

Communication

Toward Optimization of Wood Industry Wastewater Treatment in Microbial Fuel Cells—Mixed Wastewaters Approach

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Abstract: Microbial fuel cell (MFC) has the potential to become a promising sustainable technology of wastewater treatment. Usually, the investigations on MFCs are aimed at maximized power production in the system. In this article, we focused on the optimization of wood industry wastewater treatment in MFC, in combination with municipal wastewater as a source of microorganisms. We investigated the influence of different external resistance (2000 Ω , 1000 Ω , 500 Ω , and 100 Ω) on power density and wastewater treatment efficiency (chemical oxygen demand (COD) removal) in 1-month MFC operation time. We found that the highest COD removal was for MFCs under $R = 1000 \Omega$ after 22 days of MFC operation, while the highest current density was obtained for the lowest applied resistance. The results imply that wastewater treatment parameters such as resistance and time of MFC operation should be a subject of optimization for each specific type of wastewater used, in order to maximize either wastewater treatment efficiency or power production in MFC. Thus, optimization of power production and COD removal efficiency in MFCs need to be run separately as different resistances are required for maximizing these two parameters. When COD removal efficiency is a subject of optimization, there is no universal value of external resistance, but it should be set to the specific wastewater characteristics.

Keywords: microbial fuel cell; wastewater treatment; wood industry wastewater

1. Introduction

Microbial fuel cell technology (MFC) enables sustainable treatment of various wastewaters, e.g., domestic [1], brewery [2], distillery [3], hospital [4], pharmaceutical [5], paper [6], wine [7], sewage sludge [8], mine drainage wastewater [9], or pulping wastewater [10]. Due to the presence of microorganisms with different metabolism (fermenters, methanogens, or electrogens), it is possible to decompose organic matter present in wastewater into simpler compounds that are used by electrogenic species for current production (Figure 1). An obvious gain from the MFC technology application is current produced in the system, but equally valuable is effective wastewater treatment that accompanies current production. Application of MFCs to wastewater treatment allowed not only for organic matter removal (measured by chemical oxygen demand-COD removal efficiency) but also for reducing content of nitrogen, phosphorus, and various, often toxic, metals (e.g., Cr, Cu, Fe, Co) in wastewater [11–14]. In case of some wastewaters, COD or metal removal efficiencies exceeded 90% what enables direct reuse of the MFC-treated water [15].

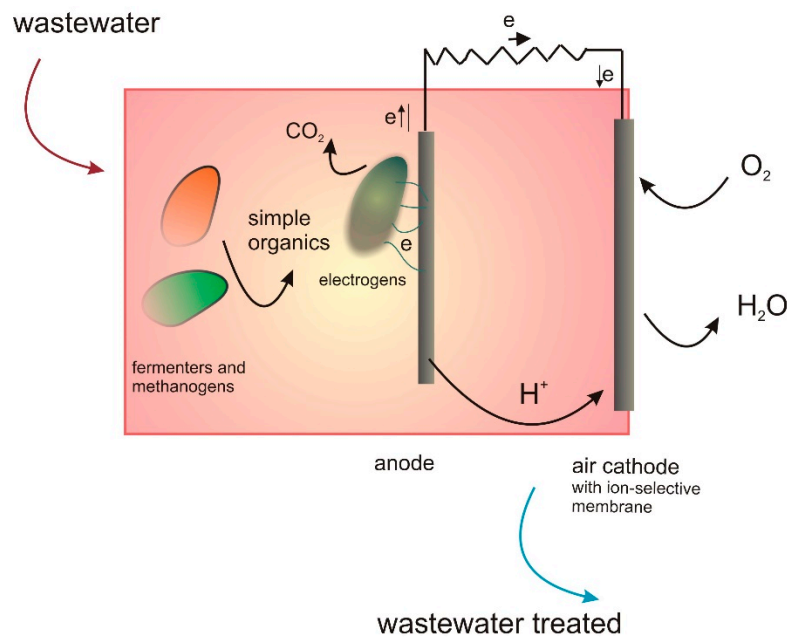


Figure 1. Schematic presentation of wastewater treatment accompanied by current production in air cathode single-chamber microbial fuel cell (MFC).

Though various industrial wastewaters were treated in MFCs, wood industry wastewater has been ignored as this sector is commonly thought of as environmentally benign and based on sustainable substrate—wood. Meanwhile, the plants producing wood-based panels (e.g., OSB—oriented strand board, PB—particleboard, MDF—medium density fiberboard) use between 0.1 and 1.5 m³ of water per 1 m³ of panel produced [16]. According to the Food and Agriculture Organization of the United Nations, global production of wood-based panels reached 416 mln m³ in 2016, which is a 24% increase compared to 2012 [17]. Thus, we can estimate that the global wood-based panel industry can generate up to ca. 600 mln m³ of wastewater every year. Using MFC technology would allow to treat wood industry wastewater in situ and reuse treated water in the producing plant. This approach reduces high costs of treatment of high-COD wastewater and decreases water consumption in the plant.

As much attention is paid to maximizing power production in MFCs, a lot is known about factors influencing power production in these systems. Power produced is dependent on the reactor geometry, surface area of anode and cathode (doubling cathode area can increase power production by 62%, but doubling anode area increases power only by 12%) [18], solution conductivity (the higher solution conductivity, the higher power produced), or COD load [15]. However, much less we know about factors that influence wastewater treatment efficiency. An exception is the hydraulic retention time, which seems to be the key parameter influencing wastewater treatment efficiency in the MFC of fixed architecture [19]. External resistance is a very important parameter that affects the amount of current produced in the system as it drives anode potential and influences the competition between electrogenic and non-electrogenic species [20,21]. However, the influence of external resistance on wastewater treatment efficiency and power production is not clear as discrepant results were obtained for various wastewaters (Table 1). Lyon et al. investigated domestic wastewater-fed MFCs where microbial community structure changed with external resistance so that the power was produced on a comparable level [22]. Applied various external resistances had no influence on maximum power and current produced in the reactors [22]. In MFC fed mixed wastewaters (brewery and domestic), external resistance had slight influence on power production, but the highest current was produced for the lowest applied resistance [23]. Simultaneously, the highest COD removal efficiency was observed for the highest external resistance. Zhang et al. reported that lowering external resistance resulted in an enhanced sulfide and vanadium removal rate in sulfide-rich wastewater [24]. The investigations showed

40% increase in V^{5+} reduction between the highest and the lowest resistance tested. An opposite effect was observed for cellulose degradation—increasing external resistance caused increase of cellulose degradation, which was a result of changing microorganisms' metabolism [21]. In another work, changing MFC external resistance in wide range from 2 Ω to 5000 Ω had no influence on COD removal efficiency [25].

In this work, we investigated the influence of different external resistance on wood industry wastewater treatment efficiency throughout MFC operation. The results of the investigations are needed for optimization of the procedures for wood industry wastewater treatment in MFC technology.

Table 1. Influence of external resistance on wastewater treatment efficiency and power production in MFCs fed various types of wastewater.

Wastewater Type	MFC Type	Power Density	Current Density	Wastewater Treatment Efficiency	Reference
distillery wastewater	dual chamber	R = 5000 Ω ; 280 mW/m ² R = 100 Ω ; 25 mW/m ²	R = 5000 Ω ; 5.9 mA/m ² R = 100 Ω ; 53 mA/m ²	not investigated	[26]
brewery wastewater:domestic wastewater (1:100)	dual chamber	R = 50 k Ω ; <3 mW/m ² R = 25 k Ω ; ca. 3 mW/m ² R = 10 k Ω ; ca.3 mW/m ² R = 1 k Ω ; ca. 10 mW/m ² R = 100 Ω ; 3 mW/m ²	R = 50 k Ω ; 8 mA/m ² R = 25 k Ω ; 13 mA/m ² R = 10 k Ω ; 25 mA/m ² R = 1 k Ω , 130 mA/m ² R = 100 Ω ; 274 mA/m ²	R = 50 k Ω ; Δ COD = 60% R = 25 k Ω ; Δ COD = 85% R = 10 k Ω ; Δ COD = 87% R = 10 k Ω ; Δ COD = 88% R = 0.1 k Ω , Δ COD = 92%	[23]
domestic wastewater fed acetate	single chamber	R from 10 Ω to 10 k Ω , ca. 30 mW/m ² for all applied resistances	R from 10 Ω to 1000 Ω , ca. 150 mA/m ² for all applied resistances	not investigated	[22]
high sulfide wastewater	dual chamber	not investigated	not investigated	V^{4+} increased from 116.2 to 177.3 mg/L when R decreased from 1000 Ω to 50 Ω	[24]
malodorous surface water	single chamber	not investigated	not investigated	R = 2 Ω ; R = 30 Ω ; R = 5000 Ω ; maximum Δ COD = 86% for all tested resistances	[25]
artificial brackish water	dual chamber	not investigated	R = 10 Ω ; 1.65 mA/m ² R = 100 Ω ; 1.75 mA/m ² R = 188 Ω ; 0.8 mA/m ² R = 1875 Ω ; 0.3 mA/m ² R = 4690 Ω ; 0.1 mA/m ²	no influence of external resistance on COD removal (maximum Δ COD = 30% vs control)	[27]
synthetic wastewater	dual chamber	R from 5 Ω to 200 Ω ; maximum 170 mW/m ² for R = 50 Ω	R from 5 Ω to 200 Ω ; maximum current 1.8 A/m ² for R = 5 Ω	R from 5 Ω to 200 Ω ; maximum nitrate removal rate for R = 10 Ω (54.80 \pm 0.01 gm ⁻³ d ⁻¹)	[28]
wine wastewater	dual chamber	R = 600 Ω ; 191 mW/m ² R = 900 Ω ; 80 mW/m ² R = 1400 Ω ; 40 mW/m ²	R = 600 Ω ; 360 mA/m ² R = 900 Ω ; 203 mA/m ² R = 1400 Ω ; 112 mA/m ²	R = 600 Ω ; Δ COD = 59% R = 900 Ω ; Δ COD = 41% R = 1400 Ω ; Δ COD = 50%	[7]
wood industry wastewater:domestic wastewater (1:1)	single chamber	R = 100 Ω ; 14 mW/m ² R = 500 Ω ; 25 mW/m ² R = 1000 Ω ; 112 mW/m ² R = 2000 Ω ; 175 mW/m ²	R = 100 Ω ; 443 mA/m ² R = 500 Ω ; 269 mA/m ² R = 1000 Ω ; 400 mA/m ² R = 2000 Ω ; 354 mA/m ²	R = 100 Ω ; Δ COD = 66% R = 500 Ω ; Δ COD = 65% R = 1000 Ω ; Δ COD = 94% R = 2000 Ω ; Δ COD = 80%	this work

2. Materials and Methods

2.1. MFC Construction and Operation

In all experiments, single-chamber MFCs prepared according to the design described by Logan et al. were used [29]. The MFCs volume was 28 mL, and anode was carbon fiber brush (2 cm long, 2.5 cm in diameter) and air cathode (of 7 cm² area). The cathodes were made of carbon paper with four PTFE diffusion layer, Pt as catalyst, and Nafion as a binder as described earlier [30]. The mixture of 1:1 v/v wood hydrothermal wastewater (WHTW) with municipal wastewater in 50 mM phosphate-buffered saline (PBS) was added to the MFC. WHTW was collected from the basin for wood hydrothermal treatment at 65 °C in a plywood manufacturing plant. The wastewaters were used as the source of bacteria and substrate without additional supplementation. MFCs were operated in fed-batch mode at 37 °C under various external load: 2000 Ω , 1000 Ω , 500 Ω , and 100 Ω . MFC fed WHTW in 50 mM PBS under constant R = 1000 Ω was used as the control. The solution was changed when voltage dropped below 50 mV [31]. When the solution was replaced, ca. 5 mL of solution was left in reactors for the next cycle of treatment. Current and power production were determined by measuring voltage (V) every 20 min across applied external resistance with the use of automated measuring system connected with a computer. Current (I) was calculated from Ohm's law ($I = V/R$), and power (P) was calculated as

P = IV. The current density and power density were normalized to the projected surface area of the cathode. The maximum current and power values were calculated from voltage peaks.

2.2. Chemical Characteristics of Wastewater and COD Removal Efficiency Determination

Fe was determined colorimetrically [32], and determination of chlorides was made with the use of potentiometric chloride ion-selective electrode. Determination of the amount of suspension was made according to standard method PN-72/C-04559 [33]. Determination of solids was made according to standard method PN-75/C-04541 [34]. Wastewater conductivity was measured by ELMETRON CC-105 conductivity meter.

The COD was determined by dichromate method that measured the oxygen equivalent of the amount of organic substances oxidized by potassium dichromate in a concentrated sulphuric acid solution [35]. Silver nitrate was added as a catalyst, and mercury sulfate was added to reduce interferences from the oxidation of chloride ions by dichromate.

The efficiency of the chemical oxygen demand removal (ΔCOD) was calculated from the formula:

$$\Delta\text{COD} = \frac{\text{COD}_0 - \text{COD}_t}{\text{COD}_0} \times 100\%$$

where COD_0 is the initial COD, and COD_t is the COD after time t .

The coulombic efficiency (CE) was calculated from the equation:

$$\text{CE} = \frac{M \int_0^t I dt}{n v F (\text{COD}_0 - \text{COD}_t)}$$

where M is the molecular weight of oxygen, I is current, F is Faraday's constant, n is 4—the number of electrons exchanged per mole of oxygen, and v is the anolyte volume.

3. Results and Discussion

3.1. Influence of External Resistance on Current Production in MFCs

WHTW is one of the wastewaters generated in plywood production during wood hydrothermal treatment. The chemical composition of WHTW differs between particular plants as is dependent on wood hydrothermal treatment nexus, e.g., wood species used, temperature, time of wood treatment, or frequency of changing water in the basin [36]. Generally, WHTW is acidic (pH usually ca. 5), highly polluted wastewater with COD ranging from 3000 mg/L to even above 11,000 mg/L. It contains products of wood hydrolysis as well as those extracted from wood, celluloses, hemicelluloses, and lignin. In these investigations, we used wastewater sampled from a basin where beech wood was treated hydrothermally at 65 °C. The chemical composition of WHTW used for investigations was as follows: COD = 4900 mg O₂/L, pH = 5.6, conductivity 4.35 mS, Fe 78 mg/L, chlorides 37 mg/L, dry residues 476 mg/L, total suspension 500 mg/L. We decided to use reactors of well-known architecture that had been employed in our previous studies as well in many other research groups. The approach allowed for reliable comparison of the obtained results and elimination of the influence of electrode and reactor configuration on MFC work parameters [37].

After introducing WHTW to reactors, a very low voltage was generated, which resulted in production of current density not exceeding 20 mA/m² throughout 1-month observation time (Figure 2—control). These observations were different from our previous work where we used WHTW sampled from another plant where wood hydrothermal treatment conditions were different (lower temperature and much longer time of wood treatment in the basin, COD = 3300 mg O₂/L) and then obtaining power production 318 mA/m² was possible [36]. This confirmed that wood industrial wastewater differs much between particular plants and its treatment in MFC needs optimization in each specific plant separately. Taking into account our previous findings, we mixed WHTW with municipal

wastewater that was earlier found to stimulate development of a new microbial consortium, enabling power production and treatment of WHTW [38]. In order to investigate the influence of the external resistance on current produced in MFCs, we introduced mixed wastewaters to the reactors kept under different external resistance: 100 Ω , 500 Ω , 1000 Ω , and 2000 Ω (Figure 1). Mixing wastewaters resulted in even ca. 13-fold increase in current production when compared to the control, which was coherent with our previous observations [36]. The MFC work parameters were observed for 1 month after which current density decreased, which was due to cathode fouling and is characteristic for air-cathode MFCs of the used design [39]. The lowest current density was obtained for 500 Ω and the highest for 100 Ω , ca. 440 mA/m². Maximum power density produced was 178 mW/m² (ca. 4 W/m³) for R = 2000 Ω , which was comparable to the power produced from domestic wastewater (1.7 to 3.7 W/m³) in single-chamber MFC [40,41]. Maximum CE obtained here was low and did not exceed 11%. Similar observation was made for the municipal wastewater-fed MFC where CE was below 12% [42]. Low coulombic efficiencies are commonly reported for real wastewaters where fermentation and methanogenesis compete with electrogenesis [43]. Low CE means that large amount of organic matter is oxidized via fermentation or aerobic respiration alternative to electrogenesis, which was earlier reported especially for the air-cathode MFCs and is caused by oxygen leak through air cathode [44].

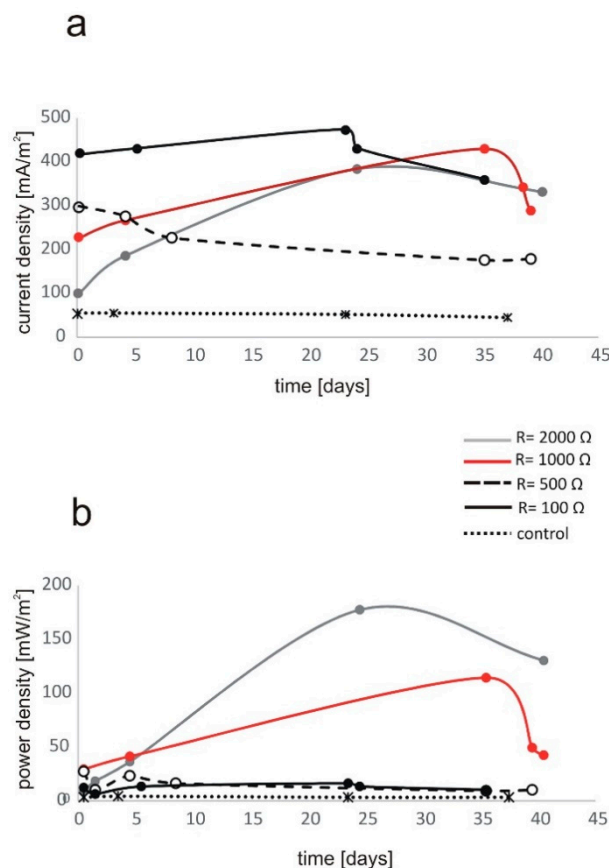


Figure 2. Maximum current density (a) and maximum power density (b) under different external resistances in MFC fed mixed wastewaters in time (control—MFC fed WHTW, R = 1000 Ω).

3.2. Influence of External Resistance and Time on Wastewater Treatment Efficiency

Wastewater treatment efficiency in MFCs is usually linked with the power produced in the system. Higher power produced in MFC is often identified as higher wastewater treatment efficiency. However, it is not always true as enhanced power production does not necessarily yield higher effectiveness of wastewater treatment at the same time (Table 1). Previous reports show that changing temperature from 23 $^{\circ}$ C to 30 $^{\circ}$ C allowed for ca. 20% enhancement of power produced in MFC but did not influence

domestic wastewater treatment efficiency, which was below 30% at both temperatures [45]. An increase of oxygen flow rate to the anode chamber decreased both power production and CE in flat plate MFC but instead increased COD removal efficiency [46]. Thus, having in mind optimization of wastewater treatment in MFC we need to search conditions that, firstly, allow for enhancing treatment efficiency and, secondly, enhance power production during treatment process.

Our previous investigations showed that COD removal of undiluted wood industry wastewater in MFC is low, not reaching 40% [36]. However, COD removal efficiency was remarkably enhanced to ca. 87% when WHTW was mixed with municipal wastewater. Thus, in the present work, COD removal was observed in MFCs fed mixed wastewaters (WHTW: municipal wastewater) under four different resistances through 33 days (Figure 3). After 15 days of MFC operation, COD did not change in MFCs under 2000 Ω , which indicated no wastewater treatment occurred during this time. For all other investigated resistances, wastewater treatment ran yet during first 15 days of MFC operation. The highest COD reduction during this time (3520 to 2160 mg/L) was observed for the reactors under R = 1000 Ω . In all cases, treatment efficiency increased after 20 days of MFC operation, but the highest COD drop was observed after 22 days for R = 1000 Ω where COD decreased below 200 mg/L.

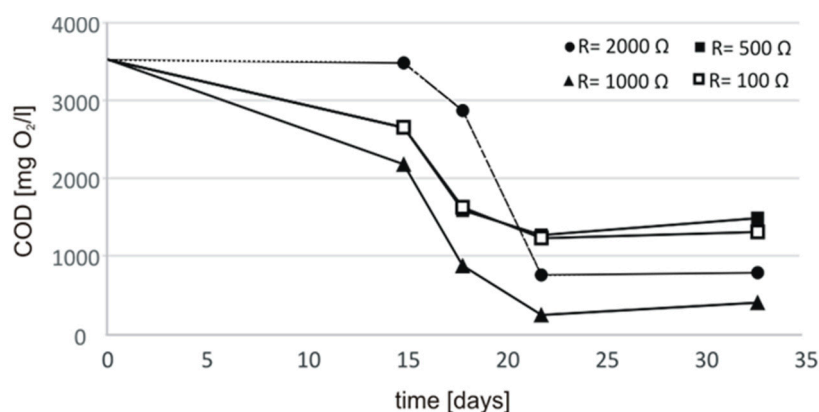


Figure 3. Changes of chemical oxygen demand (COD) in time in MFC fed mixed wastewaters under various external resistances. Analyses were made in duplicates, differences between measurements did not exceed 1%.

The COD removal efficiency increased along with time during the first month of MFCs operation for all tested resistances (Figure 4). The highest Δ COD (94%) was achieved for R = 1000 Ω after 22 days of MFC operation. At the same time, the lowest Δ COD (ca. 65%) was for R = 100 Ω and 500 Ω . Our investigations suggest that lower power production does not strictly correlate with lower wastewater treatment efficiency as though power density under 1000 Ω was lower ca. 40% from power density obtained under 2000 Ω , Δ COD was higher under 1000 Ω resistance. For the highest applied resistance, wastewater treatment start-up time was also elongated in comparison to lower resistances. External resistance has been proved to be an important parameter during wastewater treatment as increasing resistance from 100 Ω to 1000 Ω stimulated ca. 30% increase of COD removal efficiency. These results are different from the previous studies where the highest wastewater treatment efficiency in MFC was described for low resistance 220 Ω [47]. Our investigations demonstrate that wastewater treatment in MFCs requires individual optimization in each case regarding external resistance as various wastewaters are colonized by different microorganisms that adapt differently to available substrate and resistance conditions. External resistance affects significantly anode communities what were earlier observed for two-chamber MFC where growth of methanogenic bacteria was stimulated by increasing external resistance [20].

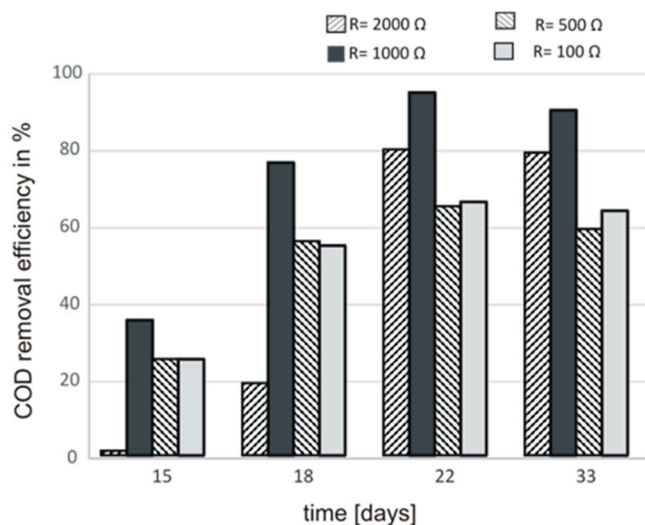


Figure 4. Influence of external resistance on COD removal efficiency of MFC fed mixed wastewaters. Analyses were made in duplicates, differences between measurements did not exceed 1%.

4. Conclusions

External resistance is an important parameter affecting wastewater treatment efficiency. Our investigations show that peak COD removal efficiency does not always correlate with peak power produced in MFCs, as the highest wastewater treatment efficiency was obtained for external resistance under which power density was not maximized. It has been demonstrated that for the maximized wastewater treatment efficiency in MFCs, the optimum external resistance is case-specific for the wastewater to be treated. Thus, power production optimization and COD removal efficiency optimization in MFCs need to be run separately as different resistances are optimal for maximizing each of these two parameters. When COD removal efficiency optimization is needed, no general setting exists, thus the most effective external resistance must be found for a given wastewater type used.

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