

Research Paper

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Proposal for a Binder Blended with PTFE and Japanese Sumi Ink for the Cathode in Microbial Fuel Cells

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Abstract

Various issues, such as population growth, agriculture, and urban expansion, have led to the continuous degradation of water resources. To solve this problem, sustainable water treatment technologies are required. In water treatment, microbial fuel cells (MFCs) have attracted attention because of their low cost and low risk of cross-contamination. However, the performance of membrane-less MFCs degrades over time because of microbial adhesion to the cathode. In this research, we proposed using a blend of polytetrafluoroethylene (PTFE) and Japanese Sumi ink to prevent the adhesion of microbial cells to the cathode. The hydrophobic properties of PTFE prevented the adhesion of microbes on the cathode surface. In addition, the Sumi ink improved the electrical conductivity and physical stability of the cathode. It was confirmed that the hydrophobic properties of PTFE and the conductive and physical stability of Sumi ink could be fully exploited by adjusting the composition ratio of PTFE and Sumi ink. As a result, the power density of 35.1 $\mu\text{W}/\text{cm}^2$ and stable operation for at least 114 days were obtained.

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Keywords

Ink; Cathode; Binder; PTFE; Sumi ink

1. Introduction

Water resources are continuously degraded due to various issues, such as population growth, agricultural and urban expansion, land use change, and overdevelopment caused by economic development (Mittal et al., 2023). Therefore, there is a need for efficient and sustainable water treatment technologies with low energy costs and a low risk of cross-contamination (Fan et al., 2023). Among various water treatment technologies, biological treatment technology is widely used because it uses microbial metabolism to decompose contaminants in water, offering the advantages of low cost, simple operation, and prevention of cross-contamination (Jin et al., 2023). Microbial fuel cells (MFCs) are systems in which anodes receive electrons generated by microorganisms when they decompose organic matter in their metabolism process and utilize the electrons for electricity generation (Nguyen et al., 2022). Thus, the MFCs have been applied in wastewater treatment.

When membrane-less MFCs were used in wastewater treatment, biofilm formation at the cathode degraded the performance of the MFCs (Xu et al., 2019). Therefore, methods to inhibit the formation of biofilms, such as making

the cathode bactericidal properties, have been studied (Lai et al., 2022). Polytetrafluoroethylene (PTFE) has been widely considered for use as a good binder for the cathode electrode because of its low microbial affinity caused by its hydrophobic properties and high oxygen reduction reaction (ORR) performance (Santoro et al., 2013, Narayanasamy & Jayaprakash, 2020). However, since PTFE is a non-conductive material, using PTFE increases the internal resistance of the electrode and causes a decrease in cathodic performance (Sanchez-Pena et al., 2021).

The previous experiment found that Japanese Sumi ink can be used as a conductive binder for low-cost and easy-to-fabricate electrodes (Hirose et al., 2023). Japanese Sumi ink is a colloidal solution in which carbon black is dispersed, making it a conductive material. However, because the main composition of Sumi ink is carbon black, it has a characteristic of bacterial affinity and may facilitate the formation of biofilms at the cathode.

This study considered blending PTFE and Japanese Sumi ink to make the binder for the cathode electrode. The purpose of this blending is twofold. First, Sumi ink helps improve the conductivity of the blending material at a low cost (Sumi ink: Approx. \$0.81/180ml). Second, PTFE prevents the adhesion of microorganisms to the blend material. Moreover, this paper examined the ratio of PTFE and Sumi ink in the cathode material to find the optimum blend for improving the performance of the cathode in the membrane-less MFC. In addition, rice husk charcoal was used as the primary material for the electrodes. Because rice husks are industrial waste generated during rice harvesting, the cost of electrodes can be expected to decrease.

2. Materials and methods

2.1. Preparation of cathode

To fabricate the cathode, a solution was prepared by mixing alkali-treated rice husk charcoal (Tokorozawa Ueki Bachi Center, Ltd., Saitama, Japan) with Sumi ink (Daiso Industries Co., Ltd., Hiroshima, Japan), and PTFE (D-210C, DAIKIN INDUSTRIES, LTD., Osaka, Japan). Three types of electrode solutions were prepared by changing the PTFE and Sumi ink ratio, as shown in Table 1. Block-shaped electrodes were made in the same way as in the previous experiment (Hirose et al., 2023) (the prepared solution was poured into a mold and dried). The size of the cathode was 1 cm².

Table 1. Composition of PTFE, Sumi ink, and rice husk charcoal in the fabricated cathodes.

Cathode	Sumi ink (ml)	PTFE (ml)	Rice husk charcoal (g)
PTFE 0	3	0	0.6
PTFE 40	1.8	1.2	0.6
PTFE 80	0.6	2.4	0.6

2.2. MFC setup and operation

The schematic of the MFC is shown in Fig. 1. In this research, a floating membrane-less single chamber MFC type was used. The anode and cathode were separated by medium with a distance of 2 cm.

The anode of the MFC was fabricated by using 2.4 ml of Sumi ink and 0.6 g of rice husk charcoal. The size of the anode was 1 cm², the same as the cathode. To simulate the operation of the MFC in wastewater, MFC was placed floating in a solution of 60 ml of Luria-Bertani (LB) medium diluted in 300 ml of tap water (COD: 2976 mg/l) plus 1 ml of muddy water collected from a rice field (34°59'42.7986" N, 135°57'16.1892" E, Shiga, Japan) as a microbial source. LB medium was prepared by dispersing 2.5 g yeast extract, 5 g tryptone, 5 g NaCl, and 0.03 g NaOH (to maintain pH 7) in 500 ml purified water.

The MFC was operated indoors (25 ± 1 °C) with an external resistor of 10 kΩ connected between the two electrodes. Measurements were continued over five cycles by replacing the new aqueous LB solution when most of the organic matter had been degraded, and the output of the MFC dropped.

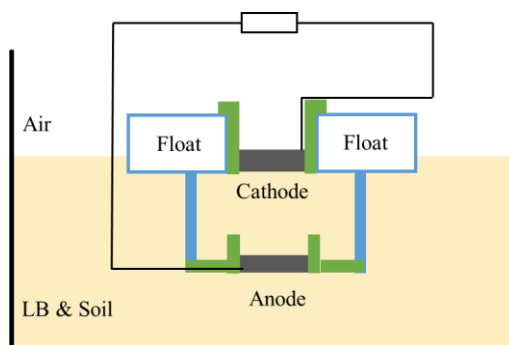


Figure 1 Schematic of the float-type microbial fuel cell.

2.3. Evaluation Method

Scanning electron microscope (SEM) and cathodic potential were used to evaluate the amount of microbial deposition on the cathode surface. The surface of the cathode taken out after the MFC operation test was observed by SEM (S-4300, Hitachi, Ltd., Japan). The cathodes were dehydrated and fixed using ethanol, and then the surfaces were sputtered with gold to prepare samples for SEM observation. Ag/AgCl was used as the reference electrode to measure the cathodic potential.

To evaluate the performance of the MFC, the power density curve was measured, and the internal resistance was calculated. The power density curve was obtained by measuring the voltage corresponding to the value of the external resistance (10-3000 Ω) and calculating it based on the voltage and resistance. However, a 10 Ω external resistor was connected for 2 hours to consume excess electrons in the MFC before measuring the voltage. The internal resistance was determined from the resistance at the highest power density in the power density curve.

3. Experimental Results and Discussion

3.1. Hydrophobicity evaluation of cathode surface

A contact angle test was performed to confirm the effect of changing PTFE content on the hydrophobicity of the cathode surface. Fig. 2 (a), (b), and (c) show the cathodes immediately after creation, and their contact angles were 0°, 100°, and 138°, respectively. PTFE 40 and PTFE 80 have obtuse contact angles, indicating that their surfaces are hydrophobic. These results confirmed that by adding PTFE to the cathode, the hydrophobicity of the cathode could be increased. Also, by comparing Fig. 2 (a), (b), and (c), it was confirmed that the higher the PTFE content, the higher the hydrophobicity.

The contact angles of (a'), (b'), and (c') in Fig. 2 were 0°, 99°, and 136°, respectively. Comparing these with (a), (b), and (c), there was no significant change in contact angle. This confirms that the hydrophobicity was not significantly degraded by placing the cathode in water.

3.2. Microbial formation on the cathode surface

Comparing Fig. 3 (a), (b), and (c), the higher the content of PTFE in the binder, the lower the amount of microbial adhesion on the surface. Fig. 4 shows the cathode potential monitoring in the fifth MFC operation cycle. From this monitoring, the cathode potential is improved by PTFE. This is thought to be due to PTFE reducing the amount of microbial adhesion on the cathode surface thanks to its hydrophobic characteristic.

From the data shown in Fig. 2 - 4, it is considered that the higher the content of PTFE in the binder, the more hydrophobic the cathode surface becomes, the fewer microorganisms adhere to it, and the higher the cathode potential.

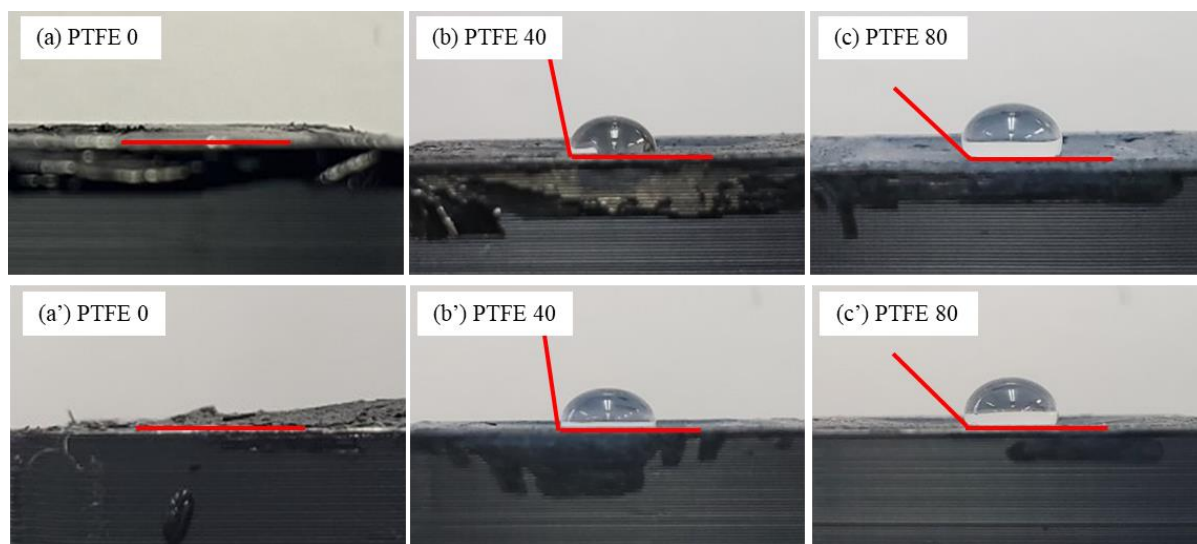


Figure 2 The contact angle of the cathode surface immediately after fabrication (a - c) and after 40 days in water (a' - c').

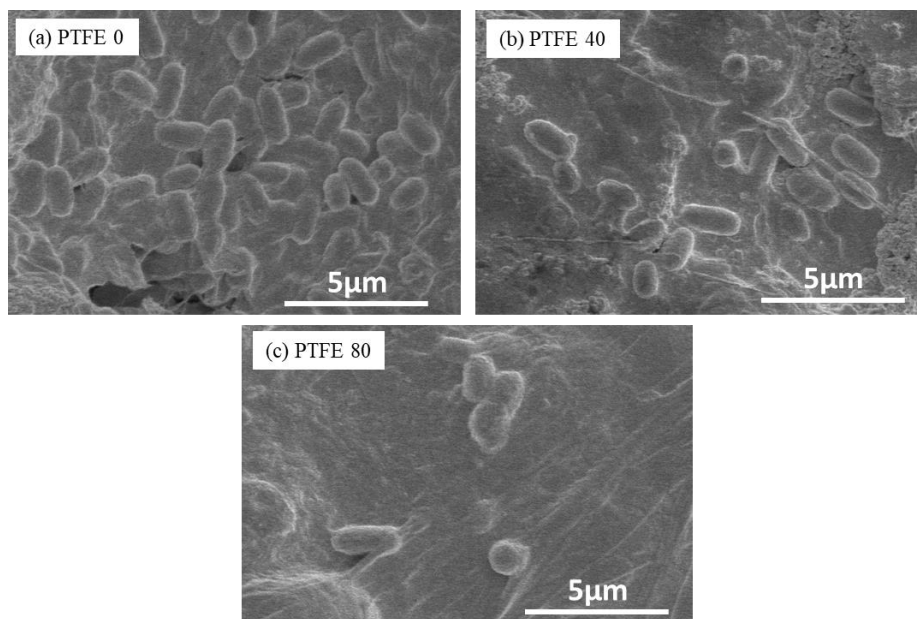


Figure 3 SEM images of (a) PTFE 0, (b) PTFE 40, and (c) PTFE 80 surfaces after MFC operation.

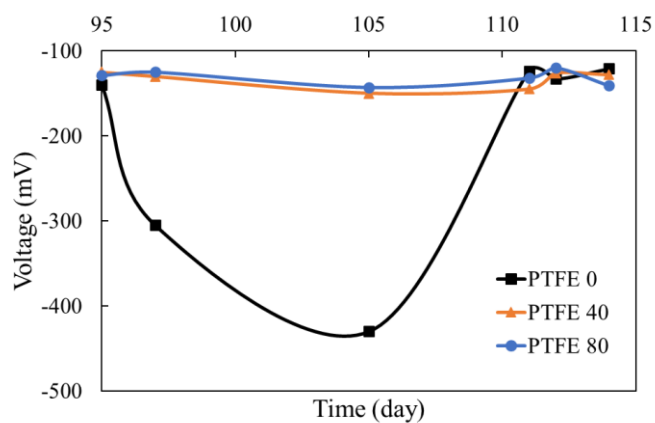


Figure 4 Cathode potential versus Ag/AgCl reference electrode during the fifth cycle of operation.

3.3. Evaluation of MFC performance

For 0%, 40%, and 80% PTFE content in the cathode, the obtained power densities of the MFCs were $6.5 \mu\text{W}/\text{cm}^2$, $35.1 \mu\text{W}/\text{cm}^2$, and $21.7 \mu\text{W}/\text{cm}^2$, respectively, as shown in Fig. 5. By blending PTFE with Sumi ink, power density was increased by up to 440% (PTFE 40 cathode compared with PTFE 0 cathode). This result confirms the effectiveness of the PTFE and Sumi ink binder blend on the performance of the cathode and the MFC. Furthermore, the internal resistance of the MFCs determined from these power density curves is shown in Table 2. The internal resistance of PTFE 40 was 0.46 times that of PTFE 80. It was found that the lower the PTFE content in the binder, the higher the Sumi ink content, leading to lower internal resistance.

In addition, Fig. 6 shows that the MFC with PTFE 0 and PTFE 40 had a stable power density for at least 114 days. On the other hand, the MFC with PTFE 80 showed a significant decrease in power density in the fifth cycle. This may be due to the low content of Sumi ink in the cathode, which affected the physical stability of the electrode. The above results suggest that PTFE 40 maximized the hydrophobicity of PTFE and the conductivity and physical stability of Sumi ink, resulting in high MFC performance.

Table 2 The internal resistance of each MFC using cathodes with different PTFE content.

	PTFE 0	PTFE 40	PTFE 80
Internal resistance	300 Ω	700 Ω	1500 Ω

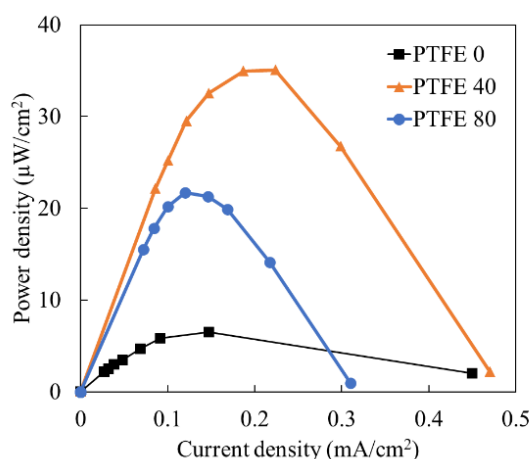


Figure 5 Power density curve of MFC after 80 days of operation.

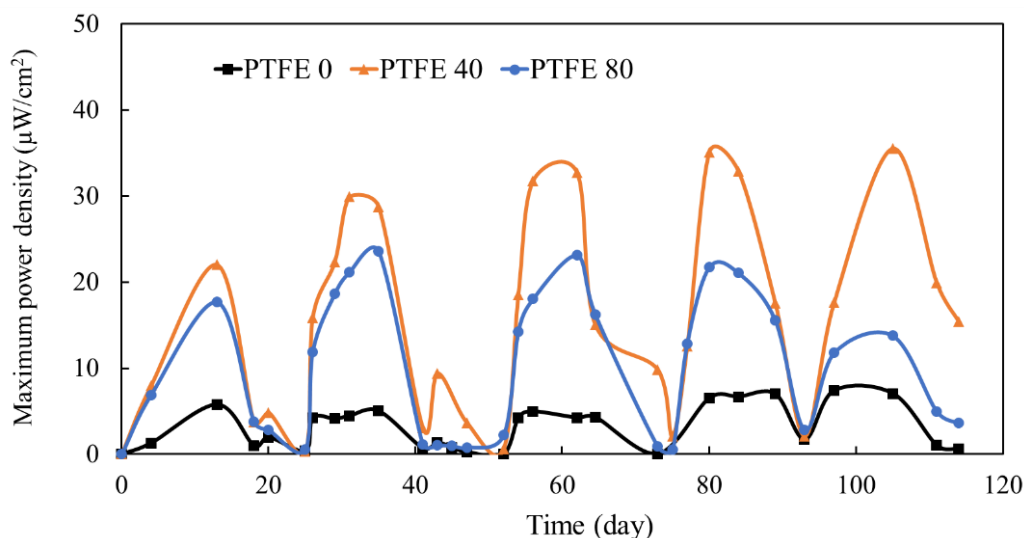


Figure 6 The maximum power density of MFC over time.

4. Conclusion

In this study, we proposed using a PTFE and Japanese Sumi ink blend as a binder for the cathode in MFCs used in wastewater. The proposed binder increased power density by up to 440%. The cathode with the binder with 40% PTFE content exhibited stable performance for at least 114 days. When the content of PTFE in the binder was increased, the hydrophobicity of the cathode surface increased, and the amount of microbial adhesion decreased. However, due to a decrease in the content of Sumi ink, an increase in internal resistance and a decrease in long-term stability were observed. These results suggest that the hydrophobic properties of PTFE and the conductive and physically stable properties of Sumi ink can be benefited by adjusting the ratio of them in the cathode binder blend.

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Not applicable.

Conflict of interest:

The authors declare that there is no competing interest.

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