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Functional study of cover holes for air cathodic protection and retention of cathodic function by ball-shaped cover

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Abstract

As the world's electricity consumption increases in proportion to population growth, research on renewable energy sources to replace fossil-fuel-based power generation is underway around the world. Microbial fuel cells (MFCs) are attracting attention as a renewable energy source that can generate electricity while decomposing pollutants in wastewater. Among these, single-chamber MFCs have low maintenance costs and are suitable for independent operation in outdoor environments. However, the air cathodes widely used in single-chamber MFCs are susceptible to degradation due to moisture. In this study, we proposed a simple method to protect the air cathode from moisture using a spherical frame. The spherical MFC we constructed prevented the inflow of wastewater whilst achieving stable, continuous operation for three months. Compared to an MFC where the cathode was contaminated by wastewater, the proposed spherical MFC exhibited an average output voltage 300 mV higher and a maximum power density approximately 13.9 times greater. The maximum power density of 14.27 $\mu\text{W}/\text{cm}^2$ is 42% lower than that of floating-type MFCs reported in previous studies, indicating room for improvement. However, as the spherical MFC frame also functions as a protective cover, it can contribute to the manufacture of more practical MFC devices.

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Keywords

Microbial fuel cells; Air cathode; Ball-shapes; Sustainable Energy

1. Introduction

In recent years, the world's population has continued to grow and is estimated to reach 9.4-10.1 billion by 2050 (Danan et al., 2021). As the population increases, the consumption of electricity, which is indispensable for our daily lives, is also increasing every year (Klyuev et al., 2021). Accordingly, the world's consumption of fossil fuels is also increasing (Holechek et al., 2022). However, the increased use of fossil fuels has also led to increased emissions of greenhouse gases such as CO₂, CH₄, and N₂O, which in turn contribute to global warming (Filonchuk et al., 2024). Given these current conditions, research is underway worldwide to replace fossil fuels with sustainable energy sources. In this context, fuel cells are attracting attention as an alternative power generation technology to fossil fuels.

Fuel cells convert chemical energy into electrical energy and thermal energy by oxidizing hydrogen at the anode and reacting oxygen at the cathode with protons and electrons transferred from the anode, resulting in the reduction of oxygen to produce water (Mekhilef et al., 2012). Since water is the only substance generated before and after the reaction, research is progressing on this system as a very clean power generation system with little environmental

impact. Among fuel cells, microbial fuel cells (MFCs) are attracting attention as a very clean renewable energy source because they can simultaneously treat wastewater and recover electrical energy. While most fuel cells use an anode to reduce hydrogen, MFCs use organic matter as the reducing material, making them extremely safe fuel cells. The anode is a bio-metabolizing microorganism that produces electrons and protons from the organic matter in the wastewater, and the oxygen taken in at the cathode is oxidized using the electrons and protons sent from the anode, thereby operating as a battery (Kižys et al., 2023).

MFCs are very simple and can generate electricity because they consist mainly of an anode, a cathode, and wastewater containing organic matter. There are two types of MFCs: single-chamber MFCs, in which the anode and cathode exist in the same chamber, and dual-chamber MFCs, in which the anode and cathode exist in different chambers separated by an ion exchange membrane. Although each type has its own merits, the single-chamber type is considered more suitable for practical MFC applications because it avoids the need for aeration and cathode liquid collection, is easier to maintain, and can be automated more easily (Huang et al., 2023). Among cathodes, air cathodes are those that do not use aeration. This is a simple structure that allows oxygen to be taken in by exposing the cathode to air, and it is also very easy to handle. One structure of MFCs that makes effective use of air cathodes is the floating microbial fuel cell (FMFC). This is a type of MFC that floats on wastewater and generates electricity autonomously; it is characterized by an anode that is constantly immersed in the wastewater and a cathode that is exposed to the outside air (Nguyen & Taguchi, 2020). However, it is widely known that if the cathode gets wet due to disturbances such as rain or waves and the air cathode surface is covered, the potential drops (Li et al., 2023). As structurally similar issues arise in FMFCs, cathode protection is a critical challenge for ensuring long-term stable operation. If the air cathode is completely sealed with a cover to deal with this problem, oxygen deficiency will occur, which will also degrade performance (Mateo et al., 2015). Therefore, to use air cathodes in a real environment, it is necessary to both protect them from moisture and exchange oxygen.

Therefore, we propose a new type of FMFC, the Floating Spherical MFC (FBMFC), with a spherical frame that enables floating, cathodic protection, and air exchange with a simple structure without increasing the number of materials. The effect of the hole in the top of the frame was verified by making a separate cover and pouring water stained with methylene blue over the cover.

2. Materials and methods

2.1 Preparation of electrode material

Rice husk smoked charcoal (RHC) carbonized by the smoking method was used as the electrode material; RCH was stirred in 0.6 mol/L NaOH solution for 24 hours and subjected to alkaline etching treatment. After etching, the RHC was washed with tap water and dried at 60°C for at least 48 hours. After that, it was ground into a powder using a mortar. Co-MnO₂ catalyst was used for the cathode in addition to RCH. The catalyst was synthesized by a simple redox method: KMnO₄, Co(NO₃)₂, and carbon black were mixed in sequence and dried to obtain the catalyst. This is a commonly proposed redox method of preparation (Xia et al., 2020).

2.2 Electrode Preparation

The composition of the electrode materials is shown in Table 1. Anodes were prepared by mixing powdered RCH (0.45 g) with India ink (1.75 mL). The cathode was prepared by mixing powdered RCH (0.2625 g) and sumi ink (1.625 mL) with Co-MnO₂ catalyst (0.375 g). Electrodes were prepared by pouring each electrode material solution into a mold and drying in a dryer at 60°C until solidified (Soichiro et al., 2023). The molds were made of PLA resin. After drying, the electrodes were removed from the molds, the Nafion membrane was placed between the anode and cathode as shown in **Figure 1**, and the assembly was solidified with adhesive; the molds were fixed with screws to prevent any gap between the anode, cathode, and Nafion membrane.

In this way, the anode and cathode were made into one block of electrodes (Soichiro et al., 2023).

Table 1. Composition of electrode materials

Material	Anode	Cathode
RHC(g)	0.45	0.2625
India ink (ml)	1.75	1.625
Co-MnO ₂ catalyst (g)	-	0.375

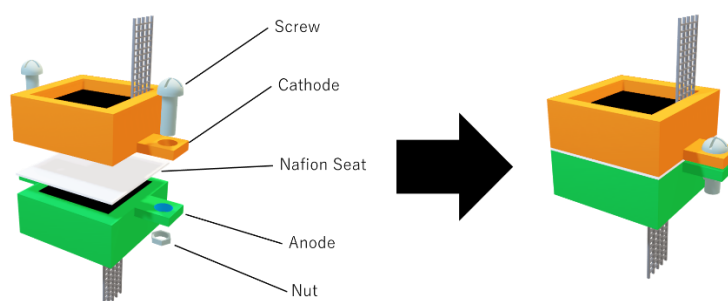


Figure 1. Structure of anode-cathode integrated electrode.

2.3 Preparation of the frame

The frame was made of PLA resin, as was the mold for the electrode. It consists of a lower part on which the electrode is mounted and a hemispherical upper part with small holes. The upper part was drilled with three holes of 500 μm in diameter, vertically aligned in eight directions (Figure 2).

A drill was used to drill the holes. The electrode and the lower part of the frame were fixed with adhesive as shown in Figure 3. The purpose of these holes was to allow the exchange of oxygen used in the cathode reaction whilst preventing the inflow of wastewater from the outside. After the upper parts were completely glued together, three coats of waterproof paint were applied to prevent water from penetrating into the frame.

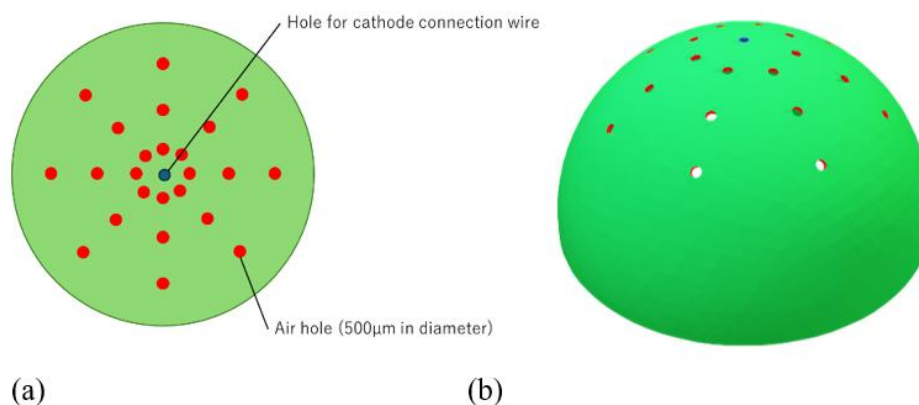


Figure 2. Structure of the ball-shaped MFC, including the plan view of the holes in the upper cover and the corresponding CAD image.

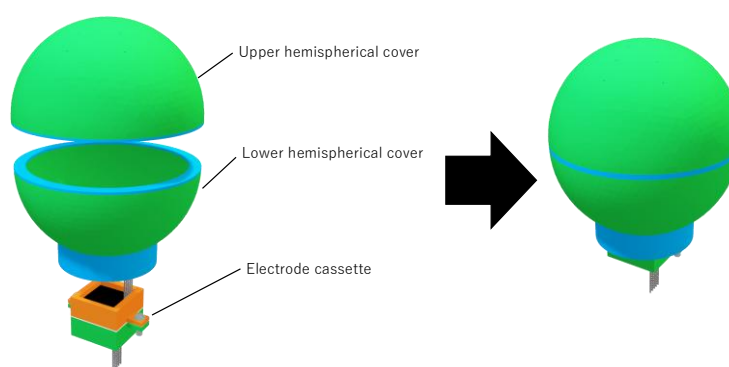


Figure 3. Structure of FBMFC.

2.4 Composition of MFC

As shown in **Figure 4**, the MFC created is a single-chamber type. The chamber was filled with 2 L of 10% LB medium solution. Microorganisms were collected from Japanese paddy field soil.

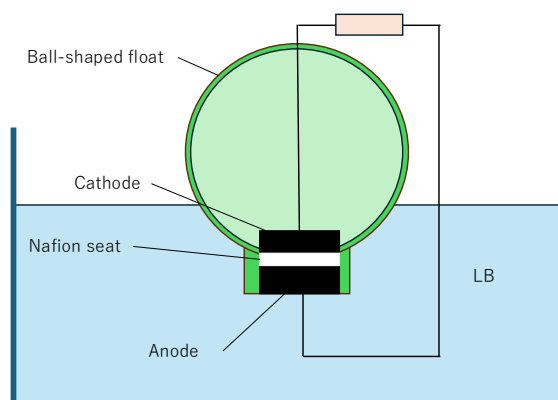


Figure 4. Setup of FBMFC.

2.5 Measurement Method

An external resistor of 10 k Ω was connected to the MFC. Experiments were performed in an incubator controlled at 27°C. To see the reduction in cathode performance and output voltage due to wastewater inflow, enough LB medium was added until a voltage reversal was observed. Output voltage was automatically measured every 15 minutes by DAQ over a two-month period. The anode and cathode electrode potentials were measured using AgCl electrodes. After the voltage reversal, the medium was replaced, and the output voltage of each MFC was restored. The current densities and voltages were then measured using a multimeter. The measurements were calculated by dividing the power and current, respectively, by the effective area of the anode.

2.6 Evaluation of the effectiveness of the small holes in the upper part of the frame

To evaluate the effectiveness of the holes drilled in the upper part of the MFC, a cover with a hole of 500 μm , the same size as the one used in this experiment, and a cover with a hole of 1500 μm , three times the size of the hole in the cover used in this experiment, were prepared. The top of the covers was covered with 500 mL of colored water, and the amount of water passing through the holes was observed.

3. Results and Discussion

3.1 Long-term stability of MFC power generation

Figure 5 shows the change in voltage of the MFC when an external resistor ($10\text{ k}\Omega$) is connected. To assess the change in cathode performance due to wastewater inflow, two FBMFCs were prepared for this experiment: an FBMFC that floated stably without wastewater inflow and an FBMFC with wastewater inflow. As a result, the FBMFC with no wastewater inflow generated electricity stably for 90 days after installation, while the FBMFC with wastewater inflow showed a significant decrease in potential from 41 days after the inflow was confirmed, and the anode and cathode potentials reversed on the 63rd day.

Figure 6 shows the time shift of the anode and cathode potentials of the FBMFC. While both anode and cathode potentials are stable regardless of wastewater inflow, the cathode potential significantly decreased after 41 days of wastewater inflow, and a comparison between days 23 to 37, when the cathode potential stabilized after installation, and days 64 to 87, when the cathode potential decreased after degradation, shows that the average potential before inflow was -134.0 mV , 134.0 mV , while the average potential after the inflow was -442.8 mV , a drop of more than 300 mV . This indicates that the inflow of wastewater into the cathode caused biofilm to settle on the cathode.

These results indicate that cathodic protection is essential for stable, long-term power generation in MFCs, and that once wastewater enters the MFC, its performance is significantly degraded and recovery is not expected.

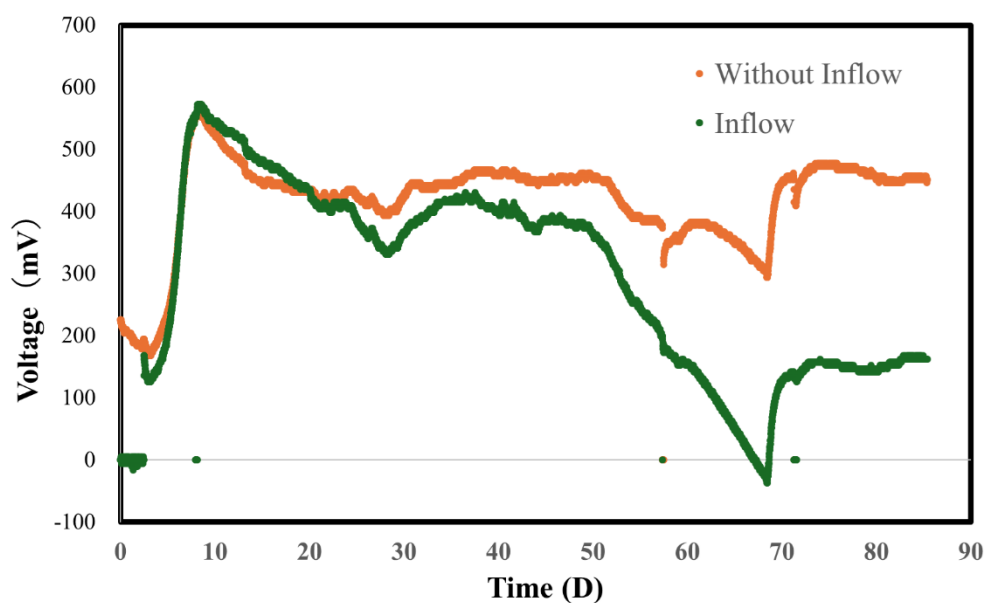


Figure 5. Comparison of time shift of FBMFC voltage with and without wastewater inflow.

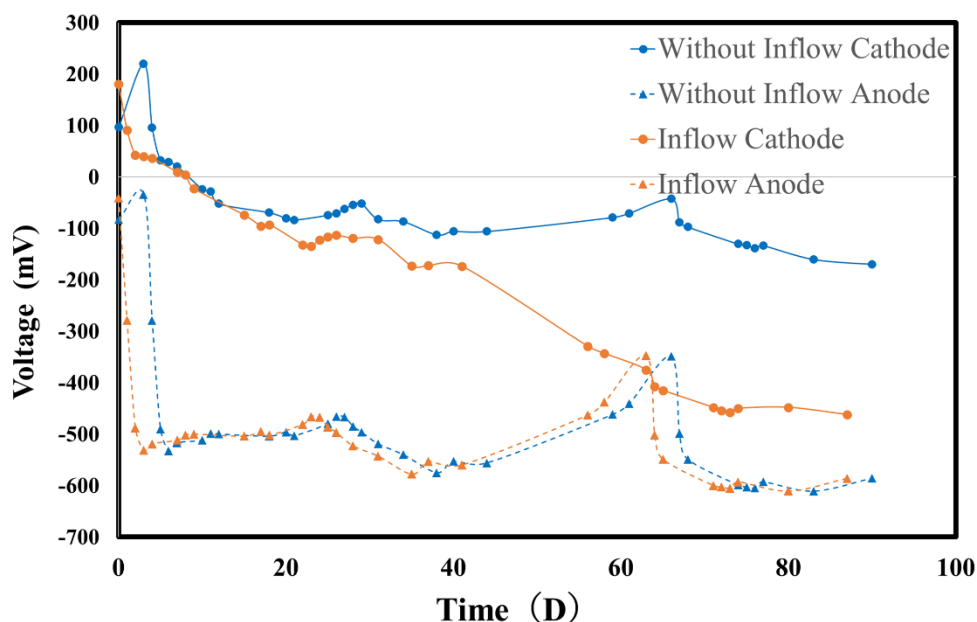


Figure 6. Comparison of time shift of anode and cathode electrodes of an FBMFC with and without wastewater inflow.

3.2 FBMFC Performance

Figure 7 shows the power density curve of the FBMFC. The FBMFC with no wastewater inflow had a maximum current density of $14.27 \mu\text{W}/\text{cm}^2$, while the FBMFC with confirmed wastewater inflow had a maximum power density of $1.03 \mu\text{W}/\text{cm}^2$, a 93% reduction in power density. The internal resistance of the two did not differ by 2000Ω , indicating that the inflow of wastewater significantly reduced the performance of the FBMFC. The maximum power density of a float-type MFC using a similar electrode preparation method is $24.8 \mu\text{W}/\text{cm}^2$ (Soichiro, H. et al., 2023). This power density is 1.74 times higher than that of the MFC created in this study; the decrease in the maximum power density of the FBMFC compared to the reference study can be attributed to the increased resistance due to the use of the Nafion membrane.

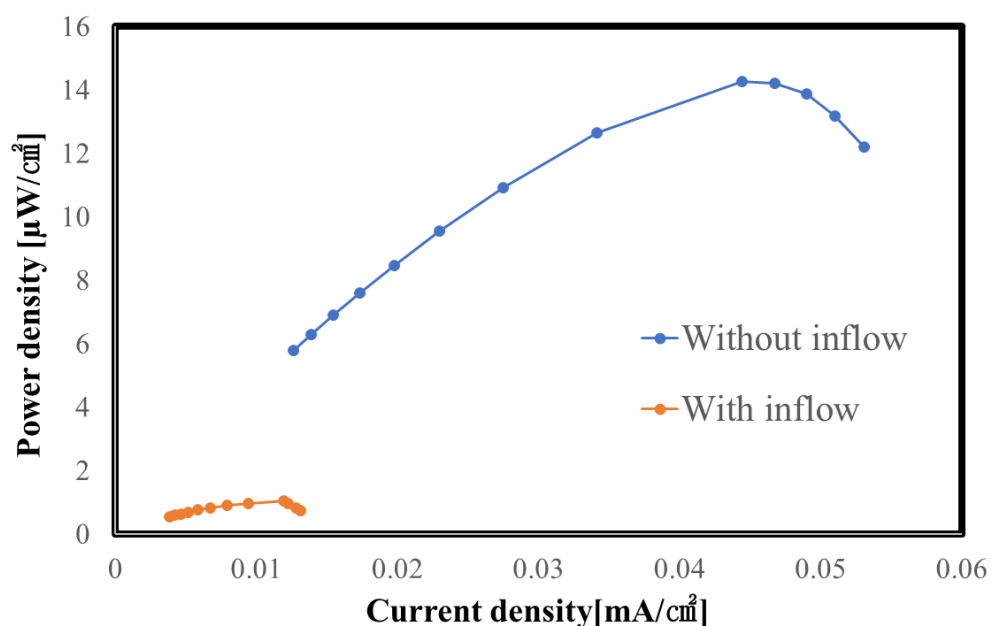


Figure 7. Comparison of maximum power density of FBMFC with and without wastewater inflow.

3.3 Evaluation of the effectiveness of FBMFC cover holes

The right side of **Figure 8** shows a cover with 500 μm holes, and the left side of Figure 8 shows a cover with 1500 μm holes. Under these two covers, a container was glued to catch the liquid leaking from the holes. From the top of the container, 500 mL of tap water stained with methylene blue was poured for 12 seconds. Then, almost no inflow of water could be observed in the cover with 500 μm holes, whereas an inflow of wastewater could be observed in the cover with 1500 μm holes. This indicates that even a hole of 500 μm can protect the cathode from rain and waves.



Figure 8. Comparison of the effectiveness of cathodic protection by the size of holes in the upper ball-shaped cover.

3.4 Discussion

Next, we examine the effectiveness of the spherical cover proposed in this study from the perspective of air cathode protection. The experimental results confirmed that preventing cathode contamination is essential for maintaining the functionality of the air cathode. Furthermore, it was found that the cover proposed in this study suppresses water ingress from the external environment. Consequently, the spherical cover is considered a structure that reduces the risk of cathode contamination and helps maintain stable power generation. Furthermore, as the cover used in this study is made of ABS resin—a material renowned for its durability and chemical stability—it is considered to offer a certain level of safety even when used in natural environments. Moreover, as this structure can be easily fabricated using a 3D printer and does not require complex processes, it can be manufactured at low cost. In light of the above, it is suggested that the spherical cover proposed in this study, as a simple and low-cost air cathode protection technology, is applicable to the long-term stable operation of MFCs in outdoor environments.

4. Conclusions

In this study, the frame of the MFC was made spherical, and a 500 μm hole was made in the top of the sphere in order to generate stable power while floating for a long period of time. As a result, it was found that the FBMFC that had no inflow of wastewater remained afloat for three months and generated an average of 444.55 mV of electricity stably for a long period of time. However, for FBMFCs with wastewater inflow, the cathode voltage dropped 289 mV from the stable state, and the overall power generation dropped by an average of 72.8%. In addition, experiments to evaluate the effectiveness of cover holes showed that even a 500- μm hole is sufficient to protect the cathodes inside the cover from rain and waves, but a 1500- μm hole would allow wastewater to enter the cover. These results demonstrate that the spherical cover proposed in this study is effective for cathode protection and, consequently, that FBMFCs can generate electricity stably over long periods even in environments where practical application is anticipated, such as rivers, lakes, and wastewater treatment facilities. Furthermore, they demonstrate that even a hole diameter of 500

μm —the smallest diameter reproducible in the laboratory is sufficient to allow oxygen exchange to the air cathode whilst adequately protecting it from external influences. As this structure protects the air cathode simply by perforating the cover, it does not require complex manufacturing processes. Consequently, it is expected to contribute to streamlining industrial processes in FBMFC production and to expanding more practical MFC technology.

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