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COMPARATIVE EVALUATION OF UP-FLOW MICROBIAL FUEL CELLS: PROTON EXCHANGE MEMBRANE AND EARTHEN CYLINDER DESIGN

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Abstract

The investigation was centered on the continuous production of bioelectricity using an innovative up-flow bio-cathode microbial fuel cell (MFC). MFC-1 was equipped with a commercially available proton exchange membrane (PEM) and was tested at different organic loading rates (OLRs). The results showed that MFC-1 had a maximum volumetric power density of 10.04 W m^{-3} at an OLR of 0.923 kg COD m^{-3} d^{-1}, and consistently achieved COD removal efficiency above 90% at all OLRs. MFC-2, on the other hand, had an inner anode chamber made of an earthen cylinder instead of a polymer membrane and achieved better performance. MFC-2 had a maximum volumetric power density of 14.59 W m^{-3} at an OLR of 0.923 kg COD m^{-3} d^{-1}, which was a 46% increase over MFC-1. Additionally, MFC-2 had a lower internal resistance (69 Ω) compared to MFC-1 (96 Ω), indicating its superior COD removal and power generation capabilities relative to MFC-1.

Keyboard: Earthen cylinder, Microbial fuel cell, Organic loading rate, and Power density

1. Introduction

The world's need to move away from fossil fuels due to environmental concerns, such as pollution and climate change, has driven a concerted effort towards renewable energy solutions. Developing new, effective clean-energy solutions is essential for maintaining sustainability, competitiveness in the economy, and energy security—all of which promote long-term, sustainable growth in the economy. One such promising innovation is the MFC, which not only treats waste-water but also converts the biochemical power stored in biological matter into electrical energy making it a long-term energy generation method. However, the widespread adoption of MFCs faces hurdles, including their currently insufficient power densities for most envisioned applications and the significant costs associated with commercial deployment. Presently, practical applications are mostly limited to sediment MFCs, who energize electronic monitoring equipment with organic substances found in sea sediments [1].

To overcome these challenges, MFC performance must be improved. Various factors affect power generation in MFCs, such as substrate characteristics, biomass concentration, electron transfer rates, circuit resistance, proton mass transfer, cathode performance, ionic strength, pH, temperature, and reactor design. A diverse range of MFC configurations has been explored, including single-chamber air cathode MFCs, single electrode/membrane assemblies in flat plate MFCs, tubular air cathode MFC systems, up-flow fixed-bed biofilm reactors [2,3,4,5], and stacked MFCs arranged as flat plates or linked in series. Efforts to optimize these factors are crucial for realizing the full potential of MFCs as a viable renewable energy source [6,7].

Biocathodes are highly effective components in MFCs that offer many benefits over abiotic cathodes. Firstly, biocathodes reduce the MFCs cost by using microorganisms as catalysts for

electron transfer, getting rid of the requirement for synthetic electron mediators or costly metal catalysts. This reduces both the construction and operational expenses of MFCs. Secondly, biocathodes aid in denitrification processes [8], which helps in removing nitrogen compounds from the system. Thirdly, biocathodes can generate oxygen internally through photosynthetic reactions facilitated by microorganisms like algae, eliminating the need for an external oxygen supply. Finally, the metabolic activities of microorganisms within biocathodes offer the potential for the production of valuable by-products or the removal of undesirable compounds, further enhancing the functional versatility of MFC systems [9].

The efficiency of power generation in MFCs depends on the proton exchange membrane (PEM) used. The PEM must allow proton transport to the cathode while preventing the passing of other substances such as the substrate or the acceptor of electrons (O_2). Common PEM materials used in MFCs include salt bridges, Ultrex, and Nafion [10,11]. Among these, Nafion has been extensively researched due to its high ionic conductivity (10^{-2} S cm^{-1}). However, it is expensive and sensitive to the permeability of oxygen. Recently, Behera et al. [15] demonstrated an affordable alternative by using an earthen pot for MFC construction. It acts as the anode chamber and the proton exchange medium simultaneously, significantly reducing manufacturing costs. This innovative approach offers a practical and economical solution for MFC implementation [12,13,14].

In this study, we examined an innovative up-flow cylindrical MFC configuration's performance, featuring an outer cathode chamber and an inner concentric anode chamber, complemented by a biocathode. A widely accessible PEM was used to conduct evaluations on the MFC under various OLRs. Subsequently, we compared the efficiency of this MFC, operating at its optimal OLR, including a different MFC variation where the earthen cylinder was used to build the inside anode chamber instead of a PEM. The anode effluent flows into the cathode to serve as the cathodic electrolyte during regular operation. To assess maximum power generation capabilities, we employed permanganate as an electron acceptor, effectively mitigating cathodic limitations for comparative analysis.

2. Materials and methods

2.1 Experimental configuration

The study involved the application of two up-flow dual-chamber microbial fuel cells on a laboratory scale. The polyacrylic plastic MFC-1, shown in Fig. 1, had a cylindrical shape with an inner anode chamber that was concentric (5 cm inner width) and an outer cathode chamber that had an inside diameter of 10 cm. Four PEMs, each with a surface area of 10 cm^2 (Nafion-117), were employed to separate the anode and cathode chambers, resulting in a total surface area membrane of 40 cm^2.

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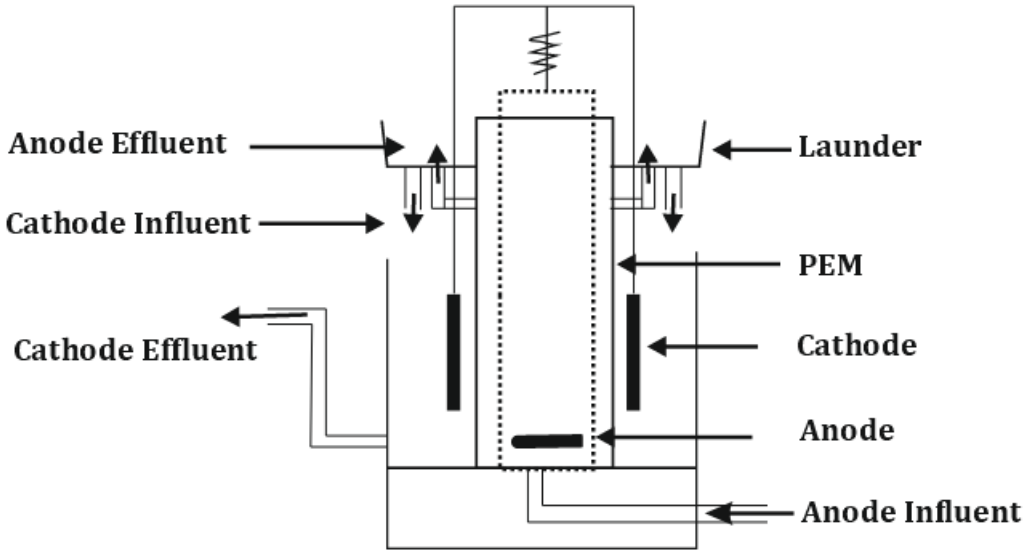

Fig. 1. A schematic design of the MFC-1 used in the investigation was shown. In MFC-2, an earthenware cylinder was used in place of the original polyacrylic plastic interior anode chamber.

In contrast, MFC-2 featured a concentric inside anode chamber (7 cm inner diameter) crafted from an earthenware cylinder, with no utilized PEM. The surface used for proton exchange was the 5 mm-thick clay cylinder wall. The earthen cylinder was composed of accessible local soil with elemental composition including Na (1.15%), Mg (1.52%), Al (20.50%), Si (53.52%), K (4.74%), Ca (1.15%), Ti (0.94%), and Fe (16.48%).

Both MFCs had a working volume of 600 mL in the anode chamber. MFCs, the anode electrode was made of coated stainless-steel mesh electrodes with an overall surface area of 360 cm^2, while the cathode electrode was made of graphite plates with a total surface area of 250 cm^2. The electrodes had an exterior of 100 ohms resistance and were externally linked via copper wires.

2.2 MFCs of Operation

The study used a synthetic wastewater solution with sucrose as a carbon source and a chemical oxygen demand (COD) of approximately 500 mg L^{-1}. The sucrose medium was supplemented with specific components per gram of COD, including NaHCO3 (1500 mg), NH4Cl (318 mg), CaCl2 . 2H2O (250 mg), MgSO4 . 7H2O (64 mg), K2HPO4 (27 mg), and KH2PO4 (9 mg). Additionally, trace metals were introduced as FeSO4. 6H2O (10.00 mg L^-1), MnSO4 (0.526 mg L^{-1}), ZnSO4 . 7H2O (0.106 mg L^{-1}), H3BO3 (0.106 mg L^{-1}), and CuSO4 . 5H2O (4.5 mg L^{-1}) [16]. For the first phase, to keep the sludge loading rate (SLR) at 0.1 kg COD kg VSS^{-1} d^{-1}, anaerobic sludge obtained from the bottom of a septic tank was heat-treated at 100°C for 15 minutes to inhibit methanogens [17]. This sludge was put into the reactors at a volume of 250 mL. The influent feed's pH remained between 7.1 to 7.9 throughout the experiment.

The MFCs were operated at ambient temperatures ranging from 26 to 34°C. MFC-1, constructed with a polyacrylic plastic structure, operated initially in continuous mode at OLRs ranging from 0.6 to 2.0 kg COD m^{-3} d^{-1}, with corresponding HRTs of 20 to 6 hours. MFC-2 featured an earthen cylinder replacing the inner anode chamber and was operated at the optimal OLR determined

for MFC-1 to assess electricity generation and substrate degradation. Once the MFCs reached steady-state conditions, the researchers evaluated their maximum power production capacities by introducing 0.21 g L^{-1} of potassium permanganate (KMnO4) as a catholyte to address the potential cathodic limits.

2.3 Statistical Analysis

Suspended solids (SS), volatile suspended solids (VSS), and chemical oxygen demand were monitored following the standardized methods outlined by the American Public Health Association (APHA) [18]. Energy-dispersive X-ray (EDX) analysis was used to determine the elemental composition of the clay cylinder material using a scanning electron microscope (SEM) fitted with an EDX detector. Voltage and current measurements were performed using a digital multimeter integrated with a data acquisition unit sourced from Agilent Technologies in Malaysia. Power calculations were derived from the formula P = IV, where 'I' represents current, and 'V' denotes voltage. Following the stabilization of MFC performance, polarization curves were generated utilizing a 5 KU changing resistor. The MFC of the internal resistance was determined by analyzing the slope of the voltage-current plot [19].

The Coulombic efficiency (CE) was calculated by comparing the actual charge generated, determined by integrating the recorded electrical current over time, with the expected charge based on the consumed COD. To estimate the theoretical charge, the formula

$$CE = \frac{F*n*w}{M}$$

where F is Faraday's constant, n represents the number of moles of electrons produced per mole of substrate, w stands for the daily COD load consumed in grams, and M is the molar mass of acetate. [20].

3. Results and discussion

3.1 Organic matter removal

MFC-1 was operated at varying organic loading rates by adjusting the HRT while maintaining a consistent influent COD concentration of approximately 500 mg L^{-1}. Synthetic feed was added after the anode compartment was inoculated with anaerobic mixed communities. Anaerobic treatment of the synthetic wastewater occurred in the anode chamber, followed by subsequent aerobic treatment in the cathode chamber. The MFC required approximately 20 days to achieve steady-state conditions, described as steady performance going forward, when initially run at an OLR of 0.6 kg COD m^{-3} d^{-1} (Fig. 2) was implemented, followed by incremental increases in the loading rate. In MFC-1, the anode chamber achieved an average COD removal efficiency of 87.5 ± 1.6% at this initial OLR, which represented the highest efficiency observed among all experiments. This superior performance was attributed to the combination of the lowest OLR and the longest hydraulic retention time (HRT) of 20 hours during this phase.

A decrease in COD removal efficiency in the anode chamber was noted as the OLR increased. At an OLR of 2.0 kg COD m^{-3} d^{-1}, the average COD removal efficiency decreased to 70.6 ± 2.4% in the MFC. The total COD removal effectiveness varied from 92% to 95.5% for all OLRs after further aerobic treatment in the cathode chamber (Fig. 2). The aerobic treatment in the cathode chamber promoted substrate breakdown, resulting in over 90% COD efficiency in MFC-1 at all OLRs, despite the diminishing organic removal rate in the anode chamber with increasing OLR. These

results highlight the MFC configuration's potential as a dependable and efficient wastewater treatment technology that produces consistently high-quality effluent.

3.2 Electricity production

After a 22-day inoculation period, MFC-1 was able to attain consistent values of 7.20 mA for short-circuit current and 670 mV for open circuit voltage, at an organic loading rate (OLR) of 0.6 kg COD m^{-3} d^{-1}. As the OLR increased, electricity generation in MFC-1 also rose. Calculated to the net anode amount, the highest volumetric power was 8.02 W m^{-3} and 8.40 W m^{-3} for OLRs of 0.6 and 0.705 kg COD m^{-3} d^{-1}, respectively. At an OLR of 0.923 kg COD m^{-3} d^{-1}, MFC-1 exhibited the highest voltage and current outputs compared to other OLRs, has a maximum volumetric power of 10.04 W m^{-3} with a maximum OCV of 655 mV and SC of 9.20 mA. However, the amount of power generation decreased to 9.75 W m^{-3} with a subsequent rise in OLR to 2 kg COD m^{-3} d^{-1}.

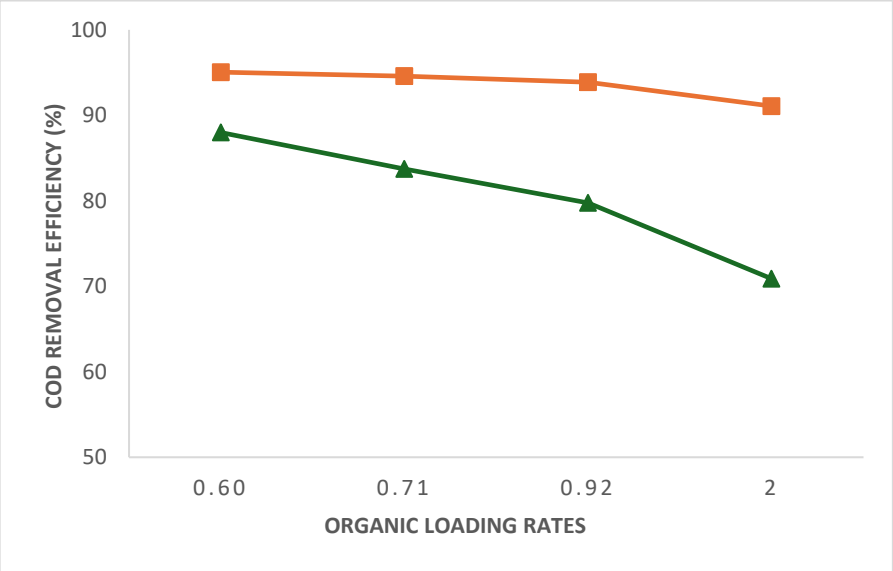

Fig 2. The removal of COD from the anode chamber and MFC-1 as an entire at different OLRs.

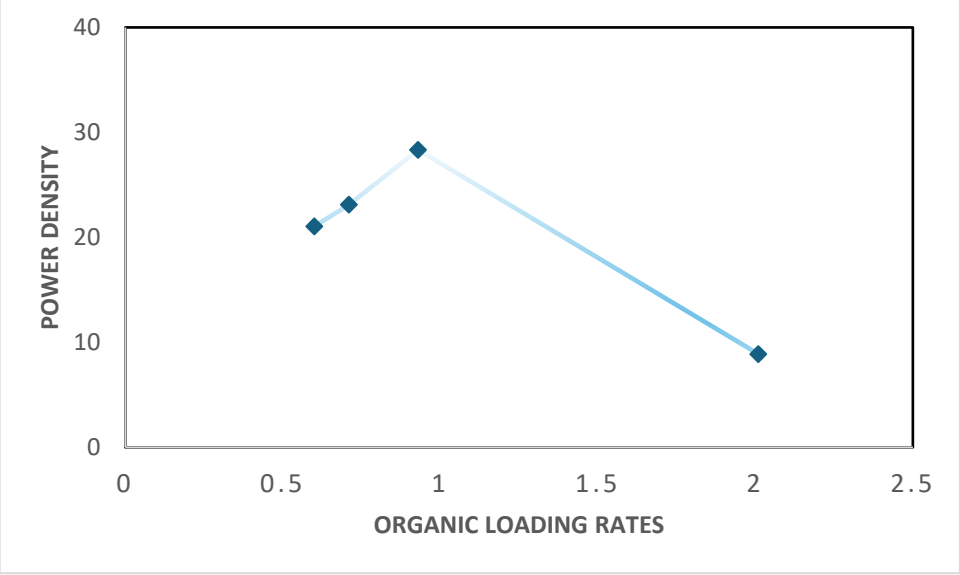

Fig 3. Power production in MFC-1 across varying OLRs with a 100 Ω.

As seen in Fig. 3, the sustained power generation was observed to be 21.24 mW m^{-2} and 23.05 mW m^{-2} at OLRs of 0.65 and 0.706 kg COD m^{-3} d^{-1}, respectively, measured over a 100 Ω external resistance. Notably, the highest sustainable power generation recorded among all OLRs tested was 28.27 mW m^{-2} (equivalent to 3.19 mA and 0.319 V) at an OLR of 0.926 kg COD m^{-3} d^{-1}. However, the output of sustainable electricity dropped to 8.92 mW m^{-2} when the OLR was increased to 2.1 kg COD m^{-3} d^{-1}.

In prior studies, Kim et al. [21] investigated a longitudinal cylindrical MFC operated at various OLRs, ranging from 0.042 to 0.412 kg COD m^{-3} d^{-1}, with sucrose serving as the basis. Their findings indicated an increase in power generation with higher OLRs within this range. Similarly, Mohan et al. [22] investigated how applied OLR affected power output in a two-chambered MFC running at OLRs between 0.517 and 1.033 kg COD m^{-3} d^{-1}. Power generation in their study reached its highest at an OLR of 0.574 kg COD m^{-3} d^{-1} and then began to decline as the OLR increased. Consistent with these observations, our study demonstrated an increase in power production up to an OLR of 0.926 kg COD m^{-3} d^{-1}, followed by a decline beyond this threshold. This decline beyond an OLR of 0.926 kg COD m^{-3} d^{-1} implies that the increased rate of organic loading exceeded the electrogenic bacteria's ability to oxidize.

3.3 Electricity production and Organic matter removal

MFC-2 was run continuously at an OLR of 0.926 kg COD m^{-3} d^{-1} to compare its performance to that of an earthen cylinder MFC without PEM and PEM-MFC (MFC-1). MFC-2 required around 14 days to reach a steady state, which was defined by consistent power generation and COD elimination. Remarkably, the MFC-2 anode chamber's average COD removal efficiency of 81.8 ± 1.8% was higher than the MFC-1 average COD efficiency of 79.4 ± 2.2% at the same OLR. Post-aerobic treatment in the cathode chamber, the overall COD removal efficiency was 92.5% in MFC-1 and notably higher at 95.7% in MFC-2.

The highest power density and volumetric power of MFC-2 were 243.29 mW m^{-2} and 14.59 W m^{-3}, respectively, due to its peak OCV of 715 mV and maximum SC of 12.25 mA. This earthenware cylinder MFC's performance using a low-concentration substrate, demonstrates its potential, comparable to MFCs employing metal catalysts on the cathode or specialized electrogenic cultures and mediators [23]. MFC-2 maintained 48.30 mW m^{-2} of power density and 2.89 W m^{-3} of volumetric power with a 100 Ω resistance. (equivalent to 4.17 mA and 0.417 V) [14].

The Coulombic efficiency of MFC-2 and MFC-1 were 19.8% and 14.2%, respectively. Remarkably, the earthenware cylinder of MFC outperformed the PEM-based MFC (MFC-1) in power generation. The earthen cylinder material facilitated efficient proton transfer, contributing to accelerated substrate degradation in MFC-2 and consequently higher COD removal efficiency compared to MFC-1. Moreover, the high number of protons and electrons in MFC-2 led to increased electron production and increased OCV and Coulombic efficiency due to quicker oxygen removal from the cathode chamber [15]. MFC-1 is net energy efficiency varied depending on the applied organic loading rate, from 0.8% to 2.52%. Notably, during the trial, the earthen cylinder MFC continuously demonstrated greater energy efficiency in comparison to the PEM-based MFC (MFC-1). Specifically, at an OLR of 0.926 kg COD m^{-3} d^{-1}, the energy generation efficiency across a 100 Ω external resistance was 1.09% for MFC-1 and 2.04% for MFC-2. During polarization, MFC-1 achieved its higher energy efficiency of 1.16% at a 70 Ω external resistance, while MFC-2 attained a peak efficiency of 2.54% at a 60 Ω external resistance.

In comparison, Kim et al. [21] reported overall energy efficiency in a longitudinal cylindrical MFC ranging from 0.1% to 6.5% depending on the influent sucrose loading. Nonetheless, the current investigation showed that the economical earthen cylinder MFC, which lacked PEM and used advanced electrode materials with noble metal catalysts, had a maximum energy efficiency of 2.54%.

3.4 Polarization

Research on polarization was conducted on both MFCs, wherein the external resistance was systematically varied from 5000 Ω to 10 Ω. The resulting polarization curve revealed a peak power density of 28.98 mW m^{-2} at an external resistance of 100 Ω for MFC-1,

Table 1. A comparison between MFC-1 and MFC-2 electricity generation

MFC	OCV (V)	Voltage across 100 Ω	Internal Resistance (Ω)	Current (mA)	Current with 100 Ω	Power Density with 100 Ω	Power/ Vol with 100 Ω
MFC- 1	0.655	319	95	9.20	3.19	28.27	1.49
MFC - 2	0.715	417	67	12.25	4.17	48.3	2.89

while MFC-2 exhibited a higher power density of 59.60 mW m^{-2} at an external resistance of 60 Ω. The internal resistance of each MFC was determined by analyzing the slope of the voltage-current plot, yielding values of 96 Ω for MFC-1 and 69 Ω for MFC-2. Remarkably, the earthen cylinder MFC demonstrated a lower internal resistance compared to its PEM-based counterpart, which contributed to its superior power generation performance.

3.5 Assessment of MFC efficiency using permanganate as the cathodic electron acceptor

Upon achieving operational stability with consistent substrate degradation and voltage generation, by using KMnO4 with anode chamber discharge as the cathodic electron acceptor, changes were produced. Peak power density and volumetric power in MFC-1 after KMnO4 addition were 448.75 mW m^{-2} and 26.92 W m^{-3}, respectively.

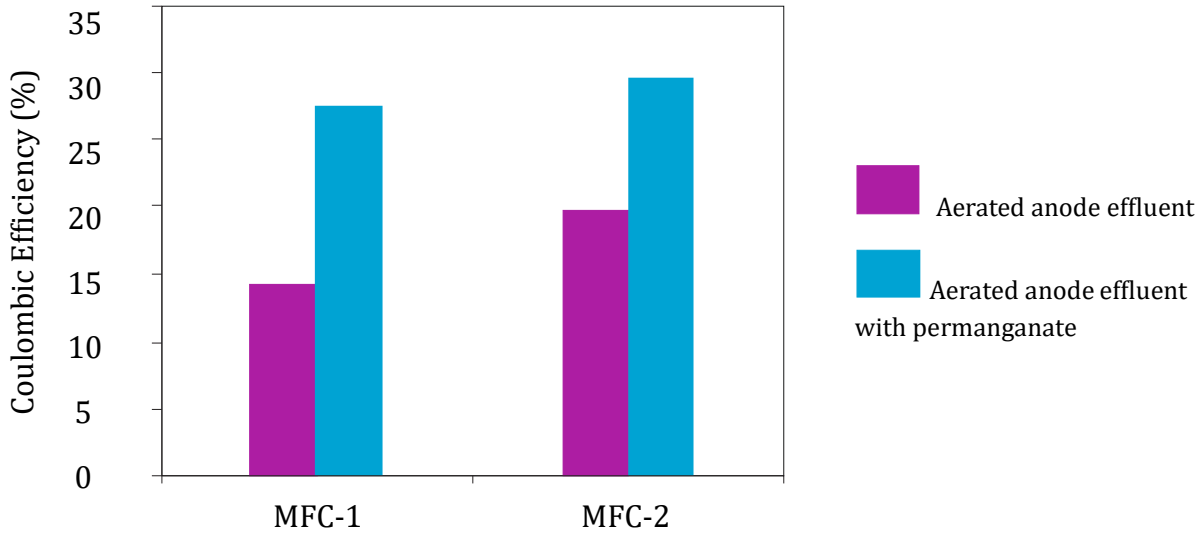

Fig 5. (a) Evaluation of Coulombic Efficiency performance between MFC-1 and MFC-2.

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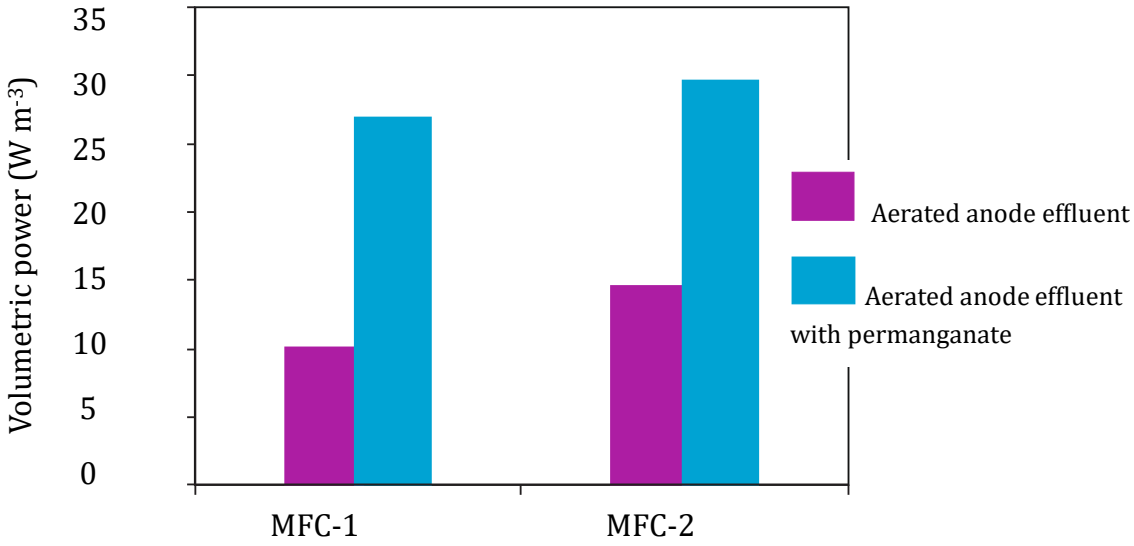

Fig 5. (b) Evaluation of Volumetric power performance between MFC-1 and MFC-2.

At an OLR of 0.926 kg COD m^{-3} d^{-1}, accompanied by an OCV of 0.897 V and SC of 17.99 mA (Fig. 5b). The Coulombic efficiency escalated from 14.2% (pre-KMnO4 addition) to 27.6% post-KMnO4 (Fig. 5a). Similarly, MFC-2 is highest power density and volumetric power were 29.67 W m-3 and 480.18 mW m-2, respectively, after KMnO4 inclusion. After adding KMnO4, the Coulombic efficiency in MFC-2 rose from 19.8% to 29.7%. There is an important potential difference between the anode and cathode, which is responsible for the increased power density after KMnO4 addition due to its enhanced redox potential. Moreover, the current production surged post-KMnO4 addition, potentially due to a shift in reaction equilibrium leading to enhanced electron harvesting at the anode facilitated by greater electron consumption at the cathode [24]. The significant enhancement in the Coulombic efficiency post-KMnO4 addition, particularly in MFC-1 with a 94.4% increase, implies that charge transfer was a critical limiting factor in its performance, ameliorated by KMnO4 incorporation. Conversely, the relatively modest performance improvement post-KMnO4 50% increase in Coulombic efficiency is added in MFC-2 suggesting its superior operation compared to MFC-1 when oxygen served as the cathodic electron acceptor. This underscores the potential of MFCs fabricated with an earthen cylinder to outperform those when oxygen serves as the cathodic accepting electron in PEM.

4. Conclusion

The investigation focused on the relationship between power production and the organic loading rate in the up-flow cylindrical MFC. The MFC configuration proved highly effective in treating wastewater, achieving over 90% COD removal efficiency across all loading rates. The earthen cylinder material showed better proton transfer capabilities, producing better results in terms of eliminating organic materials and producing power, compared to MFCs that use expensive proton exchange membranes. This makes the earthen cylinder a cost-effective alternative to PEMs in MFC fabrication. The excellent performance of low-cost earthen cylinder MFCs could lead to commercial viability for this technology.

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