

Review

A Perspective Review on Microbial Fuel Cells in Treatment and Product Recovery from Wastewater

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Abstract: 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. Since traditional wastewater treatment has various limitations, sustainable implementations of MFCs might be a feasible option in wastewater treatment, green electricity production, biohydrogen synthesis, carbon sequestration, and environmentally sustainable sewage treatment. In MFCs, the electrochemical treatment mechanism is based on anodic oxidation and cathodic reduction reactions, which have been considerably improved by the last few decades of study. However, electricity production by MFCs remains a substantial problem for practical implementations owing to the difficulty in balancing yield with overall system upscaling. This review discusses the developments in MFC technologies, including improvements to their structural architecture, integration with different novel biocatalysts and biocathode, anode, and cathode materials, various microbial community interactions and substrates to be used, and the removal of contaminants. Furthermore, it focuses on providing critical insights and analyzing various types, processes, applications, challenges, and futuristic aspects of wastewater treatment-related MFCs and thus sustainable resource recovery. With appropriate planning and further studies, we look forward to the industrialization of MFCs in the near future, with the idea that this will lead to greener fuels and a cleaner environment for all of mankind.

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



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1. Introduction

In the current era of technological development, the depletion of non-renewable resources and the simultaneous environmental damage caused by their utilization are crucial global concerns. The overexploitation of energy resources has caused various

negative impacts, including greenhouse emissions, rising global temperatures, and severe climate change. The dependency and the negative environmental impacts of fossil fuels for energy consumption have prompted the urgent requirement to focus on the development of alternate technologies and sources that could prove advantageous in meeting energy demands together with environmental protection. Along with this energy deficit, the treatment of wastewater containing contaminants is also one of the rising issues from many in most parts of the world [1–5]. A large share of the total energy load drained by the wastewater treatment sector has been identified as a renewable resource and could be used to address both issues of energy generation and treatment together with the recovery of nutrients [6–12]. Accomplishing the need to treat wastewater and generate energy, microbial fuel cells (MFCs) are one of the recent techniques that could be utilized [13–18]. MFCs are proficient in converting chemical energy from civil and industrial wastewaters together with the generation of electrical energy through the application of specific electroactive bacteria (EAB). EAB could act on the reactions of substrate oxidation and oxidant reduction, thus aiding in the generation of energy and minimizing wastewater treatment costs [19–24]. Domestic wastewater has been reported to hold 13 kJ/g of chemical oxygen demand (COD) for chemical energy i.e., about nine-fold of the energy required for treatment [25,26]. Thus, if the energy included in wastewater could be efficiently retrieved, then this could lead to no external energy input requirement for operating wastewater treatment plants (WWTPs) [25–27]. However, if a fraction of the energy could be retrieved, then it could benefit in the reduction of economic costs of wastewater treatment. MFCs' performance can also be increased by managing operational parameters, i.e., the organic loading rate (OLR), hydraulic retention time (HRT), pH, and applied electric resistance [28–32]. Although the process as well as design management of MFCs are being improved and modified by introducing developments in mathematical models, operational understanding of the current techniques still needs to be achieved [33,34].

In the current scenario, the commercialization of MFCs is restricted due to various bottlenecks associated with this, including limited efficiency and high operational and maintenance costs. The practical application of this technology in various areas is still limited due to system instabilities, scaling-up limitations, competitive microbial reactions, and thereby a limited generation of power [16,19–23,25–31,33,34]. The treatment of wastewater with the application of MFCs is still obscured at different points and needs to be investigated. For example, the advances realized at a low scale have been not translated to a larger scale, suggesting that an additional understanding of the progress is still mandatory.

Rapid advancements in MFC research have resulted in the publication of various illuminating reviews on organic biomass resources, the assessment of various configurations, specialized themes such as resource recovery, robustness, and repeatability, and waste-to-energy transformation using MFC technologies [35–39]. To add to previous studies, the present review focuses on the robust applications of MFCs and their types in wastewater treatment and sustainable resource recovery, with great attention to the working principles and the parameters affecting their scaling up. Furthermore, insights into various types, processes, applications, challenges, recent advances, and futuristic aspects of wastewater treatment-related MFCs are comprehensively provided.

2. 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 [40–45]. 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 [46].

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. A few examples of intracellular mediators include NADPH, NADH, cytochromes, and so forth. Certain synthetically obtained mediators include thionine, methyl blue, methylene blue, neutral red, hydroxy-1,4-naphthoquinone, and so forth. Synthetic mediators might be integrated, but they have limited applications due to their toxicity. Recent studies suggest that the direct transfer of the electrons from the surface of the cell to the anode surface with increased stability and coulombic efficiency (CE) plays an important role in the operation of MFCs [47]. CE is the analysis that refers to the transfer of electrons in a system to carry out an electrochemical reaction. In MFCs, CE measures the number of coulombs recovered as electrical current and is dependent on the types of microorganisms involved, the substrate used, the type of wastewater, the design of MFCs, and the experimental protocol [29].

The microbes that have been reported to transfer the electrons efficiently to the anode directly are *Rhodospirillum rubrum* [48], *Shewanella putrefaciens* [49], *Geobacter metallireducens* [50], *Geobacter sulfurreducens* [51], and *Aeromonas hydrophila* [52]. 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 [53,54].

2.1. 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.

2.1.1. Single-Compartment MFCs

Single-compartment MFCs are composed of one anode compartment, and the cathode is exposed. Oxygen (O₂) supply is not required in this design due to the presence of an exposed cathode, rendering these MFCs simple and cost-effective [55]. 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 [53]. 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 [47]. 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 [56].

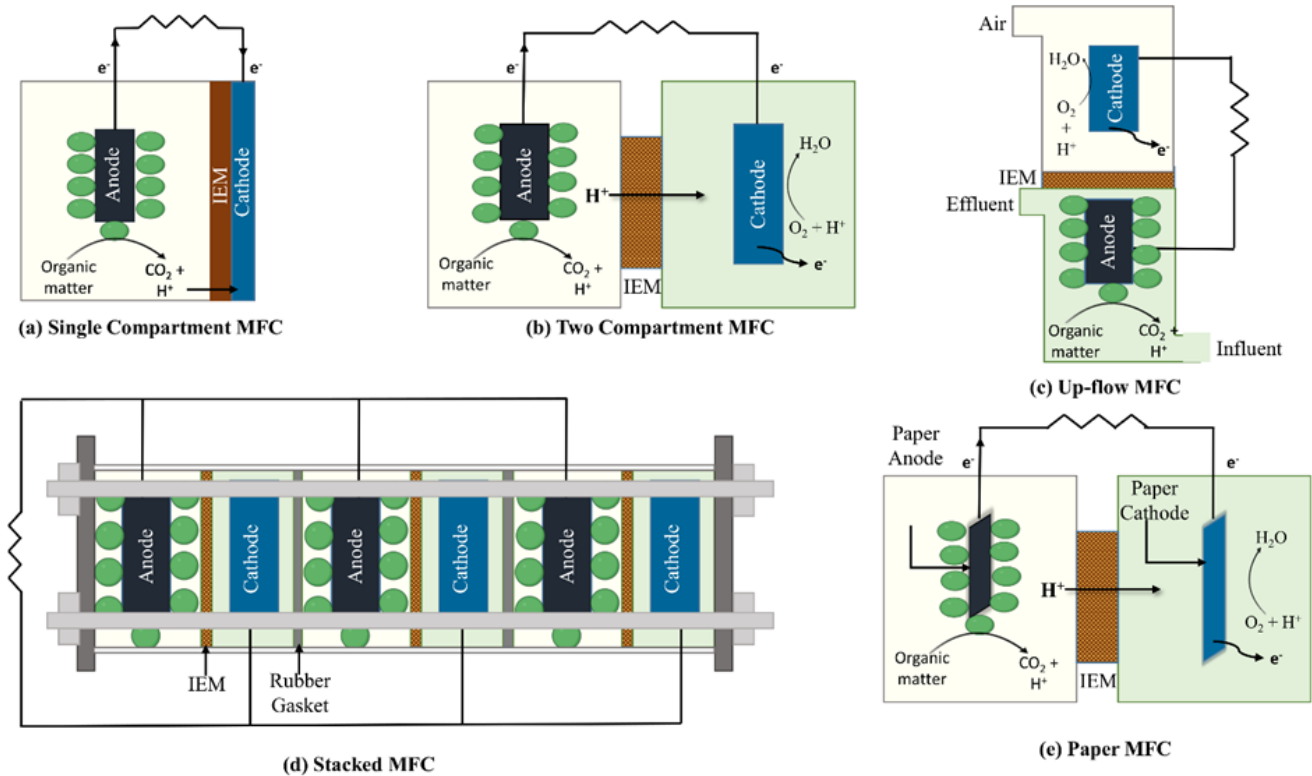


Figure 1. Different types of MFCs (IEM: ion exchange membrane).

2.1.2. 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 [56]. 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 $4\text{ }^\circ\text{C}$ for use [47].

2.1.3. 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 [57]. 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 [47,57].

2.1.4. 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 [16]. 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 [47,58].

2.1.5. 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 [47,59].

2.2. 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 [60]. 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 [61]. Various substrates that are used in MFCs include pure compounds and complex mixtures of organic matter that are present in wastewater [62]. The most frequently utilized substrates are glucose, brewery effluent, acetate, synthesis wastewater, lignocellulosic biomass, landfill leachates, starch processing wastewater, inorganic substrates, and dye wastewater [40].

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*, *Rhodospirillum rubrum*, *Geobacter metallireducens*, *Desulfobulbus propionicus*, *Geobacter sulfurreducens*, *Geothrix fermentans*, *Pseudomonas aeruginosa*, *Escherichia coli*, *Desulfovibrio desulfuricans*, *Klebsiella pneumoniae*, *Ochrobactrum anthropi*, *Shewanella oneidensis*, *Geopsychrobacter electrodiphilus*, *Rhodospseudomonas palustris*, and *Pichia anomala* [47]. The mechanism of electron transport in the plasma membrane for the generation of electricity is represented in Figure 2.

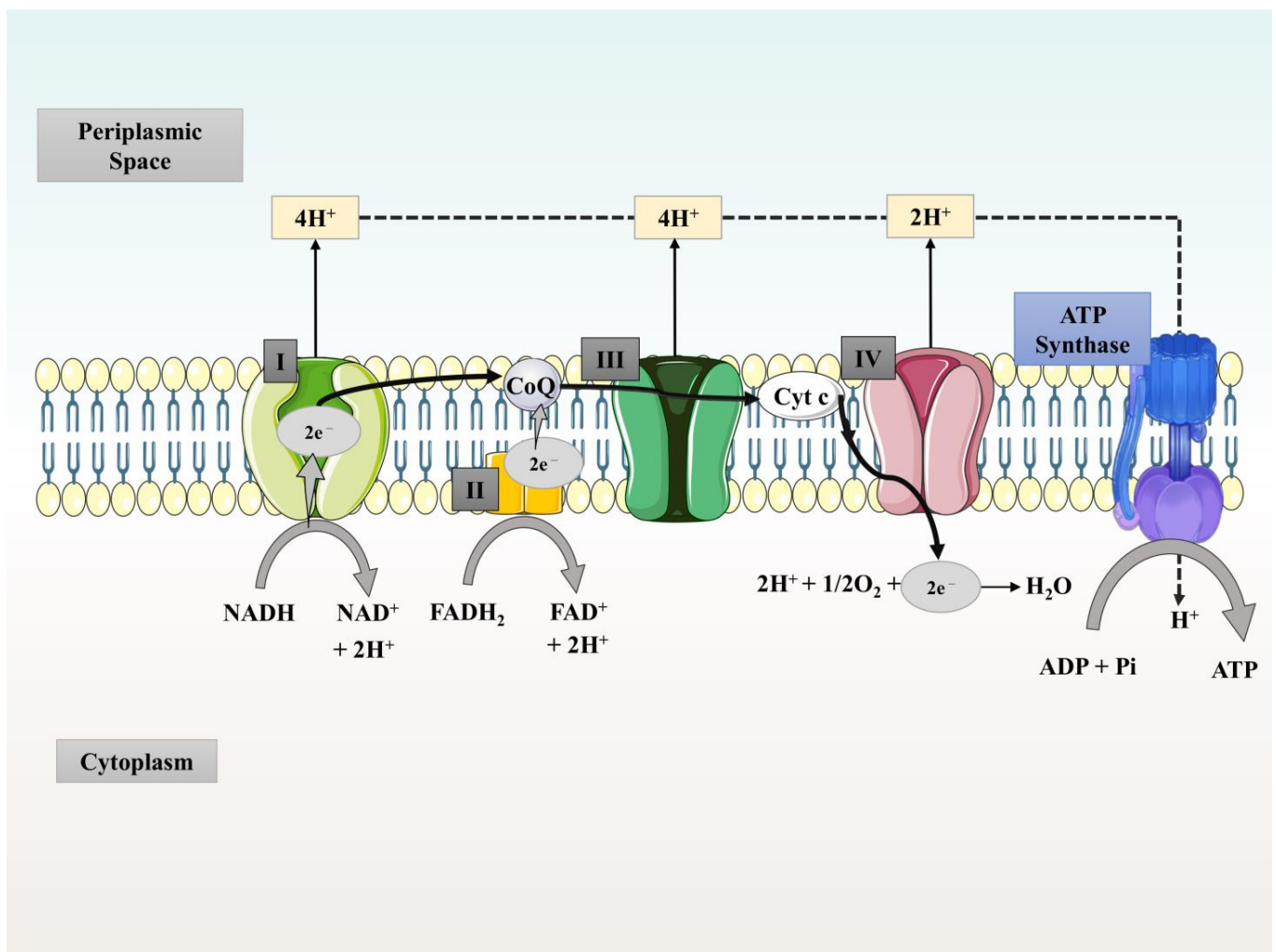
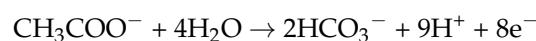


Figure 2. Mechanism of electron transport in the plasma membrane.

3. Working Principle/Treatability of MFCs and Generation of Energy

As discussed in the preceding sections, MFCs consist of electrodes, i.e., an anode and cathode together with a microorganism, a substrate/anolyte, and an external circuit. Figure 3 illustrates the working principle of a typical MFC. The reaction that occurs in the MFC is generally represented by the oxidation of acetate, as shown below [26]:



The EAB operates as a biocatalyst that further results in the substrate oxidation and transfer of electrons to the anode by direct transfer [63]. Figure 4 depicts proton and electron generation via exoelectrogens and proton transportation across the PEM membrane in MFCs. The indirect transfer can proceed through soluble electron shuttles that initiate the extracellular electron transfer to the anode [64]. MFCs can be composed of electron transfer mediators or a non-mediator. Synthetic materials comprised of dye-based materials, such as phenothiazine, phenazine, indophenol, and thionine, can also be utilized as mediators. Furthermore, in the reaction, electrons produced in the anode chamber move through the external circuit to reach the cathode and generate a current as a result [63,65]. The cathode chamber contains electron acceptors (e.g., O_2) that enable reduction reactions, as described below [66]:



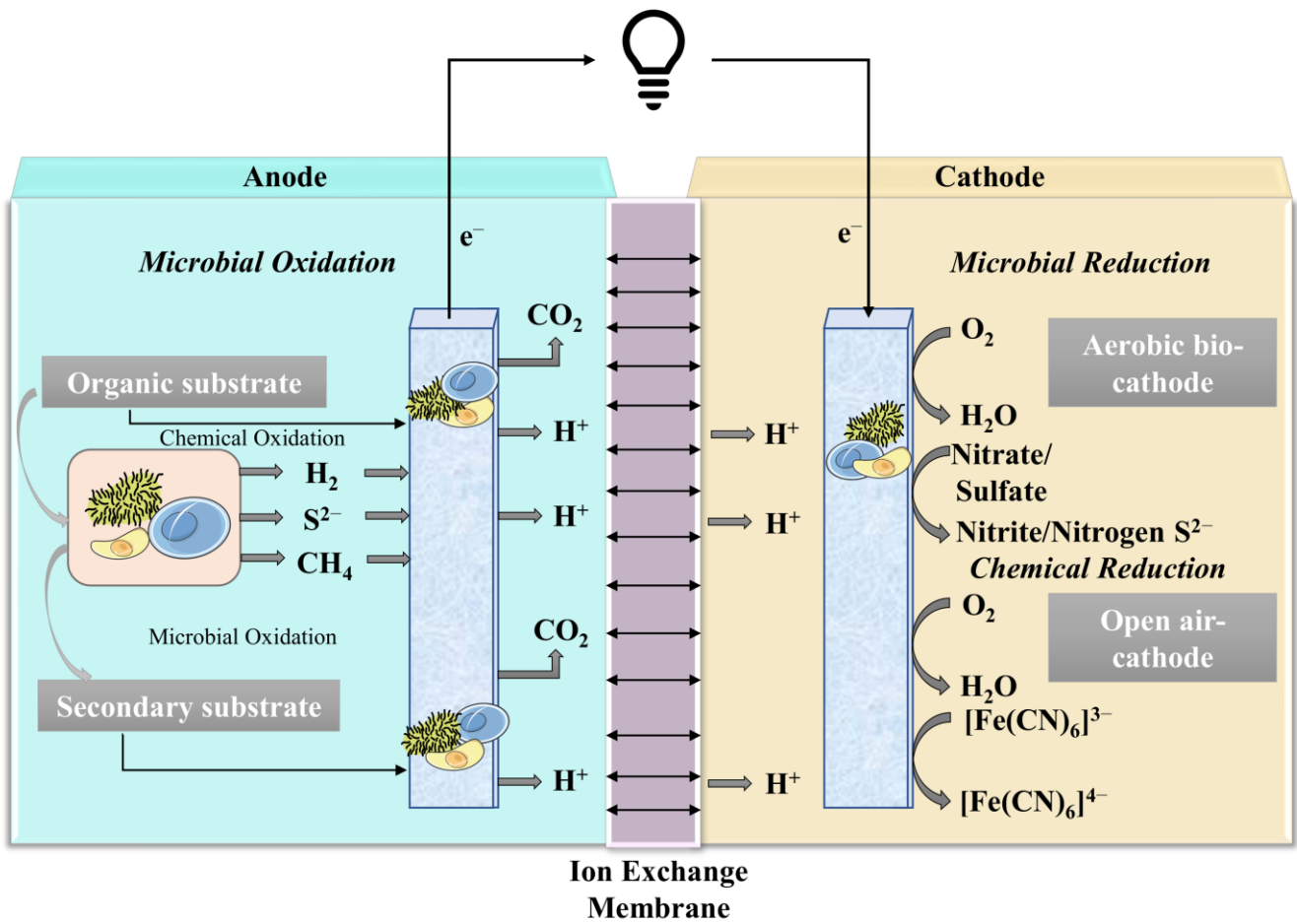


Figure 3. Working principles of MFCs.

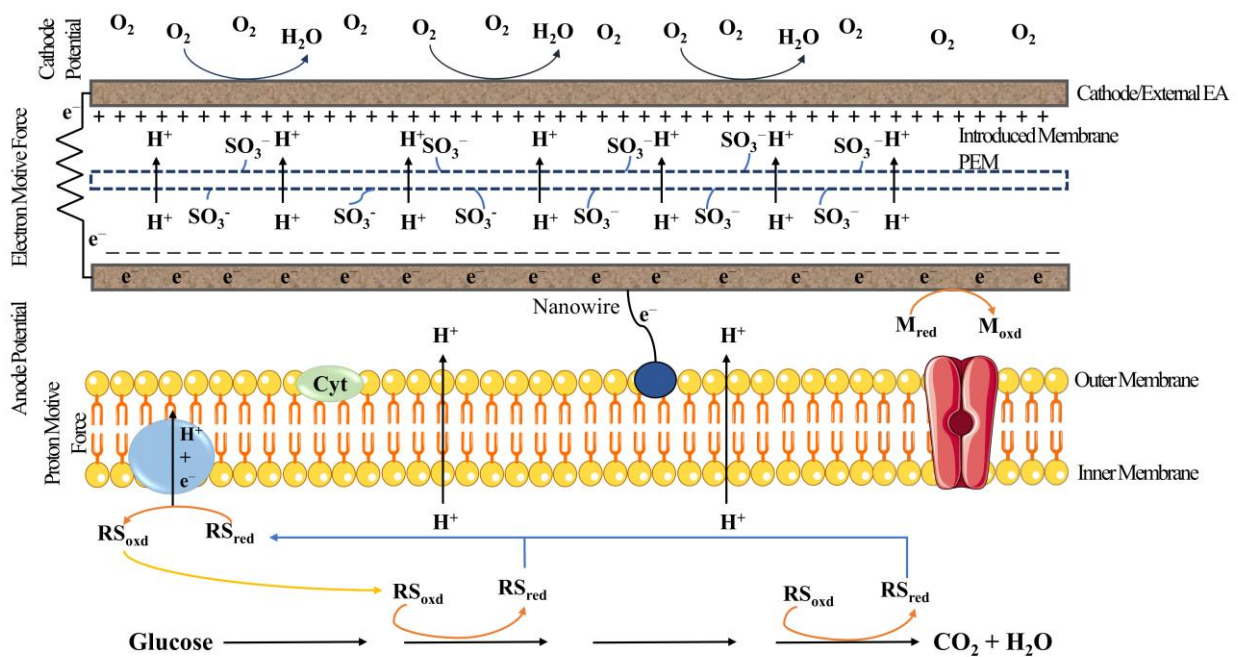


Figure 4. Illustration depicting proton and electron generation via exoelectrogens and proton transportation across the PEM membrane in MFCs.

As the electrons move through the external circuit, it causes the flow of protons through the PEM, which then react with oxygen in the cathode chamber to form water molecules. The reaction is shown below [67]:



Wastewater that is obtained from domestic, agricultural, and industrial sources consists of a variety of materials that can be used as a renewable fuel source for MFCs. MFCs can be designed and developed to function as self-sustaining wastewater treatment technologies that can enable them to be independent of external power sources to supply energy. This technology offers several economic benefits, including energy efficiency, the production of less sludge or even anaerobic digestion processes, minimum or no toxic byproducts, the recovery of economically important products from wastewater such as nutrients and electricity, easy operation under varying conditions, and a reliable and sustainable technology [26].

The effectiveness of MFCs is affected by a combination of factors that mainly includes biological, physicochemical, electrochemical, and operational parameters. The biological factors that affect the performance of MFCs include the numbers, types, and catalytic activity related to microorganisms that are incorporated. The energy dissipation at the anode can be imputed for the loss of electrochemical activity of the respective microorganisms as well as the anode overpotential transport loss [68,69]. The physicochemical factors that can interfere with the functioning of MFCs are the types and efficacy of the electrode surface area, electrolytic resistance [70], the transfer rate of the proton through the PEM, a reduction reaction occurring at the cathode [71], and external resistance incorporated across the electrodes [64,72,73]. Electrochemical factors that can manipulate MFC performance include ohmic resistance, internal resistance, diffusion resistance, and charge transfer resistance [26].

The surface area of the PEM also affects power generation in an MFC [26]. When the PEM has a smaller surface area as compared to the surface area of the anode and cathode, it leads to an increased MFC internal resistance, thus constraining power generation [56]. Internal resistance in the MFC is directly proportional to the distance between the cathode and the anode. The PEM is significant for enhancing MFC performance in double-chamber MFCs, as it also determines the operational expenses and maintenance costs. Many of the prior MFC investigations have been performed using commercial Nafion as the PEM. As a membrane with lower resistance and higher ion selectivity is desirable, UltrexTM, a cation exchange membrane with low resistance, is preferred over Nafion. The carbon paper can be used to function as a separator and could reduce the internal resistance and thereby the MFC expenditures. Ceramic materials are both viable and cost-effective substitutes for extensively used membranes [74]. Operational parameters include organic loading rates [75], as well as the type and concentration of the substrate utilized. However, the evaporation of the substrate/anolyte remains a difficulty for MFC performance and requires further research [26].

4. 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 [76]. 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 [77]. 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 [78].

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 [79]. The maximum power density, maximum CE, and COD removal were reported as 20.2 W/m³, 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/m² as compared to the unmodified control (552 mW/m²). 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 [80].

The substrate is regarded as the most important biological aspect of MFC, since it determines how much electricity will be generated [70]. 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%	[81]
2	Agriculture wastewater (Human feces wastewater)	Two-chambered	-Anode: carbon paper -Cathode: carbon paper with 40% platinum -Anode: graphite fiber brush.	70.8 mW/m ²	TCOD: 71.0%, SCOD: 88.0%, NH ₄ ⁺ : 44.0%	[82]
3	Domestic and olive mill wastewater	Single-chambered air cathode	-Cathodes 7 cm ² (total exposed surface area)	124.6 mW m ⁻²	TCOD: 65.0% BOD: 50.0%, CE: 29%	[83]
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%	[79]
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%	[84]
6	Industrial wastewater	Dual chambered anaerobic MFCs	Anode and cathode	260 mW/m ²	TCOD: 87%, SCOD: 79%, TSS: 72%	[85]

Table 1. Cont.

S. No.	Inoculum and Substrate	Type of MFC	Electrode Material	Power Density/Current Density/Voltage	Treatment Efficiency	Reference
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%)	[86]
8	Arabitol	Single-chambered MFC	Co substrate in a single chamber	0.68 mA/cm ²	COD: >91%	[87]
9	Cysteine	MFC with carbon paper electrodes (11.25 cm ²) dual chamber	Co-substrate	36 mW/m ²	-	[86]
10	Common effluent treatment plant (CETP) wastewater	H-type, dual chamber, mediator-less MFC	graphite plates	0.6 V	COD: 50%	[88]
11	Sodium benzoate (0.721 g/L)	H-type, dual chamber, mediator-less MFC	graphite plates	0.8 V	COD: 89%	[88]

4.1. Factors Affecting Performances of MFCs during Wastewater Treatment

As discussed in the previous section, 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 [70].

4.1.1. 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 [77]. 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/m³ and 54 Ω, respectively, were reported [89].

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	[90]
2	Graphite brushes	Anode	Easy to construct and more specific area	Clogging issues	[91]
3	Carbon cloth	Anode	Large porosity relatively	Not cost efficient	[92]
4	Carbon paper	Anode	Easy to construct wire connection	Brittle	[93]
5	Carbon felt	Anode	Enormous surface area	Elevated resistance	[94]
6	Reticulated vitreous carbon	Anode	High electrical conductivity	Delicate and large resistance	[48]
7	Stainless steel	Anode	High conductivity, cost efficient, and easily accessible	Low surface area, compatibility issues, can get corroded	[95]
8	Pt-based catalyst	Cathode	High surface area and low potential for the oxygen reduction reaction	pH sensitivity, sulfide poisoning, and non-sustainability	[96]
9	Non-Pt-based catalyst	Cathode	pH control, no sulfide poisoning, and non-sustainability	Compromised electron transfer	[97]
10	Carbon Nano tubes	Cathode	High surface area and power density	Voltage losses	[98]
11	Palladium	Cathode	Excellent catalytic properties and low cost	Very low oxygen reduction reaction overpotential for catalytic hydrogen production	[99]
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	[100]
13	Anaerobic biocathode	Cathode	Prevention of loss of electrons via anodic end	Biofilms catalyze the reduction of chemically active species	[54]
14	Cathode with metal-free catalyst	Cathode	Cheap materials, catalytic activity, stability	Superior electrocatalytic activity, with lower overpotential and prolonged stability for ORR	[97]

4.1.2. 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 [101]. A study has demonstrated the effect of pH on the production of electricity and contaminant dynamics through MFCs [102]. 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.

4.1.3. 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, coulombic efficiency, COD removal, and internal resistance of a two-chamber MFC [103]. 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.

4.1.4. 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 [104]. 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 [105]. 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 [106].

5. 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 [5,107–109]. 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 adsorp-

tion amount of 115 g/m² [110]. 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 [111,112]. However, algal MFC provides an advantage over other technologies due to enhanced power generation, with a maximum power density of 36.4 Mw/m² [110]. 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 [113–115]. MFCs have also resulted in 98.3% and 89.6% Cu²⁺ and Pb(II) removal rates, respectively [116,117]. However, for cesium removal from wastewater, MFCs have shown undesirable results owing to high electrical resistance and low potential [118].

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 [119]. Researchers have worked on the development of a lucrative MFC system for silver metal recovery from silver (Ag) ion-containing wastewaters [120]. 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 (NH₄) and magnesium (Mg) on phosphorus precipitation in artificial wastewater.

Phosphorus was precipitated as struvite, and NH₄ and Mg were added to the effluent. After dissolving the precipitated phosphorus on the cathode with MES buffers and Milli-Q water, the phosphorus was able to be recovered [121]. A three-chamber MFC-based nitrogen recovery technique from synthetic wastewater was demonstrated in a study [122]. The ferric nitrate was a single-electron acceptor in the cathode and was also utilized to evaluate the NO₃-N recovery efficiency in the event of NO⁻³ as the main anion in the cathode. Overall, the results showed it was possible to recover approximately 47% NH⁺⁴-N in the anode chamber and 83% NO⁻³-N in the cathode chamber. In another study, dual-compartment MFCs were constructed and operated continuously with various influent concentrations of ammonium-nitrogen (5–40 mg/L). The influence of ammonium on organics removal and output of the energy and the recovery of nutrients was examined. Overall, the experimental results showed that the phosphate recovery rate gained in MFC was influenced by ammonium concentration [123]. Researchers have evaluated the work performance of the MFCs using mixed consortia and isolated pure cultures of *Firmicutes* and *Proteobacteria* species from biofilm for electricity production and nutrient recovery. The study revealed that the microbes utilize less than 10% of total phosphorus for their growth; meanwhile, 90% was able to be recovered as struvite [124].

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/m³ and 11.76 W/m³ of power, respectively, and in the second stage, MFC was used for nutrient recovery [25]. Table 3 shows innovative approaches to improve MFC function and outcome.

Table 3. Innovative approaches to improve MFC systems and productivity.

S. No.	MFC System	Outcome	Ref
1	Synthetic polymeric tubular MFC having affinity binding group for removal of volatile fatty acids and inorganic compounds	Effective removal ($\geq 98\%$) of pollutants, up to 95% biodegradation of the toxic compounds	[125]
2	CW-MFC system developed for electroactive and textile dye wastewater treatment through microbial community	Bioaugmentation and dynamic removal of pollutants through electroactive bacterial community	[126]
3	Screening of fruit waste for MFCs	The energy production rate of orange waste was maximally up to 357 mV voltage output, followed by banana and mango fruit waste	[127]
4	Estimated the power generation capacity of sodium citrate-treated MFCs	Significantly improved biocatalytic activity of anode with maximum electrical energy output	[128]
5	Study the MFC coupled effect with stacked 12 vertically-arranged constructed wetlands	Reduced COD level, uptake of free N and P, electricity generation	[73]
6	Microalgae can be used in MFCs	Efficient for CO ₂ uptake, effective removal of N and P, symbiotic microalgae–bacterial interactions for power generation	[129]
7	Anode–cathode catalysts immersed in biomolecule solutions (monosaccharides, nitrogen and amino acid)	51% COD, 20 mL methane gas was achieved at 20.5 °C temperature	[10]
8	MFC operated through bioelectrochemical nutrient from human urine as a self-power generating system	Endured power and electrical current generation at a rate of 3 A/m for over two months and simultaneously increased concentration of N and K by a factor of 1.5–1.7	[123]
9	Evaluated the effect of ammonia concentration on MFC power generation and efficiency	A high concentration of ammonia in the influent negatively affected the ammonium recovery and poor uptake of phosphorus by MFCs	[72]
10	Estimated the treatment efficiency of membrane-less MFCs by simulating core of a shallow un-planted horizontal subsurface flow-constructed wetland system	Effective for domestic wastewater treatment with 25% efficiency	[72]
11	Applicability of lingo-cellulosic low-cost material for MFCs	Maximum power generation through the high electro-osmotic force and high pH at the cathode with significant recovery of elements	[130]
12	Evaluated the effect of nitrate and sulphate components on MFC microbial component activity	Nitrate does not show any effect on cathode and anode microbial flora. However, the bacterial community of <i>Desulfovibrio</i> showed dominant growth on the cathode (32.9%) after the addition of sulfate	[131]
13	Studied the long-term processing of multi-layered MFC for brewery effluent	Maximum removal efficiency for COD up to $94.6 \pm 1.0\%$ but system failure due to long-term processing	[132]
14	Algae cathode MFCs for landfill leaching at different concentrations of pollutants of 5–40%	Enhanced removal of nitrogen and phosphorus with power generation	[133]
15	200 L modularized MFC system consisting of 96 MFC modules	The cost-effective system generates ~200 mW power, 75% of the total COD, and 90% of the suspended solids removal	[7]
16	Electro-chemicals disruption of pollutants	Non-toxic metabolites	[134]
17	Two-chamber MFC for wastewater treatment at a rate of 84 L/hr and COD of 3000 mg/L	COD conversion of 91.9%, electricity generation of 26.4 kWh for the feed of 84 L/hr	[135]
18	Constructed wetland reactor and a microbial fuel cell reactor(CW-MFC) for digestion	MFC digestion rate for 98–100 L/hr and 74% electricity generation	[136]
19	microbial bioelectrochemical systems (BES) co-culture <i>Pseudomonas aeruginosa</i> and other strain	Highest electrochemical activity	[137]
20	MFC system with passive aeration method for waste treatment	Cost-effective approaches for electricity generation by the 80% organic compound removal	[138]
21	Comparative study of a single-chamber (MFC-1) and double-chamber (MFC-2) MFC for wastewater treatment	Effective removal of solutes, maintenance of COD level and electricity production	[139]

6. Recent Advancements in MFC Technology

MFCs are a relatively new promising technology for producing renewable energy while treating wastewater. Despite the various advantages that MFCs provide, this technology still has significant drawbacks that prevent its expansion and widespread use in potential areas of application. Recent developments in MFC technology have involved significant research on novel materials to be applied in the primary system components (anodic/cathodic electrodes and separator) to overcome these system bottlenecks, mainly low power density levels from wastewater substrates. Nanostructured materials have attracted a lot of attention for the fabrication of advanced electrodes and separators due to their improved properties such as high specific surface area, increased transfer rates, and in many cases, low cost and ease of fabrication [140]. Metal nanoparticles such as copper, gold, platinum, and palladium; silver, quantum dots, and metal-oxides such as 480 CeO_2 , TiO_2 , ZnO , SiO_2 , and MnO_2 ; graphene (2D nanomaterials), carbon nanotubes, and nanocomposites (multiphase materials) are some examples of nanomaterials for electrodes that can increase the function of MFCs [141]. The addition of nanosized fillers to the nanocomposite matrix can improve the performance of anode materials. Multiphase materials that include at least one component phase with a size smaller than 100 nm are included in nanocomposites. To operate as a suitable anode material in MFCs, nanocomposites are created to enhance a material's distinctive functionalities (such as mechanical strength, electrical conductivity, and chemical stability) [37]. The power density of MFCs may be increased to more than 2000 mWm^{-2} by utilizing nanocomposite anodic materials such as graphene oxide and stainless steel-based nanocomposite [142]. Carbon nanotubes (CNTs) are a novel form of carbon material that has received considerable interest from scientists due to their distinctive fiber structure, extremely high mechanical strength and toughness, large specific surface area, good thermal stability and chemical inertia, strong conductivity, and distinctive one-dimensional nanometer scale [143]. Through layer-by-layer self-assembly, Roh et al. [144] changed carbon paper to use multi-walled carbon nanotubes as the anode. The internal resistance of the modified carbon paper with CNTs in the MFC was 258Ω , which was much lower than the equivalently operated MFC with carbon paper (1163Ω), as compared to plain carbon paper. From a starting power density of 241 mW/m^2 , the maximum power density reached 290 mW/m^2 . The results might be explained by the electrical double-layered region expanding due to the CNT's inclusion, which increased the surface area and average pore diameter. Today, due to its exceptional qualities, graphene is regarded as a top competitor for a highly effective and affordable material for MFCs. Since it has a high surface area and high electrochemical conductivity, graphene is frequently utilized as an anode material in MFC. The graphene-modified stainless steel mesh anode in MFC has a power density of 2668 mW m^2 [145]. For the benefit of enhancing the MFC's performance, a new modified anode based on multi-walled carbon nanotubes was designed. *E. coli* grew effectively on MWCNT-, MWCNTCOOH-, and MWCNT-NH₂-doped anodes as opposed to bare carbon fiber anodes. As a result, 560.4 mW/m^2 is the greatest power density that could be measured using an anode modified by MWCNT-COOH [146]. Therefore, these kinds of adjustments contribute to enhancing the MFC's power generation and stability. As MFC can produce power by treating wastewater, nanomaterial electrode materials offer a viable technique for high hydrogen generation.

7. Challenges and Future Prospects

Various research papers and studies suggest that MFC technologies have been demonstrated as ecologically sustainable techniques to generate power while also removing pollutants from different forms of wastewater. However, there are a few significant issues, including economical aspects, development of designs that offer maximum output, and so forth, with MFC technologies. Thus, they have never been considered a significant competitor in the field of renewable energy or the wastewater treatment sector. Nevertheless, MFCs are probably sufficient in terms of net energy production, rather than utilizing energy obtained from the oxidation of organic materials using wastes and inorganic car-

bon under certain conditions. MFC systems have the benefit of being able to transform chemical energy directly into electrical energy through biological processes, allowing them to physiologically adapt to the treatment of a wide range of chemical substrates at different concentrations. MFC technology can be used by research groups to facilitate a better understanding of electrochemical, biochemical, microbial, and material surface responses under regulated conditions, which has had a favorable influence. The main focuses of their study are comprised of how materials, chemical compounds, and feedstock substrates, among other things, might affect them [45]. This strategy allows us to better understand the potential issues affecting MFCs' larger scale applications.

To achieve commercial success of MFC technology, measures should be taken to minimize the high cost of operation and ameliorate poor power production [147]. Concerning configuration and treatment capabilities, MFCs have a capital cost that is 30 times greater than a standard activated sludge treatment system utilized for household wastewater [148]. The utilization of expensive electrode materials, such as catalyst, current collector, and separator materials, leads to the high-level capital cost of MFCs. In MFCs, bacteria can transfer electrons to the anode and protons into the solution, which causes a negative anode potential (approximately 0.2 V). The most promising cathode oxidants are oxygen and air, and both have a maximum theoretical potential of 0.805 V. Pt-catalyzed cathodes facilitate a maximum achievable potential of +0.3 V in MFC. The highest limits of power density in the case of MFCs were anticipated to be between 17 and 19 W/m², assuming negligible internal resistance or first-order kinetics characteristic of microorganisms in biofilms.

The poor power density is due to a combination of factors, including solution conditions, high internal resistance, substrate degradability, and the dynamics of biofilm. To improve currents and voltage output, the stacked MFCs could be used in parallel or series; however, parallel connections may enhance currents and power density, and CE is also substantially higher in parallel than in series. The stacked MFCs in a direct series can increase voltages, which is difficult to achieve with a chemical fuel cell due to the impact of the external circuit on the microbial consortia. The voltage reversal in stacked MFCs and its impact on the enhancement of the performance in a direct series can be a topic of investigation for future study. The larger voltage could be accomplished by connecting arrays of MFCs to the charge capacitors in parallel and further discharging them in series. However, this technique can raise the cost and power consumption [148].

Although MFCs are considered a new trend, further studies should concentrate on minimizing the limiting factors and comprehending the metabolic process involved to select high electrochemically active microorganisms. This entails creating a thick conductive biofilm and fine-tuning the operational parameters. The applicability of MFCs in wastewater treatment is directly determined by the design and architecture of the reactor [149,150]. Investigations on energy utilization and storage are required to develop a power utilization and collecting system that would speed up the commercial deployment of MFCs as well as reduce the elimination of hazardous substances. Figure 5 depicts green chemistry approaches for wastewater treatment through the MFC system. Owing to remarkable advancements in electrode materials and inoculation patterns, MFC stacks can withstand the critical challenges of ionic short circuits and voltage reversal, which have been significant hurdles to practical use [151,152].

Further research is needed to address the following issues: enhanced power generation, regulated microbial performance in the unit, creation of new full-scale MFC models to reduce possible losses for optimal performance, and reduction of costs. Finally, the integration of MFCs with other wastewater treatment technologies can enhance treatment efficiency and hence reduce the overall power consumption to a great extent.

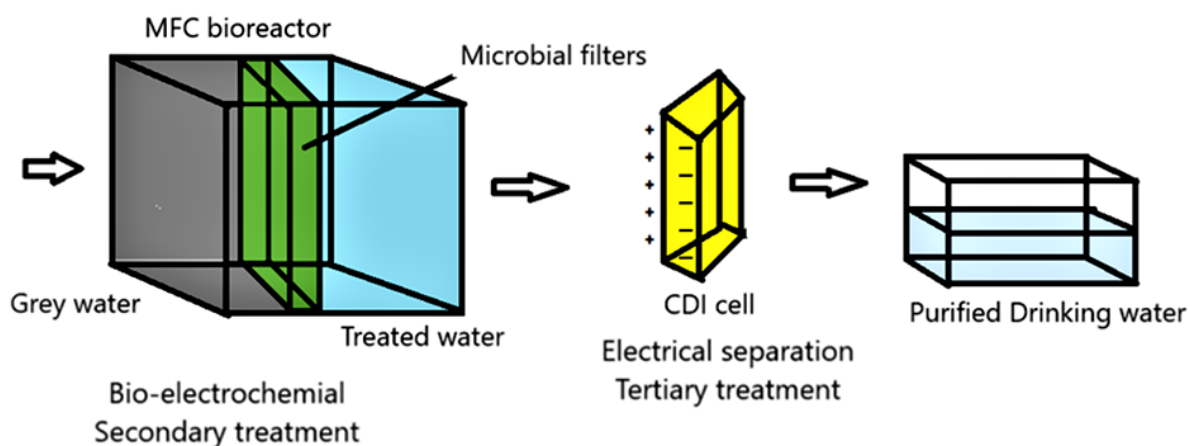


Figure 5. Green chemistry approaches for wastewater treatment through MFC system.

8. Concluding Remarks

This review article has attempted to summarize the applications of MFCs in wastewater treatment and green bioelectricity generation. Recent developments in MFC technologies have been comprehensively discussed, including improvements in their structural architecture, integration with different novel biocatalysts and biocathode, anode, and cathode materials, various microbial community interactions and substrates to be used, and the removal of contaminants. Furthermore, limitations restricting the commercialization of fuel cells such as the cost and efficiency of treatment, electrode performance, power density, and high maintenance expenditure have been highlighted. MFCs are widely acknowledged as a decent and potent solution for integrating bioenergy production and wastewater treatment. MFC technologies are ecologically favorable techniques, and power output may be enhanced by utilizing cellulosic materials, which is beneficial to the system. As a result, enhancements in power densities, COD removal, pollutant degradation, and a widespread requirement to generate electricity without CO₂ emissions might indicate that MFCs are increasingly practicable for power production. Thus, it can be concluded that MFCs may be commercialized in large-scale industries by enhancing power density and overall efficacy while decreasing resource budgets, as well as coordinating a boundless accomplishment in energy production and sustainable wastewater treatment.

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