Microbial Fuel Cell and Wastewater

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Contributor: N. Evelin Paucar

In recent years, wastewater has been considered as a renewable resource of water, nutrients, and energy. Domestic wastewater is estimated to contain 13 kJ/g of COD of chemical energy, which is nine fold more than the energy required to treat it (Heidrich et al., 2010; Yang et al., 2018). Therefore, if its energy were effectively recovered, no external energy input would be required to operate WWTPs.

Keywords: energy generation ; nutrient recovery ; nutrient removal ; wastewater ; microbial fuel cell

1. Introduction

Water, energy, and food are essential for all living forms to survive and thrive, and they are inseparably linked. Although humans have made great strides in securing those resources, the world is facing an uphill battle due largely to the increasing human population and climate change. By the next decade, the world is expected to face 40% freshwater and 36% energy shortages [1][2], together with increasing demand for food [3][4] and treatment of wastewater.

In recent years, wastewater has been considered as a renewable resource of water [5], nutrients, and energy [5][6][7][8][9][10]. Domestic wastewater is estimated to contain 13 kJ/g of COD of chemical energy, which is nine fold more than the energy required to treat it (Heidrich et al. , 2010; Yang et al., 2018). Therefore, if its energy were effectively recovered, no external energy input would be required to operate WWTPs [11][12].

The increase of atmospheric pollution partly due to the emission of sulfur and nitrogen oxides during fuel combustions may induce irreparable damages to the earth [13][14]. To overcome the energy and environmental crisis caused by the utilization of fossil fuels, a new energy revolution based on renewable resources is beginning to take shape, with electricity as the backbone of energy.

The discharge of wastewater containing high levels of nutrients and organics to a receiving water body is a potential cause of eutrophication and hypoxia in the water environment [15][16]. Therefore, nutrients such as phosphate (PO 43−) and ammonium (NH 4+) are being removed or recovered in WWTPs using methods that require large energy input in order to meet the discharge requirements [17].

2. Microbial Fuel Cell

The EAB act as a biocatalyst for the oxidation of substrate and transferring electrons to the anode [18][19][20]. Microscopic observations have revealed that EAB proliferates over the anode surface to form a multi-layered biofilm [21][22]. The EAB in the monolayer biofilm that is in direct contact with the anode typically utilizes outer-membrane redox proteins and cytochrome cascades to transfer electrons directly to the anode [23]. On the other hand, the microbes in the outer layers develop nanowire structures to connect with the anode surface or use other microbes via an extracellular conductive matrix to transfer electrons, known as interspecies electron transfer [24][25]. In addition to the direct electron transfer, the indirect transfer can also occur via soluble electron shuttles or mediators that transfer extracellular electrons to the anode [26][27]. Based on electron transfer mediators, MFCs can be divided into mediator and non-mediator (or mediator-less) microbial fuel cells. There are two main types of mediators added to microbial fuel cells. The first category is synthetic mediators, mainly dye-based substances, such as phenazine, phenothiazine, indophenol, and thionine [28]. The second type is those synthesized by microorganisms and used by the same organisms or by other organisms for transferring electrons. For instance, Pseudomonas aeruginosa strain KRP1 synthesizes mediator substances such as pyocyanin and phenazine-1-carboxamide [29].

The cathode chamber contains electron acceptors (e.g., O 2) to facilitate reduction reactions, typically given as [30]: 2H 2O + O 2 + 4e − → 4OH − (3)

While electrons flow through the external circuit, protons pass through the PEM to react with oxygen to form water molecules in the cathode chamber [31][32]: 2H + + 2e − + O 2 → 2H 2O (4)

Given the facts that domestic, agricultural, and industrial wastewaters contain various substrates that can serve as a renewable fuel source for MFCs [33][34] and that MFCs have the potential ability to capture a large faction of chemical energy from wastewater [35][12], MFCs can be self-sustaining wastewater treatment technologies that require no external power sources [36]. Specifically, as compared to the conventional wastewater treatment, the MFC technology offers the following potential advantages: (a) Energy-saving—MFCs require no or reduced aeration [37][38][39][40][41][42][43]; (b) Production of less sludge [38][43][44][45][46][47]—MFCs produce less sludge compared to the conventional activated sludge processes [39][40] or even anaerobic digestion processes [37][41][42]. In an MFC, a large fraction of the organic mass in wastewater is converted to electrical energy at a high conversion efficiency [39][40][41][42][43][46][48] with faster reaction kinetics [38]; (c) No generation of harmful toxic byproducts [49][50] such as trihalomethanes (THMs) produced in the chlorination of wastewater [51]; (d) Ability to recover valuable products from wastewater; i.e., electricity [43][46][48] and nutrients [11][15][52][53]; (e) Easy operation under the different conditions [49] such as various temperatures [39] even at low temperatures [37][41][42], various pH values, and with diverse biomass [39]; (f) Clean and efficient technology [49][50]. MFCs can produce electricity with less environmental burdens and a low carbon footprint [54][37][45].

Despite the inherent limitations of MFC technology, overall, it possesses several advantages over conventional wastewater treatment methods, and thus, it is gaining recognition as a potential sustainable wastewater treatment technology. The new advanced electrode materials such as 2D nanomaterials are expected to promote the development of electromicrobiology [50].

3. Energy Generation by MFCs

The typical biological factors are the types, numbers, and catalytic activity of the microorganisms in the MFC. The energy losses at the anode can be attributed to the loss of electrochemical activity of the microorganisms [55] and the anode overpotential transport loss [56]. The physicochemical and electrochemical factors include, but are not limited to, the types and effective surface area of the electrode, electrolytic resistance [50], rate of the proton transport through the PEM, rate of the reduction reaction at the cathode [38][57], and external resistance applied across the electrodes [58][59][60]. The organic loading rates [12][61][62][63], and type and concentration of the substrate are the operational parameters. The intricate interdependence of these factors and parameters makes the optimization of the MFC difficult. For instance, the rate of substrate conversion can be affected by the total amount of electroactive bacterial cells, a phenomenon of mixing mass transfer, bacterial growth kinetics [64][65][66], organic loading rate per biomass (grams of substrate per gram of biomass per day), transmembrane efficiency for the proton transport [67], and total potential of the MFC [64][68].

Internal resistance is one of the major electrochemical factors that affect MFC performance. The internal resistance can be divided into ohmic resistance, charge transfer resistance, and diffusion resistance [69][70]. The ohmic losses occur due to the resistances of the electrodes, PEM, and electrolytes [71]. On the other hand, the charge transfer and diffusion resistance take place in the interface between the electrodes and the surrounding electrolyte [72][73]. The power generation in an MFC is affected by the surface area of the PEM [74]. If the surface area of the PEM is smaller than that of the electrodes (anode and cathode), the internal resistance of the MFC will increase to limit power output [71]. Internal resistance is also a function of the distance between the cathode and anode. For the optimal design, the anode and cathode should be situated as close as possible.

In a dual-chamber MFC, the higher COD loading to the anode chamber can lead to membrane fouling adversely affecting its performance [75]. In contrast, lower COD loadings could facilitate higher electricity generation [63][76]. It has also been found that at low OLRs, MFCs require more time to reach their maximum performance (i.e., maximum current density and maximum power density) [77]. The Coulombic efficiency can be optimized by improving the electrode surface area per reactor volume [46][78].

In an MFC, the external resistance regulates the flow of electrons and consequently regulates the power generation efficiency. In other words, the lower resistance facilitates the electron flow from the anode to the cathode, supporting the microbial electron respiration on the anode, thus enhances the substrate removal efficiency [61][59]. On the other hand, the higher resistance reduces electron flow towards the cathode maintaining a high potential difference, thus enhances the power harvest [58][60][79]. The low voltage at a high external resistance may be due to the slower speed of electrons used on the cathode, compared to its transfer rate [61]. The maximum power density is achieved when the internal and external resistances are equal [80][81]. Various factors such as the distance between the electrodes, electrode material, ionic strength of the anolyte and catholyte, substrate properties, operation modes, and MFC design affect the internal

resistance of MFCs. The optimization of these factors can improve the MFC performance. In general, a single-chamber MFC exhibits lower internal resistance than the dual-chamber MFC: such information should be taken into consideration when designing MFC systems [69].

4. Nutrient Removal and Recovery

The removal of nutrients from the WWTP effluent can reduce the eutrophication potential in the receiving water environment. In comparison with energy-intensive nutrient removal technologies currently employed in the conventional WWTPs, MFCs have the advantage that they generate electricity. The effectiveness of different types of MFCs for the removal and/or recovery of nutrients from various wastewaters and operational conditions are summarized in **Table 1** .

Table 1. MFCs for nutrient removal/recovery from different types of wastewaters and operational conditions.

Type of Wastewater	System Type/Operation Mode	Initial Wastewater Characteristics	COD Removal/HRT	NH_4^+-N Removal/Recovery	PO_4^{3}-P Removal/Recovery	Reference
Synthetic wastewater	Mediator-less dual chamber MFC 2-stage feed-batch mode (Two sets of dual-chamber H-type bottles, operated for 120 days)	COD: 1.5 g/L pH: >8	70–90% HRT = 48 h	----------	38% recovery	[53]
Primary effluent of municipal wastewater	200 L Modularized MFC system (96 tubular MFC modules of 2 L/each) Continuous mode, operated for one-year	TCOD: 155 ± 37 mg/L SCOD: 73 ± 23 mg/L NH_4^+: 25.7 ± 5.5 mg/L TSS: 72.9 ± 16.6 mg/L pH: >8	>75% HRT = 18 h	68% removal	~20% biomass uptake	[7]
Dairy industrial wastewater	Catalyst-less and mediator-less membrane dual chamber MFC. Continuous mode.	COD: 3620 mg/L NH_4^+: 174 mg/L Total P: 187 mg/L NH_3: 167 mg/L TSS: 1430 mg/L VSS: 647 mg/L BOD_5: 2115 mg/L pH: 8.5–10.3	90.46%	69.43% removal	Removal efficiencies: 31.18% dissolved phosphorus, 72.45% phosphorus in suspended solids	[61]
Untreated human urine	3-stage MFC system in a continuous mode (System of MFCs that fits urinals).	NH_4^+: 363 mg/L PO_4^{3-}: 202 mg/L	20% HRT = 18 min for individual MFCs for 5 days	20% removal 7% recovery	82% removal 78% recovery	[15]
Domestic wastewater	Algal biofilm MFC. Continuous mode.	COD: 186.8 327.9 mg/L Total N: 25.3–52.5 mg/L Total P: 2.9–8.3 mg/L	81.9% HRT = 12 days	TN removal: 95.5% 50% recovered by harvested algae	TP removal: 96.4% 62% recovered by harvested algae	[11]
Effluent drain of vegetable oil industry	Dual chambered MFC. Batch mode at 35 °C.	pH: 5.7 TDS: 517 mg/L TSS: 252 mg/L	60–90% 40–80% at 25 °C HRT = 72 h	----------	73.6% removal 56.9% at 25 °C	[82]

Type of Wastewater	System Type/Operation Mode	Initial Wastewater Characteristics	COD Removal/HRT	NH_4^+-N Removal/Recovery	PO_4^3-P Removal/Recovery	Reference
Digestate coming from an anaerobic digester	MET (MFC or MEC) coupled with struvite crystallization using seawater bitterns (SWB). Single chamber, air-cathode MFC batch mode.	NH_4^+: 1943 ± 53 mg/L PO_4^{+-}: 60 ± 3 mg/L COD: 7.2 ± 1.6 g/L	44.7 ± 1.6%	MFC: 10.1 ± 0.5% removal Further removal by precipitation: 14.7 ± 0.6%	MFC: 35.8 ± 1.2% removal Further removal by precipitation: 83.1 ± 3.7%	[83]
Pre-hydrolyzed human urine	Electrodialysis system embedded in an MFC. Continuous mode.	NH_4^+: 7.8 g/L PO_4^{+-}: 0.33 g/L TCOD: 9.5 g/L pH = 8.8	40–65 days	1.2% recovery	0.002% recovery	[6]
Synthetic domestic wastewater	Photoautotrophic H-type MFC. Continuous mode.	Inoculated microalgal biomass: 0.75 g/L	93.2% HRT = 11.8 h	95.9% removed in anodic chamber. 27.7–50.0% removed/recovery in cathodic chamber by microalgae.	82.7% removed in anodic chamber. 37.1–67.9% removed/recovery in cathodic chamber by microalgae.	[84]
Synthetic domestic wastewater	Double-chamber MFC. Continuous mode.	COD: 300–600 mg/L OLR: 435–870 mg COD/L.d	90% (from a wide range of organic loading rate (435 to 870 mg COD/L.d). HRT = 0.69 d	Removed in anode chamber: 14% at OLR of 435 mg COD/L.d and 75.13% at OLR of 870 mg COD/L.d. Recovered in cathode chamber: 85.11% at OLR of 435 mg COD/L.d and 24.34% at OLR of 870 mg COD/L.d.	Removed in anode chamber: 12.43% at OLR of 435 mg COD/L.d and 71.5% at OLR of 870 mg COD/L.d. Recovered in cathode chamber: 24.4% average recovery.	[12]
Synthetic municipal wastewater	Double–compartment MFC. Continuous mode.	COD: 300 ± 15 mg/L NH_4^+-N: 5–40 mg/L OLR: 435 mg COD/L.d	>85% for wide range of NH_4^+-N concentrations (5 to 40 mg/L). HRT = 0.69 d	Removal: ~14% at 5 mg. NH_4^+-N/L and ~14.10% at 40 mg.NH_4^+-N/L. Recovery: 85.11% at 5 mg. NH_4^+-N/L and 15.33% at 40 mg. NH_4^+-N/L.	Removal: ~12.45% at 5 mg. NH_4^+-N/L and 13.33% at 40 mg.NH_4^+-N/L. Recovery: 83.23% at 5 mg. NH_4^+-N/L and 80.5% at 40 mg.NH_4^+-N/L.	[52]
Synthetic urine-containing wastewater	Three-chamber resource recovery MFC. Batch mode.	COD: 24.60 mg NH_4^+: 0.10 mg TN: 20.20 mg PO_4^{+-}: 0.90 mg pH: 6.9	97% HRT = ~3 days	40% of NH_4^+ removed. 98% of TN removed. 42% of TN recovered in middle chamber	99% removed. 37% recovered in middle chamber.	[8]
Synthetic municipal wastewater	Two-chambered MFC. Continuous mode.	COD: 300 ± 15 mg/L pH: 7.00 ± 0.02 OLR: 435–870 mg COD/L.d	>90% (from a wide range of organic loading rate (435 to 870 mg COD/L.d) and HRT = 0.69-0.35 days.	Removed in anode chamber: 13%-15% at different OLR (435–870) mg COD/L.d) and different HRT (0.69 d-0.35 d). Recovered in cathode chamber: ~85% at different OLR (435–870 mg COD/L.d) and different HRT (0.69 d-0.35 d).	Removed in anode chamber: 12–14% at different OLR (435–870 mg COD/L.d) and different HRT (0.69 d–0.35 d). Recovered in cathode chamber: ~83% at different OLR (435–870 mg COD/L.d) and different HRT (0.69 d-0.35 d).	[10]

To date, only a few studies have been undertaken to recover nutrients using MFCs [6][8][10][12][52][85][86]. The recovery of P and N by MFCs has been accomplished mainly by the formation of struvite, NH 4MgPO 4.6H 2O [83][86][87][88][89][90]. Struvite has been demonstrated to be slow-release fertilizer [91] and has a commercial value [92][93][94][95].

In a dual-chamber MFC, nutrient removal usually occurs in the anode chamber and recovery in the cathode chamber [12][52][84]. Almatouq & Babatunde [53] investigated the P recovery and electricity generation using a two-stage, mediator-less dual-chamber MFC system, which was operated in a fed-batch mode. In the first cycle, synthetic wastewater was fed to the anode chamber to remove organics (measured as COD). At the end of the first cycle, the effluent from the anode chamber was filtered and fed to the cathode chamber to recover P as struvite. In their study, 8 mM of NH 4Cl and 8 mM of MgCl 2 solutions were added to the cathode chamber at a rate of 6 mL/day. When the COD concentration was increased from 0.7 to 1.5 g/L, the P recovery efficiency increased from 7% to as high as 38%. The reported power density is 72 mW/m 2 [53]. The COD concentration and aeration rate were shown to be the key factors that affect the P recovery and electricity generation. Since the dual-chamber MFC creates an alkaline environment around the cathode, it provides better nutrient recovery efficiencies [96].

Human urine typically contains 9 g of NH 4+ -N/L, 0.7 g of PO 43− -P/L, and other constituents, and has been used as an electrolyte in an MFC for nutrient recovery system [97][98]. In a study by You et al. [15] nutrients were recovered from human urine in a form of struvite, while generating electricity, using a 3-stage single-chamber MFC/struvite extraction system. The first and third stage MFCs generated 14.32 W/m 3 and 11.76 W/m 3 of power, respectively. The second stage MFC was used for nutrient recovery. The hydrolysis reaction of urea was accelerated in the first stage. In the second stage, magnesium was added to form struvite. In the third stage, after the completion of struvite precipitation, the supernatant was treated for additional power generation and COD removal. In their work, 78% of PO 43− -P and 7% of NH 4+ -N were recovered as struvite. Overall, 82% of PO 43− -P and 20% of COD were removed from human urine. Lu et al. (2019) developed a three-chamber MFC (called a recovery resource MFC or RRMFC) and used it to remove organics and salts, simultaneously recovering nutrients from synthetic wastewater containing urine. The RRMFC consisted of three chambers (anode, middle-recovery, and cathode chambers), and was operated in a batch mode for 33 cycles (~3 days per cycle). Synthetic urine wastewater was fed to the anode chamber where organics were oxidized, and urine was hydrolyzed. Deionized water was fed to the middle chamber where PO 43− and NH 4+ were precipitated as struvite. The effluent of the anode chamber was fed to the cathode chamber for power generation. In their system, the removal efficiencies of COD, NH 4+ , total N, and PO 43− reached 97%, 40%, 98%, and 99%, respectively. At the same time, the RRMFC recovered 42% of total N and 37% of PO 43− in the middle chamber. The NH 4+ mass increased from 0 to 9.01 ± 2.12 mg in the middle chamber, indicating that a large amount of NH 4+ migrated from the anode chamber to the middle chamber through the PEM. Similarly, PO 43− migrated from the cathode chamber to the middle chamber with the effect of the electric field. The decrease of the PO 43− concentration in the cathode chamber may be due to struvite precipitation under the alkali conditions [8]. A fraction of PO 43− may also be removed by microbial assimilation in the anode chamber [99]. The RRMFC produced the maximum currents of 1.30 ± 0.30 mA and maximum power density of 1300 mW/m 2 of the anode surface at an external resistance of 10 Ω. The RRMFC did not require any external energy input for its operation (Lu et al., 2019). Freguia et al. [6] used an MFC/electrodialysis-hybrid system for nutrient recovery from human urine. The fresh urine was left to hydrolyze before the supernatant was collected and used as a feed to the microbial electro-concentration cells. In their study, only about 5% of the influent flow passed through the PEM resulting in a poor nutrient recovery (i.e., the recovery of 1.2% of N and 0.002% of P). It is noteworthy that, if they were designed as an on-site system, their processes not only generate power and recover nutrients, but also save a large amount of water that is necessary to flush and transport urine to a central treatment facility.

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