

Electricity generation and winery wastewater treatment using silica modified ceramic separator integrated with yeast-based microbial fuel cell

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Abstract

The ceramic separator has been interested in low-cost alternative proton exchange membranes in a microbial fuel cell (MFC). In this study, the silica-modified ceramic separator has been integrated with the yeast-based MFC for electricity generation and phenol treatment from the winery wastewater. The 30% (w/w) silica powder was mixed with the 70% (w/w) natural clay. The modified ceramic plates (0.2, 0.5, and 1.0 cm of thickness) were prepared at 680°C and used for MFC operation. As an anolyte, synthetic winery wastewater (2,000 mg COD/L and 100 mg/L phenol) with 5% (v/v) ethanol was used. The ethanol-tolerant yeast *Pichia* sp. ET-KK was used as an anodic catalyst. The results showed the maximal power density of 0.212 W/m² and phenol removal of 95.05% were reached from the 0.2-thick ceramic plate integrated MFC. This study demonstrated that the silica-modified ceramic separator has a high potential for enhancing electricity generation in the yeast-based MFC.

Keywords: Microbial fuel cell; electricity generation; phenol removal; winery wastewater; ceramic membrane

1. Introduction

Winery wastewater is one of the complex wastewaters that mainly contains various organic compounds such as sugar, organic acid, glycerol, ethanol, esters, phenolic compounds, and ethanol. It contains 320–49,105 mg/L of chemical oxygen demand (COD), 10–415 mg/L of total nitrogen (TN), and 2.1–280 mg/L of total phosphorous (TP) affecting the groundwater pollution where it is discharged [1,2]. Rodriguez-Caballero et al. stated that winery wastewater contains a high ethanol concentration [3] and some previous studies showed that ethanol establishes up to 90% of the COD content in winery wastewater. In wastewater, the contaminated ethanol can inhibit the growth of various microbe and increase the water pollutant. Consequently, the ethanol tolerant yeast has been interested in being used under high ethanol-containing conditions [4].

Microbial fuel cells (MFC) convert the chemical energy in the organic materials to electrical energy throughout the microbial metabolism without combustion. It can be applied for wastewater treatment and simultaneous electricity generation. The study of Hou et al. showed that the MFC coupled with phenol degrading microbe has a high potential for phenol degradation from the wastewater as the result of the decreasing

toxic pollutant from the wastewater [5]. Nevertheless, the yeast-based MFC has also generated less electrical energy than the bacterial-based MFC. Boas et al. on the other hand demonstrated that the yeast *Zygosaccharomyces bailii* can produce flavins, an electroactive metabolite that can play an important role in ethanol degradation [6]. On the other hand, the yeast-based MFC can be applied for electricity generation and high-value by-product production [7,8]. The performance of MFC is governed by its components and operating conditions. Proton exchange membranes, such as Nafion, play an important role in proton transport in a dual-chamber MFC. However, it has several negative outcomes, like high cost, high oxygen diffusion, and anolyte crossover [9]. The ceramic separator has been developed as a low-cost alternative separator for application in an MFC. The previous study showed a high energy conversion efficiency and constant power performance from the ceramic separator MFC [10].

Raychaurhuri et al. showed that the membrane containing silica exhibits the improving performance of proton diffusion and enhances electricity generation [11]. Moreover, previous studies have shown various materials that have been used for modified ceramic plates, such as rice husk ash [12], chitosan [13], iron oxide [14], polypropylene [15], and others.

In this study, the silica-modified ceramic plates were developed for integration with MFC for winery wastewater treatment (phenol removal under ethanol-contaminant wastewater) and electricity generation with constant power performance.

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2. Materials and Methods

2.1. Microbe

The ethanol-tolerant yeast *Pichia* sp. ET-KK (TSU_MFCY001) was gained from the Microbial fuel cell & Bioremediation Laboratory, Faculty of Science, Thaksin University. It was maintained on the sucrose-yeast agar (10% (w/v) sucrose, 1% (w/v) yeast extract, and 15% (w/v) agar) and kept at 4°C until being used in this experiment (figure 1).

For liquid culture preparation, the colony of yeast was inoculated into a sucrose-yeast medium (10% (w/v) sucrose and 1% (w/v) yeast extract) via the stainless wire loop and incubated at a room temperature for 5 days with 150 rpm shaking.



Fig. 1. The colony of *Pichia* sp. ET-KK

2.2. Ceramic plate preparation

The silica-modified ceramic plate was prepared according to the modified method of Raychaurhuri et al. [16]. Briefly, the 70% (w/w) of locally ceramic soil was mixed with the 30% (w/w) of silica powder. The modified ceramic plate was molded to prepare with various thicknesses of 0.2, 0.5, and 1.0 cm. The ceramic plates were dried at 80°C for 7 days and baked in a muffle furnace at 680°C for 30 minutes.

2.3. MFC operation

Figure 2 show the construction of dual chamber in which the 7.50 cm² of graphite plated electrodes were prepared according to Kim et al. [17]. The anode and cathode chamber were made from the 50 mL cell culture flasks. The copper wire was used to connect the electrodes. The ceramic separators were inserted between the anode and cathode chamber. The 1 M KMnO₄ solution was used as a catholyte. The synthetic winery wastewater (2,000 mg/L of total COD and 100 mg/L of phenol) with 5% (v/v) ethanol was prepared according to Welz and Rose-Hill [18].

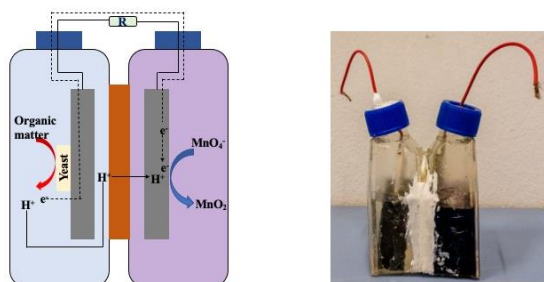


Fig. 2. The dual-chamber MFC used in this experiment

The 10% (v/v) ethanol tolerant yeast *Pichia* sp. ET-KK (1×10^8 cell/mL) and 90% (v/v) of the synthetic winery wastewater were added to the anode chamber. Here, a 1,000-Ω resistor was connected and incubated for 5 days to immobilize a yeast on the electrode surface.

For operation, the anolyte was fed-out and replaced with 40 mL of synthetic winery wastewater. The opened-circuit voltage (OCV) was collected every 6 hours for 5 days. The closed-circuit voltage (CCV) was monitored between 1 – 5,000 Ω external resistances and the electrochemical properties were calculated according to Ohm’s law [19].

The CCV was determined at the stationary phase of the bio-electrical cycle. The current, power, current density and power density were calculated as follows:

$$I = V/R \quad (1)$$

$$P = IV \quad (2)$$

$$CD = I/A \quad (3)$$

$$PD = P/A \quad (4)$$

where I is the current (A), V is the CCV (V), R is the external resistance (Ω), P is the power (W), CD is the current density (A/m³ or A/m²), PD is the power density (W/m³ or W/m²), and A is the working volume (m³) or electrode area (m²).

2.4. Phenol removal

The winery wastewater (influent and effluent) was collected from the anodic chamber of the dual-chamber MFC. The wastewater was filtered through the filter paper (Whatman no.1) using a vacuum pump. The filtrate was measured the phenol content using a colorimetric method according to Chaijak et al. [20]. The Folin-Ciocalteu reagent was used, and the absorbance of the reaction was monitored at 760 nm. The phenol removal was calculated.

3. Results and Discussion

The maximal OCV of 790.0±5 mV was gained from the yeast-based MFC with the 0.2 cm of thickness silica modified ceramic separator, followed by the 0.5 cm and 1.0 cm of the thickness of 730.0±5 mV and 480.0±10.0 respectively. The thinnest membrane can produce the highest voltage owing to it providing the shortest distance for proton transportation. The results are displayed in figure 3.

According to Ohm’s law, the MFC with high voltage potential can generate high current and power. The highest CD of 29.500±1.000 A/m³ (1.573±0.000 A/m²) and the highest PD of 3.980±0.150 W/m³ (0.212±0.000 W/m²) were generated from the 0.2 cm thick modified ceramic plate. While, the internal resistance of the yeast-based MFC of 500 Ω was presented in the polarization curve (figure 4).

However, the 0.5 cm thick modified ceramic plate produced the highest CD of 1.33±0.10 A/m³ and the highest PD of 0.03±0.00 W/m³ (figure 5). Figure 6 shows the polarization curve of the dual-chamber MFC with a 1.0 cm thick ceramic plate. The maximum CD of 0.50±0.00 A/m³ and the maximum PD of 0.0005±0.0000 W/m³ were generated. Similar with the study by Khalili et al., the result indicated that the thinnest

ceramic separator could provide the highest electricity generation by the MFC [21].

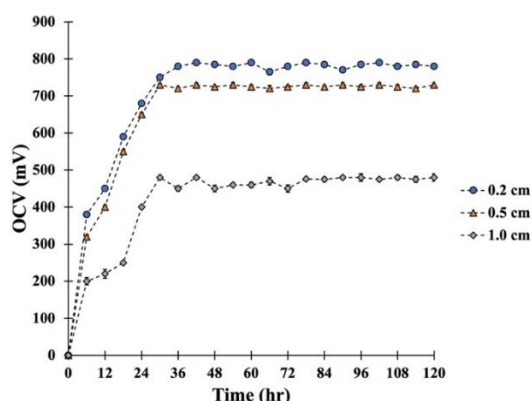


Fig. 3. The OCV of the dual-chamber MFC with different thicknesses of modified ceramic plate

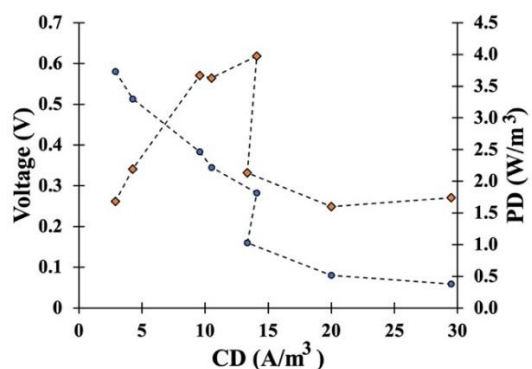


Fig. 4. The polarization curve of dual-chamber MFC with 0.2 cm thickness ceramic plate

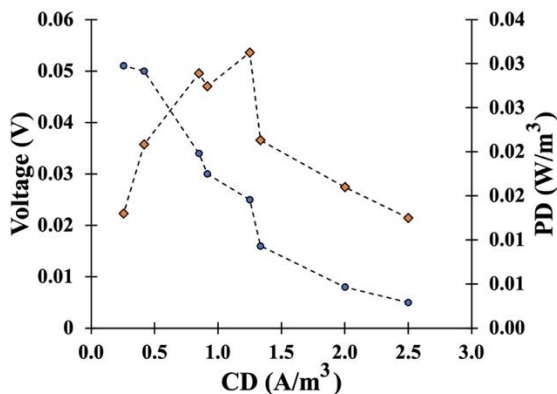


Fig. 5. The polarization curve of dual-chamber MFC with 0.5 cm thick ceramic plate

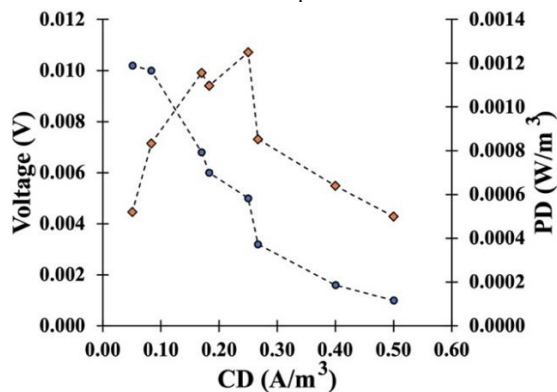


Fig. 6. The polarization curve of dual-chamber MFC with 1.0 cm thick ceramic plate

The phenol removal of 95.05±0.03%, 94.02±0.10%, and 90.10±0.30% were achieved from the yeast-based MFC with 0.2, 0.5, and 1.0 cm of silica-modified ceramic separator, respectively.

The MFC technology with an ability to convert contaminants to electrical energy can be enhanced with a low-cost ceramic separator. The ceramic is porous, thus allowing the direct exchange of protons [22]. The 5% (w/w) goethite modified ceramic separator integrated with MFC produced a maximal PD of 0.112 W/m² [23]. Table 1 depicts a comparison of ceramic-separator MFC. According to the data, the modified ceramic separator has been used as a proton exchange membrane in various wastewater sources. The data suggested that the modified ceramic membrane can provide a higher electricity generation than other ceramic membranes. Moreover, the previous study by Alftessi et al. showed that the silica sand ceramic separator can be used for the high efficiency of wastewater treatment and power generation [28].

Table 1. Review of ceramic separator MFC

MFC type	Ceramic material	Wastewater/ Microbe	PD (W/m ³)	Reference
Dual-chamber	Silica modified ceramic	- Winery wastewater - <i>Pichia</i> sp. ET-KK	3.980	This study
Dual-chamber	Montmorillonite modified ceramic	- Synthetic wastewater - Anaerobic mixed sludge	0.084	[23]
Dual-chamber	Fire clay ceramic	- Urine wastewater - Exoelectrogens	-	[24]
Dual-chamber	Nafion coated ceramic	- Synthetic wastewater - Sewage sludge	0.084	[25]
Dual-chamber	Native clay ceramic	- Domestic wastewater - Sewage sludge	0.48-20.18	[26]
Dual-chamber	Chitosan/Montmorillonite modified ceramic	- Domestic wastewater	0.229	[27]

Moreover, the MFC has been used for the brewery wastewater. In Liu et al., the winery wastewater was treated by the MFC integrated with Betaproteobacteria, which played an important role in electricity generation. The maximal voltage of 0.63 V was generated [29]. On the other hand, the maximal power density of 0.29 W/m³ was caused when the initial COD concentration was 1-10 gCOD/L [30].

Vilas Boas et al. confirmed that the yeast *Zygosaccharomyces bailii* can be used for ethanol-contaminated winery wastewater treatment using an H-type dual-chamber MFC system. However, the power density of yeast-based MFC has not been analyzed [31]. Table 2 showed the comparison of winery wastewater treatment using the MFC.

Various processes have been applied for phenol removal from industrial and agricultural wastewater. The results of Luo et al. indicated that the MFC can enhance phenol degradation and electricity generation [37]. On the other hand, the cube-type dual-chamber MFC has been used for phenol removal and electricity generation from phenol-acetone wastewater. The maximal output voltage and phenol removal of 250 mV and

95.12% were gained [38]. Moreno et al. showed the batch-operated MFC with graphite-granule electrode can produce the maximum power output of 0.78 W/m³ when the phenol removal was gained [39]. Table 3 presents the comparison in the use of MFC for phenol removal and electricity generation from the wastewater 3.

Table 2. Reviews of winery wastewater treatment using MFC

MFC type	Initial COD (g/L)	Power output (W/m ³)	Power output (W/m ²)	Reference
Dual-chamber	2.00	3.980	0.212	This study
Dual-chamber	-	-	0.275	[32]
Dual-chamber	6.85	-	0.420	[33]
Dual-chamber	1.00	-	0.105-0.465	[34]
Single-chamber	140-230	-	0.111-0.262	[35]
Dual-chamber	-	0.014	-	[36]

Table 3. Reviews of phenol removal using MFC

MFC type	Wastewater	Phenol removal (%)	Reference
Dual-chamber MFC	Winery	95.05	This study
Photoelectrocatalytic combined MFC	Refractory	96.00	[40]
Dual-chamber MFC	Industrial	41.00	[41]
Single-chamber MFC	Synthetic	78.80	[42]
Single-chamber MFC	Synthetic	94.50	[43]
Dual-chamber MFC	Synthetic	81.97	[44]

The results indicated that the dual-chamber MFC with the silica-modified ceramic separator can be successfully used for wastewater treatment in terms of phenol removal under microbial inhibitor (ethanol) contaminated conditions and electrical power generation.

As shown in table 2, this study provides a higher power output than other studies carried out in the dual-chamber and single chamber MFC owing to the thin modified ceramic separator membrane where the winery wastewater has been used as a substrate.

Moreover, the *Pichia* sp. ET-KK indicated that it had a high potential for phenol removal when being used for whole-cell biocatalyst in the anodic chamber (in table 3) owing to it can be tolerant to contaminated ethanol in the effluent.

4. Conclusion

This study has demonstrated that the 30% (w/w) silica powder modified ceramic plate has a high potential for use as an MFC proton exchange membrane. Furthermore, the modified ceramic separator MFC combined with the ethanol tolerant yeast was successfully used for phenol degradation from winery wastewater as well as electricity generation. This MFC system can be developed for the use with various wastes and scaled up owing to its low structural cost. However, the larger scale will be studied in further work.

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