

Residence time distribution in microbial fuel cell and its influence on COD removal with electricity generation

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Abstract

The liquid flow characteristics in a microbial fuel cell (MFC) were determined using a residence time distribution (RTD) test, and its effects on the performance of the MFC were investigated in terms of electricity generation and chemical oxygen demand (COD) removal. A membrane-less MFC was used with two different anode structures; normal graphite felt disk (“Normal” MFC) and perforated graphite felt disk (“Perforated” MFC). The RTD results showed that there exists nonideal flows such as channeling and tailing in the “Normal” MFC and the flow characteristics were much better in the “Perforated” MFC with the improved electricity generation. COD removal rate was similar between the MFCs. These results show that the flow characterization is an important area of study for the optimization of an MFC.
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1. Introduction

Residence time distribution (RTD) is the mixing characteristics in a reactor [1]. The RTD test has been widely used to examine the flow nonideality in a reactor. It is useful and simple tool to analyze flow property to develop a flow mathematical model, and to predict the performance of a reactor [2]. Flows characterization using the RTD test has been extensively studied for immobilized microbial reactors [3,4]. The analysis of nonideal flow in reactors is often neglected or not considered properly in waste-water treatment processes [5].

A microbial fuel cell (MFC) is a bioreactor that can generate electricity coupled to the oxidation of fuel through microbial actions [6]. The previous results showed that an MFC could be operated using electrochemically active microorganisms without the aid of mediators [6–9]. The mediator-less MFC systems have been studied for the removal of organic

contaminants in waste-water and the concomitant production of electricity [10–15]. Waste-water treatment process using MFC is promising since this process converts the major part of the chemical energy of the contaminants to electricity reducing the generation of excess sludge [6,13].

A membrane-less microbial fuel cell (ML-MFC) has been proposed as a waste-water treatment device [13]. The ML-MFC is a tubular type reactor consisting of one compartment packed with several anodes and cathodes, and is operated by a up-flow mode. The fluid elements travel upwardly along the ML-MFC. The anode part occupies the bottom half of the ML-MFC where the fuels are oxidized, and the reaction products and unused reactants flow from the anode to the cathode region to take part in the cathode reactions in this design. The flow characteristics in the anode region, which can be depicted as an immobilized cell packed bed reactor, may affect overall MFC performance. An optimal configuration for the anode of an ML-MFC is thought to be an ideal plug flow reactor. Therefore, the performance of ML-MFCs can be improved through understanding of the flow characteristics in its anode.

In the present study, the flow patterns were compared between ML-MFCs packed with normal graphite felt disk

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and perforated graphite felt disk, and the performances of the MFCs were analyzed in terms of electricity generation and chemical oxygen demand (COD) removal.

2. Materials and methods

2.1. Membrane-less microbial fuel cell

An ML-MFC described by Jang et al. [13] was modified and used for continuous COD removal and electricity production (Fig. 1). The diameter and the height of the modified ML-MFC were 6 and 7.5 cm, respectively. The empty volume of the ML-MFC was 212 ml. Two different graphite felt types, the normal graphite felt sheet (6 mm thick, GF series, GEE Graphite Limited, Dewsbury, UK) and the perforated graphite felt sheet, which were fabricated from the normal graphite felt sheet by perforating eight holes with the diameter of 1 cm into the normal one, were used as anode packing materials. They are referred to as the “Normal” and the “Perforated” MFC. In each MFC, eight sheets of anode packing material were placed being touched each other at the lower part of the ML-MFC as the anode. Two sheets of platinum-coated graphite felt sheet were positioned on the upper part of the ML-MFC as the cathode. Platinum powder was spray-coated onto normal graphite felt at the density of 0.1 mg platinum/cm² electrode area according to the method described previously [16]. A perforated polyacrylic plate was fixed between the anode and cathode. The plate was put in place to achieve a direct contact with both of anode and cathode, minimizing the distance between the anode and the cathode as well as ensuring electrical insulation. Between the two platinum-coated graphite felt sheets, used as the cathode, an aerator made of sintered glass was inserted to provide air required for cathode reaction. The void volume of the “Normal” and the “Perforated” MFC were 100 and 115 ml, respectively. The void volume of the anode was 55 ml for the “Normal” MFC and 70 ml for the “Perforated” MFC.

2.2. RTD test

RTD tests were performed to analyze the flow characteristics in the anode region of the MFCs with naked electrode.

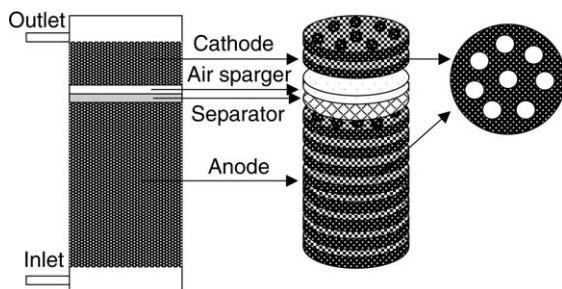


Fig. 1. Schematic diagram of ML-MFC configuration.

The 100 mM NaCl solution as tracer was fed into the anode in a step input using a peristaltic pump (505S, Watson-Marlow, Falmouth, Cornwall, UK) to obtain a *F*-curve that is a time-course of the response to step input of the tracer. The response was recorded as C/C_0 (effluent concentration/influent concentration). The conductivity was monitored to measure the tracer concentration in the effluent from the anode region using a conductivity meter (CON 5 Meter, LaMotte, Chestertown, MD, USA). The relationship between the concentration and the conductivity was linear at the concentration up to 100 mM. The feeding rate of the tracer was varied from 0.15 to 0.68 ml/min, similar to the feeding rates to operate the ML-MFC. The effluent was collected in an aliquot of 3 ml when it left the anode region. The *F*-curve obtained was fitted to a non-linear equation (Logistic 3 parameter) provided by Sigma plot[®] 7.1 (SPSS Inc., Chicago, IL, USA). *C*-curve was obtained by differentiating the non-linear equation, which was chosen to give a correlation coefficient over 0.99.

2.3. Waste-water

Artificial waste-water (AW) containing glucose and glutamate was used throughout the study [17]. AW was autoclaved at 121 °C for 15 min before being added with filter sterilized glucose and glutamate solutions to the final COD values of 100–400 mg/l. AW was made and maintained under nitrogen atmosphere in a carboy, which was connected to a nitrogen-containing gas-tight bag (SKC Inc, Valley View Road, PA, USA) to avoid air diffusion due to negative pressure.

2.4. ML-MFC operation and analyses

After the RTD tests, the ML-MFCs were enriched and operated under the various operating conditions as described earlier [13]. The ML-MFCs were installed in a temperature-controlled chamber maintained at 35 °C. For the enrichment as a start-up process, the anode compartment of the ML-MFC was loaded with freshly collected sludge from a sewage works using a syringe and kept for 24 h. The AW of 100 mg COD/l was fed into the anode continuously using a peristaltic pump (505S, Watson-Marlow) equipped with Marprene II tubing (Watson-Marlow) at the rate of 0.15 ml/min for enrichment, and air was introduced into the cathode compartment as an oxidant at the rate of 100 ml/min.

After the enrichment, MFCs were operated continuously in a similar way with varying the feeding rates as specified in the text. The voltage between the anode and the cathode was measured using a multimeter (Model 2700, Keithley Instruments Inc., Cleveland, OH, USA) linked to a differential multiplexer (Model 7701, Keithley Instruments Inc.). Data were recorded digitally on a personal computer via IEEE-488 interface card (Model PCI-488 Keithley Instruments Inc.) every 5 min. The multimeter was controlled using TestPoint software (Capital Equipment Co. Cleveland, OH, USA). The potential difference measured was

converted to current according to the relationship of potential = current \times resistance. All experiments were performed using three ML-MFCs in each experimental set. The mean values or typical results were presented. The potential was recorded at different resistances from 3 Ω to 11 K Ω to analyze the MFC performance through the polarization curve method. The chemical oxygen demands of the effluent were determined using a COD assay kit (Ultra-Range COD reagent, Hack, CO, USA). All assays of COD were performed in triplicate and mean values were presented.

3. Results and discussion

3.1. RTD curves

ML-MFCs with naked electrodes were fed with 100 mM NaCl in a step-input mode and the conductivity of the effluents from the anode part was monitored (Fig. 2A). The time and the corresponding conductivity were normalized with the hydraulic retention time (τ) and the conductivity of the NaCl solution used. In the case of the “Normal” MFC, the tracer concentration increased before the effluent volume reached the void volume ($t/\tau < 1$). Around two t/τ , a long tailing was also observed. These show that short-circuiting and/or channeling phenomena exist in the anode region of the “Normal” MFC.

When the “Perforated” MFCs were used in a similar experiment, the rapid increase in tracer concentration was observed around $t/\tau = 1$, and the C/C_0 value reached to 1 earlier than that of the “Normal” MFC. These results show that the flow in the “Perforated” MFC is closer to a plug flow than that in the “Normal” MFC. It is thought that the holes on the graphite felt disk reduce the flow resistance of the graphite felt.

A non-linear equation (Logistic 3 parameter, Sigma plot[®] 7.1) was found to fit the F -curves well, which was differentiated to obtain the C -curves, the normalized response to the pulse input of tracer, according to Levenspiel (1972). The R^2 values were over 0.99 in both fittings. The C -curves obtained were used for the further analysis (Fig. 2B). These C -curve data could be fitted to the axial dispersion model developed to describe the large extents of dispersion [2] given by the following equation;

$$C/C_0 = \frac{1}{2\sqrt{\pi\theta(D/uL)}} \exp\left[-\frac{(1-\theta)}{4\theta(D/uL)}\right] \quad (1)$$

where θ is the normalized time t/τ (dimensionless); t , the time (s); τ , the theoretical retention time (s); D , the coefficient of axial dispersion (m^2/s); u , the flow velocity (m/s); L is the characteristic length (m).

The expression of uL/D is identified as the Peclet number (Pe) in Eq. (1). It is a dimensionless number, which represents the ratio of the rates of transport by convection to those by diffusion or dispersion. The inverse is called as dispersion number. The estimated Pe values by the above flow model were 38.5 for the “Perforated” and 5.7 for the “Nor-

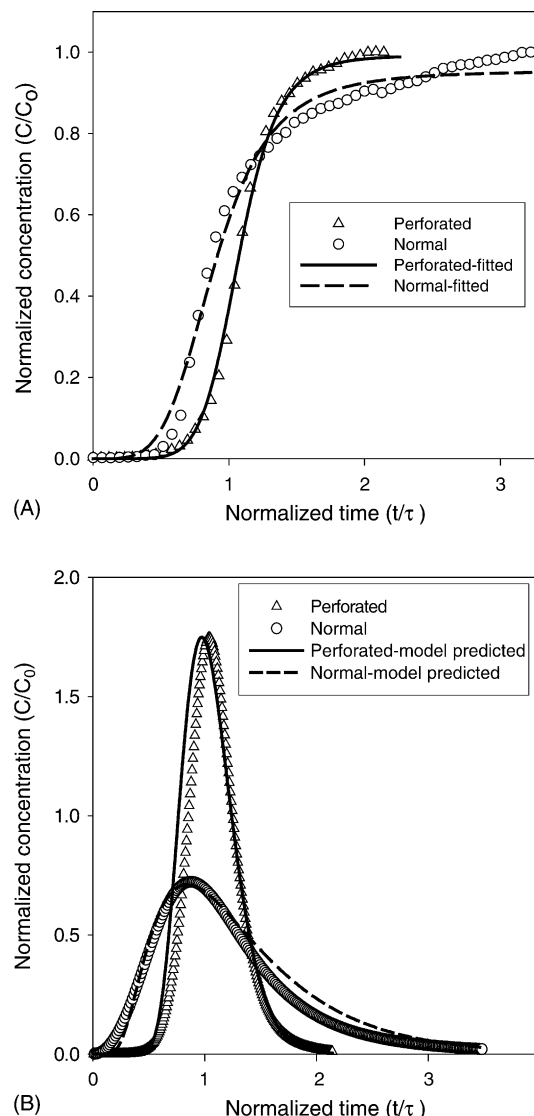
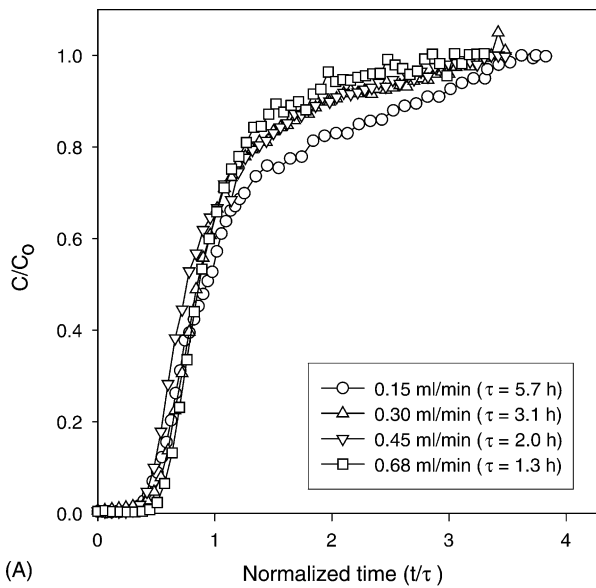


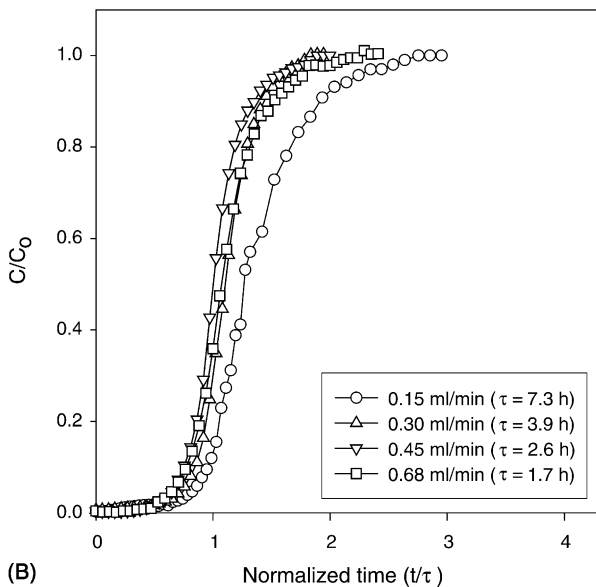
Fig. 2. RTD curves in anode of up-flow tubular mediator-less microbial fuel cells with different anode structure. (A) F -curve (B) C -curve. The tracer was fed at the flow rate of 0.45 ml/min giving the corresponding hydraulic retention time of 2.0 for the “Normal” MFC and 2.6 h for the “Perforated” MFC, respectively.

mal” MFC. These indicate that the mass transfer in the MFC is achieved dominantly by convection, and its tendency is more outstanding in the “Perforated” MFC.

The effect of the flow rate on the F -curve was examined (Fig. 3). In both the “Perforated” and the “Normal” MFC, flow rate did not change the F -curve patterns significantly at the flow rate from 0.30 to 0.68 ml/min, but the response was slightly retarded at a low flow rate of 0.15 ml/min compared to those of the higher flow rates. This result shows that the flow rate in the tested range does not significantly affect the extent of dispersion, which is determined by flow velocity gradient, turbulent diffusion, and molecular diffusion, and that the different RTD pattern shown in Fig. 2 was not due to the different hydraulic retention time.



(A)



(B)

Fig. 3. Effects of flow rate on F curve pattern in MFCs with different anode structure. (A) "Normal" MFC, (B) "Perforated" MFC.

3.2. Start-up process

The MFCs were inoculated with fresh sludge, and fed continuously with the AW of 100 mg COD/l at the rate of 0.15 ml/min to enrich electrochemically active microbial consortium, monitoring the current (Fig. 4). The "Perforated" MFCs showed a higher initial current, rapid increase in current and higher steady state current than the "Normal" MFCs. The better performance of the "Perforated" MFCs might be due to the even flow property. The electrochemically active microbes in the inoculum cover more electrode surface in the "Perforated" MFCs than the "Normal" MFCs to give a higher initial current. For the same reason the higher steady state current was obtained in a shorter period of time. These

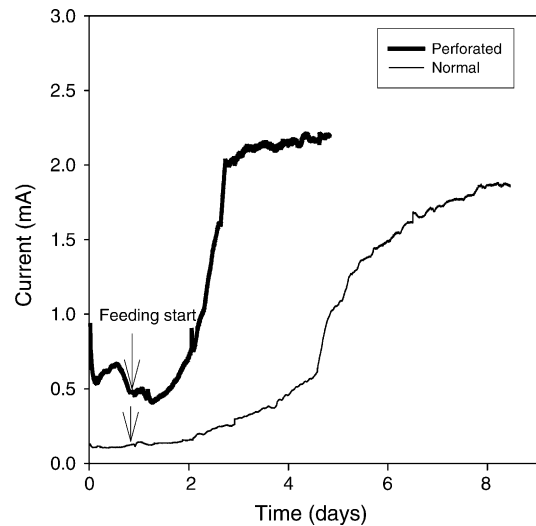


Fig. 4. Enrichment of electrochemically active microbes using MFCs with different anode structure. The AW of 100 mg COD/l was fed at the flow rate of 0.15 ml/min.

show that the performance of an MFC depends heavily on the liquid flow characteristics through the anode.

3.3. Electrochemical property

The MFCs were operated using various resistances to compare their electrochemical properties such as overvoltages through the polarization curves (current versus voltage) method (Fig. 5). The curves were similar to that of MFCs with membrane using platinum coated graphite felt as cathode, where activation and ohmic overvoltages were observed [15]. Both "Normal" and "Perforated" MFCs showed similar curve patterns in the low and medium current region. The

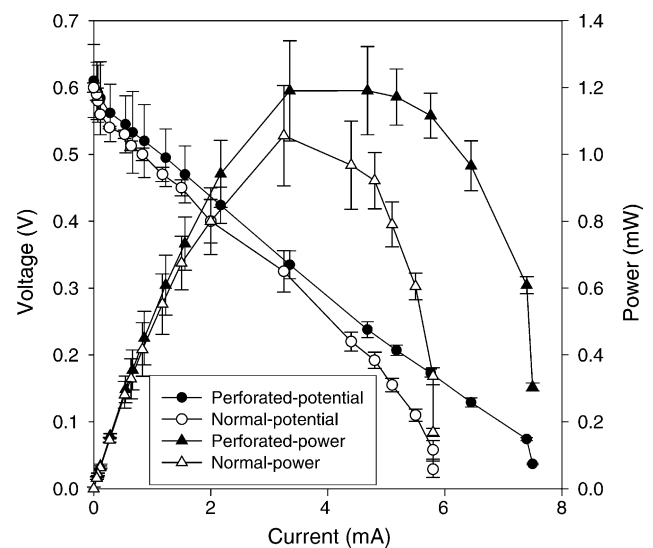


Fig. 5. Polarization curves of MFCs with different anode structure. The AW of 300 mg COD/l was fed at the flow rate of 0.30 ml/min.

voltage drops were linear to current increase up to 5 mA in “Normal” and 7 mA in “Perforated” MFC. In the “Normal” MFCs, the voltage dropped more sharply with the increase in the current over about 5 mA, while the voltage in the “Perforated” dropped rapidly over 7 mA. These voltage drops are due to the overvoltage caused by the mass transfer loss of fuel [18]. These results show that the flow condition in the anode region affects the electrochemical performance of MFCs.

The maximum power generated in both ML-MFCs was similar within the standard deviations because the maximum power was obtained at the medium current where the voltage drop was similar.

3.4. Dynamic property

The current was monitored with the change in the AW concentration from 200 to 300 mg COD/l at a feeding rate of 0.15 ml/min to test the dynamic property (Fig. 6). The “Perforated” MFC showed a better dynamic response. It took about 3 h in “Perforated” and 17 h in “Normal” MFC, respectively to reach a new steady state. The difference in the response time is probably due to the different flow properties shown above. It is known that the organic compounds not used in the anode region are transferred to the cathode region in an ML-MFC, where they are oxidized through the aerobic bacterial respiration consuming oxygen [13]. Because the aerobic bacteria have higher affinity for oxygen than the cathode [16], organic compounds leaving the anode region can inhibit the cathode reaction. For this reason uneven flow should be avoided. The oxygen limitation to the cathode reaction can cause instability of the MFC and lead to a process failure.

When MFCs were as a continuous BOD sensor [17,19–23] a short response time is a prerequisite [23]. The response time could be reduced through optimization of the feeding rate and cell design. The improvement of the flow property is another area of interest for the purpose of reducing the response time.

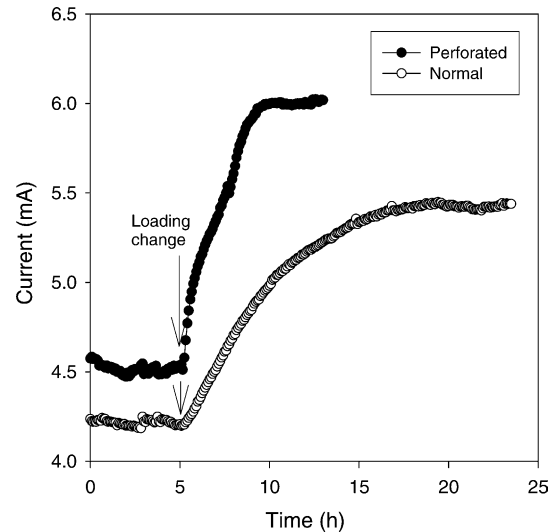


Fig. 6. Dynamic responses of MFCs with different anode structure to the change in AW load. The AW was fed at the flow rate of 0.15 ml/min. The COD concentration of the AW was changed from 200 to 300 mg/l at the time indicated by arrows.

3.5. COD removal and electricity generation

The current was monitored during changes in fuel concentration and feeding rate to examine the effect of the COD loading rate on current generation and COD removal (Table 1). COD removal efficiency and coulombic yield were calculated based on the overall ML-MFC including anode and cathode compartment. In all tested loading rates, the “Perforated” MFCs showed better performance in terms of current generation and coulomb yield. The difference in current generation between the two MFCs was more significant at high loading rates. The maximum current generated from the “Perforated” MFCs was 7.8 mA, which was 34.4% higher than that from the “Normal” MFCs. With the increase in the fuel concentration from 100 to 400 mg COD/l at the feed-

Table 1
Effect of waste-water loading rate on performance of ML-MFCs

AW concentration (mg COD/l)	Feeding rate (ml/min)	Graphite disk type	Current (mA)	Effluent COD (mg/l)	Coulomb yield (%)
100	0.15	Normal	1.9 ± 0.3	9.7 ± 0.1	70.3
		Perforated	2.2 ± 0.2	9.6 ± 0.1	85.4
200	0.15	Normal	4.1 ± 0.1	10.1 ± 0.3	71.9
		Perforated	4.5 ± 0.1	10.3 ± 0.2	78.9
	0.30	Normal	6.5 ± 0.4	10.4 ± 3.9	57.0
		Perforated	7.0 ± 0.3	10.6 ± 2.1	61.4
	0.45	Normal	5.9 ± 0.6	10.7 ± 0.1	34.7
		Perforated	7.6 ± 0.5	12.6 ± 0.1	44.7
300	0.15	Normal	5.5 ± 0.5	17.6 ± 1.6	65.1
		Perforated	5.8 ± 0.8	14.7 ± 0.5	68.2
	0.30	Normal	5.8 ± 1.4	11.9 ± 1.7	33.3
		Perforated	7.8 ± 0.9	13.5 ± 0.3	43.5
400	0.15	Normal	5.2 ± 1.9	12.4 ± 0.2	44.7
		Perforated	6.8 ± 0.5	12.5 ± 0.2	50.0

Table 2
Comparison of MFC performances

Mode of operation	Membrane usage	Cathode system	MFC volume ^a (ml)	Volumetric COD removal rate ^b (kg/m ³ d)	COD removal efficiency (%)	Maximum volumetric power ^b (mW/l)	Coulomb yield (%)	Reference
Continuous	None	Aeration	212	0.10–0.59	>90	7.6	44–85	This work
Continuous	None	Aeration	6280	0.02–0.15	>90	0.01	<10	[13]
Continuous	Yes	Air-saturated water	52	1.2–4.1	>90	51	63–92	[15]
Continuous	Yes	Air-breathing	388	0.13–0.70	40–80	1.7	3–12	[14]
Batch	None	Air-breathing	28	0.144	75	12.5	9–12	[24]
Batch	Yes	Air-saturated water/ferric cyanide	90	0.22–0.97	65–89	97	65–89	[12]
Batch	Yes	Air-saturated water/ferric cyanide	420	0.34	>90	0.13	81	[11]

^a The volume including both of anode and cathode compartment.

^b These values were computed based on the empty MFC volume including both of anode and cathode compartment.

ing rate of 0.15 ml/min, current from the “Perforated” MFCs increased from 2.2 to 6.8 mA but the “Normal” MFCs generated a lower current with the 400 mg/l fuel than with the 300 mg/l.

The coulomb yield varied from 43.5 to 85.4% whilst the COD removal efficiency was over 90% in all cases. The “Perforated” showed higher coulomb yield than the “Normal” MFC, and the yield was higher at lower loading rate. These results show that at high loading rates a considerable amount of fuel passed through the anode to the cathode where they were oxidized by aerobic bacteria reducing the coulomb yield. The lower coulomb yield in the “Normal” MFCs shows that more fuel was consumed at the cathode probably due to the uneven flow through the anode region.

The improvement in MFC performance through the use of perforated anode is probably due to the even flow in the anode region without compromising the space required to form biofilm and microbial clumps as a bacterial community in the anode region. An extra advantage of using a perforated anode is to void clogging the MFC when applied to wastewater containing suspended solid.

The performance of the “Perforated” MFC was compared with those of other MFCs reported (Table 2). The maximum COD removal rate of 0.59 kg/m³ d was obtained, which was about 4 times higher than that of previous ML-MFC [13]. The maximum volumetric power of 7.6 mW/l in the present work was over 700 times higher than that of previous one operated in continuous mode [13]. The coulomb yield was considerably improved also.

The coulomb yield of the ML-MFCs used in the present work was comparable with other continuous MFCs using a membrane but the COD removal rate and power generation were one or two order lower than those with membranes. Although the performance of ML-MFCs is not as good as those with membranes, the former have some advantages over the latter. The ion-exchange membrane is expensive and it can foul during use.

The performance of an MFC depends on various factors [10] including the size of the cell [23] and the distance between the anode and cathode in the case of an ML-MFC

[13]. The ML-MFC could be developed as a viable wastewater treatment process through its optimization.

4. Conclusion

Studies were made on the flow property through the anode of MFC using the RTD technique and on its effects on the performance of MFCs for the first time. It was shown that the hydrodynamic characteristics affected the MFC performance. The RTD test showed that the “Perforated” MFC has better flow characteristics than the “Normal” MFC. With the improved flow characteristics “Perforated” MFC showed higher current and coulomb yield than the “Normal” MFC at the COD loading rates ranging from 0.10 to 0.59 kg/m³ d but COD removal efficiency was similar. It is concluded that the flow characteristics are an important factor to be considered in MFC design.

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