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Energy advantage of anode electrode rotation over anolyte recirculation for operating a tubular microbial fuel cell

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ABSTRACT

Mixing plays a key role in both electricity generation and organic removal in microbial fuel cells (MFCs) via affecting substrate distribution and internal resistance. Herein, two mixing methods, anode electrode rotation and anolyte recirculation, were investigated in terms of energy consumption and production. Anode electrode rotation could increase the maximum power density and COD removal by 81.5 and 45.7%, respectively, when the rotating speed increased from 0 to 45 rpm. Likewise, anolyte recirculation also improved the power density and COD removal by 43.1 and 30.1%, respectively, at an increasing rate from 0 to 300 mL min^{-1}. The enhancement of electricity generation became less significant at a high mixing level, likely because that substrate supply was relatively sufficient and other factors posed more effects on electricity generation. The MFC with anode electrode rotation achieved a higher energy balance (e.g., 0.254 kWh kg COD^{-1} at 35 rpm) than the one without any mixing (0.124 kWh kg COD^{-1}), while anolyte recirculation led to a lower or even negative energy balance compared to that with no mixing. The results of this study have demonstrated energy advantages of anode electrode rotation and encouraged further exploration of energy-efficient mixing methods for MFC operation.

1. Introduction

Microbial fuel cells (MFCs) have been intensively studied in the past decade for bioelectricity generation from low-grade substrates such as wastewater [1]. Much progress has been made in understanding fundamental issues such as microbiology [2] and electrochemistry [3] and optimizing systems via reactor architecture and scaling up [4]. However, there are still challenges to address, which hinder MFC development towards practical application. In particular, a thorough understanding of energy performance of MFCs including both energy production and energy consumption has not been well achieved [5]. To better describe energy performance, a parameter - normalized energy recovery (NER) was proposed for data presentation in either kWh m^{-3} (based on the volume of the treated wastewater) or kWh kg COD^{-1} (based on the removed chemical oxygen demand - COD) [6]. Energy production by MFCs is relatively straightforward and can be estimated by using the data of power production and operating time. However, the majority of MFCs studies have failed to report energy consumption, thereby making it difficult to evaluate whether MFCs are really energy efficient precisely. The major energy consumers in a typical MFC include aeration and pumping system (for feeding and recirculation) [7]. A recent review paper has discussed the importance of energy consumption and provided detailed analyses of energy consumption of several major bioelectrochemical systems based on literature data [8].

One of the key factors to MFC operation and also its energy performance is “mixing”, which is to increase the distribution of substrates. The enhanced substrate distribution can decrease concentration overpotential through reducing the concentration gradients of reactants/products adjacent an electrode, thereby benefiting electricity generation [9]. Typical methods for mixing include magnetic stirring (e.g., bench-scale MFCs) and anolyte recirculation (e.g., larger-scale MFCs). It has been well demonstrated that optimized mass transfer of substrates and mediators can lead to a significant increase in power output. For example, in a tubular MFC, a higher anolyte recirculation rate could result in a less negative energy balance due to a relatively higher energy production [10]. It was reported that enhanced mass transfer by applying a higher recirculation rate in an MFC can help cultivate electrochemically active biofilm that is thicker and denser for better attachment, resulting in the increased maximum power density from 50 to 160 W m^{-3} [11]. A recent study of an 85-L MFC found that recirculating the anolyte increased the maximum power density from 0.101 ± 0.006 to 0.118 ± 0.006 W m^{-2}, a 17% improvement

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compared to a static flow condition [12]. To better understand the effect of liquid flow on substrate distribution and thus electricity generation, a computational fluid dynamics model was developed and used to predict the optimal flow to improve substrate distribution [13].

Another mixing method that is studied much less frequently is the electrode rotation. "Rotation" has been employed in a wastewater treatment technology - rotating biological contactors (RBC) for providing mixing and substrate supply [14]. The similar concept was adopted by researchers to develop a rotatable bioelectrochemical contactor that could alternate the anode and the cathode electrodes in the air and liquid [15], but it is more common to rotate one of those electrodes for enhancing mixing. In an early study of rotating electrode, a sediment MFC with a rotating cathode electrode increased the maximum power density by 69% [16]. Similar improvement was obtained in another study that had the power output increased from 486 ± 11 to 879 ± 16 mW m^{-2} with applying 20 rpm cathode rotating speed [17]. To reduce energy consumption, cathode rotation may be driven by a hydraulic flow [18], though this has not been demonstrated yet. Rotating an anode electrode at a very slow speed of 3 rpm could increase power production by 1.4 times compared to the MFC without anode rotation [19]. Despite improved electricity generation with rotation, it is unclear whether it is energetically favourable to perform rotation because it also requires energy input. In this study, we have investigated the effects of anode electrode rotation on MFC energy performance, and compared it to a commonly used mixing method – anolyte recirculation. This is the first time that such a comparison was conducted from the aspect of energy performance in MFCs.

2. Materials and methods

2.1. MFC setup and operation

A tubular MFC was fabricated according to a previous study [20] with details shown in Fig. 1 (cell sizing ID = 90 mm, L = 400 mm). A tube made of cation exchange membrane (CEM, CMI-7000, Membrane International Inc., Glen Rock, NJ, USA) was used to separate the anode and cathode chambers. The liquid volume of the CEM tube (anode chamber) was about 800 mL and the cathodic volume was 1200 mL. The anode electrode was a carbon brush (Gordon Brush Mfg. Co., Inc., CA, USA). Before use, the carbon brush was pre-treated by being soaked in acetone for 12 h and then heat-treated in a muffle furnace (Model 550 Isotemp Series, Fisher Scientific, Pittsburgh, PA, USA) at 450 °C for 30 min. The anode electrode was rotated by a variable speed motor. The cathode electrode was a piece of carbon cloth (25 cm × 16 cm) coated with 4 mg cm^{-2} activated carbon powder as a catalyst for oxygen reduction.

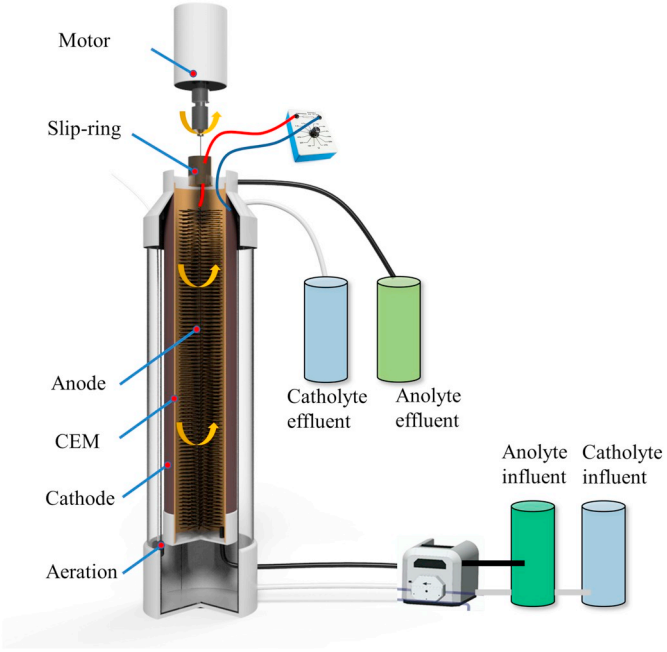

Fig. 1. Schematic diagram of the tubular MFC with a rotating carbon brush anode.

The MFC anode chamber was inoculated with anaerobic sludge from a local wastewater treatment plant (Peppers Ferry, Radford, VA, USA). The MFC was operated in a continuous mode and at a room temperature of 21 ± 2 °C. During the first 10 days, the external resistance of the system was changed from 5000 to 10 Ω gradually to slowly generate a higher current. After the MFC system was maintained at 10 Ω for 3 days, the tests for data collection started. Synthetic domestic wastewater was prepared containing (per L DI water): 0.5 g sodium acetate, 0.1 g NH_4Cl, 0.5 g NaCl, 0.015 g $MgSO_4$, 0.02 g $CaCl_2$, 1 g $NaHCO_3$, 0.53 g KH_2PO_4, 1.07 g K_2HPO_4, and 1 mL trace element [21]. The cathode was fed with 0.05 M NaCl as a catholyte at the flow rate of 1.67 mL min^{-1} (12 HRT). The anode rotating speed was controlled at 15, 25, 35, or 45 rpm. In the experiment of anolyte recirculation, the anolyte was recirculated at 0, 150, or 300 mL min^{-1}.

2.2. Measurements and analysis

The COD concentration was measured by using a colorimeter (DR89, Hach Company, Loveland, CO, USA). The voltage over an external resistor of 10 Ω was monitored every 2 min by a data logger (2700, Keithley Instruments Inc., Cleveland, OH, USA). Polarization tests were conducted by using a potentiostat (Reference 600, Gamry Instruments, Warminster, PA, USA) at a scan rate of 0.5 mV S^{-1}. Power density and current density were calculated based on the liquid volume of the anode chamber.

The estimate of energy balance was conducted by calculating the difference between energy production and consumption. Energy production was expressed in kWh kg COD^{-1} based on the amount of organic substrates removed in the MFC [6]. The detail calculation method of Reynold's number and theoretical energy consumption (anode electrode rotation and anolyte recirculation) were provided in the Supplementary Materials.

3. Results and discussion

3.1. MFC performance with anode electrode rotation

The effects of anode electrode rotation on the MFC performance was investigated by varying the anode rotating speed from 0 to 45 rpm. As shown in Fig. 2A, the operating current density of the MFC increased from 14.95 ± 0.28 A m^{-3} (0 rpm) to 16.23 ± 1.30 A m^{-3} (15 rpm) and eventually to 24.22 ± 1.04 A m^{-3} (45 rpm). This represents an improvement of current generation by 62% from 0 to 45 rpm. The rotation speed played a significant role in medium (anolyte) turbulence. This reflected in the estimated Reynolds number N_{Re} that increased from 783 (15 rpm) to 2351 (45 rpm). A higher Reynolds number is able to obtain a more negative anode potential, indicating a better performance of electrochemically-active bacteria [22]. This is related to the increased diffusion of anolyte containing the substrate, trace elements and other ions into the anode biofilm. Meanwhile, fluid motion also increases the diffusion of protons and other metabolites out of the anode biofilm, thereby minimizing a localized pH gradient within the anode biofilm [23]. In addition, a higher Reynolds number of the anolyte is able to decrease the anode resistance and transport resistance for ions through ion exchange membrane, resulting in a higher current density [24]. When the rotation stopped, current density quickly decreased to 18.01 ± 1.96 A m^{-3} and then recovered to 24.01 A m^{-3} after the anode rotation re-started at 35 rpm, which demonstrated that rotation speed played the significant role in current density increasing. The

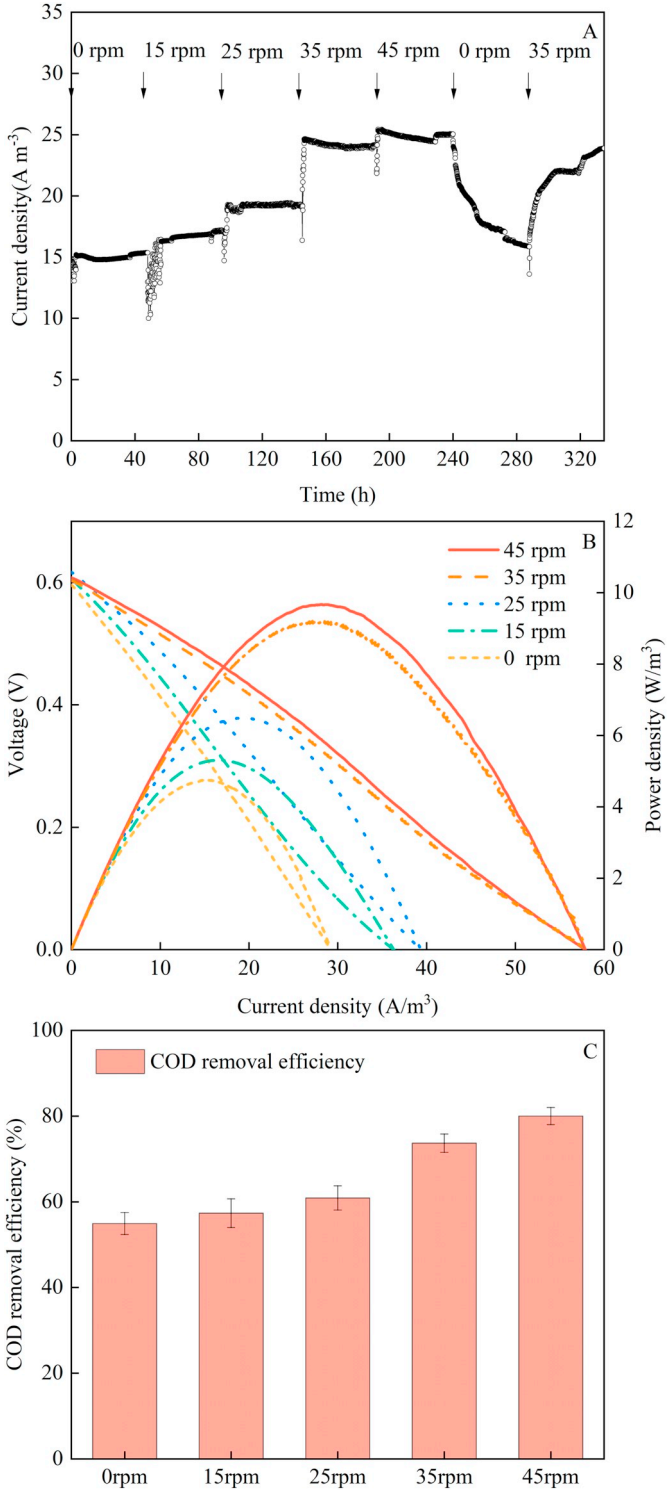

Fig. 2. Effects of anode electrode rotating speeds on MFC performance: (A) operating current density; (B) polarization curves; and (C) COD removal efficiency.

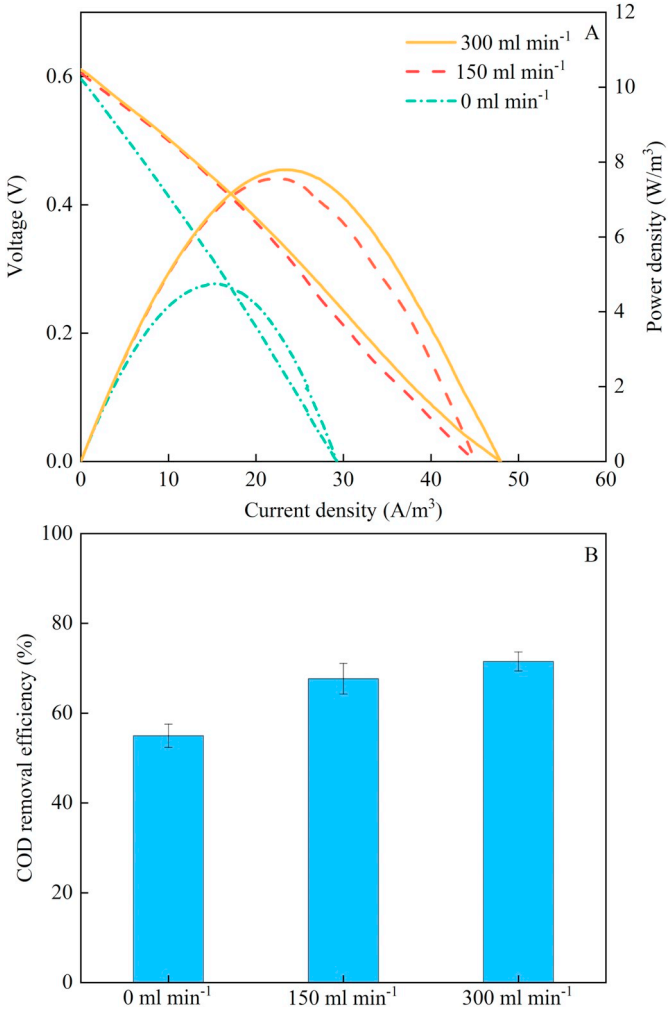

Fig. 3. Effects of anolyte recirculation rates on MFC performance: (A) polarization curves; and (B) COD removal efficiency.

performance of electricity generation was further evaluated with polarization tests (Fig. 2B). Like current generation, the maximum power density increased from $5.03 \pm 0.43\ W\ m^{-3}$ (0 rpm) to $9.13 \pm 0.72\ W\ m^{-3}$ (45 rpm). The increase of the maximum power density from 0 to 35 pm was significant ($p < 0.05$), but the difference between 35 rpm ($8.95 \pm 0.48\ W\ m^{-3}$) and 45 rpm ($9.13 \pm 0.72\ W\ m^{-3}$) was insignificant ($p > 0.05$). Those results have demonstrated that increasing anode electrode rotation could enhance electricity generation of the MFC and this enhancement became less significant at a high rotating speed (> 35 rpm).

The rotation of the anode electrode has also affected organic removal in the anode. Although the COD removal efficiency was not obviously different between 0 rpm ($54.9 \pm 2.6\%$) and 15 rpm ($57.4 \pm 3.4\%$, $p > 0.05$), further increasing the rotating speed gradually improved COD removal to $60.9 \pm 2.8\%$ (25 rpm), $73.7\% \pm 2.1\%$ (35 rpm), and $80.0 \pm 2.0\%$ (45 rpm) (Fig. 2C). The corresponding COD removal rate was 0.43 ± 0.02 (0 rpm), 0.43 ± 0.01 (15 rpm), 0.49 ± 0.01 (25 rpm), 0.57 ± 0.02 (35 rpm), and $0.63 \pm 0.02\ kg\ COD\ m^{-3}\ d^{-1}$ (45 rpm). Those results suggested that the increasing anode rotating speed has benefited COD degradation by microorganisms likely via improved substrate distribution. It is interesting to notice that the COD removal increased significantly from $73.7 \pm 2.1\%$ (35 rpm) to $80.0 \pm 2.0\%$ (45 rpm), while the increase of power output was insignificant. This was possibly because that anode electrode rotation enhances the mass transfer, thereby increasing the activities of electrochemically-active bacteria as well as other ordinary heterotrophic organisms (OHO). In particular, other OHO may consume COD significantly without contributing to electricity generation [25]. This was partially supported by the difference in COD removal under an open circuit condition (no electricity generation): $52.9 \pm 2.3\%$ (or $0.41 \pm 0.02\ kg\ COD\ m^{-3}\ d^{-1}$) at 45 rpm and $34.7 \pm 2.4\%$ (or $0.27 \pm 0.02\ kg\ COD\ m^{-3}\ d^{-1}$) at 0 rpm. In addition, the results may also indicate that at a higher rotating speed the substrate supply may not be a key limiting factor to electricity generation; that is,

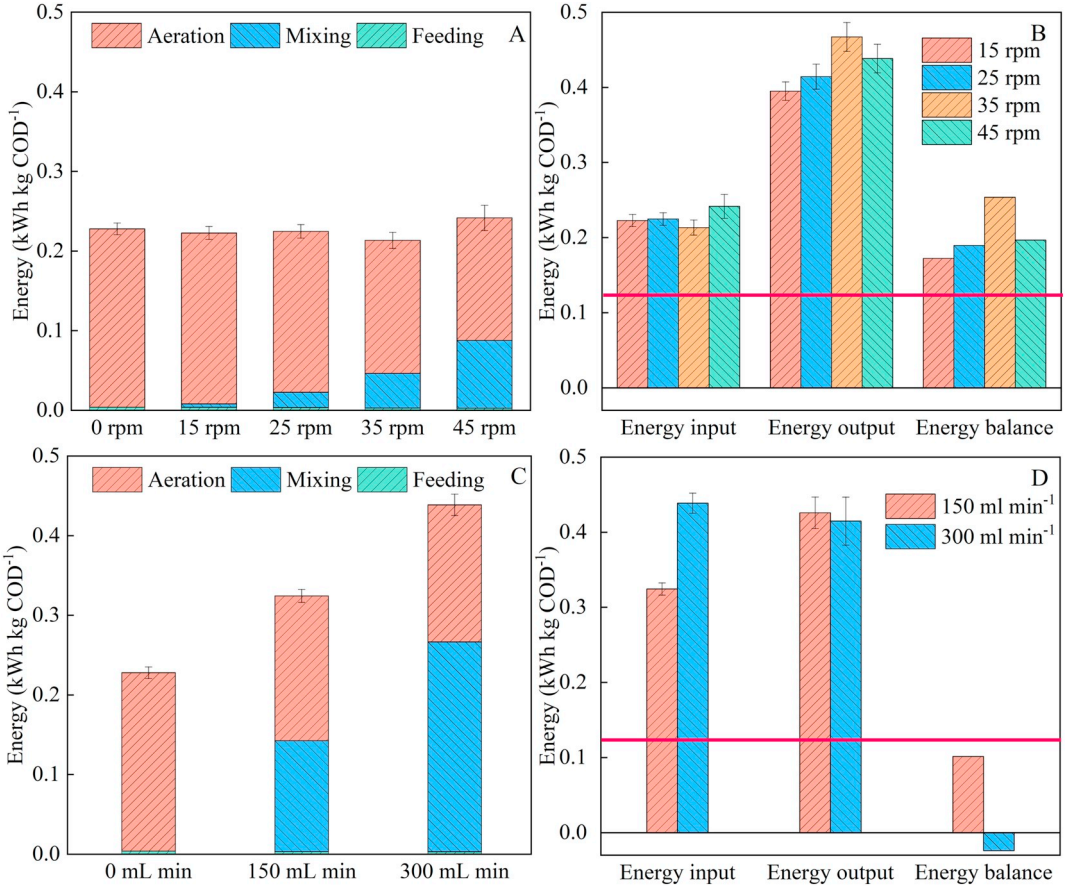

Fig. 4. Energy performance and comparison of mixing methods: (A) energy consumption by anode electrode rotation; (B) energy balance of the MFC with anode electrode rotation; (C) energy consumption by anolyte recirculation; and (D) energy balance of the MFC with anolyte recirculation. Red lines in panel B and D indicate the energy balance without mixing. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

electrochemically active microorganisms cannot convert all of the available substrates to electricity, thereby limiting further increase of current generation.

3.2. MFC performance with anolyte recirculation

For the purpose of comparison, the MFC was also operated with anolyte recirculation (no electrode rotation) at three recirculation rates, 0, 150 and 300 mL min^{-1}. As expected, the operating current density across 10 Ω increased from 14.95 ± 0.28 A m^{-3} (0 mL min^{-1}) to 18.77 ± 0.27 A m^{-3} (150 mL min^{-1}), and eventually to 20.39 ± 0.53 A m^{-3} (300 mL min^{-1}). The polarization curves show that the recirculation rate of 300 mL min^{-1} resulted in the highest maximum power density of 7.71 ± 0.40 W m^{-3}, much higher than 5.03 ± 0.43 W m^{-3} (0 mL min^{-1}) (Fig. 3A). However, the difference between 300 mL min^{-1} (7.71 ± 0.40 W m^{-3}) and 150 mL (7.49 ± 0.37 W m^{-3}) was insignificant ($p > 0.05$). It has been reported that high shear rates can result in stronger aggregation and attachment of microbes [26]. The biofilm in an MFC with the high shear rate is not only thicker but also denser than that in the low shear enriched MFC, resulting in improved MFC performance [27]. However, there is a limiting level named "tensile strength" at which an excessively high a shear force will cause detachment of the biofilm [28]. In this study, the maximum power density was not reduced at the high mixing strength (45 rpm and 300 mL min^{-1}), indicating that the shear force strength employed here has not reached the tensile strength.

The COD removal was also increased from 54.9 ± 2.6% (0.43 kg ± 0.02 COD m^{-3} d^{-1}) at 0 mL min^{-1} to 67.7 ± 3.4% (0.53 ± 0.03 kg COD m^{-3} d^{-1}) at 150 mL min^{-1} and then to 71.5 ± 2.1% (0.56 kg ± 0.02 COD m^{-3} d^{-1}) at 300 mL min^{-1} (Fig. 3B). Those results confirmed the importance of recirculation (mixing) to electricity generation via better substrate supply. However, the improvement of electricity generation by recirculation would be limited at a relatively high recirculation rate, indicating that electricity generation is limited by factors other than mass transfer under a condition of sufficient mixing.

3.3. Comparison of energy performance

In this section, energy performance (production, consumption, and balance) of the MFC with anode electrode rotation or anolyte recirculation was described separately, followed by a comparison between the two. The MFC with anode electrode rotation consumed 0.228 ± 0.007, 0.223 ± 0.008, 0.225 ± 0.008, 0.213 ± 0.010, and 0.242 ± 0.016 kWh kg COD^{-1}, as the rotating speed increased from 0 to 45 rpm (Fig. 4A). Aeration was the main energy consumer, making 63.7 to 98.3% of overall energy consumption. The contribution by anode electrode rotation increased from 2.0 to 35.2% with the increasing rotating speed. The feeding pump had a very minor contribution to energy consumption. The energy production of the MFC increased from 0.395 ± 0.012 kWh kg COD^{-1} (15 rpm) to 0.467 ± 0.019 kWh kg COD^{-1} (35 rpm). Further increasing the rotation speed to 45 rpm slightly decreased the energy production to 0.438 ± 0.019 kWh kg COD^{-1}, likely related to the faster increase in COD removal than that of electricity generation. The overall energy balance of the MFC with anode rotation was all higher than 0.124 kWh kg COD^{-1} at 0 rpm (Fig. 4B), indicating that anode electrode rotation could enhance the energy performance of the MFC.

The MFC with anolyte recirculation required 0.228 ± 0.007, 0.324 ± 0.008, and 0.439 ± 0.013 kWh kg COD^{-1} when the recirculation rate was 0, 150 and 300 mL min^{-1}, respectively (Fig. 4C). The contribution of aeration became much smaller at 56.1% (150 mL min^{-1}) and 39.2% (300 mL min^{-1}), compared to that with anode electrode rotation. The contribution of anolyte recirculation to energy consumption increased from 43.0% to 60.1% with the increased recirculation from 150 to 300 mL min^{-1}. The energy production of the MFC was 0.426 ± 0.021 kWh kg COD^{-1} at 150 mL min^{-1} and 0.415 ± 0.032 kWh kgCOD^{-1} at 300 mL min^{-1} (Fig. 4D). However, the energy balance of the MFC was 0.101 kWh kg COD^{-1} (150 mL min^{-1}) and −0.024 kWh kg COD^{-1} (300 mL min^{-1}), both of which was below that with no recirculation (0.124 kWh kg COD^{-1}). At the 300 mL min^{-1}, the MFC had a negative energy balance, due to more energy consumption by the high recirculation rate than its energy production.

Comparison of energy performance between anode electrode rotation and anolyte recirculation is straightforward. The MFC with anode electrode rotation has achieved a higher energy balance than the benchmark (the one without any mixing, indicated by the line in both Fig. 4B and D), while anolyte recirculation led to a lower or even negative energy balance. Thus, anode electrode rotation could potentially have an energy advantage over anolyte recirculation, and this advantage benefits from both lower energy consumption and higher energy production with anode electrode rotation.

Despite the promising results, we should also note the limitations of the findings that would warrant further investigation. For example, substrate supply could be affected by the concentration of organic compounds and a higher organic loading rate may lower the requirement of mixing. Thus, the energy benefits of anode electrode rotation should also be evaluated under a high-strength organic input. Anode electrode rotation has a higher requirement for equipment (e.g., motors) when MFCs are deployed in modules containing multiple anode electrodes, compared to hydraulic recirculation that can be conducted by fewer pumps. The energy benefits of anode electrode rotation may be further improved through coupling with proper reactor designs by installing baffles/channels that can optimize flow.

4. Conclusions

This study has evaluated two mixing methods in the aspect of energy performance and demonstrated the advantage of anode electrode rotation over anolyte recirculation. Increasing the mixing via either anode electrode rotation or anolyte recirculation could improve both electricity generation and organic removal, but the enhancement of electricity generation became less significant at a stronger mixing intensity. The tested anode electrode rotation speeds resulted in positive energy balance, higher than that in the absence of mixing. However, anolyte recirculation had a large energy consumption and led to lower (or even negative) energy balance than the benchmark value. This study has demonstrated that the rotating anode electrodes to provide mixing will require less energy input and generate more energy, although such benefits need further evaluation by considering factors such as organic concentration, MFC modularization, and hydraulic flow pattern.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.elecom.2019.106529.

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