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Effects of electrode distance and mixing velocity on current density and methane production in an anaerobic digester equipped with a microbial methanogenesis cell

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ABSTRACT

The microbial methanogenesis cell (MMC) has been studied to enhance organic removal efficiency and methane production in an anaerobic digester (AD). However, its applicability remains limited without practical approaches to scale-up the design for commercialization. Internal resistance within MMC is closely related to the transfer of hydrogen ions between electrodes. We analyzed the effects of various electrode distances and mixing velocities on the current density and methane production in a single AD equipped with an MMC. As the distance between electrodes increased from 1 cm to 5 cm, methane production and current density decreased to 51% and 92%, respectively. Although an increase in mixing velocity decreased the internal resistance, this effect was not significant below a certain distance. For larger distances, an increase in mixing velocity not only increased current density by a factor of approximately 2.5, but also enhanced methane production by a factor of approximately 1.4.

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Introduction

Recently, microbial electrochemical technology (MET), including the microbial electrolysis cell (MEC) and microbial methanogenesis cell (MMC), has attracted increasing attention as a technology that enhances organic removal efficiency in a reactor and maximizes the production of biogas such as hydrogen and methane [1,2]. However, current evaluations of MET are not positive, because its applicability is limited and it is not scalable for commercialization. This is because of the persisting problem of high internal resistance, which has inevitable economic consequences such as requirements for over-potential and high-cost electrode materials [1]. The types of internal resistances that apply to MET can be divided into ohmic resistances (charge transfer resistance and electrolyte resistance) and mass-transfer resistances [3]. The sizes of the reactors in most previous MET studies have been very small (<1 L), resulting in reduced internal resistance due to the minimization of the distance between the anode and cathode. The small size mainly minimizes the resistance arising from

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Abbreviations: MMC, Microbial Methanogenesis Cell; MET, Microbial Electrochemical Technology; MEC, Microbial Electrolysis Cell; MFC, Microbial Fuel Cell; AD, Anaerobic Digester; CFD, Computational Fluid Dynamics.

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mass transfer between the electrodes. Compared with a single chamber, a double chamber is more sensitive to the internal resistance of the membrane and the internal resistance between the electrodes is dependent on the membrane type or material; however, reports suggest that this increased sensitivity also has limitations [3-5]. In previous studies, the membrane has been a major contributor to the internal resistance in a double chamber. Clauwaert et al. [6] reported that, under the same conditions, the internal resistances of an MEC without a membrane and an MEC with a membrane were 1.1 and 2.2 Ω , respectively. In addition, the diffusion coefficient of hydrogen ions in a solution at 30 °C was 6.64×10^{-5} and 1.03 \times 10 $^{-5}$ cm $^2/s$ with and without a membrane (Nafion[®]), respectively. As the diffusion resistance caused by the membrane was approximately six times that without a membrane, early research moved toward the investigation of double-chamber MEC rather than single-chamber MEC [7,8]. Thus, the internal resistance in MET is surely an important factor that limits its upscaling and efficiency.

Different methods to minimize the internal resistance have been reported, such as minimizing the electrode distance, selecting electrodes with high current density, increasing the ionic strength and electrical conductivity, selecting various types and materials of electrodes, and enhancing the internal mass transfer based on mixing [1,9,10]. The main factor affecting internal resistance is the distance between electrodes, and most research has been conducted to minimize this distance. Wang et al. [11] reported that when electrodes were changed to cuboid shapes in an H-shaped MEC reactor, the distance between electrodes could be reduced from 14 cm to 4 cm or less, which resulted in an increase of hydrogen gas production by approximately 25%. In addition, Cheng et al. [12] reported that, when the electrode distance was reduced from 3 cm to 2 cm in an MFC reactor, the internal resistance could be reduced from 35 Ω to 16 Ω , and Liu et al. [13] reported that decreasing the electrode distance from 4 cm to 2 cm decreased the internal resistance from 116 Ω to 77 Ω . Cheng and Logan [14] compared electrode distances of 1.0, 2.0, 3.0, and 3.5 cm in a single-chamber MEC reactor and found that the electrode distance and current density were inversely proportional to the amount of hydrogen gas produced. An et al. [15] also reported that, when the electrode distance increase in solid oxide fuel cell (SOFC), the current decreases because the local current density between the electrodes decreases. Clearly, the electrode distance should be minimized to reduce internal resistance, but this decreases the size of the reactor. In other words, electrode distance is related directly to reactor scale, and it is believed that reactor scale is the first factor that must be resolved prior to commercialization. To date, few research findings have presented reasons or specific solutions for the variation of internal resistance with respect to electrode distance; most studies have focused solely on reducing the internal resistance by changes to the type, material, and structure of the electrodes. The principal factor limiting internal resistance in a single-chamber MEC is the transfer of hydrogen ions from the oxidation electrode to the reduction electrode [16]. One way to enhance the transfer rate of hydrogen ions is to improve the mass transfer by improving fluid mobility via mixing in the reactor [6,17,18]. If a hydrogen ion, produced

from an anode in a reactor where the electrode distance is not minimized, exists in a fluid with strong mobility, the fluid mobility would enhance hydrogen ion mobility toward the cathode and reduce internal resistance.

The total mass transfer flux of hydrogen ions can be expressed as the sum of the mass transfer caused by convective flux in the bulk and the mass transfer caused by molecular diffusion as follows [19,20]:

$$N_{\rm H} = C_{\rm H} \times V_{\rm H} \left(-D_{\rm V} \cdot d \left(\frac{C_{\rm H}}{b} \right) \right), \tag{1}$$

where N_H is the total flux of hydrogen ions in kmol/m²s, C_H is the hydrogen ion concentration in kmol/m³, V_H is the hydrogen ion velocity in m/s, D_v is the diffusion coefficient in m²/s, and d (C_H /b) is the concentration gradient with b denoting the path length vertical to the area where diffusion occurs in units of m.

As expressed by Eq. (1), mass transfer in the solution can be divided into convection and molecular diffusion. The mass transfer arising from convection is related to the driving force that is dependent on mixing and temperature, while molecular diffusion is related to the concentration gradient. If hydrogen ions were distributed with the negligible concentration, then increasing the driving force by mixing could improve their mass transfer. In other words, if the mixing velocity between the electrodes were increased via mixing, the mass transfer rate and flux of the hydrogen ions would increase, making it possible to reduce the internal resistance.

This study investigated the effect of mixing velocity on internal resistance based on electrode distance by analyzing the electrical characteristics and amount of methane produced in different MMC reactors with the same applied voltage.

Materials and methods

Characteristics of seed sludge and substrate

Seed sludge was inoculated to both an anaerobic digester (AD) and MMC reactors after collection from an AD in a local city food waste treatment plant. For the inoculated sludge, acetate was provided at 1.172 g-acetate/d to meet the organic loading rate of 2.0 kg-COD/m³d in each reactor, and nutrition batches manufactured to the specifications listed in Table 1 were injected to the substrate.

Configuration of the reactor

As shown in Fig. 1, both the diameter and height of the sequencing batch reactor (SBR) were 100 mm, the total volume was 785 mL, and the working volume was 625 mL. Graphite carbon coated with nickel was used as the anode, whereas graphite carbon coated with copper, iron, and nickel was used as the cathode. For the cathode, a complex metal catalyst solution was prepared by dissolving 30.125 g $MnSO_4$ ·H₂O, 19.75 g KMnO₄, 0.5684 g iron phthalocyanine (FePc), and 0.5761 g copper phthalocyanine (CuPc) in 1 L of distilled water, and stirred for 2 h. The prepared solution was heated for 1 min

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