH. With a rapid decrease in pH, fermentation becomes unstable, and methane production is inhibited. Usually, the temperature during AD is maintained in the mesophilic (35 °C) or thermophilic (55 °C) range. Thermophilic AD, which achieves a high digestion rate of substrates, has been used with high organic loads to prevent the inhibition of methanogens by an organic overload. Nonetheless, thermophilic AD has lower stability than mesophilic AD does (Cerrillo et al., 2016) because the microorganisms are more sensitive to high ammonia concentrations (Cerrillo et al., 2017). To solve these problems, various researchers proposed some countermeasures. For example, pretreatment of lignocellulosic substrates involving physical, chemical, and/or biological approaches can improve the efficiency of gas production (Zheng et al., 2014). Besides, straw addition for co-fermentation, which balances pH with  $\text{NaHCO}_3$  or  $\text{CaCO}_3$ , has been applied to food waste to alleviate the inhibition due to high oil and salt concentrations and low pH.

Microbial electrolysis cells (MECs) have been recently developed for hydrogen generation from organic matter (Kadier et al., 2014); this approach is more efficient than production of hydrogen from water (Zhang and Angelidaki, 2014). A MEC system is similar to that of a microbial fuel cell (MFC), except that it involves a sealed cathode and external voltage (Logan et al., 2008). In MECs, electrochemically active microorganisms are the dominant populations at the anode and convert organic compounds to protons,  $\text{CO}_2$ , and electrons (Rozenal et al., 2006). Electrons generated by these microorganisms are transferred to the anode and subsequently to the cathode through an electrical circuit where hydrogen is generated (Croese et al., 2014). There are two ion-selective membranes (anion exchange membrane and cation exchange membrane), which generally serve for preventing the reaction between oxygen and the generated hydrogen (Rozenal et al., 2007), similar to those used in water electrolyzers. Between these two types of membrane, the anion exchange membrane is considered more efficient because of lower internal resistance, which is attributed to lower resistance to the transport of ions through the membrane (Sleutels et al., 2009). The use of MECs in fermentation promotes substrate decomposition, thereby resulting in hydrogen production (Dhar et al., 2015).

The aim of using MECs is to promote hydrogen production, whereas methane can be produced in AD. Thus, various strategies to inhibit methanogens in MEC-AD, e.g., exposure to oxygen (Tice and Kim, 2014), alamethicin, or a chemical inhibitor (2-bromoethanesulfonate) (Catal et al., 2015) for hydrogen production have been proposed.

Nevertheless, methane production still proceeds even though 50 mM 2-bromoethanesulfonate is used (Rago et al., 2015). Thus, methane, which is also a source of renewable energy, is considered an alternative option for energy recovery from waste by means of MEC-AD. Moreover, the degradation rate of recalcitrant compounds or complex wastewater is higher in MEC-AD (Mahmoud et al., 2014). Stability and robustness of AD are also enhanced through coupling with a MEC. In stable MEC-AD, a linear correlation ( $R^2 = 0.99$ ) between the electric signal and concentration of substrate was observed with a MEC-type sensor (Jin et al., 2017). MEC-AD increases the chemical oxygen demand (COD) removal rate and accelerates VFA degradation, which inhibits methanogens and thus decreases methane production (Li et al., 2011).

Although acceleration of substrate decomposition and an increase in biogas production have been achieved in MEC-AD, the relations among the microbial community, stable MEC-AD process, and biogas production have not been summarized. The objective of this review is to discuss the advantages, current challenges, and prospects of MEC-AD and to encourage large-scale application of this method to waste management in the future.

## 2. Microbial electrolysis cells can accelerate substrate degradation and alter microbial community to increase biogas production during anaerobic digestion

### 2.1. Substrate degradation and methane production are accelerated at suitable voltage

Usually, diverse substrates are used in AD systems, such as acetate, glucose, waste-activated sludge, and food waste. One research group constructed a reactor using 20 mL of waste-activated sludge (pH 6.8, total suspended solids  $1.75 \times 10^4 \text{ mg L}^{-1}$  and volatile suspended solids  $1.38 \times 10^4 \text{ mg L}^{-1}$ ) as substrate and supplemented the system with 150 mL of acetate ( $10 \text{ g L}^{-1}$ ) to detect the COD removal rate and methane production. The results showed that the COD removal efficiency was 56.5% and the cumulative methane yield reached  $147.1 \pm 29.2 \text{ mL}$  in 72 h (Bo et al., 2014). In another study, investigators utilized glucose ( $2.5 \text{ g L}^{-1}$  in 50 mM PBS) as a carbon source for methane production and obtained an average methane production rate of  $0.027 \text{ (m}^3 \text{ CH}_4\text{) m}^{-3} \text{ d}^{-1}$  (Cai et al., 2016).

The MEC is a promising technology for the removal of organic pollutants and for biogas production (Logan and Rabaey, 2012). COD removal rates reach 75% within 184 h when swine wastewater is treated with a MEC (Wagner et al., 2009). Many studies revealed higher degradation rates of different substrates in MEC-AD systems than in an AD system (Cai et al., 2016; Lu and Ren, 2016). Removal efficiency of total COD (TCOD) increases from 10.7% to 31.5%, when MECs are employed to manage waste-activated sludge (MEC-WAS) at a concentration of  $10 \text{ (g COD) L}^{-1}$  (Sun et al., 2015). Furthermore, the protein removal rate shows a 1.9-fold increase in a closed MEC-WAS system compared to an open-circuit control system. During utilization of acetate for biogas production, a significant increase in MEC-AD is observed, with the removal rate of COD reaching 100% (1.7-fold higher than that in the AD process) in 72 h (Bo et al., 2014). In addition, the lowest carbon dioxide content ( $2\% \pm 1.4\%$ ) in total biogas is obtained in a MEC-AD reactor with an applied voltage of 1 V as compared to that obtained in an AD reactor ( $43.2\% \pm 2.3\%$ ) or MEC-AD reactor with applied voltage 0.4 V ( $\sim 10\%$ ). This finding indicates that different processes occur at different applied voltages. For the majority of MEC-AD systems, fixed voltage is applied to each system, and improvements in COD removal rates and biogas production have been observed. Nonetheless, most studies have not tested whether the voltage used for MEC-AD systems is optimal or not. Choi et al. explored the influence of various voltage values (0.5, 0.7, 1, and 1.5 V) on a MEC-AD fermentation process for methane production. In this study, the effluents of anaerobic digestion and growth media were mixed as substrates in a volume ratio of 1:1 (Choi et al., 2017). The percentage of soluble COD

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