Influence of Very High Flow Rates on Performance of Biofilter-Microbial Fuel Cells

Songyot Mongkulphit, Petch Pengchai, and Nattawoot Suwannata

***Abstract*—Scaling-up microbial fuel cells for continuous-flow wastewater treatment is the conventional challenge for numbers of researchers. Here in this study, we constructed large volume biofilter-microbial fuel cells (BMFCs) by applying graphite electrodes to the low-cost biofilters. Very high flow rates of 625-2,667 mL/hr (15-35 L/day) were applied to the BMFCs to explore their influence on wastewater treatment and electricity generation. An effect of hydraulic retention time (HRT) was expunged from the experiment by using various chamber volumes under the same HRT of 5 hrs. The result revealed that 32-73 % of COD removal, 5-14% of TN removal, 13-18% of TP removal, and 16.2-53 mW/m^2 of power output could be achieved by the BMFCs. Higher flow rates led to higher pollutant removal rates and higher power densities under the linear regression equations with determination coefficients (R^2) of 0.81-0.99. As the power density was the linear function of the pollutant removal rates (R^2 = 0.93-0.97), the increasing shear rate which accorded with the increasing flow rate was considered as the key factor to enrich biomass and provoke electrogenic activity in an anode chamber. Therefore, the highest pollutant removal rates and highest power density were observed at the highest flow rate.**

***Index Terms*—Biofilter-microbial fuel cell, flow rate, shear stress, COD.**

I. INTRODUCTION

Among numbers of water supply and wastewater treatment technologies which require more than 2% of the world's electrical energy [1], microbial fuel cell (MFC) is an alternative technology which can produce electrical power while removing pollutant from wastewater. A conventional MFC consist of anodic and cathodic compartments. In an anodic chamber, microorganisms oxidize organic fuel, such as wastewater, and release electrons and protons during their metabolism. The released electrons which were successfully transferred to an electrode (anode) will then flow through an external electrical circuit to another electrode (cathode) at a cathodic compartment. Meanwhile, the released protons will diffuse from an anolyte through a proton exchange membrane to the cathodic compartment. The electrons, protons, and oxygens at the cathodic compartment were then combined together and become molecules of water according to a reduction reaction. As MFCs can be operated efficiently at ambient temperature without any toxic by-products and their fuel to electricity conversion is not limited by the Carnot cycle because the chemical energy is converted directly into electricity instead of incurring partial heat losses [2], variety of researches has been done to develop this prospective technology into practical use. An MFC demonstration cell containing 1 L solution, 4 graphite brush anodes, and two cathodes was used to run a fan for more than 1 year without constant feeding [3]. A mobile phone was charged by feeding real neat urine to membrane-less MFCs made of ceramic material and plain carbon-based electrodes [4]. In year 2017, an MFC treatment system called “EcoVolt” was firstly brought to market application by an American company [5]. Recently, a cascade comprising 4 membrane-less MFCs modules was used to directly power a microcomputer and its screen with 158-mW electrical power [6]. This is the first time that a MFC system has been reported to directly and continuously power a small application without any electronic intermediary [6].

Although the scientists have successfully improved power density (a proportion to an anode area) of a single cell MFC from less than 1 mW/m^2 [7] to 6.9 W/m^2 or over 1 kW/m^3 under optimal conditions [3], there are still barriers for the practical use. The first constraint is their high installation cost due to an application of proton exchange membrane as a chamber separator and platinum as a cathode catalyst [7]. Thus, the membrane-less-tubular MFC without the addition of mediators were of our interest due to its economic advantage and wastewater treatment capacity in scaled-up MFC. The second constraint of MFCs is their low power output due to ohmic losses, activation losses and concentration losses, etc. [7]. When volume of an MFC increases, the output power reduces proportionally [8]. Maximum attainable voltage of any single cell MFC with oxygen as an electron acceptor remains less than 1 V [9] with low electrical current of 0.01-1000 order of micro ampere [10]. Therefore, the typical volumes of a single cell MFC reactor were tens to hundreds of milliliters and the improvement of MFC volumetric power densities was usually done by increasing total surface area of the electrodes per reactor volume [3]. However, for the real application in wastewater treatment, MFCS should be appropriately operated in conditions of higher operational volume and continuous flow. Thus, an influence of wastewater flow rate on MFC performance is needed to be clarified, especially in

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Manuscript received January 20, 2020; revised December 13, 2020. This work was supported in part by the Kurita-AIT Research Grant from Kurita Water and Environment Foundation (KWEF) and Doctoral Research Grant from Faculty of Engineering, Mahasarakham University. Put a space between authors' initials.

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doi: 10.18178/ijesd.2021.12.3.1320

case of large chamber volume and high substrate flow rate.

It has been proved by several researches that an increase of the flow rate leads to an increase of the power output [11]. However, some experts have found that MFCs underperformed at very high flow rates. Power densities of 0.78-0.91 mW/m^2 were generated during 1.5 mL/h flow rate whereas obviously lower power densities of 0.33-0.56 mW/m^2 were detected under 125 mL/h condition [12]. Similarly, another work indicated that for the range of 9-180 mL/h flow rate, the best performance in power generation (37.3 mW/m^2), COD removal efficiency (always higher than 74%), and columbic efficiency (41.9%) was seen at the lowest flow-rate [11]. Longer hydraulic retention time (HRT) which associated with lower flow rate was addressed as the key point for higher power-generation [12]. However, in terms of shear rate effect, it was proved that high shear rate (120 s^{-1}) which in accordance with high flow rate caused 2 to 3 times higher power output than that in a case of low shear rates, 0.3 s^{-1} [13]. Biomass and biofilm analyses showed that the anodic biofilm in the MFC was highly enriched under high shear rate conditions [13]. These facts supported an assumption that high influent flow rates that associated with high shear stress may provide high power output if the HRT is sufficient for microorganism metabolism in an anodic chamber. This study was aimed to prove the above assumption with the following unique points, 1) the application of very high flow rates (625-2,667 mL/hr or 15-35 L/day) to large single-chamber MFCs (4.16-9.72 L), 2) the isolation of shear stress effect from that of HRT by using various chamber volume under the same HRT, 3) the very low-cost MFC configuration, 4) the concern of total nitrogen and total phosphorus as well as COD removal rates.

II. METHOD

A. Reactor Construction

Five membrane-less, air-cathode, single-chamber MFCs were constructed from 10-cm diameter polyvinyl chloride pipe with different chamber volume of 4.16 L, 5.56 L, 6.95 L, 8.33 L, and 9.72 L as shown in Fig. 1.

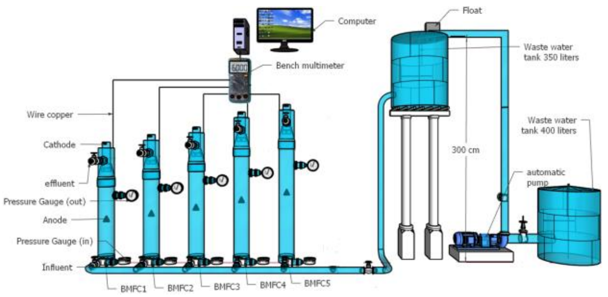

Fig. 1. BMFCs used in this study.

Influent ports and effluent ports were made in the base and top of the MFCs. For each MFC, a triangular anode made of graphite plate (projected surface area of 5.2 cm^2) was place in the middle of the chamber. An air-cathode made of graphite plate (projected surface area of 20.78 cm^2) was placed at the water surface of an effluent port (adapted from the configuration used by another Thai famous researcher [14]). Copper wire in plastic tube sealed with hot glue stick was used to connect the electrodes to electrical circuits. Bundles of 20 nylon ropes were filled inside a chamber to create a pack-bed filter with media bed porosity of 75%. Predominant microorganism of anodic compartment which could remove pollutant as well as produce electricity should grow up and attached to the media. Since an anode chamber could also function as biofilter, these reactors were called Biofilter Microbial Fuel Cells (BMFCs).

B. Synthetic Wastewater

As the highest open circuit voltage (1.29V) ever reported in the MFC system was observed in landfill leachate fed MFCs [15], we selected synthetic landfill leachate modified from a literature [16] as our treatment scope. One-liter of our synthetic landfill leachate consisted of 0.538 mL acetic acid (CH_3COOH), 0.385 mL propionic acid ($C_3H_6O_2$), 12 mg $MgSO_4$, 221.7 mg $CaCl_2$, 24.92 mg Na_2CO_3, 184.6 mg $(COO{\cdot}NH_4)_2{\cdot}H_2O$, 110.8 mg NaCl, and 0.076 mL Trans metals solution (TMS). NaOH solution was added to the synthetic landfill leachate for pH 7 adjustment. TMS was prepared from $CuCl_2$ 40 mg, $(NH_4)_2SO_4{\cdot}NiSO_4{\cdot}6H_2O$ 50 mg, $(NH_4)_2Fe(SO_4)_2{\cdot}6H_2O$ 2000 mg, $BaCl_2{\cdot}2H_2O$ 50 mg, $MnSO_4{\cdot}4H_2O$ 500 mg, 96% sulfuric acid (H_2SO_4) 1 mL and certain volume of distilled water to make 1-L solution [16].

C. Experiment Set up

The experiment included start-up phase and treatment phase. In start-up phase, the media beds of BMFCs were individually filled with the inoculum composing of photosynthetic bacteria (PB) mixture prepared from Siam Rhodo PB liquid fertilizer, and real landfill leachate collected from Nong Pling municipal landfill site, Mahasarakham province, Thailand at ratio of 1:22. After the letting the pack-bed filters soaked in the inoculum for 59 days, BMFCs were then continuously fed with the real landfill leachate (COD 15,755.9 mg/L, BOD 550 mg/L, total nitrogen (TN) 74.04 mg/L, total phosphorus (TP) 45.11 mg/L, pH 9.4) with recirculation ratio of 100% for 12 days. Applied feeding rates were different, i.e. 15 L/day for BMFC1, 20 L/day for BMFC2, 25 L/day for BMFC3, 30 L/day for BMFC4, 35 L/day for BMFC5 under the same HRT of 5 hrs. Some researchers from France and India indicated that not less than 80 % COD removal from low strength wastewater (1900 ± 200 mgCOD/L) could be achieved using a high rate anaerobic filter at HRT of 4 hr [17]. Based on our previous work, HRT of 5.4 hr. was proved to be enough for 80% COD removal by an up-flow anaerobic filter [18]. Further, the Indonesia researchers reported the BOD removal efficiency of anaerob biofilter reactor as 10.81-66.39 % for 24-hr HRT, 69.62-72.85 % for 6-hr HRT, and 57.34-88.36 % for 3-hr HRT [19]. Thus, the HRT of 5 hours should be sufficient for our BMFCs. At the end of the inoculation, real landfill leachate was continuously fed into individual BMFC again for 10 days without recirculation to confirm that the BMFCs were available for wastewater treatment and power generation.

In the treatment phase, the synthetic landfill leachate (COD 1,781.33 mg/L, TN 91.12 mg/L, TP 19.28 mg/L) was fed to each BMFC reactor with the flow rate as previously described for 10 days. An external resistor was selected to connect an anode to a cathode of each BMFC based on the result of polarization experiment. Real-time monitoring of voltage was done for each BMFC throughout the experiment using a bench multimeter (GDM-8255A, Good Will Instrument Co., Ltd.) Samples of influent and effluent were collected and analyzed for COD using closed reflux titrimetric method, TN using alkaline peroxodisulfate digestion method [20], TP using Vanadomolybdophosphoric Acid Colorimetric Method indicated as APHA Standard Methods 4500-P [21], pH using pH meter, and oxidation reduction potential (ORP) using ORP meter.

D. Polarization Experiment

Polarization experiment was done for each BMFC every 7 days by connecting anode to the cathode through 9 various external resistances, i.e., 20,000; 10,000; 7,500; 2,200; 1,000; 560; 250; 150, and 10 ohms. Voltages (V) across each external resistance (R_{ext}) were measured and applied to equation $P = VI = V^2/R_{ext}$ to calculate an electrical power (P) transferred to the R_{ext}, whereas I is the electrical current. The R_{ext} which provide maximum P for each BMFC was determined and applied in the BMFC operation.

E. Michaelis-Menten Kinetics

To compare the pollutant removal rates ($\nu, mg \cdot L^{-1} \cdot hr^{-1}$) to the conventional theory and estimate the possible maximum removal rates ($\nu_{\max}, mg \cdot L^{-1} \cdot hr^{-1}$), our experimental data were plotted according to the Lineweaver-Burk plots based on the Michaelis-Menten kinetics equation $1/\nu = (1/\nu_{\max}) + (k_m/\nu_{\max})(1/C_s)$, where $C_S, mg/L$ referred to initial pollutant concentrations [22].

F. Coulombic Efficiency

Based on voltage data recorded during the treatment phase, average coulombic efficiency ($CE, \%$) of each BMFC was calculate using equations $CE = C_p \times 100 / C_{Ti}$, where C_p is the total coulombs calculated by integrating the current over time [23]. C_{Ti} is the theoretical amount of coulombs that can be produced from either sodium acetate (i = a) or sodium butyrate (i = b), calculated as $C_{Ti} = Fb_iS_iV/M_i$, where F is Faraday constant (96,485 C / mol ·electrons), b_i is the number of moles of electrons produced per moles of substrate ($b_a = 8, b_b = 20$), S_i is the substrate concentration, and M_i is the molecular weight of substrate [23].

G. Electrical Energy

Over all energy produced by each BMFC was calculated by integrating the electrical power over time [23].

III. RESULTS

A. Relationship between Flow Rates and Pollutant Removal

Under the condition of 5-hr HRT, 8.0 to 8.5 pH, and -363.0 to -333.4 mV ORP, BMFCs removed 32.8 ± 6.8 % to 73 ± 3.7 % of COD, 4.6 ± 1.6 % to 14.7 ± 7.7 % of TN, and 12.9 ± 0.73 % to 17.6 ± 3.5 % of TP from the influent (See Fig. 2). Without an influence of decreasing HRT, higher flow rates were proved to enhance the COD, TN, and TP removal rates as shown in Fig. 3. Possible reason for this fact could be similar to that was previously described. Higher flow rates provided higher shear rate [13] to the filter media in anodic chambers. Due to well diffusing condition which corresponded with the high shear rate, biofilms in the BMFCs was highly enriched [13] and then resulted in high pollutant removal rates.

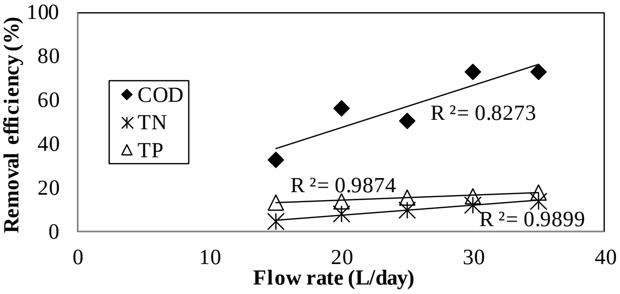

Fig. 2. Average removal efficiencies of BMFCs as functions of flow rate (15-35 L/day, 5-hr HRT).

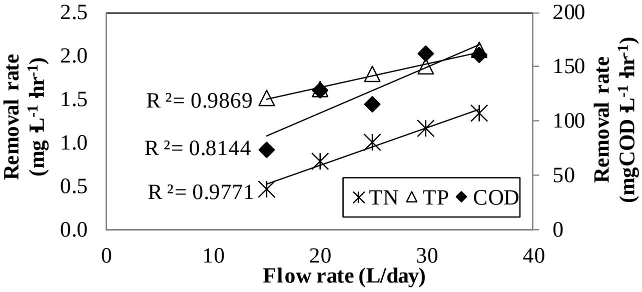

Fig. 3. Average removal rates of BMFCs as functions of flow rate (15-35 L/day, 5-hr HRT).

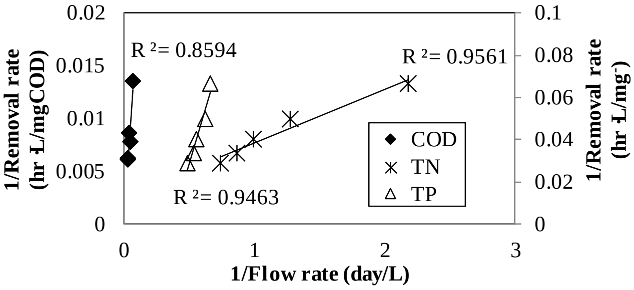

Fig. 4. Lineweaver-Burk plots of Michaelis-Menten kinetics.

High determination coefficients (R^2 = 0.85-0.96) of the Lineweaver-Burk plots of Michaelis-Menten kinetics in Fig. 4 indicated that COD, TN, and TP removal processes of our BMFCs were consistent with the conventional theory of biological enzyme reaction. As the removal rates of COD (R_{COD}), TN (R_{TN}), and TP (R_{TP}) were related to each other with high correlation coefficients (0.92 for R_{COD} and R_{TN}, 0.85 for R_{COD} and R_{TP}, and 0.98 for R_{TN} and R_{TP}), those pollutants uptake processes in BMFCs could occur either by the same microorganism or different microorganisms that involved in mutualism or commensalism relationship. Denitrifying phosphorus accumulating organisms (DPAOs) is an example of our assumption. According to the regression equation based on the above data, $R_{TN} = 0.6181R_{TP} + 1.1871$ ($R^2 = 0.9556$), it can be said that every 1 mg of TP was removed along with 0.62 mg of TN. This ratio is comparable with 0.66, the ratio of maximum denitrification rate (12.73 mgNO$_2^-$-N g MLSS^{-1} h^{-1}) to phosphorus uptake rate (18.75 mgPO $_4^{3-}$-P gMLSS^{-1} h^{-1}) of DPAOs at low nitrite level reported by Chinese researchers [24]. Accordingly, DPAOs might be one of the main microorganisms in the operation of BMFCs. However, further study is needed to prove this assumption.

B. Relationship between Flow Rates and Power Generation

According to the polarization results at the end of the start-up phase (169th-215th hr.) and during the treatment phase (312nd-358th hr.), maximum electrical power generated by each BMFC was provided by the same external resistor of 7,500 ohms. Therefore, a cathode and an anode of each BMFC were connected to a 7,500-ohm resistor throughout the treatment phase. Voltages measured during this closed-circuit condition (closed circuit voltages; CCVs) and the average power densities were shown in Fig. 5 and Fig. 6 respectively. The results demonstrated that power densities were linearly consistent with the flow rates and pointed out at BMFC5, a reactor with the highest flow rate condition, as the highest power generator (53 ± 1.16 mW/m^2).

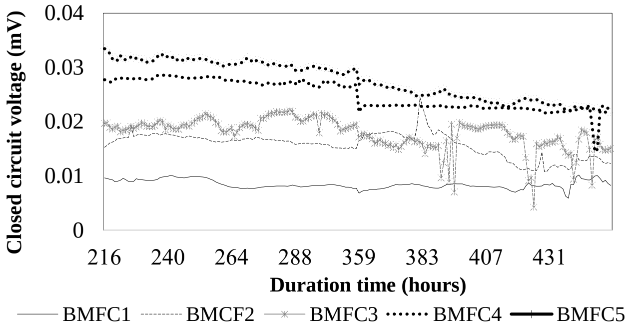

Fig. 5. Closed circuit voltages of each BMFC (7,500 ohm-external resistor).

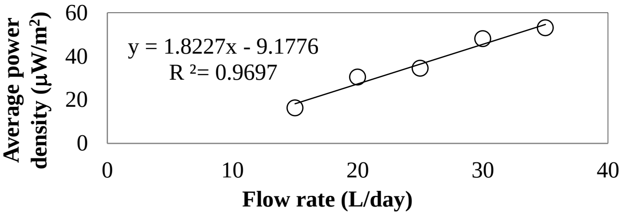

Fig. 6. Average power density of each BMFC as a function of flow rate.

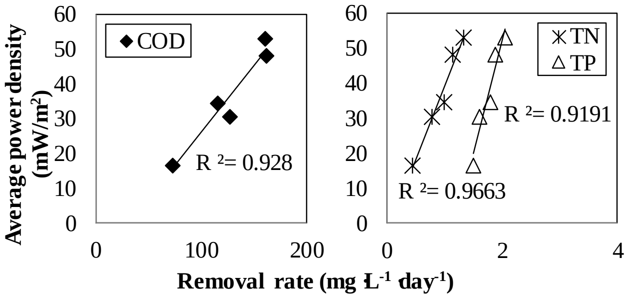

Fig. 7. Average power density of each BMFC as functions of removal rates.

The linear functions were also derived in Fig. 7. Increased COD, TN, and TP removal rates caused the increase of power densities. This result conformed to an explanation that the increased flow rates provided greater shear rate which resulted in the enrichment of biomass in an anodic chamber [13], thus biologically removal process in the BMFCs become more effective with greater number of electrons released from microorganisms. Therefore, high power outputs were consistent with high flow rates. Comparing the average power density of BMFC5 (53 ±1.16 mW/m^2) to the range of power densities reported in literature (30-3,750 mW/m^2) [25], our BMFCs appeared to be the low-ranking power generator. However, in terms of very high flow rate and large volume condition, it can be said that the BMFCs generated high power output comparing to those reported in other works, such as 0.78-0.91 mW/m^2 at 1.5 mL/h flow rate and 0.33-0.56 mW/m^2 at 125 mL/h [12], and 37.3 mW/m^2 at 9-180 mL/h flow rate [11]. Comparing to the following simple design and low-cost MFC configurations in other literatures, our BMFCs are also competitive. A single chamber microbial fuel cell containing 8 graphite anodes and a single air cathode built by the experts of the Pennsylvania State University generated maximum electrical power of 26 mW/m^2 while removing 80% of COD in local wastewater [23]. Researchers of Noshirvani University, Iran presented a three-baffle, two-chamber, membrane-less MFC which had 66% maximum COD removal efficiency and 40.43 mW/m^2 maximum power density [26]. A membrane-less and mediator-less MFC created by Malasian researchers removed 68.57% of COD with power output of 18.42 ± 5.84 mW/m^2 for mixed culture [27].

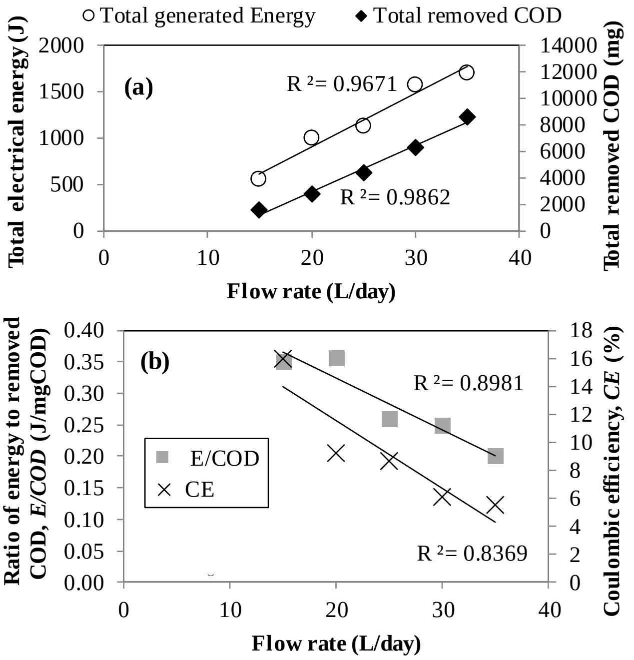

Fig. 8. (a) Total electrical energy and total removed COD as functions of flow rate (b) Ratio of total energy to total removed COD and coulombic efficiency as functions of flow rate.

Total electrical energy (J) generated by each BMFC and total removed COD (mg) throughout the treatment phase were plotted in Fig. 8 (a). Similar to the previously described trends, higher flow rates provided higher energy and larger amount of removed COD. However, in terms of energy generation efficiency, the opposite trend was revealed. Ratio of total generated energy to total removed COD varied inversely with the flow rates (see Fig. 8 (b)). Although the power output and COD removal rates were obviously enhanced by higher flow rate, the derived energy was not as high as it should be based on the theoretical estimation from removed COD. This aspect led to the inverse relationship between CEs and flow rates as shown in Fig. 8 (b). As our BMFCs are membrane-less, it was possible for high flow rate to blow away certain number of electrons from an anode chamber to a cathode compartment via the water flow (short circuit). Thus, the electrons that were transferred from the anode to the cathode via electrical circuit could decrease at high flow rate and this resulted in low CE value. Although it is difficult to prevent the short circuit in our BMFC configurations, there are several ways to improve the CE value. French researchers found that CEs increased from 8% to 25% when HRT was increased from 6 to 48 hr. in reactor A [28]. The replicates confirmed the same trend, with CEs rising from 4% to 16% when HRT increased from 14 to 41 hr. [28]. Researchers of the Pennsylvania State University reported that the reduction of oxygens diffusion from an air-cathode compartment to an anode chamber by the application of diffusion layers could improve the CEs from 13-20 % to 20-27 % [29]. Accordingly, longer HRT and the reduction of oxygen diffusion at the connection point of anode and cathode compartments should be considered in our future study.

IV. CONCLUSION

At very high flow rates of 15-35 L/day and constant HRT of 5 hrs, our large-volume BMFCs showed linear relationships between flow rates and COD, TN and TP removal rates (R^2 = 0.81-0.99). Generated power density and total electrical energy also varied linearly due to the change of flow rates (R^2 = 0.97-0.99). The highest flow rate of 35 L/day provided best performance with COD removal rate of 160.78 mgCOD L^{-1} hr^{-1}, TN removal rate of 1.34 mgTN L^{-1} hr^{-1}, TP removal rate of 2.06 mgTP L^{-1} hr^{-1}, power density of 53 ±1.16 mW/m^2, and total electrical energy of 1,706.95 J. The increasing shear rate due to the increase of flow rate was considered as the key factor for this high performance. However, in terms of CEs, the lowest value of 5.5 % was derived at 35 L/day flow rate. The short circuit caused by high flow rate was the possible explanation. In order to improve the CE, longer HRT and the reduction of oxygen diffusion to the anode chamber should be applied to our future study.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

Songyot Mongkulphit carried out all experiments and analyses. Petch Pengchai supervised the project and drafted the manuscript, which was revised by all authors. Nattawoot Suwannata, the co-supervisor of the project, helped with the discussion in the electrical aspects. All authors read and approved the final manuscript.

ACKNOWLEDGMENT

We thank Natchanan Jokloy, Deelers Chankumung, Kanokpan Jaikhan, and Sasiyaporn Pitimol for their technical assistance. We also appreciated Asst. Prof. Dr. Chontisa Sukkasem for the precious suggestion on our works.

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