Microbial Fuel Cells in Treatment from Wastewater

Subjects: Water Resources

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The treatment of wastewater is an expensive and energy-extensive practice that not only ensures the power generation requirements to sustain the current energy demands of an increasing human population but also aids in the subsequent removal of enormous quantities of wastewater that need to be treated within the environment. Thus, renewable energy source-based wastewater treatment is one of the recently developing techniques to overcome power generation and environmental contamination issues. In wastewater treatment, microbial fuel cell (MFC) technology has demonstrated a promising potential to evolve as a sustainable approach, with the simultaneous recovery of energy and nutrients to produce bioelectricity that harnesses the ability of electrogenic microbes to oxidize organic contaminants present in wastewater.

Keywords: energy production ; microbial fuel cells ; resource recovery ; wastewater treatment

1. General Features, Types, and Designs of MFCs

MFCs are relatively new and are one of the promising technologies that facilitate the simultaneous resolution of energy needs and environmental concerns [1][2][3][4][5][6]. MFCs are equipped with the production of biohydrogen, biosensors, and in situ power sources that are utilized for bioremediation collectively with the treatment of wastewater facilities. The rationale that facilitates the use of MFCs for wastewater treatment includes the process of the direct conversion of energy obtained from substrate to electricity, production of controlled activated sludge, insensitivity to the operating environment at low temperatures, their ability to be used without treatment of gas and input of energy for aeration, and their utilization in areas with limited electrical infrastructures [7].

The amount of energy generated through MFCs mainly depends on their design, the distance between electrodes, the electrode utilized, the proton exchange membrane (PEM), the mediators, the substrate, and the microorganisms involved along with certain external influences. MFCs are composed of different designs, including single-chambered, double-chambered, stacked designs, etc. PEM, which is the main component in MFCs, plays a crucial role, as its area in comparison to the electrode surface area affects the power production. PEM is composed of Nafion, cellophane, agar, etc. Mediators are the compounds that are involved in the transportation of electrons from microorganisms to the electrode surfaces and thus induce power density.

The microbes that have been reported to transfer the electrons efficiently to the anode directly are *Rhodoferax ferrireducens* [8], *Shewanella putrefaciens* [9], *Geobacter metallireducens* [10], *Geobacter sulfurreducens* [11], and *Aeromonas hydrophila* [12]. Microbes require nutrients to operate properly, and those nutrients can be provided using certain waste sources as substrates, such as swine waste, dairy waste, as well as combined industrial waste, and so forth [13][14].

1.1. Types of MFCs

The types of MFCs are differentiated depending upon the presence of an anode chamber, cathode chamber, electrode assembly, and PEM or salt bridge. Different MFCs classified according to their design and mechanism of operation are illustrated in **Figure 1**.

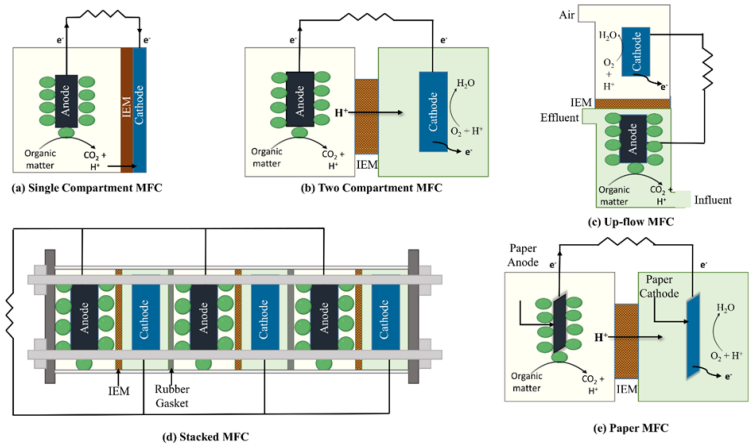

Figure 1. Different types of MFCs (IEM: ion exchange membrane).

1.1.1. Single-Compartment MFCs

Single-compartment MFCs are composed of one anode compartment, and the cathode is exposed. Oxygen (O_2) supply is not required in this design due to the presence of an exposed cathode, rendering these MFCs simple and cost-effective [15]. The basic design allows for batch or continuous operation, as well as a rapid scale-up process. A single-compartment microbial fuel cell (MFC) was designed by availing non-conductive polycarbonate plates that were closed using a system of screw and bolt [13]. Porous carbon paper incorporated as an anode and carbon cloth together with a platinum catalyst has been employed to work as a cathode. Nafion membrane can be used as the PEM, and copper wire is utilized to join electrodes and external circuits [16]. The application of ceramics in MFCs has been reported to be beneficial for the advancement of MFCs' functioning, as ceramic is a feasible material for conventional ion exchange that is of low cost and provides a natural and stable environment for the bacteria as well as an efficient system for energy harvesting [17].

1.1.2. Two-Compartment MFCs

This type of MFC is comprised of two compartments for the anode and cathode, respectively. The anode and cathode compartments are divided by the placement of either a PEM or a salt bridge. This design positions the required microbes, microbe-specific medium, and the electrode in the anode chamber. The cathode chamber includes freshwater or buffers as catholyte, along with an electrode and an O_2 supply. In a two-compartment design, MFCs' electrodes are fabricated using stainless steel mesh, copper, graphite, carbon paper, and carbon and graphite fiber brush [17]. To achieve anaerobic conditions in the anode compartment, a continuous nitrogen supply may be required in some cases. This type of MFC can be constructed by incorporating two borosilicate glass bottles, a clamp system for connecting the glass bridge of two chambers, and a PEM (Nafion) to separate the chambers. The carbon paper is applied to function as the electrode for the anode as well as the cathode. The cathode is embedded with platinum, and lake sediments can be used as an inoculum for microbes. However, due to the expense, Pt-free catalysts such as Pd-Cu, manganese oxides, activated carbon-nickel, and so forth are alternatively being used. Microbes for this design can be cultured in a mineral salt medium (MSM) and can be further stored at 4 °C for use [16].

1.1.3. Up-Flow MFCs

Up-flow MFCs are a continuous mode design, and they apply an injection of wastewater into the system from the bottom with high force towards the upward direction. The effluent can then flow to exit the system from the top [18]. The design is tubular in structure and manufactured with polyacrylic plastic without the use of a PEM. The anode and cathode for this structure are composed of graphite felt, and the separators are formed of glass beads and glass wool. Artificial wastewater containing glucose and glutamate can be utilized as a fuel source. In this design, aeration is provided by cathode layer aerators. The electrodes and the external circuit are connected using platinum wire. Excluding PEM from the design enables it to be used in a continuous mode and parallelly reduces the expenditure. Although this design has immense potential for future applications, it is deficit in terms of net energy efficiency, as the amount of energy that is required to pump the wastewater in MFC is greater than the generated energy [16][18].

1.1.4. Stacked MFCs

This design is comprised of multiple MFCs that have been stacked together parallelly. As multiple MFCs are used, it contributes to higher output efficiency and higher COD removal [19]. This constructed design is composed of six separated continuous MFCs piled together. The anode and cathode of this design are made of graphite granules, and Ultrex

CMI7000 PEM is used as a separator. It has been observed that parallel connections applied in cells possess a better performance in comparison to series connections due to high efficiency and COD removal [16][20].

1.1.5. Paper MFCs

Paper MFCs provide several benefits that mainly include economic effectiveness, chemical resistance, and ease of disposal. The design consists of an anode and cathode with electrodes composed of graphite particles. The particles of graphite are deposited onto the paper through four separate pencil hits. PEM made from parchment paper is used, and crayons can also be used to substantially increase the hydrophobicity. Microbes such as *Shewanella oneidensis* can be introduced in the anode chamber together with appropriate growth media [16][21].

1.2. Substrate and Microorganisms That Are Used in MFCs

The substrate is an important factor that influences the activity of microbes present in the biofilm of the anode and thereby MFCs' performance and generation of electricity [22]. It plays a vital role in providing nutrients and energy to microbes. The substrate can be called anolyte, which is a liquid solution present inside the chamber of the anode [23]. Various substrates that are used in MFCs include pure compounds and complex mixtures of organic matter that are present in wastewater [24]. The most frequently utilized substrates are glucose, brewery effluent, acetate, synthesis wastewater, lignocellulosic biomass, landfill leachates, starch processing wastewater, inorganic substrates, and dye wastewater [1].

According to studies, only a few microorganisms are capable of transferring electrons to the anode, and these types of microorganisms are known as exoelectrogenic bacteria. Single microbes, as well as their mixtures composed of both exoelectrogenic bacteria and non-exoelectrogenic bacteria, can be introduced as a biofilm. The exoelectrogenic bacteria can transfer electrons to the anode using mediators, nanowires, and direct contact with the electrode. Non-exoelectrogenic bacteria utilize mediators that are produced through exoelectrogenic bacteria and thus transfer the electrons to the electrode. Certain exoelectrogenic bacteria used in MFCs include *Shewanella putrefaciens*, *Desulfuromonas acetoxidans*, *Clostridium butyricum*, *Rhodoferax ferrireducens*, *Geobacter metallireducens*, *Desulfobulbus propionicus*, *Geobacter sulfurreducens*, *Geothrix fermentans*, *Pseudomonas aeruginosa*, *Escherichia coli*, *Desulfovibrio desulfuricans*, *Klebsiella pneumonia*, *Ochrobactrum anthropi*, *Shewanella oneidensis*, *Geopsychrobacter electrodiphilus*, *Rhodopseudomonas palustris*, and *Pichia anomala* [16]. The mechanism of electron transport in the plasma membrane for the generation of electricity is represented in **Figure 2**.

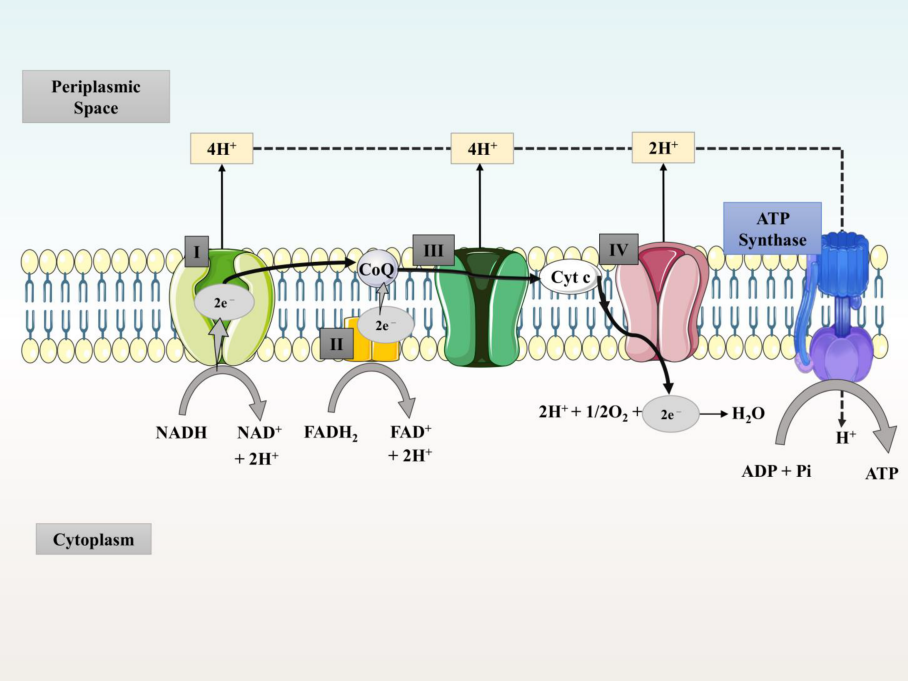

Figure 2. Mechanism of electron transport in the plasma membrane.

2. Application/Performance of MFCs in Wastewater Treatment

Significant developments have been made to improve MFCs' performance together with their application by introducing considerable efforts in the exploration of separators, electrode materials, design of the reactor, and various methods to analyze wastewater other than the power generation and cost-effectiveness [25]. MFC performance is majorly studied through power density that is based on the surface area of the anode or cathode. The anode should be non-toxic to

microorganisms, chemically inert, cost-efficient, and long-lasting [26]. Anodes in MFCs play a key role in durability, power output, and easier functioning. The anode should consist of a wide surface area to allow for bacterial adherence and high electrical conductivity to facilitate the transfer of charge, as well as improved current collection competency. The surface area of the anode is essential in fostering and sustaining bio-catalytic activity and can be changed to enhance its suitability for microorganisms, ultimately improving the transfer of electrons from bacteria to the surface of the anode. The amount of bacterial adhesion is proportionally related to power generation. Thus, more bacterial adhesion will result in more power generation with reduced electrical loss [27].

It has been demonstrated that the construction of a novel annular single-chamber microbial fuel cell (ASCMFC) utilizing stainless-steel mesh and a graphite-coating anode will provide high power density with dairy wastewater substitute [28]. The maximum power density, maximum CE, and COD removal were reported as 20.2 W/m^3, 91%, and 26.87%, respectively. In another study, researchers evaluated a modification for the anode to improve the MFC's performance through electrochemical oxidation. It was reported that CM-N (carbon mesh nitric acid) MFC was capable of 81.7% recovery. The maximum power density observed in the nitric acid-modified anode was reported to be 792 mW/m^2 as compared to the unmodified control (552 mW/m^2). Furthermore, the CE is significantly enhanced after modification to 24% from 14% (the unmodified MFC), and the efficiency to remove COD was also observed to be higher than the unmodified one [29].

The substrate is regarded as the most important biological aspect of MFC, since it determines how much electricity will be generated [30]. Researchers have employed municipal wastewater, swine wastewater, starch-processing wastewater, food-processing wastewater, and chocolate factory wastewater to generate energy using MFC. **Table 1** lists the few most frequent substrates and their effects on their performance.

Table 1. Applications of MFCs in the treatment of different types of wastewaters.

S. No.	Inoculum and Substrate	Type of MFC	Electrode Material	Power Density/Current Density/Voltage	Treatment Efficiency	Reference
1	Swine wastewater manure	Two-chambered	Carbon cloth	13 mW/m^2	TCOD: 83%, CE: 0.3%	[31]
2	Agriculture wastewater (Human feces wastewater)	Two-chambered	-Anode: carbon paper -Cathode: carbon paper with 40% platinum	70.8 mW/m^2	TCOD: 71.0%, SCOD: 88.0%, NH_4^+: 44.0%	[32]
3	Domestic and olive mill wastewater	Single-chambered air cathode	-Anode: graphite fiber brush. -Cathodes 7 cm^2 (total exposed surface area)	124.6 mW m^{-2}	TCOD: 65.0% BOD: 50.0%, CE: 29%	[33]
4	Dairy wastewater (COD of 1000 mg/L) inoculated by activated sludge from the dairy WWTP	Annular single chambered	-Graphite-coated stainless-steel mesh anode -Cathode: carbon cloth type B	20.2 W/m^3	COD: 91%, CE: 26.87%	[28]
5	Synthetic wastewater	Up-flow constructed wetland (UCW-MFC)	-anode: graphite -cathode: magnesium	15.1 mW/m^2	COD: 92.1%, NH_4^+: 93.2%, NO_3^-: 81.1%, CE: 1.64%	[34]
6	Industrial wastewater	Dual chambered anaerobic MFCs	Anode and cathode	260 mW/m^2	TCOD: 87%, SCOD: 79%, TSS: 72%	[35]
7	Acetate	Single-chambered MFC	Substrate as a source of carbon to stimulate electroactive bacteria	506 mW/m^2	CE (72.3%), butyrate (43.0%), propionate (36.0%), and glucose (15%)	[36]
8	Arabitol	Single-chambered MFC	Co substrate in a single chamber	0.68 mA/cm^2	COD: >91%	[37]

S. No.	Inoculum and Substrate	Type of MFC	Electrode Material	Power Density/Current Density/Voltage	Treatment Efficiency	Reference
9	Cysteine	MFC with carbon paper electrodes (11.25 cm^2) dual chamber	Co-substrate	36 mW/m^2	-	[36]
10	Common effluent treatment plant (CETP) wastewater	H-type, dual chamber, mediator-less MFC	graphite plates	0.6 V	COD: 50%	[38]
11	Sodium benzoate (0.721 g/L)	H-type, dual chamber, mediator-less MFC	graphite plates	0.8 V	COD: 89%	[38]

2.1. Factors Affecting Performances of MFCs during Wastewater Treatment

The MFC performance and efficiency are determined by several factors that include microbial electron transfer, fuel oxidation, oxygen supply, circuit resistance, proton transfer via the membrane, reduction at the cathode site, concentration, and pH. These factors can be reinforced and modified over time for better output [30].

2.1.1. Electrode Properties

Electrode performance and power output are influenced by electrode material properties. **Table 2** represents different anode and cathode materials that could be used in MFCs as well as their advantages and disadvantages. The material used for the anode should support a broad surface area, as well as good electrical conductivity and stability. However, because of the high-power output per unit surface area, graphite felt, carbon cloth, carbon felt, carbon mesh, and graphite fiber brush are frequently utilized as electrodes. Furthermore, reports on platinum (Pt)-based cathodes and biocathodes suggest an increase in MFCs' power input by increasing catalytic activity using oxygen or reducing over potential. However, the cathodes mentioned are not economically friendly [26]. The performance of the cathode electron receiver determines the power and voltage density in MFCs. Researchers have studied electrical functioning of MFCs with potassium permanganate, ferricyanide solution, and dissolved oxygen as cathode electron receivers. The highest power density and smallest internal resistance of 4.35 W/m^3 and 54 Ω, respectively, were reported [39].

Table 2. Anode and cathode materials used in MFCs.

S. No.	Material Used	Anode/ Cathode	Advantages	Disadvantages	Reference
1	Graphite rods	Anode	High conductivity, chemical stability, low cost, and easy to handle	Surface area is difficult to increase	[40]
2	Graphite brushes	Anode	Easy to construct and more specific area	Clogging issues	[41]
3	Carbon cloth	Anode	Large porosity relatively	Not cost efficient	[42]
4	Carbon paper	Anode	Easy to construct wire connection	Brittle	[43]
5	Carbon felt	Anode	Enormous surface area	Elevated resistance	[44]
6	Reticulated vitreous carbon	Anode	High electrical conductivity	Delicate and large resistance	[8]
7	Stainless steel	Anode	High conductivity, cost efficient, and easily accessible	Low surface area, compatibility issues, can get corroded	[45]
8	Pt-based catalyst	Cathode	High surface area and low potential for the oxygen reduction reaction	pH sensitivity, sulfide poisoning, and non-sustainability	[46]
9	Non-Pt-based catalyst	Cathode	pH control, no sulfide poisoning, and non-sustainability	Compromised electron transfer	[47]
10	Carbon Nano tubes	Cathode	High surface area and power density	Voltage losses	[48]

S. No.	Material Used	Anode/ Cathode	Advantages	Disadvantages	Reference
11	Palladium	Cathode	Excellent catalytic properties and low cost	Very low oxygen reduction reaction overpotential for catalytic hydrogen production	[49]
12	Aerobic biocathode	Cathode	Production of methane, ethanol, and formic acid via microbes and application as a biosensor for BOD detection	Loss of electrons through oxygen	[50]
13	Anaerobic biocathode	Cathode	Prevention of loss of electrons via anodic end	Biofilms catalyze the reduction of chemically active species	[14]
14	Cathode with metal-free catalyst	Cathode	Cheap materials, catalytic activity, stability	Superior electrocatalytic activity, with lower overpotential and prolonged stability for ORR	[47]

2.1.2. pH

In MFC, protonic generations occur at an anodic end with facilitation of the smooth flowing electrons to interact with the oxygen molecule for the production of water. Anode acidification occurs due to the continuous loop operation due to incomplete proton transport through the membrane. On the other hand, the cathode is alkalized due to the lower efficiency of proton replacement. These constraints eventually hinder the effectiveness of MFCs, resulting in a pH concentration gradient. An increase in the pH of the cathode compartment reduces current production, thus lowering the operating pH required to achieve higher power production [51]. A study has demonstrated the effect of pH on the production of electricity and contaminant dynamics through MFCs [52]. Reports suggest that the production of power was the highest (0.66 Wm^{-3}) with a pH of around 9.5 for the air cathode chamber. For 30 days, the MFC operation was in continuous control mode and enhanced the performance of the cell in terms of power output, which was reported to be 1.8 Wm^{-3} at the optimum pH. The study demonstrated that physical ammonium loss and organic matter removal were directly influenced by pH.

2.1.3. Temperature

Temperature also has a considerable impact on MFCs' performance in terms of removing COD and generating electricity. MFCs' kinetic properties and thermodynamic properties are highly dependent on temperature. An increase in temperature leads to an increase in the power density but a decrease in the ohmic resistance. A study has examined the effects of a change in temperature on the electrode potential, power density, columbic efficiency, COD removal, and internal resistance of a two-chamber MFC [53]. It was reported that as the internal resistance of the MFC increases, the power density acquired is simultaneously reduced. The data for CE were observed to be 8.65% at 30 °C, 8.53% at 37 °C, and 13.24% at 43 °C, respectively. These findings illustrate that MFCs are capable of operating at a wide range of temperatures.

2.1.4. Aeration

Aeration, along with the presence of oxygen in the cathode, is another key characteristic of MFC function, since organic catalysts such as Pt, iron, and Al are known to require large amounts of oxygen to carry out the reduction process as the electron acceptor of the cathode. However, since an air-purging pump is required within the cathode, this technique raises the cost of an MFC. Different aeration rates were used to evaluate the efficacy of MFCs. To study the influence of anode aeration on electricity generation, an air-cathode MFC that had earlier been embellished anaerobically in the anode was subjected to aeration intermittently and steadily [54]. Except for a loss in CE, intermittent aeration had almost no impact on electricity production. An electricity with 0.35–0.41 V was generated with a wide range of dissolved oxygen concentrations (D.O) at rate of (0.1–4.0 mg/L). The study revealed that the maximum voltage output was minimally affected by anode aeration, but CE was dramatically lowered. In another study, power generation was increased dramatically in the anode MFC due to aeration. The results showed that the maximum voltage generation of aerated, aerobic, and anaerobic anodes was 183, 150, and 68 mV, respectively [55]. The aeration flow rate also plays a significant role in bioelectricity generation. However, it has been demonstrated that power generation does not increase proportionally with the increase in aeration flow rate. In another study, the optimal aeration flow rate to accomplish the maximal power generation was reported to be 600 mL min^{-1} due to the presence of adequate oxygen to serve as the terminal oxygen acceptor for electricity production [56].

3. Different Products' Recovery from Wastewater Using MFCs

To date, various wastewater treatment technologies are in practice to achieve sustainable goals along with greater treatment efficiencies [57][58][59][60]. MFCs, although being an eco-friendly approach, require a high amount of investment, together with a maintenance cost, which eventually leads to compromised economical aspects of their development. This increased cost is mainly due to the involvement of separator materials and expensive electrodes. The development of cost-effective bioelectrodes and abiotic electrodes has been a promising aspect in minimizing the cost of MFCs' establishment and maintenance but requires more research for designing. MFCs based on decentralized wastewater are cost efficient due to the reduction in the transportation cost for wastewater and less energy consumption. Moreover, they promote the recovery of additional valuable substances found in wastewater, such as gold, heavy metals, and silver, thus rendering them economically viable.

MFCs have shown excellent efficiencies for the removal and recovery of heavy metals from wastewater. For instance, an algal (*Chlorella* sp.)-based MFC with nickel-foam/graphene electrodes achieved up to 95% Cd(II) removal efficiency with a maximum adsorption amount of 115 g/m^2 [61]. Other studies using ligand-based sensor materials have also achieved significant Cd(II) adsorption capacities of 167.33 mg/g and 176.19 mg/g, respectively [62][63]. However, algal MFC provides an advantage over other technologies due to enhanced power generation, with a maximum power density of 36.4 Mw/m^2 [61]. Similarly, for the detection of wastewater metal contaminants such as cesium, copper, and lead, functionalized adsorbents that can achieve excellent adsorption capacities have been developed [64][65][66]. MFCs have also resulted in 98.3% and 89.6% Cu^{2+} and Pb(II) removal rates, respectively [67][68]. However, for cesium removal from wastewater, MFCs have shown undesirable results owing to high electrical resistance and low potential [69].

The wastewater generated from food industries, animal houses, and agriculture is high-strength wastewater containing high amounts of nitrogen (N) and phosphorus (P) components that can be further utilized as fertilizers that are vital in agricultural activities [70]. Researchers have worked on the development of a lucrative MFC system for silver metal recovery from silver (Ag) ion-containing wastewaters [71]. Silver metal recovery was achieved with efficiencies as high as 99.91 ± 0.00% to 98.26 ± 0.01% with an output rate of 69.9 kg silver/kWh energy output. This was obtained using a batch-fed cathode and continuously fed anode systems with an initial silver concentration of 200 ppm. Another study investigated the utilization of tetrachlorocuprate as an electron acceptor of an MFC for the discovery of requirements that can affect the cost-efficient recovery of gold. The highest MFC efficiency was found to be around 57% for Au (III), with a concentration of 200 ppm, and the Au recovery efficiency and remaining concentration were reported to be 99.89 ± 0.00% and 0.22 ± 0.00 ppm, respectively. An air-cathode single-chamber MFC was constructed to analyze the effects of ammonium (NH_4) and magnesium (Mg) on phosphorus precipitation in artificial wastewater.

It has been reported that the ammonium content is approximately 9000 mg/L in human urine, and it was 8100 mg/L after urine hydrolysis was utilized with different MFCs models, such as continuous mode, for treatment. Through the application of MFCs, nutrients were able to be recovered from human urine also in a form of struvite together with electricity generating. A three-stage single-chamber MFC/struvite extraction system was utilized to recover nutrients. In the first and third stages, MFCs were reported to generate 14.32 W/m^3 and 11.76 W/m^3 of power, respectively, and in the second stage, MFC was used for nutrient recovery [72].

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