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# Scalable multi-electrode microbial electrolysis cells for high electric current and rapid organic removal

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## HIGHLIGHTS

- New stack design was examined for multi-electrode microbial electrolysis cells.
- The stacked design successfully enhanced the electric current density in MECs.
- High current density (520 A $m^{-3}$) was achieved without precious metal catalysts.
- The COD removal rate increased to 129 mg-COD $L^{-1}$ $hr^{-1}$ with increasing flow rate.
- All individual electrodes in the stack contributed to the total current generation.

## ARTICLE INFO

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## ABSTRACT

Microbial electrolysis cells (MECs) can be used to produce hydrogen gas from wastewater. A novel multi-electrode stack design was proposed and examined under various operating conditions to maximize electric current in MECs without precious metal catalysts. For the cathode in the electrode stack, stainless steel mesh generated higher electric current than activated carbon cloth. The electric current density increased in proportion to the number of electrode pairs as the maximum current density was 520 A $m^{-3}$ in MEC-10 (10 electrode pairs), 270 A $m^{-3}$ in MEC-5 (5 electrode pairs), and 45 A $m^{-3}$ with a single electrode pair. The stacked MEC was not ideal for fed-batch operation due to the short inter-electrode distance (~2 mm); consequently, continuous-recycle and -flow operation resulted in the high electric current generation. During continuous-flow operation, individual electrodes in MEC-10 and MEC-5 showed a variation in electric current capacity (0.9–2.7 mA for 0.6 mL $min^{-1}$). The COD (chemical oxygen demand) removal rate increased from 45.7 to 128.8 mg-COD $L^{-1}$ $h^{-1}$ with increasing flow rate from 0.1 to 0.6 mL $min^{-1}$. These findings indicate that the stacked multi-electrode design can magnify the current generation and COD removal rate in MECs.

## 1. Introduction

Microbial electrolysis cells (MECs) are an emerging technology to treat wastewater and simultaneously produce hydrogen gas [1]. A MEC reactor consists of the anode and cathode. Organic substrates are oxidized by exoelectrogenic bacteria to release electrons to the anode while water is reduced at the cathode to hydrogen gas. To drive the electrode reactions, a small voltage (0.13–1.23 V) needs to be applied to the MEC; as a result, the energy recovered as $H_2$ gas is usually much greater than the energy consumed for the applied voltage [2,3]. In MEC operation, the rate of organic removal and hydrogen gas production is represented by the magnitude of electric current. There are many factors that govern the electric current generation in MECs, such as electrode materials, electrode size, electrode catalysts, inter-electrode distance, and material transport near electrodes, which is often determined by hydrodynamic conditions. The inter-electrode distance has been minimized in previous studies by developing the sandwiched electrode assembly that significantly decreases the internal resistance of bioelectrochemical systems (BES) [4–6]. The separators such as carbon cloth [6,7], glass fiber [8,9], and spacers [10] are commonly used in the sandwiched electrode assembly design. In addition, the specific surface area of electrodes (electrode surface area per volume of reactor) also affects the electric current generation. For instance, a recent study employed bundles of 73,000 graphite fibers in a 0.125 L MEC (specific anode surface area was 2530 $m^2$ $m^{-3}$) and demonstrated substantially high electric current generation at 1470 A $m^{-3}$ [11]. However, the bundled electrode design can hardly be scaled up for practical applications of MECs. In another study, high current generation (2.82

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A $m^{-3}$) was demonstrated in a relatively large scale MEC reactor (6.6 L) with a specific anode surface area of 6.1 $m^2 m^{-3}$ [2]. A maximum current density of 180 A $m^{-3}$ was demonstrated by increasing the specific anode surface area to 100 $m^2 m^{-3}$ using carbon felt in MECs [12]. Carbon cloth was also utilized in MEC construction to maximize the specific electrode surface area and the resulting current density was also relatively high at 65.3 A $m^{-3}$ and this current density was higher than that produced in another MEC with the same electrode materials but reduced specific surface area from 12 to 6 $m^2 m^{-3}$ [2,13]. The multi-electrode design is an effective way to increase the specific surface area of electrodes. A multi-electrode MEC with 8 separate electrode pairs produced a maximum current density of 74 A $m^{-3}$ [14]. However, this MEC design used graphite fiber brushes as the anodes and thus required a certain reactor space for the brushes. As a result, the specific surface area was limited to 64 $m^2 m^{-3}$. Therefore, in this study, we proposed and examined a new MEC electrode design that satisfies the three key requirements: high specific surface area; low internal resistance; and easy scalability. Various anode materials have been examined in the BES, including graphite brushes [14,15], graphite granules [16], carbon cloth [1,13], and carbon felt [12,17,18]. Graphite brushes can provide a large specific surface area for the growth of exoelectrogenic bacteria. Graphite granules also provide a large surface area and relatively high electric current generation (66 ± 2 to 156 ± 1 A $m^{-3}$) [16]. Unlike graphite brushes or granules, carbon cloth or carbon felt can substantially decrease the inter-electrode distance without electric short circuiting problems. By decreasing the inter-electrode distance, the internal resistance dropped from 580 to 220 Ω by using carbon cloth [19]. Porous anode materials were also examined for BES scale-up [20,21]. However, the highly porous activated carbon materials were found to be non-ideal for stacked electrode design compared to carbon cloth materials because of potential limitation of substrate transport in the activated carbon pores.

The catalytic capability of the cathode is also an important factor that governs the MEC operation efficiency. For MEC cathode preparation, biocathodes or biocatalysts [22,23] are an alternative way to eliminate the use of expensive Pt catalysts [24]. The concept of biocathode was proposed based on the utilization of a large variety of microorganisms that can produce hydrogen gas [25]. The biocathode application resulted in a substantially higher current generation (3.3 A $m^{-2}$) than that in the control MEC (0.3 A $m^{-2}$; cathode without biofilms) in a previous study [23]. Thus, we examined the biocatalytic effect in our newly designed MEC reactors.

In this study, a new design of stacked electrodes was proposed and examined in MECs to improve the electric current generation and organic removal rate. The repeated stack with narrow inter-electrode distance allowed significantly large specific electrode areas and the low resistance. The stack of multiple electrode pairs can result in slow transport of reactants and products of the MEC electrode reactions, making the MEC performance limited by mass transfer. The mass transfer limitation can be reduced by employing continuous flow operation at a high flow rate. Continuous flow operation is usually designed for practical applications in wastewater treatment and energy production while fed-batch operation is widely used for lab-scale MEC studies.

We also examined various MEC operating conditions, such as fed-batch, continuous flow with effluent recycle, and continuous flow without effluent recycle to optimize the electric current generation and reactor operation for the stacked MEC design. For continuous flow operation, an increased flow rate can reduce the mass transport limitation for a high electric current generation. At a high flow rate, however, the exoelectrogenic microorganisms growing on the anode can be washed away. In addition, individual electrodes in a stacked design can perform differently depending on their location and other operating conditions, such as flow rate. Since there are no previous studies on the performance of individual electrodes under continuous flow conditions, we focused on providing the clear understanding of individual electrode performance in stacked-electrode MECs. Other specific objectives of this study are to: investigate the start-up of the stacked electrode MECs; evaluate the performance of the MECs under fed-batch, continuous-recycle, and continuous-flow operation conditions; examine two stackable cathode materials (carbon cloth and stainless steel mesh); maximize the electric current generation and organic removal rate in 3 stacked MECs (1, 5, and 10 electrode pairs); and study the performance of individual electrodes on electric current generation.

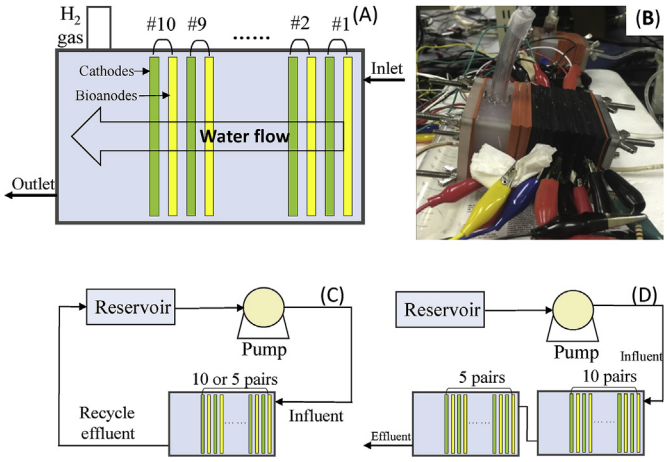


**Fig. 1.** (A) Schematic diagram of MEC-10 design; (B) Photograph of MEC-10; (C) Schematic diagram of continuous recycle operation; (D) Schematics for continuous flow operation.

## 2. Material and methods

### 2.1. Stacked MEC construction

A polypropylene block with a cylindrical hole (7 $cm^2$ in cross section) was used to build each MEC reactor. Four MEC reactors were built in this study: one MEC with 10 electrode pairs (MEC-10 as shown in Fig. 1A and B); one with 5 electrode pairs (MEC-5); and two MECs with a single electrode pair. Activated carbon cloth (ACC100, Evertech Envisafe Ecology, Taiwan) was used as the anode after treating in a surfactant solution [26]. The stainless steel mesh (304 stainless steel, 200 × 200 mesh, McMaster Carr, USA) was used as the cathode without any precious metal catalysts in MEC-10, MEC-5, and one of the MECs with a single electrode pair. For the other MEC with a single electrode pair, the cathode was the same material as the anode (activated carbon cloth). Two rubber gaskets and one plastic mesh were sandwiched between the anode and cathode to avoid potential electric short circuits between the electrodes as previously described [27]. The mean distance between the electrodes as well as between the electrode pairs was 2.8 mm spaced by the two rubber gaskets. Each electrode pair was operated and monitored independently. The effective volume of MEC-10 and MEC-5 were 40 and 35 mL, respectively. The volume of the MECs with a single pair of the electrodes was 28 mL. The upper and lower parts of each electrode were cut off by 0.3 cm from the edge to allow water and gas flow, resulting in 5.42 $cm^2$ of the surface area per electrode. Therefore, the specific surface area for MEC-10 (10 electrode pairs) was 136 $m^2 m^{-3}$ and 77 $m^2 m^{-3}$ for MEC-5 (5 electrode pairs).

### 2.2. Reactor operation

Effluent from existing lab-scale MECs that were originally started with primary clarifier effluent from a local wastewater treatment plant was used to inoculate the constructed MECs during the start-up period. Thus, the experiments were conducted with mixed-culture MECs. The feed solution was prepared with sodium acetate (1 or 2 g $L^{-1}$