Performance of Microbial Reverse-Electrodialysis Cells for Power Generation at Different External Resistance

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ABSTRACT: In this study, the effect of external resistance on the microbial reverse-electrodialysis cell (MRC) performance using organic-rich wastewater as an electron donor was examined. The optimum of external resistance was determined to be 300Ω. In such condition, the power density of 1.53 ± 0.198 W/m², substrate removal of 52 ± 2.3%, Coulombic efficiency of 70 ± 2.6%, energy recovery of 3.0 ± 0.4%, and energy efficiency of 53 ± 7.1% were obtained in the MRC. The differences in power density at different external resistances were mainly due to the changes in internal resistance and ion flux efficiency in the MRC. The external resistance affected substrate removal and Coulombic efficiency through the length of batch cycle time, and current density exchanged as well as the Tafel slope. Furthermore, the proper external resistance applied to the reactor created high power production; thus, high energy efficiency and energy recovery were achieved. These results demonstrated that selecting proper external resistance was an essential key for a successful MRC operational.

KEYWORDS: Renewable energy, bioelectrochemical systems, salinity gradient energy, energy efficiency, energy recovery

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Introduction

Bioelectrochemical systems (BESs) is considered as one of the most promising technologies that can be used to extract the energy in organic/inorganic wastes as electrical power or chemicals.1 Over the last decade, microbial fuel cells (MFCs) have gained extensive attention from researchers, representing over 75% publication of the BESs research area in 2016.2 Microbial fuel cells are unique devices of BESs that can convert the chemical energy stored in biodegradable materials into direct electrical power while simultaneously treating wastewater.3 In this system, the direct electrical power is generated as a result of simultaneous processes oxidation of biodegradable materials by exoelectrogens and oxygen reduction in the anode and cathode electrodes, respectively. However, in the MFC pilot scale using organic materials of wastewater as an electron donor, to date, the highest power densities reached approximately 7 to 60 W/m² as normalized by volume of the reactor.4-6 The operational strategy with series mode has been done to improve the performance of the reactor in terms of voltages and power densities.7 In such a condition, the reactors have a significant performance in treating wastewater; however, this is not the case for power production.

Salinity gradient power (SGP) is a method to convert the Gibb free energy from the mixing of water with different salinity concentrations into electrical power.8 Reverse-electrodialysis cell (RED), as a representation of SGP is the most recommended technology to harness the energy from salinity gradients.9 In such a system, the cell comprises several pairs of anion- and cation-exchange membrane, which stacked alternatingly each other and situated in between an anode and a cathode chamber. When the solutions with different salinity concentrations were introduced to the flow channel between these membranes, electrochemical potential energy was generated across the membranes as a result of selective ion transport from the solutions.10 The corresponding potential energy is theoretically around 0.1 to 0.2 V for each pair of membranes for 15 to 150 salinity ratios of seawater and river water.11 This potential can rise by increasing the number of cell pairs; however, efficiency and operational power production become lower than expected due to the hydrodynamic and ohmic losses as well as overpotential.12 For instance, in the pilot applications (125-500 cell pairs), the power output of the RED stack reached 700 W and decreased to 330 W. It occurred when the system operational shifted from the artificial brine and brackish water to real solutions with the expected energy output of 1 kW.13 Hence, a different approach is needed to optimize the RED’s potential as a power source.

Recently, several studies have been demonstrated to prove that the combination of a small number of RED stacks with an MFC could synergistically enhance the potential electrical power generation compared with the individual systems.14,15 In such a system, when 5 membrane pairs of RED stacks were
used for the electrical power production, the maximum voltage and power density reached 1.3 V and 4.3 W/m², respectively. The values were higher than those obtained from MFC alone (0.5 V, 0.7 W/m²). Confirmed by further research, when a microbial reverse-electrodialysis cell (MRC) used ammonium bicarbonate in the RED stack and domestic wastewater as an electron donor, the result showed that even with a single pair of RED stacks, the performance of reactor significantly improved. In addition, the energy efficiencies (35%-42%) and energy recoveries (10%-21%) of the corresponding system were relatively comparable with the RED systems (14%-35% and 9%) than MFCs, as those 2 figures for MFCs were generally low (<7.2% and <6.5%).

In this study, the effect of external resistance on the MRC reactor performance using organic-rich wastewater as an electron donor was examined. Because it has high organic content, food waste leachate as a representation of organic-rich wastewater is highly potential to be used as a substrate for the microorganism in the MRC reactor. Preliminary studies were conducted in the fed-batch mode using a different number of membrane pairs to determine the optimum cell pairs. In further experiments, the reactor was operated under various external resistances. The reactor performance was evaluated by measuring power density, substrate removal, Coulombic efficiency (CE), energy recovery, and energy efficiency.

**Materials and Methods**

**Reactor setup**

The MRC reactor was constructed as previously described with minor modifications. The reactor consisted of an anode chamber, a cathode chamber, and a small number of the RED stack (Figure 1). The anode and cathode chambers were constructed using poly(methyl methacrylate) with a working volume of 30 mL. A heat-treated graphite fiber brush, 3 cm in diameter and 2.5 cm long, was used as the anode electrode following the method from a previous study and placed vertically in the middle of anode chamber. The wet-proofed (30%) carbon cloth (1071 HCB; AvCarb, United States) was used as a cathode electrode with a modification of 4 polytetrafluoroethylene diffusion layers on air-side and a Nafion binder mixed with platinum catalyst on solution-side. The Ag/AgCl reference electrodes were assembled on each of the chambers to measure the electrode potentials. The RED stack consisted of 3 cell pairs (except as noted) made of alternation anion and cation exchange membrane (Selion AMV and CMV, Asahi Glass, Japan). Each membrane had an essential area of 12.25 cm² (3.5 cm × 3.5 cm). These membranes were separated by silicone gaskets and a nylon mesh spacer with a thickness of 0.18 mm, forming flow channels for alternating high-concentration (HC) and low-concentration (LC) solutions.

**Solutions**

The anode was enriched with exoelectrogens in a single chamber of an MFC at fix external resistance 1000Ω and fed with 1 g/L of sodium acetate (CH₃COONa). The MFC reactor was fed with gradual increased concentrations of NaCl (0.10-0.35 M) to acclimate the exoelectrogens to chloride ions. The adapted anode was transferred to the MRC after reproducible maximum voltages on the MFC were stable at least for 3 consecutive batch cycle.
The anolyte solution was prepared by dissolving the following chemicals in 1 L of deionized water: 1.90 g CH₃COONa, except as noted, 0.31 g NH₄Cl, 0.13 g KCl, 6.60 g NaH₂PO₄·2H₂O, 8.19 g Na₂HPO₄, as well as 12.5 g minerals and vitamins. A 1.0 M NaHCO₃ solution was used as catholyte (conductivity ~5.06 mS/cm; pH ~8.03). The HC solution was 35.0 g/L NaCl (~53.6 mS/cm), whereas the LC solution was 0.70 g/L NaCl (~1.39 mS/cm), creating a salinity ratio of 50. Leachate, as a representation of organic-rich wastewater, was used as an electron donor (substrate). The characteristic of leachate is as follows: soluble biochemical oxygen demand (COD) = 2100-2300 mg/L; nitrogen (NH₃) = 130 mg/L, nitrate (NO₃⁻) = 6.5 mg/L, phosphate (orthophosphate) = 86.4 mg/L, conductivity = 13.91 mS/cm, and pH = 7.15.

Reactor operation
Preliminary experiments were conducted to determine the optimum membrane pairs in the RED stack. The MRC reactor was operated using different numbers of the RED stack (1, 3, and 5 cell pairs). The reactor was fed with 1.9 g/L of CH₃COONa and connected to 1000Ω external resistor. In further experiments, the MRC reactor was operated with different external resistances (150Ω, 300Ω, 650Ω, and 1000Ω). Food waste leachate was used as a substrate in the anode chamber and was replaced when the generated current decreased to less than 0.5 mA. In the RED stack, HC and LC were used as a feed solution and continuously supplied at a fixed rate of 1.2 mL/min during reactor operation. All of the MRC experiments were done in a constant room temperature (26 ± 1°C).

Experimental analysis and calculations
The potential difference (U) between anode and cathode was monitored and recorded every 5 minutes using a voltage recorder (VR-71; T&SD Corporation, Japan) connected to a personal computer. The current was determined as i = U/R, and the current density was normalized by the cathode projected surface area (7 cm²). The maximum power density was obtained from the polarization curve, which was determined using variable external resistance (5-40,000Ω) at 20-minute intervals. The effluent anolyte, catholyte, HC, and LC solutions were measured for pH and conductivity using a pH meter (Orion Three-star, Thermo Scientific Co., United States) and a conductivity meter (Orion Three-star, Thermo Scientific Co., United States). The COD of influent and effluent anolyte was measured according to the standard method (DR-2800; Hach Company, Indonesia). Ion flux efficiency (ηᵢᵣᵢᵢ), CE, energy recovery (rₑ), and energy efficiency (ηₑ) were described as follows:

The chemical activity of ion (αᵢ) was determined by multiplying the chemical concentration in molality by the activity coefficient (γᵢ). The extended Debye-Huckel equation was used to determine the activity coefficient:

The Debye-Huckel constants are \( A = 0.5085 \text{ kg}^{1/2}/\text{mol}^{1/2} \) and \( B = 0.3282 \text{ kg}^{1/2}/\text{A}\cdot\text{mol}^{1/2} \). The ion size parameter (\( αᵢ \)) was 0.78 Å for both sodium (Na⁺) and chloride (Cl⁻), whereas \( Kₐₙ \) and \( Kₙₚ \) are 0.105 and -0.009 kg²/mol², respectively. \( I \) is the ionic strength in molality, \( m \) is the molar concentration. The equation is valid for a NaCl solution up to 1 molality.

The ion flux efficiency (\( \eta_{iᵢᵢ} \)) is a ratio of the current generation in the MRC to the ion flux through the ion-exchange membrane and determined as:

where \( N_{Mᵢ} \) is the number of cell pairs, \( i \) is the current generation in MRC (A), \( Q \) is the flow rate of stack (L/s), and \( \epsilon \) is the molar concentration of NaCl (mol/L).

The CE was calculated based on the COD removal and the number of coulombs produced during the reactor operation as previously described:

where \( 8 \) is the number of electrons for substrate oxidation, \( i \) is current produced during the operational time (A), \( Q_{an} \) is the volume of the anolyte (L), and ΔCOD is the change of substrate during operational time (g-COD).

The energy recovery (\( rₑ \)) is the ratio of power production to the total energy provided in the MRC, and it was defined as:

where \( P \) is the power produced from the MRC (W), ΔHᵢ is the heat of combustion of the substrate (kJ/mol), \( nᵢᵢ \) is the molar concentration of substrate at the initial of operational time, \( tᵢᵢ \) and \( Xᵢᵢ \) is the flow rate of feed stack (L/s), and \( \epsilon \) is the molar concentration of NaCl (mol/L).

Energy efficiency (\( ηₑ \)) is the ratio of the power production to theoretically extractable energy provided by MRC, and it was determined as:

\[ \etaₑ = \frac{\eta_{iᵢᵢ}}{\eta_i} \]

\[ \eta_i = \frac{\Delta H_i nᵢᵢ}{P} \]

\[ rₑ = \frac{P}{\Delta H_i nᵢᵢ / tᵢᵢ + Xᵢᵢ} \]

\[ Xᵢᵢ = 2RTQ \left( \epsilon_{ᵢᵢ} \frac{\alphaᵢᵢ}{\alphaᵢᵢ} \right) \]
\[ \eta_E = \frac{P}{\Delta H_c \left( n_i^{in} - n_i^{out} \right) / \eta_B + X^{in} - X^{out}} \lim \quad (6) \]

where \( n_i^{out} \) is the mol concentration of substrate at the end of operational time, \( X^{in} \) is the theoretical energy (W) of the salinity gradient energy remaining in seawater and river water effluents.

A Tafel plot test was conducted to investigate bioelectrochemical kinetics of anode biofilm electrode using a potentiostat (DY2300; Digi-Ivy, Inc., Austin, TX, USA) in 3-electrode conventional cells. The anode, saturated calomel electrode, and the cathode served as the working electrode, reference electrode, and counter electrode, respectively.

**Results and Discussion**

**Performance of MRC using different numbers of membrane cell pairs**

The reactor was operated under fed-batch mode with different membrane cell pairs (1 MP, 3 MP, and 5 MP) and fixed external resistance (1000 Ω). As shown in Figure 2, the cell current tended to increase as the number of membrane cell pairs increased. The MRC with 5 MP produced a maximum cell current of 1.12 ± 0.09 mA, which was 1.2- and 1.9-fold higher than that of obtained using 3 MP (0.9 ± 0.02 mA) and 1 MP (0.60 ± 0.03 mA), respectively.

The polarization curve was measured by varying an external resistance in the MRC reactor to determine the maximum power density. It was observed that the maximum cell potentials produced during the polarization test increased as the number of membrane pairs increased (Figure 3A). Furthermore, adding more membrane pairs increased the maximum current density from 0.51 mA/cm² (1 MP) to 0.85 mA/cm² (5 MP). Meanwhile, consistent with previous research,²⁵ the cell potential decreased with increasing current density due to the activation losses, ohmic losses, and concentration losses during MRC operation. The power density of the MRC reactor was inclined to follow the previous results of maximum cell potentials (Figure 3B). The MRC with 1 MP produced the maximum power density of 1.19 W/m² as normalized by 7 cm² of cathode area or 30 W/m³ as normalized by anolyte volume. Meanwhile, the MRC with 3 MP produced the maximum power density of 2.31 W/m² (58.3 W/m³). Furthermore, the MRC with 5 MP achieved higher power densities, with a maximum power density of 2.75 W/m² (69.4 W/m³). However, the increase in power density as a result of an increase in the number of membrane pairs from 3 to 5 (0.44 W/m²) was smaller than that obtained when increasing membrane pairs from 1 to 3 (1.12 W/m²), suggesting that further increase in membrane pairs did not significantly increase power density.

These values of power density were relatively higher compared with power density produced by the MFC with the same operational conditions (reactor working volume and electron donor). Based on the previous study,²⁶ 2 chambers of MFC produced 0.45 W/m² of power density. Subsequently, Choi and Ahn²⁷ also reported that the MFC reactor produced 0.77 W/m³ of power density with food waste leachate as substrate and 24 mL of working volume reactor. The research from Moharir and Tembhurkar²⁸ also showed relatively the same result, claiming that the MFC reactor produced 0.29 W/m³ of power density. These results confirmed that the high power density produced in the MRC reactor is
due to the salinity energy created from the small number of membrane pairs (RED stack).

According to the above results, 3 MP was selected as the optimum operational condition as the increase in the number of membrane pairs to 5 MP did not substantially increase the reactor performance. Therefore, in further experiments, the reactor was operated using 3 MP with different external resistors (150Ω, 300Ω, 600Ω, and 1000Ω).

**Effect of external resistance on power generation**

In this section, the MRC reactors were operated for several cycles with food waste leachate as a donor electron and were connected to different external resistors (150Ω, 300Ω, 600Ω, and 1000Ω). Figure 4A shows that the power density increased from 0.82 ± 0.038 W/m² to 1.53 ± 0.198 W/m² as the external resistance decreased from 1000Ω to 300Ω. Furthermore, the power density significantly decreased to 0.34 ± 0.067 W/m² as the external resistance decreased to 150Ω. These results indicated that when external resistance dropped, power density would increase as a result. However, a fall in power density occurred when the external resistance exceeded its optimum point. The differences in power density produced were likely due to the changes in the internal resistance of the MRC reactor. Based on the slope of the polarization curve, the total internal resistance of the reactor was 544.9Ω, 507.7Ω, 372.0Ω, and 401.1Ω for the MRC with the external resistance of 1000Ω, 600Ω, 300Ω, and 150Ω, respectively. The MRC reactor with the external resistance of 300Ω achieved higher power production, compared with the other reactors, as the total internal resistance in this reactor was close to the value of its external resistance. This result was consistent with the previous study, it is stated that the reactor’s power density was maximized when the external resistance connected to it was equal or close to the total internal resistance.

To determine the influence of external resistance to the salinity energy created by the RED stack, the ion flux efficiency was measured. Ion flux efficiency is a ratio of the current generation in the MRC reactor to the ion flux through the ion-exchange membrane. Based on the concentration change in the LC effluent, the ion flux efficiency was 82 ± 2%, 88 ± 3%, 94 ± 6%, and 90 ± 5% with the average currents produced were 0.74 ± 0.038 mA, 1.04 ± 0.098 mA, 1.90 ± 0.198 mA, and 1.13 ± 0.067 mA for the MRC reactor with the external resistance of 1000Ω, 600Ω, 300Ω, and 150Ω, respectively (Figure 4B). These results showed that the ion flux efficiency increased (from 82 ± 2% to 94 ± 6%) with a decrease in the external resistance (from 1000Ω to 300Ω). This increase led to a rise in the cell current produced in the MRC reactor (0.74 ± 0.038 mA to 1.90 ± 0.198 mA). The ion flux efficiency dropped to 90 ± 5% as the external resistance fell to 150Ω; thus, the cell current produced decreased to 1.13 ± 0.067 mA. When the reactor was operated with low external resistance, the reactor generated a higher current due to the more frequent electron transfer from anode to an electrode, which then would support faster cathode reaction and high electrogenic activity. According to the above results, it can be concluded that the external resistance affected the ion flux efficiency through the magnitude of current produced in the reactor.

**Effect of external resistance on CE and COD removal**

Coulombic efficiency is the ratio of the total recovered coulombs to the theoretical amount of coulombs provided by the substrate. Generally, the CE value is influenced by substrate removal (as the soluble COD removal) in the reactor. Based on the calculation of initial and effluent of soluble COD in the MRC reactor, the substrate removal reached up to 73 ± 2.8%, 62 ± 2.1%, 52 ± 2.3%, and 41 ± 2.6% for the reactor with external resistance of 1000Ω, 650Ω, 300Ω, and 150Ω, respectively (Figure 5A). It was observed that the COD removal increased as the external resistance increased. The higher COD removal was likely a result of the longer batch cycle time of the
MRC reactor with 1000 Ω than those of other reactors. The longer the cycle time of the reactor, the more substrate was consumed by the bacteria; thus, high-soluble COD removal in the reactor was achieved.

The CE value of the MRC reactor at different external resistance can be seen in Figure 5A. The CE obtained from the MRC reactor with 300 Ω (70 ± 2.6%) and 150 Ω (70 ± 2.9%) was higher than that of the reactor with 1000 Ω (34 ± 2.7%) and 650 Ω (60 ± 2.3%). The higher CE value on the MRC reactor with 300 Ω and 150 Ω was considered to be a result of the shorter batch cycle time. Low external resistance would bring a high electron transfer rate in the reactor; thus, it probably resulted in a shorter batch cycle time. The batch cycle time would affect the CE value as the reactors were started with a mixed culture inoculum. The substrate removal by the non-exoelectrogen caused substrate loss to non-current-generating processes, which resulted in high COD removal and low CE.

To determine the electron transfer rate in the MRC reactor, the bioelectrokinetic of the anode was then analyzed using Tafel plots in terms of exchange current density (i₀) and anodic Tafel slope (ba) as shown in Figure 5B. Exchange current density is a parameter related to the bioelectrochemical activity of the anode at the equilibrium condition. Based on the Tafel plots shown in Figure 5B, the exchange current density of biofilm at 300 Ω MRC reached 23.2 mA/m², which was higher than those of other reactors (18.6 mA/m² and 12.0 mA/m² for the MRC reactors with 650 Ω and 1000 Ω, respectively). In terms of anodic Tafel slope (ba), the biofilm at the MRC reactor with 300 Ω revealed a lower value of 0.73 V/decade than the reactors with 1000 Ω and 650 Ω (0.93 V/decade and 0.84 V/decade, respectively). These results indicated that the reactor with lower external resistance could easily detach the electron from the biofilm into the anode electrode compared with the reactor with higher external resistance.

According to the results above, it can be concluded that external resistance affected the CE value of the MRC reactor through the length of batch cycle time and bioelectrokinetic anode value (exchange current density and Tafel slope). When the MRC reactor was connected to a high external resistance, the reactor would have a longer batch cycle as the exchange current density was low; thus, large COD removal and low CE value were achieved (and vice versa).

**Effect of external resistance on energy recovery and energy efficiency**

The energy recovery (r_E) is the ratio of power production to the total energy provided in the MRC. Meanwhile, energy efficiency (η_E) is the ratio of the power production to theoretically extractable energy provided by MRC. The energy efficiency and energy recovery of the MRC reactor at different external resistance are shown in Figure 6. It was observed that the energy recovery increased from 1.7 ± 0.1% to 3.0 ± 0.4% as the external resistance decreased from 1000 Ω to 300 Ω. The energy recovery then significantly decreased to 0.6 ± 0.1% when the external resistance decreased to 150 Ω. Compared with another study with 3-membrane pairs of MRC conducted by Liu et al., energy recovery results from this study were significantly lower. This low energy recovery was likely due to the relatively high feed stack flow rate used here (1.2 mL/min compared with 0.80 mL/min) compared with the lower power produced in the reactor (1.03 mW
compared with 2.4 mW). The high feed stack flow rate resulted in higher energy input creating from the RED stack (34 mW compared with 24 mW). Moreover, Kang et al.\(^\text{10}\) reported that the MRC reactor with 5 mL/min of feed stack flow rate and 2.4 mW of power produced achieved 2.0% energy recovery, which was lower than the result of the recent study. In this case, this result proves that the feed stack flow rate significantly affected the energy recovery of the MRC reactor compared with the energy produced in the reactor. In this study, the value of energy efficiency mainly depended on the power production in the MRC reactor (P varied from 0.22 to 1.03 mW) as the summation of energy created from salinity gradient (X\(\text{in}\)) and substrate loading (\(\Delta H_c x n_{\text{in}}/t_b\)) was relatively constant (0.034 W). These calculations suggested that the MRC reactor with high power production would achieve high energy recovery.

Based on the calculation of energy recovered in the system compared with the energy available on the substrate, the energy efficiency of the MRC reactor tended to follow the trend of the energy recovery result. The energy efficiency increased from 35 ± 1.6% to 53 ± 7.1% as the external resistance decreased from 1000 Ω to 300 Ω. The energy efficiency then significantly decreased to 11 ± 2.4% when the external resistance decreased to 150 Ω. The higher energy efficiency was observed in the MRC reactor with the external resistance of 300 Ω, which was 1.5, 1.3, and 4.8 times larger than the MRC reactors with the external resistance of 1000 Ω, 650 Ω, and 300 Ω, respectively. Similar to energy recovery, the value of energy efficiency in the reactor mainly depended on the power production compared with the reactor input (based on the entering minus leaving) to the reactor. The power production in the reactor varied from 0.22 mW to 1.03 mW, while the energy input was relatively constant (1.30 W). Thus, the relatively higher power production of the reactor with external resistance of 300 Ω resulted in larger energy efficiency compared with other reactors.

According to the above results, it can be concluded that the external resistance affected the value of energy efficiency and energy recovery through the magnitude of power production in the reactor. When the reactor was connected to the proper external resistance, the reactor created high power production; thus, the high energy efficiency and energy recovery were achieved.

**Conclusions**

The applied external resistance was optimized to determine its effect on the MRC reactor performances. Based on the evaluation of the reactor performance, the optimum condition was achieved when 300 Ω of external resistance was applied. In such conditions, the power density of 1.53 ± 0.198 W/m², substrate removal of 52 ± 2.3%, CE of 70 ± 2.6%, energy recovery of 3.0 ± 0.4%, and energy efficiency of 53 ± 7.1% were obtained in the MRC reactor. The external resistance affected the power density as it changes the internal resistance and ion flux efficiency of the MRC reactor. Improper selection of external resistance would lead to high losses in power output. When the MRC reactor was connected to a high external resistance, the reactor would have a longer batch cycle as the exchange current density was low; thus, large COD removal and low CE value were achieved (and vice versa). Moreover, the proper external resistance applied to the reactor created high power production, which resulted in high energy efficiency and energy recovery. Therefore, this study shows that selecting proper external resistance was an essential key for a successful MRC reactor operational.

**Author Contributions**

AJE and SH conceived and designed the research and analysed the data and wrote the first draft of the manuscript. SS, BSR, CP, and JYP made critical revisions and approved the final version. All the authors contributed to the writing of the manuscript, revisions, and agree with manuscript results and conclusions. All authors reviewed and approved the final manuscript.

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