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Milliliter-scale Microbial Fuel Cell (MFC) Fabricated by Polyether sulfone (PES) Hollow Fiber Membrane

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ABSTRACT

Microbial fuel cell (MFC) is a technology of increasing interest recently. However, the lack of efficient and cost-effective membranes has hampered the commercialization of MFCs. polyether sulfone (PES) hollow fiber was utilized as the separator in the cylindrical ml-scale active–air MFC to treat domestic wastewater. The conventional aeration unit replaced with the silicon membrane tube that was only passed the oxygen of air across wall membranes and provides appropriate dissolved oxygen in the cathode chamber. The increase in hydraulic retention time (HRT) (from 1.43 to 3.97 min) drastically improved the power and current densities of the MFC from 306.41±17.43 to 540.98 ±32.00 mW/m^2, and from 1842.138±103.18 to 2191.83±160.06 mA/m^2, respectively. The electricity generation performance gradually continued to grow by further extending the HRT from 3.97 to 10.19 min. The maximum power and current densities of 582.97±28.61 mW/m^2 and 2404.15±140.61 mA/m^2 are obtained at the HRT of 10.19 min. Similarly, the coulombic efficiency as well as the current and voltage generation of the MFC is enhanced by the increment of HRT. The maximum open circuit voltage and current of 768±14.10 mV, and 0.418±0.004 mA (with the external resistance of 986 Ω) are yielded at the HRT of 10.19 min. The rather low response time which was obtained for this hollow fiber-MFC (450 min) shows its potential applicability as a biosensor.

1. Introduction

MFC devices have shown much promise recently because of simultaneous wastewater treatment and electricity generation and lower sludge production compared to the conventional treatment processes, which reduced the treatment costs and sludge disposal problems [1-7]. However, the high capital cost [8] and relatively low-quality effluent of MFCs [9] limited their practical application for wastewater treatment. In this respect, several attempts have been performed to replace the expensive materials of MFCs, including high-cost electrodes [10, 11] and

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separators like famous Cation Exchange Membranes (CEMs), with more efficient and cost effective separators like microfiltration (MF) and ultrafiltration (UF) membranes, nanoporous filters, different coarse pore fibers [12-18], and ceramic membranes [19-27]. However, some researchers showed that using coarse pore UF and MF membranes decreased the power density of MFC owing to high oxygen and substrate leakage between the electrodes [15].

Nevertheless, Hou et al. (2011) compared the performance of different UF, and MF membranes (with diverse cut-off molecular weights), and the Nafion membrane as the separator of MFCs. They observed that diffusion rates of oxygen and substrate between the chambers were directly proportional to the pore size of membranes. Finally, the UF membrane with a smaller pore size (1KDa cut-off molecular weight) shows higher coulombic efficiency and power density than the Nafion membrane [28]. Kim et al. (2013) also reported that using a UF membrane with an appropriate pore size as a separator in MFCs not only provides efficient electricity generation but also improve the quality of effluent considerably [29].

In this respect, Tian et al. (2015) integrated one single chamber MFC with a hollow fiber membrane bioreactor (including PVDF hollow fiber) and achieved good effluent quality and acceptable electricity production [30]. Katuri et al. (2014) also introduced one novel anaerobic electrochemical membrane bioreactor (AnEMBR) by combining the microbial electrolysis cell (MEC) with the membrane filtration using nickel-based hollow fiber membranes (Ni-HFMs). The Ni-HFM is used as the cathode for hydrogen evolution reaction (HER) and also as the membrane for the filtration of the effluent simultaneously. Finally, more than 95 percent COD (chemical oxygen demand) removal was achieved in this system [31].

In this study, we constructed a ml-scale tubular MFC using polyethersulfone (PES) hollow fiber as a separator for the first time. However, using UF hollow fiber as the separator between the chambers of active air-breathing MFC could result in low power density because of the short distance between the electrodes (low thickness of hollow fiber walls) and the excessive presence of oxygen at the anode chamber as reported for systems that combined the flat-plate design and an active air-breathing cathode [32, 33]. Therefore, we introduced one novel aeration system using a silicon membrane tube that provided the appropriate amount of oxygen in the cathode chamber. Also, this system could eliminate the probable parasitic loads of running auxiliary pumps, fans, or compressors in other aeration units and reduce the overall capital and operating costs of the MFC system significantly. In addition, the short response time of this novel PES hollow fiber-MFC introduced it as a viable option for biosensor application.

2. Materials and methods

2.1. MFC construction and operation

The anode chamber of cylindrical MFC was constructed by drilling down a hole between the two attached Plexiglas pieces with a 6 mm diameter and 10 cm length. Then, one single polyethersulfone hollow fiber (0.5 mm internal diameter and 1 mm outer diameter, %50 porosity) was inserted inside this cylinder which concurrently played the role of cathode chamber and the separator of MFC. The anode electrode was the carbon fibers (500 nm diameter, %95 carbon content, Pantex®35, Zoltek Co. Ltd., USA) that was wrapped around the hollow fiber and was tightened by copper wire as the current collector, which transmitted the produced electrons to the external circuit. The cathode electrode was the single copper wire that was placed inside the hollow fiber and also connected to the external circuit. Fig.1 presents a schematic diagram of cylindrical hollow fiber-MFC and other parts of auxiliary equipment, briefly.

Wastewater from the sewage treatment plant of Sistan and Baluchestan University (Zahedan, Iran) was continuously fed as the anolyte solution using the gravity force and adjusted to the desired flow rates by using the input valves, without considering any inoculation period or addition of any supplementary nutrition or salts.

Tap water that passed through the silicon membrane tube (internal diameter of 0.063 in, the outer diameter of 0.125 in, Cole-Parmer Inst. CO., USA) was fed as the Cathode influent, counter-current relative to the anode influent. The silicon membrane tube only passed the oxygen of air through its wall. Therefore, it oxygenated the water perfectly without any aeration unit and significantly offsetting the costs of wastewater treatment.

The carbon fibers have been replaced after each experiment to establish the same conditions for all the experiments. The experiments were conducted at ambient temperature (30-35ºC) and performed at least three times to ensure reproducibility and minimize probable human errors and environmental condition changes.

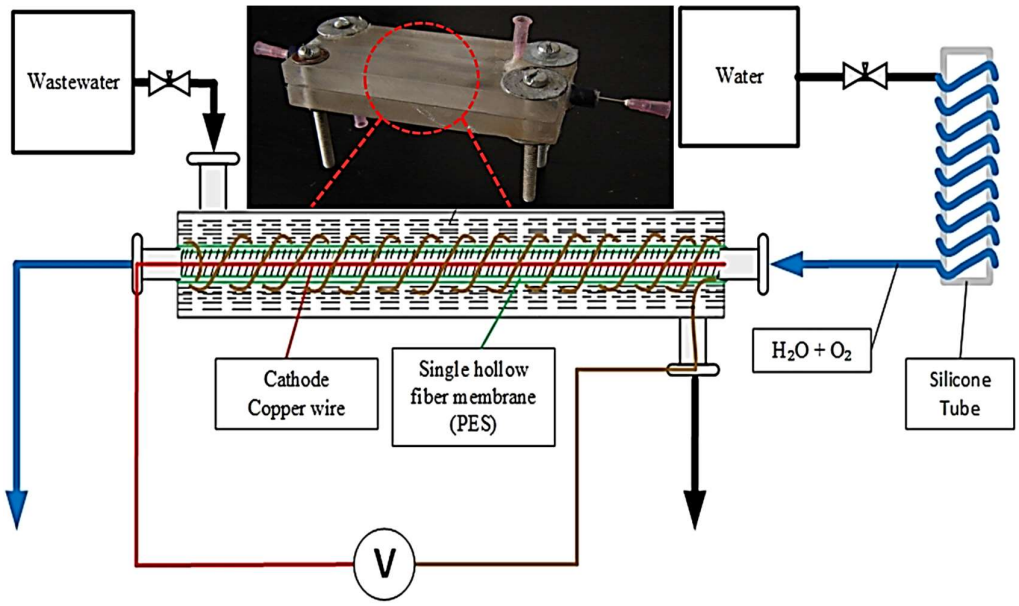

Fig. 1. The schematic representation of mililiter-scale MFC design

2.2. Analysis and calculations

The voltage (V) was monitored using a digital multimeter instrument (VC 9805, Zhangzhou Weihua Electronic Co. Ltd., Fujian, China) while the MFCs were connected to an external resistance (R_{ex}) of 986 Ω. The external resistance was varied from 17 to 46600 Ω to obtain the polarization curve. The resulting voltage at each resistor was recorded when it stabilized. The power (P=VI) and current ($I=V/R_{ex}$) values were normalized by the projected surface area of the cathode.

The coulomb efficiency (CE) was calculated by integrating the current over a certain period relative to the theoretical amount of electrons that could be produced from organic matter oxidation, based on four electrons per mole of COD [2], as described in equation 1:

$$CE=\frac{100M\int_0^t Idt}{FbV_{anode}\Delta COD} \quad (1)$$

where F is the Faraday's constant, b=4 is the number of electrons exchanged per mole of oxygen, ΔCOD is the difference between the inflow and outflow COD values, V_{anode} is the efficient volume of anode chamber, and M is the molecular weight of oxygen.

3. Results and discussions

The hollow fiber-MFC was operated in continuous mode. The influent COD was fixed at 1200 mg/l, and different organic loading rates were obtained by changing the flow rates of wastewater into the anode chamber. Table 1 presents the effect of different flow rates on the performance of MFC briefly. The open circuit voltage (OCV) and the maximum obtained current improved with increasing the hydraulic retention time (HRT) of anolyte in the anode chamber (Fig. 2(a), (b)).

On the other hand, it can be seen from these figures that the time required to reach the stabilized voltage and current (response time) for this hollow fiber-MFC configuration was around 450 min for 1200 mg/l influent COD, which was comparable to the response time (120 min to 310 min) of clayware separator MFC that used as COD biosensor in the range of 67 mg/l to 212 mg/l COD concentration. It also reported that higher COD concentrations resulted in a higher response time [34]. Therefore, this relatively low response time of the hollow fiber-MFC introduced this MFC configuration as a viable option for biosensor applicability. However, more experiments must be carried out to investigate the linearity and repeatability of the produced current of this MFC at different COD concentrations. Also, the maximum power and current densities markedly elevated as HRT increased from 1.43 min to 3.97 min, but they did not change remarkably with a further increase of HRT from 3.97 min to 10.19 min (Fig. 2(c)).

Similarly, Li et al. (2014) observed that increasing the HRT from 4 h to 6h improved the maximum power density of MFC about 1.5 times, but subsequently increasing the HRT from 6h to 8h had no evident effect on the power density [35]. Wang and Tao (2008) also reported that only moderately increasing the HRT favored electricity generation [36]. Wang et al. (2013) evaluated the performance of one novel electrochemical membrane bioreactor at various HRTs (1.6h - 14.5 h). The corresponding current and power densities improved noteworthy as HRT increased from 1.6 h to 3.6 h, but they did not have notable change afterward. Meanwhile, the coulombic efficiency enhanced from 1.8 % to 36% by increasing the HRT value [37].

Wei et al. (2013) found that increasing the HRT from 4h to 8h elevated the output voltage, but it decreased as HRT exceeded from 10h to 24h [38]. Sharma and Li (2010) also approved that at short HRTs (6.5 h < HRT < 13.1 h), the power density steadily improved with increasing the HRT, but it depleted afterward by a further increase of the HRT (13.1 h < HRT < 50 h) [39].

Therefore, at long HRTs, an insufficient substrate for electroactive bacteria (EAB) introduced the substrate as a rate-limiting factor of current generation based on the Monod equation. But at short HRTs, the oxygen reduction reaction is the limiting step. However, very high flow rates of anolyte may cause loss of the anodic biofilm layer and depletion of power and current densities as a consequence. Thus, adopting an appropriate range of HRT is necessary to have an effective electricity recovery from the substrate.

The internal resistance for 1.43 min HRT (403.82 Ω) was much lower than the other HRTs, probably due to the loss of biofilm at a high flow rate, followed by the internal resistance of 516.24 Ω for 10.19 min HRT and 560.51 Ω for 3.97 min HRT (Table 1). The lower internal resistance in 10.19 min HRT compared to that in 3.97 min HRT is probably because of lesser growth of bacteria due to low substrate condition in high HRT.

On the other hand, the higher slope of the first and the last region of the polarization curves compared to the middle part of these curves (Fig.2 d) reveals that the large portion of voltage losses in this hollow fiber-MFC design is ascribed to the activation and concentration over potentials. The relatively few ohmic losses in this MFC must be attributed to the low distance between the anode and cathode electrodes (0.5 mm) and acceptable proton conductivity of hollow fiber membranes.

Consistent with the results of Wang et al. (2013) [37], the CE improved as HRT increased. Li et al. (2014) also reported that extending the HRT resulted in a higher pollutant removal rate and boosted CE [35]. However, the maximum obtained CE was around 5.84 percent which was not a considerable quantity. Similar results were reported for MFCs that operated with real wastewater as substrate [40].

Table 1. The effect of different flow rates on the performance of MFC

Flow rate (ml/min)	HRT (min)	OCV (mV)	Peak current (mA)	Power density (mW/m^2)	Current density (mA/m^2)	CE (%)	R_{in} (Ω)
3.5	10.19	768±14.10	0.418±0.004	582.97±28.61	2404.15±140.61	5.84±0.14	516.24
9	3.97	752.1±9.45	0.397±0.011	540.98±32.00	2191.83±160.06	5.56±0.17	560.51
25	1.43	574.6±16.35	0.311±0.009	306.41±17.43	1842.138±103.18	4.07±0.13	403.82

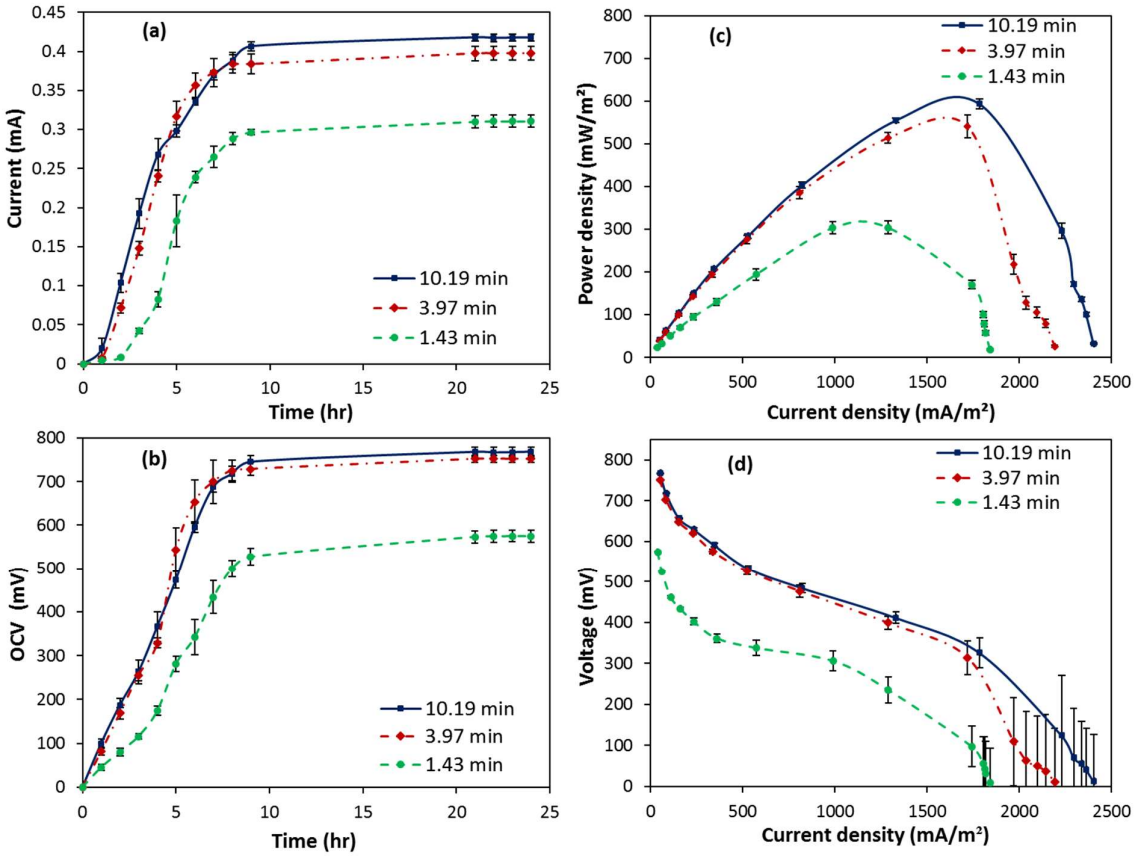

Fig. 1. The electricity generation performance of MFC; (a) the current generation versus time, (b) the open circuit voltage (OCV) versus time, (c) the power and current production during the polarization test, (d) the closed-circuit voltage during the polarization test

4. Cost analysis

One of the most crucial bottlenecks in commercializing the MFCs is the very high capital cost of this technology. Currently, a substantial portion of the MFC's capital cost (up to 90%) is attributed to high-cost separators like CEMs and Pt-based cathodes [41]. In this study, PES hollow fiber has been implemented as the separator instead of the expensive well-known Nafion membrane for the first time. The PES hollow fiber costs around 160 \$/ m^2, which is much lower than 1400 \$/m^2 for the Nafion membrane [14, 28]. Furthermore, simple carbon fiber layers have performed as the anode electrode without using a metallic catalyst like Pt. On the other hand, the oxygenation method used in this study for cathode influent water using a silicon membrane tube could significantly reduce the overall capital and operational cost of MFC systems.

5. Conclusion

The novel-designed milliliter-scale tubular PES hollow fiber-MFC has been constructed, and considerable power and current densities have been achieved. The power and current densities, voltage, current, and coulombic efficiency values improved when HRT increased from 1.43 to 3.97 min, but they did not alter considerably afterward. On the other hand, a relatively low response time was obtained for this MFC, which was in the order of magnitude of that reported for previous MFC biosensors.

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Nomenclature

MFC	Microbial fuel cell
PES	Polyethersulfone
HRT	Hydraulic retention time
CE	Coulombic efficiency
OCV	Open circuit voltage
R_{in}	Internal resistance
COD	Chemical oxygen demand

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