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## Long-term performance of a 20-L continuous flow microbial fuel cell for treatment of brewery wastewater

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### HIGHLIGHTS

- A 20 L MFC system containing two 10 MFC reactors was constructed.
- No catalysts, Nafion or ion exchange membrane was used.
- The MFC system was operated with brewery wastewater for nearly a year.
- Operational conditions were tested and the MFCs can recover from equipment failure.
- The highest COD removal efficiency is  $94.6 \pm 1.0\%$ .

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### ABSTRACT

Microbial fuel cells (MFCs) have been shown as a promising technology for wastewater treatment. Integration of MFCs into current wastewater treatment plant have potential to reduce the operational cost and improve the treatment performance, and scaling up MFCs will be essential. However, only a few studies have reported successful scale up attempts. Fabrication cost, treatment performance and operational lifetime are critical factors to optimize before commercialization of MFCs. To test these factors, we constructed a 20 L MFC system containing two 10 L MFC reactors and operated the system with brewery wastewater for nearly one year. Several operational conditions were tested, including different flowrates, applied external resistors, and poised anodic potentials. The condition resulting in the highest chemical oxygen demand (COD) removal efficiency ( $94.6 \pm 1.0\%$ ) was a flow rate of  $1 \text{ mL min}^{-1}$  (HRT = 313 h) and an applied resistor of  $10 \Omega$  across each MFC circuit. Results from each of the eight stages of operation (325 days total) indicate that MFCs can sustain treatment rates over a long-term period and are robust enough to sustain performance even after system perturbations. possible ways to improve MFC performance were discussed for future studies.

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### 1. Introduction

Wastewater treatment is an energy extensive process. Each year, wastewater treatment consumed approximately 110 TWh, which is

about 3–4% of the United States' electrical energy load. In the United States, each year \$25 billion is spent on domestic wastewater treatment, and another \$300 billion is spent on wastewater treatment plants. Based on the wastewater type and the process, wastewater treatment may require energy of  $0.5\text{--}2 \text{ kWh m}^{-3}$  of wastewater. By now, the well-established activated sludge (aerobic digestion) process has been used in most wastewater treatment systems. Although highly efficient and fast, this process is chemical- and energy-intensive with high capital and significant

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operational cost (60% of the operational cost accounts for sludge treatment and disposal). Up to 75% of the wastewater treatment plant energy is used for aeration of the activated sludge [1]. The aerobic activated sludge treatment can be standalone or it can be combined with anaerobic sludge digestion. The anaerobic digestion converts the organic carbon present in wastewater into gaseous energy carrier – methane. The latter allows energy to be generated from the treatment process, where 1 kg of methane can be harvested from 4 kg of chemical oxygen demand (COD), which corresponds to  $1.27 \text{ kWh kg-COD}^{-1}$  energy recovery, at approximately 35% of energy conversion of methane to electricity. The long treatment time, extensive methane purification, as well as, the efficiency of the methane conversion to electricity are the main drawbacks of anaerobic digestion [2].

Microbial fuel cells (MFCs) are devices that can simultaneously treat wastewater and recover energy as direct current [3–6]. MFCs use microbes as a catalyst to oxidize the organic compounds in their anode chamber and thus treat wastewater. Electricity is generated due to the ability of bacteria to interact electronically with electrode surfaces via a mechanism called extracellular electron transfer (EET) [7]. Compared to activated sludge process and anaerobic digestion, MFCs can operate at ambient temperatures and generate significantly less sludge [1]. Although the power production by MFCs is not enough to sustain energy neutral treatment yet, MFC can be integrated to the existing wastewater treatment plants to decrease the need for aeration and reduce sludge production.

During the last decade, MFC technology has attracted great research interest. However, several challenges must be overcome before these systems can be used for practical wastewater treatment applications. The ideal MFCs should have a high treatment capacity, low fabrication and maintenance cost, stable performance with a given waste stream, and a long operational lifetime.

Early MFCs were small lab-scale systems with liquid volumes less than 1 L [8,9], operated in batch-fed mode, and used synthetic wastewater or well-defined wastewater [10]. Recently, several reports have emerged about new efforts toward scaling-up MFCs. Lefebvre et al., built a membrane electrode assembly MFC with 2.9 L anode volume to treat domestic wastewater at a hydraulic retention time (HRT) of 2.4 h, and reached  $75 \pm 21\%$  removal of the total COD with a Coulombic efficiency (CE) of  $11 \pm 1\%$  [11]. Zhang et al., designed liter-scale tubular MFCs to treat primary effluent wastewater and reached 65–70% COD removal efficiency at an HRT of 11 h [12,13]. Dong et al., built a 90-L MFC by stacking anode and cathode modules into a 100-L MFC reactor vessel, and reached 87.6% COD removal efficiency while treating brewery wastewater at an HRT of 72 h [14]. Besides having a singular large reactor unit, another approach is to stack multiple small reactors together to form a bigger system. Zhuang et al., stacked 40 units of tubular MFCs to form a serpentine-type MFC with 10 L working volume, and reached 87.1% COD removal efficiency while treating brewery wastewater at an HRT of 48 h [15]. Ge et al., stacked 96 tubular MFC modules to build a MFC system with 200 L liquid volume to treat primary effluent wastewater for over a year, and reached 37.6% (anode) and 76.8% (anode + cathode) COD removal efficiency at an HRT of 18 h [16]. These efforts have shown that it might be more relevant to stack multiple MFC modules, since it allows for larger total volume capacity while each single reactor can function and be maintained independently.

Long-term continuous treatment of real wastewater with stable performance is also critical for MFC applications. COD, pH, total suspended solids, and other parameters of real wastewaters can vary greatly with weather or industrial/domestic activity, which can impact the microbial community within the MFCs and the overall system performance. Furthermore, fouling and clogging can

severely shorten the lifetime of MFCs [17,18].

Different from domestic wastewater, industrial wastewater usually contains much higher variety of pollutants and at higher amounts, requires more energy/economic input for treatment, and faces great penalty for improper disposal. Therefore, though MFC technology is still not optimized for power production, it has great potential to be used for industrial wastewater treatment, reduce the energy consumption and the cost of the overall manufactory. One example of industrial wastewater is brewery wastewater. Brewery wastewater is a broad definition. It is the wastewater generated during beer production, including many different unit operations (saccharification, fermentation, cooling, washing, etc.). The characteristics (COD, pH, DO, etc.) of brewery wastewater may vary greatly by the operations or the type of beer being produced. On average, brewery wastewater has high COD, typically in the range of  $3000\text{--}5000 \text{ mg L}^{-1}$ , which is approximately 10 times more than domestic wastewater [10]. Conventional biological methods such as anaerobic reactors can be effective for brewery wastewater treatment, but have long treatment time (15–40 days) [19] relative to aerobic solutions (8–12 h) [20]. Feng et al. and Wang et al. demonstrated the utilization of MFCs for treating brewery wastewater [21,22]. They treated brewery wastewater with COD of  $2250 \pm 418 \text{ mg L}^{-1}$  and COD removal efficiency of 85–87% [21,22]. The authors observed that the efficiency of the treatment process in terms of COD removal was not significantly influenced by temperature and conductivity but was greatly dependent on COD concentration, where higher strength wastewater showed higher COD removal rates. Wen et al. had similar conclusions for a small scale MFC treating brewery wastewater [23,24].

In this study, we constructed a 20-L continuous flow MFC system and operated it for nearly a year with brewery wastewater. To control the cost, our system was constructed with cost-effective materials and devoid of metal catalyst and Nafion (which can account up to 45% of the total cost) [25]. The long-term performance in terms of COD removal efficiency, pH, power and current densities, Coulombic efficiency, and microbial community composition were studied as a function of operational conditions. Our MFCs showed good treatment performance relative to other large-scale systems as well as stable performance over a long time (COD removal rate > 75% during the first 150 days). The diversity and relative abundance of bacteria in the present microbial population was studied and related to MFC operation. Possible improvements were also discussed.

Our study is an attempt for the development of a cost-effective MFC system for practical wastewater treatment, which will help understand the constraints of the scaling-up process and the utilization of brewery wastewater as possible waste stream. The observations performed here give valuable information for future efforts in the design and operation of MFCs for real applications.

## 2. Materials and methods

### 2.1. Brewery wastewater

Brewery wastewater can be accumulated during different stages of beer production. Therefore, the characteristics of the brewery wastewater vary greatly. In this study, brewery wastewater was collected from the equalization tank at a Brewery in Escondido, CA, USA, and stored at  $4^\circ\text{C}$  before being used. The COD of the raw brewery wastewater varied greatly from 6740 to  $20,900 \text{ mg L}^{-1}$ , depending on the type of beer being produced and the production activities. The brewery wastewater had a pH value around 4.6, which may inhibit microbial growth and activity. To maintain similar loading rates, the brewery wastewater was diluted with  $1 \text{ g L}^{-1}$  sodium bicarbonate solution. In practice, the acidic brewery

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