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Review

From Waste to Watts: Updates on Key Applications of Microbial Fuel Cells in Wastewater Treatment and Energy Production

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Abstract: Due to fossil fuel depletion and the rapid growth of industry, it is critical to develop environmentally friendly and long-term alternative energy technologies. Microbial fuel cells (MFCs) are a powerful platform for extracting energy from various sources and converting it to electricity. As no intermediate steps are required to harness the electricity from the organic substrate's stored chemical energy, MFC technology offers a sustainable alternative source of energy production. The generation of electricity from the organic substances contained in waste using MFC technology could provide a cost-effective solution to the issue of environmental pollution and energy shortages in the near future. Thus, technical advancements in bioelectricity production from wastewater are becoming commercially viable. Due to practical limitations, and although promising prospects have been reported in recent investigations, MFCs are incapable of upscaling and of high-energy production. In this review paper, intensive research has been conducted on MFCs' applications in the treatment of wastewater. Several types of waste have been extensively studied, including municipal or domestic waste, industrial waste, brewery wastewater, and urine waste. Furthermore, the applications of MFCs in the removal of nutrients (nitrogen and sulphates) and precious metals from wastewater were also intensively reviewed. As a result, the efficacy of various MFCs in achieving sustainable power generation from wastewater has been critically addressed in this study.

Keywords: microbial fuel cells; energy production; wastewater treatment; bioelectricity; waste-to-fuel

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1. Introduction

The increase in world energy demands urged researchers and engineers to overuse fossil fuels, specifically in the oil and gas industry [1–10]. These industries emit greenhouse gases into the atmosphere and their effluents are harmful pollutants to the marine environment [11–13] causing global warming and water pollution. Hence, an alternative greener and more environmentally friendly pathway as an energy source has become a must for decreasing the negative impacts of global warming and water pollution on our planet Earth. Renewable bioenergy is considered one of these alternative pathways to a greener environment [14–16]. Immense efforts are being devoted to research so as to develop biotechnologies that produce bioenergy. The most desirable pathway is to generate energy from renewable resources with a very low carbon footprint [17]. Finding a holistic approach to meeting ever-increasing energy demands in a sustainable manner has piqued the interest of both the research community and industry.

Many sources of waste, such as municipal solid waste, have been investigated and used for energy production. Waste-to-energy approaches and strategies were highlighted as a means for green energy production. Microbial fuel cells (MFCs), which use a direct conversion of waste over anodophilic microorganisms, are gaining significant potential as a means of bioelectricity production among waste-to-energy approaches [9]. In this process, a number of waste sources could be utilized, including plant, food, and animal wastes, in addition to sewage sludge, domestic, and other wastes. A large body of published work has demonstrated the potential of using MFCs for bioelectrification production [5–9]. Figure 1 demonstrates the citations count for several publishers about the topic of MFC applications in wastewater treatment. It is clearly seen from Figure 1 that Elsevier holds the largest number of citations (106,864 citations), followed by MDPI, with 33,299 citations, within the years 2017 to 2020. In this review, we focus on wastewater as a source of green, sustainable bioelectricity production.

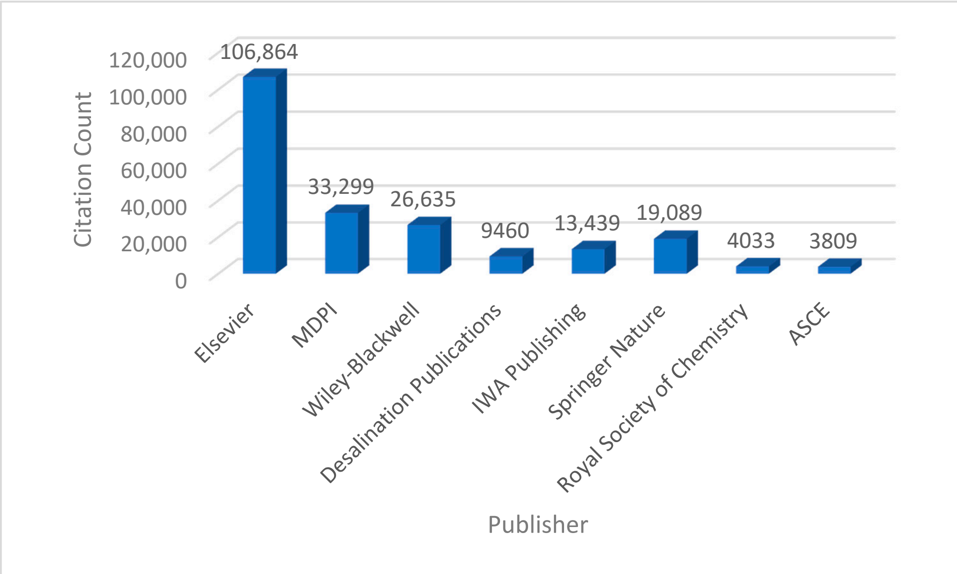

Figure 1. Citations count of MFC applications in wastewater treatment for several publishers from the year 2017 to the year 2020 (Source: Scopus).

By focusing on wastewater treatment, biotechnology is in continuous development for greener ecosystems [18–20], pollutants removal [21–28], and the production of biodegradable materials that provide a new pathway for a better wellbeing [29,30]. Wastewater is currently considered a crucial resource for recycling water [31–36] and saving energy [37–43]. The main types of wastewater include the following: industrial, storm water runoff, and domestic. Each type of wastewater has its own characteristics.

Several technologies have been proposed for the treatment of wastewater, including classical activated sludge treatment and anaerobic digestion. Such technologies have been hindered by high costs and high energy requirements, related to the cost of aeration for effective microbial growth. In addition, wastewater treatments are associated with high greenhouse gas (GHG) emissions, such as CO_2 and methane. Typically, the production of 1000 tons of waste is associated with the production of 1500 tons for each kWh of electricity produced [20]. Although biogas can be effectively produced from anaerobic treatment of wastewater, an additional separation and clean-up process is required, which adds to the energy requirements. Accordingly, current wastewater-to-energy production is deemed energy inefficient and alternative approaches are required [20]. One of the best technologies used for continuous wastewater treatment and saving energy is microbial fuel cells (MFCs). The use of MFCs in wastewater treatment has been extensively studied for the removal and recovery of harmful pollutants and contaminants, such as heavy metals, ammonia (NH_3), and chemical oxygen demand (COD), via organic matter biological degradation for the generation of electrical energy [44]. Figure 2 shows a timeline of the number of publications on MFCs in wastewater treatment. It is clearly seen from Figure 2 that the publication number increases over time. This shows a great interest in the applications of MFCs in wastewater treatment. Furthermore, the year with the greatest number of publications is 2021, with 23,600 publications on MFCs in wastewater treatment. Conversely, the year with the least number of publications is 2010, with 4950 publications on MFCs in wastewater treatment.

Figure 2. A short, post-2010 MFC timeline representing the number of MFC applications in wastewater treatment-related academic publications (Source: Google Scholar).

Microbial fuel cells (MFCs) combine electrochemical reaction pathways with microbial metabolisms [20] and convert organic energy to electrical energy directly [45], thus decreasing waste and producing electric energy. When compared to other wastewater treatment methods, MFC technology is a type of bioelectrochemical system (BES) that has recently gained popularity due to its low cost and low level of negative environmental impacts, such as sludge formation and greenhouse gas emissions [46–48]. In addition, MFCs are associated with energy production, heavy metal removal, and bio-removal of toxic waste.

MFCs, a bio-electrochemical, system typically consist of two sections: an anode and cathode chambers. In the anode chamber is the main “powerhouse” of the system, where the microorganisms are used as biocatalysts to oxidize the organic substrate.

Microorganisms hold a crucial part in these technologies, since they form electron-rich metabolites, produce redox mediators, preserve a redox gradient, and transport electrons to an electrode via direct electron transfer or through a soluble electron transfer mediator, while producing electricity as the main product [49]. Many factors affect the efficiency of the system, such as the chamber design and system operational conditions.

With this backdrop, the use of MFCs in wastewater treatments shows potential as a candidate for green sustainable wastewater treatment and energy generation. Having said that, this technology has not yet been fully realized as a practical effective alternative to current wastewater treatment technologies. This short overview focuses on highlighting the applications of MFCs in the treatment of wastewater using several types of wastewater, including municipal or domestic wastewater, industrial wastewater, brewery wastewater, and urine wastewater. In addition, this review emphasizes the application of MFCs for the removal of nutrients and precious metals from wastewater.

2. Historical Evolution of Microbial Fuel Cells (MFCs)

Generally speaking, almost all types of microbes can be successfully applied as biocatalysts in an MFC. The first MFC idea was introduced by Potter in the year 1910, in which Saccharomyces bacteria and Escherichia coli bacteria were used to produce electrical energy while using platinum electrodes [50]. This concept did not catch the interest of researchers until the 1980s, when it was found that the addition of electron mediators enhanced the power output and current density. The outer layers of most microbial organisms are made up of non-conductive lipid membranes, lipopolysaccharides, and peptidoglycans, which slow down direct electron transfer to the anode. As a result, the addition of electron mediators accelerates electron transfer from the cathode to the anode [51]. There are several synthetic exogenous mediators, including metalorganics, and dyes, including methylene blue (MB) [52], methyl orange [53], neutral red (NR) [53], thionine [54], and potassium ferricyanide [55]. However, the use of synthetic mediators in MFCs is hampered by their instability and toxicity [56]. Fortunately, a wide range of microbes, including microbial metabolites (endogenous mediators), may use naturally occurring compounds as mediators [57]. Anthraquinone and humic acids have the ability to pass electrons from the cell membrane to the anode [58]. When microbes were discovered to pass electrons directly to the anode in 1999, it was a major breakthrough [59]. These microbes have a high Coulombic efficiency and are stable in operation [60]. Unfortunately, the mediators have a high cost. Thus, MFCs that do not require a mediator are preferably used in wastewater treatment. Figure 3, below, shows the historical development of MFC technology. The following potential benefits make MFC applications appealing in general [60,61]: (1) energy from the substrate is converted directly into electricity; (2) no additional biogas treatments are required when compared to anaerobic digestion (AD); (3) the conversion of organics to energy is quite efficient; (4) there is little excess activated sludge produced, and (5) there is little reliance on external influences. Despite their benefits, MFCs face challenges in terms of the production of power density, costs, and long-term stability. Hence, the integration of MFCs with other technologies appears to mitigate these downfalls and accelerate MFC growth and commercialization in the wastewater treatment industry [49].

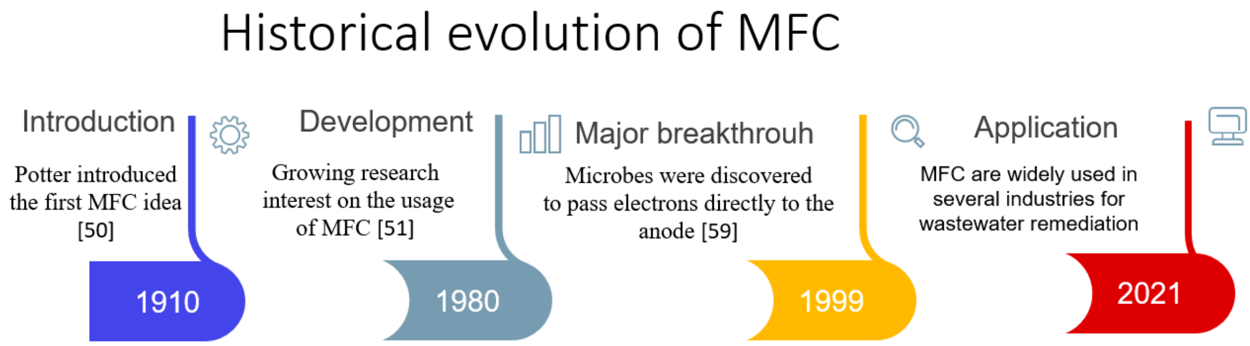

Figure 3. Historical evolution of MFCs, [50,51,59].

3. Microorganisms Applied in Microbial Fuel Cells

In addition to the physical design of the MFC system, the biocatalyst inoculation is an essential factor that will affect the generation of the bioelectricity in the system.

Several types of microorganisms are capable of transferring electrons produced by organic matter metabolism to the anode. As such, exoelectrogens generate electricity via the oxidation of the organic compounds available in the substrate. These microorganisms, as well as their substrates, are demonstrated in Table 1 below. Marine and freshwater sediment, soil, wastewater, and activated sludge are all rich in these microorganisms [61,62]. Several recent studies have focused on microbe screening and identification, as well as the enhancement of a chromosomal library for microorganisms that can produce electricity via the degradation of organic matter [63,64]. The technique behind the anodic electron transfer in MFCs is crucial to grasp the principle behind how they operate [65].

Table 1. Several microorganisms used in MFCs, with their mediators.

Microorganism	Substrate	Mediators	Reference
Proteus mirabilis	Glucose	Thionine	[66]
Saccharomyces cerevisiae	Hydrolyzed Lactose	Neutral red (NR), Methylene blue (MB)	[67]
Escherichia coli	Glucose	Neutral red (NR)	[68]
Rhodoferax ferrireducens	Glucose	Without mediator	[69]
Enterobacter cloacae	Glucose	Methyl Viologen, Methylene blue (MB)	[70]
Saccharomyces cerevisiae	Glucose	Resorufin	[71]
Enterococcus faecium	Glucose	Pyocyanin	[72]
Aeromonas hydrophila	Glucose, Acetate	Without mediator	[73]
Shewanella putrefaciens	Lactate	Without mediator	[74]
Geobacter sulfurreducens	Acetate	Without mediator	[75]
Streptococcus lactis	Glucose	Ferric Chelate complex	[76]
Activated sludge	Wastewater	Without mediator	[77]
Proteus vulgaris	Glucose, Maltose, Galactose	Thionin	[78]
Domestic wastewater	Glucose, Xylose	Humic acid	[79]
Gluconobacter oxydans	Glucose	2-hydroxy-1,4-naphthoquinone (HNQ), Resazurin, Thionine	[78]
Klebsiella pneumoniae	Glucose	2-hydroxy-1,4-naphthoquinone (HNQ)	[80]
Shewanella oneidensis	Lactate	Anthraquinone-2,6-disulfonate (AQDS)	[81]
Shewanella putrefaciens	Lactate, Pyruvate, Acetate	Neutral red (NR)	[65]
Actinobacillus succinogenes	Glucose	Neutral red (NR), Thionine	[82]
Mixed consortium	Glucose, Sucrose	Without mediator	[83]
Micrococcus luteus	Glucose	Thionine	[84]

Typically, the microbial cell membranes, consisting of materials such as polysaccharides and essential lipids, are non-conductive. The anode chamber contains the microorganisms (catalyst) and the anode electrode. It is typical to feed this chamber with the organic substrate of wastewater in addition to a redox mediator (unless it is a mediator-less MFC system) (Figure 4). Accordingly, the MFC system may be classified based on the external mediator requirements. The two most-known types are the mediator and mediator-less MFCs. In the first type, a syntactic mediator (chemical) is added to mediate the electron transfer from the microorganism to the anode. In an indirect MFC and a mediator MFC (Figure 4A,B, respectively), fermentative microorganisms require the addition of artificial mediators that can shuttle the electrons between the cell membrane and the anode. Figure 4B shows the required redox couple (oxidation–reduction) of the e-mediator. In this type, the mediator is required, as the used microorganisms in the MFC are unable to donate the electrons in a direct fashion due to the nonconductive cell surface. Many types of mediators are used, including benzylviologen, phenothiazine and others [20]. Microorganisms such as enterococcus and pseudomonas, for example, may generate their own electrode shuttles and, accordingly, facilitate the electron transfer (Figure 4A). Bacteria were reported to produce natural mediators under some stressed conditions (Figure 4B) [20]. Direct transportation of electrons in MFCs is considered one of the most important mechanisms for electron transfer (Figure 4C). Here, the electrons are typically generated during the respiration of electroactive bacteria to the anode.

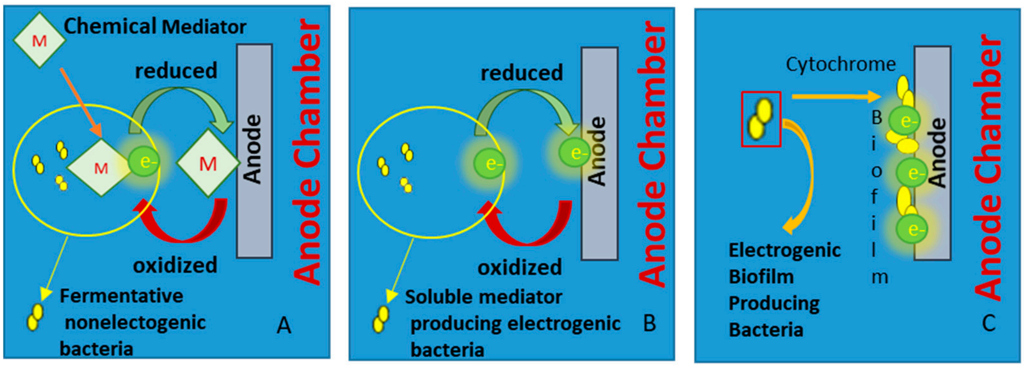

Figure 4. MFC electron transfer mechanisms: (**A**) indirect MFC; (**B**) mediator MFC; (**C**) mediator-less MFC.

When a MFC is inoculated with anaerobic sludge or marine sediments, mixed cultured microbes are present in the anode chamber. MFCs with mixed cultures have great performance most of the time. The use of complex mixed cultures allows for a much broader substrate usage. The bacteria in the MFC system can utilize a number of substrates, such as wastewaters with biodegradable organic matter that is utilized as the main energy source.

4. Features of MFCs and Operating Mechanisms

As can be seen in Figure 5, MFCs typically consist of an aerobic cathodic chamber and an anaerobic anodic chamber. Each is separated by a proton exchange membrane or an ion exchange membrane (PEM), as can be seen in Figure 5. The electrogenic bacteria's redox reaction generates bioelectricity.

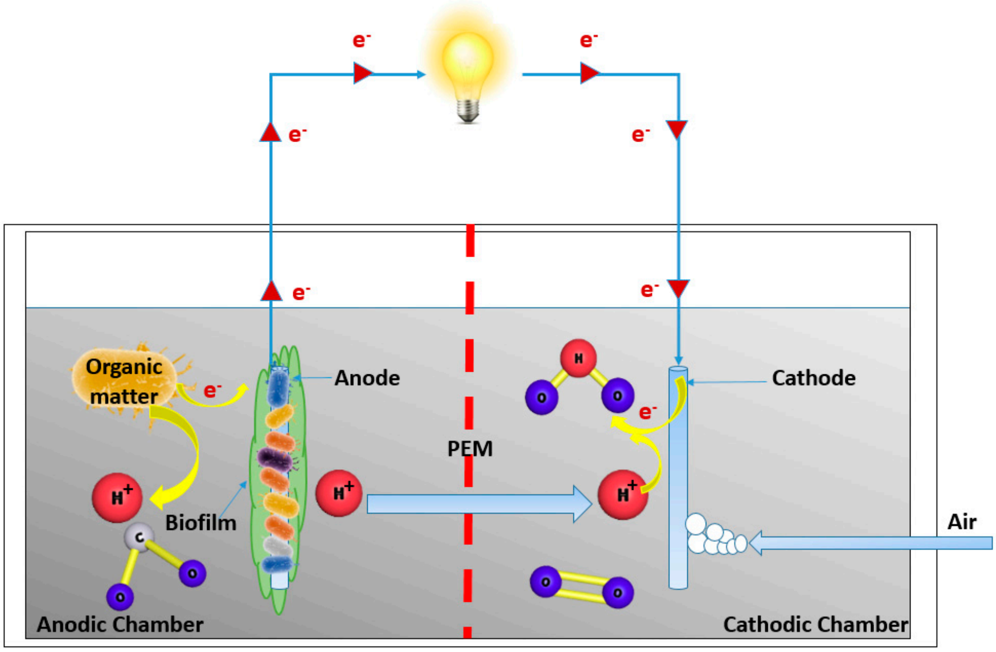

Figure 5. Schematic diagram of a microbial fuel cell (MFC).

The proton exchange membrane is a selective membrane that allows protons to diffuse from the anode to the cathode and that prohibits the diffusion of oxygen to the anode chamber [85,86]. The organic compounds, including carbohydrates found in wastewater, are oxidized in an anodic chamber as electron donors via active microorganisms, resulting in the production of electrons and protons. After that, the electrons are moved to the anode, then to the cathode in the cathodic chamber, where they produce electricity [87]. The protons then diffuse into the PEM and to the cathode chamber, where an electron acceptor, electrons, and protons combine to produce water.

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Table 2. Typical components of a microbial fuel cell.

Component	Materials	Requirement
Anodic chamber	Glass, Plexiglas, polycarbonate	Required
Cathodic chamber	Glass, Plexiglas, polycarbonate	Not required
Cathode	Graphite, carbon paper, graphite felt, Pt	Required
Anode	Graphite, carbon paper, Pt, reticulated vitreous carbon (RVC)	Required
Electrode catalyst	Pt, MnO_2, polyaniline, Fe^{3+}	Not required
Proton exchange system	Proton exchange membrane (PEM): Ultrex, Nafion	Required

In effective designs, it is essential to separate the chambers to ensure that full anaerobic conditions are maintained in the anodic section. The PEM plays an essential role in separating the two chambers, having specific selectivity for certain protons, maintaining high conductivity, and having high mechanical strength and durability for the long operation of the system. As shown in Figure 5, the PEM membrane separates the anode and cathode chambers in a typical MFC system. As indicated earlier, the PEM plays a fundamental role in power production in the cell as it controls the movement of protons from the anode compartment to the cathode one. The movement can be affected by the concentration polarization effect on the PEM membrane and can cause reduced power generation in the system. Another function of the PEM is the control of the substrate flux and O_2 diffusion towards the anode chamber.

I. Anodic reaction

$$(CH_2O)_n + nH_2O \rightarrow nCO_2 + 4ne^- + 4nH^+ \quad (1)$$

II. Cathodic reaction

$$O_2 + 4ne^- + 4\,nH^+ \rightarrow 2H_2O \quad (2)$$

In the anode chamber, the substrate is filled with exoelectrogen bacteria that grow and oxidize the organic substrates, transferring the produced electrons to the electron accepters found outside their cells. In addition, protons can be produced but are moved from the anode to the cathode compartments using deferential potential through the PEM. This is in contrast to the movement of electrons, as they require an external circuit [20].

Enhanced cell and electrode design, system optimization, and the addition of mediators could enhance the electron transfer and the system's effectiveness.

Anode materials are selected in such a way to ensure the establishment of a favorable environment for the development of active biofilms. Carbon-based materials such as cloth and different fibrous materials are utilized as anode materials especially if exoelectrogens are used in the MFC system. Other polymer-based materials such as polytetrafluoroethylene are preferred. Carbon nanotubes were also reported as means to feasibly transfer the electrons and enhance the surface area of the anode [20].

One of the most important parts of the MFC is the cathode material, as it has a main impact on the produced power in the system. Materials with high redox potential are preferred, such as graphite, Cu, Pt, and most commonly, carbon paper. The use of Pt in the cathode compartment has been shown to improve MFC system performance by increasing the reaction rate and decreasing the cathode reaction activation energy. The enhancement was reported in the first 24 h of operation in comparison to non-Pt-based cathode materials. After this time, both Pt-based and non-Pt-based cathode materials performed similarly with respect to power generation from the MFC system. Recent studies have shown that metal carbon nitrogen materials are outperforming Pt-based cathodes with respect to durability and prolonged system operation. Such materials could be easily produced using a variety of transition metals and precursors containing nitrogen and carbon.

5. Microbial Fuel Cells (MFCs) Applications in Wastewater Treatment

Microbial fuel cells have attracted great attention in recent decades as a distinguishable sustainable technology for generating energy and treating wastewater. Microbial fuel cells have proven to be a better option for wastewater treatment than conventional technologies because of their higher conversion efficiency and lower solid waste generation. In addition, they are capable of operating at any ambient temperature. Furthermore, by completely removing chemical oxygen demand (COD) and other contaminants, MFCs are fully capable of producing power densities of 4200 mW/m^3 [89–92].

The reactor configuration is a major factor that can affect the performance of the MFC. Designs can consist of single or dual chambers, as described in Figure 6. In the single-chamber design (Figure 6A), the cathode is kept in direct contact with air. The main obvious advantage of this type of design is the simple and less-costly design, as no separate cathode compartment is required. As air is in direct contact with the cathode, no aeration is required either. Separate anode and cathode chambers are required in the dual MFC design (Figure 6B), where both chambers are separated by the membrane. One of the main advantages of this design is the flexibility of the system, as each of the chambers can be operated in a batch or continued mode as required. The decision regarding the use of a single or dual design will depend on many operational and system requirements.

Table 3 shows the performance of various types of MFCs in the treatment of several types of wastewater, proving the efficiency of MFCs in their treatment applications.

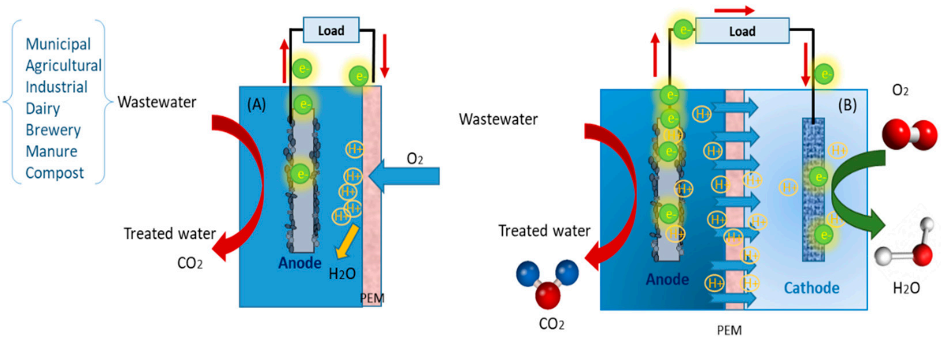

Figure 6. MFC design: (**A**) single cell and (**B**) dual cell.

Table 3. Various power outputs from different MFC configurations and wastewater feed.

MFC Configuration	Wastewater	COD Removal (%)	Power	References
50 stacked triple chambers	Municipal	70	125 W/m^3	[93]
4 single	Municipal	40	82 mW/m^2	[94]
Dual	Swine	83	13 mW/m^2	[95]
Dual	Industrial acid mine and municipal	15	1188 mW/m^2	[96]
Up-flow	Retting	70	254 mW/m^2	[97]
Single	Wood hydrothermal	94	178 mW/m^2	[98]
Flat panel	Domestic	85	6.3 W/m^3	[99]
Anaerobic baffled reactor-MFC	Fecal	93	450 mV	[100]
Single	Synthetic	57	14.41 mW/m^2	[101]
3 dual stacked	Biological treatment	84	822 mW/m^2	[102]
Up-flow MBR-MFC	Synthetic	85	44.4 mW/m^2	[103]
MBR-MFC	Synthetic	87.8	2.18 W/m^3	[104]
Single	Petroleum refinery	47	132 mW/m^2	[105]
Dual	Hospital	60	14 W/m^3	[106]
Single	Sludge	25.8	17.8 W/m^3	[107]
MBR-Dual	Paper	51.3	56.1 mW/m^2	[108]
CW-MFC	Azo dye	85.66	61.9 mW/m^3	[109]
3 chambers	Synthetic	97	111 mW/m^3	[110]
5 stacked MFCs	Synthetic	97	50.9 W/m^3	[111]
Up-flow	Seafood	83	105 mW/m^2	[112]
Up-flow CW-MFC	Synthetic	99	93 mW/m^3	[113]
Dual	Spent caustic	98	82.1 mV	[114]
Dual	Combined industrial	77.4	769 mV	[115]
2 dual in parallel	Agro-food	80	27.3 W/m^3	[116]
Dual	Brewery	82	8.001 µW/cm^2	[117]
MBR-dual	Synthetic	90	1358 mW/m^3	[118]
Dual	Yeast	90	6.1 mW	[119]
Dual	Rice mill	85.22	656.1 mW/m^3	[120]
Dual	Vegetable oil	80	5839 mV	[121]
Stacked MFC	Daily	52	290 mW/m^2	[122]
96 tubular duals	Municipal	75	200 mW	[123]
4 single	Domestic	54.2	300 mW/m^2	[124]
Single	Yogurt	87	1043 mW/m^2	[125]
Up-flow single	Dairy	94	3.5 W/m^3	[126]
Single	Tannery	88	7 mW/m^2	[127]

Traditional wastewater treatment technologies for municipal and industrial waste under aerobic conditions consume a huge amount of energy and resources. As a result, large amounts of surplus sludge are produced, which must be treated. In comparison to current technologies, using microbial fuel cells for treating wastewater has many advantages. While purifying wastewater, MFCs are capable of recovering chemical energy and converting it into electricity. Furthermore, MFC technology consumes far less energy and produces far less sludge compared to other traditional technologies. Sludge disposal is well-known to be costly, and it significantly raises water treatment costs. Microorganisms consume all the energy from organic compounds in an aerobic process, but only a small part of this energy is available to them for growth. On the other hand, the majority of the energy is converted into electrical power in MFCs. In a subsequent step, the energy produced by MFCs can be reused as feedback energy in the same wastewater treatment process. Some xenobiotic compounds can also be metabolized by MFCs.

Early in 1991, MFC technology was considered applicable and efficient for wastewater treatment. Since then, wastewater from various sources that are high in organic materials have been applied successfully as a fuel source in MFCs. Large amounts of organic compounds can be found in sewage and industrial wastewater, which can be used as fuel in MFCs [128].

5.1. Municipal or Domestic Wastewater Treatment via MFCs

The high cost and low output energy of MFCs are considered a huge concern for their potential use in wastewater treatment. Koffi and Okabe [129] synthesized serpentine up-flow MFCs with a polyvinylidene fluoride (PVDF)-based activated carbon (AC) air-cathode (MFC-PVDF/AC) that have been continuously operated for over 6 months with real domestic wastewater as a substrate. Without significant water leakage, the MFC-PVDF/ACs developed by the authors achieved average total COD removal rates (5.11–0.94 kg t COD/m^3/d) and power densities (3.96–3.01 W/m^3) that were even higher than those of MFCs fitted with Pt-based air-cathodes (MFC-Pts). In addition, the authors have created a low-voltage booster (LVB) to raise the low output voltage of MFC-PVDF/ACs, which is less than 0.4 V. The authors have successfully increased the voltage from less than 0.4 V to 4.35–5.2 V, which is capable of turning on three LED bulbs for more than 12 days. The results of this study show that the developed MFC-PVDF/AC and the LVB circuit have superb performance for municipal wastewater treatment and functional power generation, indicating that it could be successfully implemented in the wastewater treatment industry [129]. In a further study, Corbella et al. [130] evaluated the possible usage of constructed wetland microbial fuel cells (CW-MFC) as a COD evaluation method for domestic wastewater. The authors in this study used four lab-scale CW-MFCs that were set up and fed with different COD concentrations of pre-settled domestic wastewater. Two diverse anodic materials were examined under laboratory conditions (graphite rods and gravel). The results of the study show that, due to a lack of precision after several weeks of use, the CW-MFC can be applied as a method for qualitative continuous influent water quality assessment [130].

The cost of using Nafion as a proton exchange membrane in MFCs is high, and operational issues such as biofouling and fuel crossover restrict the device's use in harvesting energy from domestic wastewaters. Das et al. [131] have used poly(vinyl alcohol) (PVA) cross-linked with glutaraldehyde (GA) as a relatively low-cost and efficient membrane for MFCs, a simple route adapted to fabricate a Nafion-alternative membrane. The results of the authors' study show that the power density of the cross-linked membrane was greater than that of domestic wastewater-fed MFCs, with a maximum of 158.28 mW/m^2 for the fabricated membrane. Consequently, based on its efficiency and low installation cost, the PVA-GA membrane with antimicrobial activity, high power performance, and negligible fuel crossover shows promise as a separator in future MFCs [131].

Nutrient recovery has emerged as a viable choice for addressing the crucial issue of producing fertilizers, which is a critical component of a country's food protection. Because of its massive nutrient-rich base and quantity, municipal wastewater has the ability to be a major source of nutrients that can be recovered. As a significant candidate that can both recover nutrients and produce electricity, MFCs have been arousing interest. Ye et al. [132] designed and operated a two-chambered MFC in a continuous flow mode using artificial municipal wastewater as a substrate. The impacts of hydraulic retention time (HRT) on MFC nutrient recovery were investigated by the authors. The results of the authors' study show that the fabricated MFC in this study is a very promising strategy for removing organic matter, recovering nutrients, and producing electricity [132].

Vélez-Pérez et al. [96] evaluated the efficiency of dual-chamber microbial fuel cells in the co-treatment of industrial acid mine drainage (I-AMD) and municipal wastewater (MWW) (DC-MFC). In the anodic chamber, MWW and sewage sludge were used as inoculum-fuel. The cathode side of the chamber was fed with I-AMD. The results of the authors' study demonstrates that the organic matter removal efficiency was approximately 15%, and the wastewater alkalinity was decreased by greater than 50%. In addition, the $SO_2{}^{-4}$ concentration was decreased by up to 20% and $NO_3{}^-$ concentration was decreased by more than 90%. Moreover, several metalloid and heavy metal (HMs) removal values were shown in the cells: 84% for Cu, 77% for Al, 71% for Fe, 55% for Pb, 42% for Cd, and 42% for As. Finally, DC-MFC-A achieved a high power of 14,000 mW/m^3. The DC-MFCs were able to treat MWW, partially neutralize I-AMD, remove HMs, and generate bioelectricity all at the same time. As a result, DC-MFCs appear to be a promising bioremediation option for both MWW and I-AMD [96]. Liang [93] installed a 1000 L system of modularized MFCs for the treatment of practical municipal wastewater. The installed MFC was tested for over a year in two municipal wastewater treatment plants (MWTPs) under two distinct water flow connections to analyze their treatment capability for wastewater with both low and high initial COD concentrations. The COD removal rate in the MFC system was 70–90%, and the concentration of the COD in the MFC effluent has remained less than 50 mg L^{-1}. Additionally, when the MFC system was fed with artificial wastewater, it resulted in a maximum power density of 125 W m^{-3}; while using municipal wastewater, it produced a range of 7–60 W m^{-3}. Hence, according to the results of this study, the modularized MFC constructed has promising potential in the treatment of municipal wastewater [93].

The usage of a flat-panel air-cathode microbial fuel cell (FA-MFC) has been known to decrease the biodegradability and conductivity of domestic waste. Park et al. [133] used FA-MFCs with three anode spacing conditions and different flow rates to test the normalized energy recovery (NER) based on the volume of wastewater treated (NERV) and chemical oxygen demand (COD) removal (NERCOD). The results of the authors' study show that, at different spacings, current generation was the same; however, the removal of COD was influenced by the flow rates. Furthermore, in all anode spacing conditions, the NERV for both domestic wastewater and acetate showed strong agreement with flow rates. Independent of anode spacing, the NERCOD findings had a negative correlation with the removal rates of the COD. Consequently, this study demonstrates that FA-MFC is a very efficient candidate for an energy-efficient wastewater treatment technology [133].

Over the last decades, the use of constructed wetlands as microbial fuel cells (CW-MFCs) has been gaining popularity to increase wastewater treatment efficiency while also producing electricity. However, the knowledge about the design and operation of CW-MFCs is still lacking and needs to be studied well, specifically for real domestic wastewater treatment. Hence, Corbella [134] aimed to quantify the degree to which membrane-less MFCs improved treatment efficiency by simulating the core of a shallow, un-planted, horizontal, subsurface flow-constructed wetland. The authors set up six membrane-less MFCs in the lab and filled them with domestic wastewater in batch mode for 13 weeks. The results of the study showed that regardless of the anode material used, the best operating condition for maximizing MFC treatment performance was 220. Hence, this study shows that the use of constructed wetlands as MFCs is an efficient strategy for increasing the efficiency of domestic wastewater treatment [134]. Table 4 shows the performance of various types of MFCs in domestic wastewater treatment applications, proving the efficiency of MFCs in this treatment application.

Table 4. Performance of MFCs in domestic wastewater.

Type of MFC	Maximum Power Density (mW/m^2)	COD Removal (%)	COD Reduction (%)	Reference
Air-cathode MFC	10^3	71.0	-	[135]
Two-chamber MFC Combining four MFC reactors and an AFMBR	25	30.0	-	[136]
Single-chamber air-cathode MFC	464	40.0–50.0	-	[137]
• SEA MFC • SPA MFC	• 328 ± 11 • 282 ± 29	-	• 62.0 ± 4.0% to 94.0 ± 1.0% • 81.0 ± 5.0% to 93.0 ± 3.0%	[138]
Air-cathode MFC	420	-	44	[139]
Stackable horizontal MFC (SHMFC)	116	79.0 ± 7.0	-	[140]
Single-chamber microbial fuel cell (SCMFC)	26	80	-	[141]
Flat plate MFC (FPMFC)	72 ± 1	42	-	[142]
Air-biocathode microbial fuel cell-membrane bioreactor (MFC-MBR)	0.38	97	-	[143]

5.2. Industrial Wastewater

The microbial fuel cell (MFC) technology has piqued the interest of researchers in the last few years as a means of producing bioenergy while also treating wastewater. In order to be practical, these devices need low-cost cathode catalysts for the reaction of oxygen reduction. The use of MFCs for industrial wastewater treatment, which typically contains several contaminants, is gaining great attraction. Ortiz-Martínez et al. [144] investigated the application of mixed manganese oxides with copper and nickel synthesized by co-precipitation for use in MFC devices fed with industrial wastewater. The new catalysts were tested based on their ability to remove chemical oxygen and their power production. The oxide with the formula $NiMn_2O_4$ performed well in terms of power density, achieving 80% of the power density obtained with Pt. With no prior pretreatment, substantial COD removal from industrial wastewater was achieved after 168 h of operation [144]. Srikanth et al. [145] investigated the treatment of refinery wastewater (RW) using microbial fuel cells (MFC) in a batch mode, then continuous mode, with hydraulic retention times of 8 and 16 h (HRT), respectively. The output of the MFC was assessed by the authors in terms of power density, organics removal, particular pollutants removal, and the efficiency of energy conversion in relation to the operation mode. The results of the authors' study show that during continuous mode operation, a higher power density of 225 ± 1.4 mW/m^2 was observed, and a high substrate degradation of 84.4 ± 0.8%. Furthermore, the batch mode operation demonstrated high substrate degradation (81.8 ± 1.8%). Overall, the current study demonstrated the feasibility of using RW as a power generation substrate in MFCs, as well as its treatment [145]. In a microbial fuel cell (MFC) using a selectively enriched hydrogen-generating (acidogenic) mixed culture, the generation of bioelectricity from anaerobic chemical wastewater treatment was assessed in a further study by Venkata Mohan et al. [146]. The MFC productivity was assessed by the authors via the usage of non-coated plain graphite electrodes at two organic/substrate loading rates in terms of wastewater treatment and bioelectricity production in an acidophilic microenvironment at ambient pressures and temperatures. The results of this study show that in situ bioelectricity generation and wastewater treatment are both feasible. The applied OLR was found to affect the performance of MFCs in terms of power generation and wastewater treatment. At stable operating conditions, maximum voltages of 716 mV and 731 mV were observed by the authors. At applied 50 Ω resistance, the maximum power yield (0.73 W/kg COD_R and 0.49 W kg/COD_R) and current density (339.87 mA/m^2 and 355.43 mA/m^2) were also observed. This study shows that the designed MFC is capable of wastewater treatment and bioelectricity generation [146].

Efficient wastewater treatment and processes with long-term energy efficiency are two of the most pressing concerns in the liquid waste management industry today. Agro-industrial wastewater contains high-strength organic contaminants that, if not treated properly, can have negative consequences on the receiving water bodies. Microbial fuel cells (MFCs) combine wastewater treatment with direct chemical-to-electrical energy conversion. As a result of the organic matter content and biodegradability, wastewater from the agro-food industry appears to be especially promising [147]. Hence, Cecconnet et al. [116] aimed in their study to determine the bioelectrochemical treatability of dairy wastewater by MFCs, as well as the operational effects on MFC electrical efficiency and possible strategies for reducing overpotentials. The authors operated two parallel MFC reactors fed with undiluted dairy wastewater in continuous control for 2.5 months. The results of this study show that MFCs can treat these types of industrial effluents with high organic matter removal and recover a maximum power density of greater than 27 W/m^3. Thus, energy recovery from organic waste treatment is a viable method for pursuing renewable technologies [116].

Environmentalists are usually facing a global challenge in safely disposing polluted water. Present treatment systems are insufficiently capable of addressing wastewater contamination and meet the ever-increasing demand for water sanitation. The microbial fuel cell (MFC), on the other hand, is a modern technology that not only treats wastewater but also produces electricity. Firdous et al. [121] examined the generation of electricity by a dual-chambered MFC during wastewater treatment of vegetable oil industries in Pakistan. The microbial fuel cells were studied in the lab by the authors at two different temperatures (25 and 35 °C). The authors mentioned that, in a two-compartment MFC reactor, a proton exchange membrane separated the anaerobic anode and aerobic cathode chambers. A total of 20 wastewater samples from vegetable oil industrial effluents were obtained and treated in MFC for 72 h. The efficiency of the MFC improved as the temperature and time were increased, according to the results of this study. At 35 °C, the highest chemical oxygen demand (COD) removal efficiency reached was 80 to 90% while the maximum voltage was 5839 mV. This study demonstrates that MFCs are capable of treating industrial wastewater efficiently [121].

5.3. Brewery Wastewater

Microbial fuel cells and electro-active bacteria are used to obtain chemical energy from wastewater, making wastewater a potential renewable energy source [148–154]. Various brewery industries discharge wastewater into the environment, causing severe problems to the environment. Negassa et al. [151] demonstrated, by inoculating locally isolated microorganisms into double-chamber MFCs, the treatment of brewery wastewater while also developing bioelectricity. The authors have extracted microorganisms locally from brewery wastewater, brewery waste sludge, and food processing waste sludge. The results of the authors' study shows that the isolated microorganisms from brewery waste sludge have performed better than the bacteria isolated from brewery wastewater and food processing industry waste sludge. Moreover, MFCs with the isolated microorganisms from brewery waste sludge resulted in a maximum power densities of 0.8 W/m^3 and 0.35 W/m^3 using synthetic and real brewery wastewater, respectively. Furthermore, the maximum COD removal efficiency was 83%. This study demonstrates that a treatment of brewery wastewater should be attained via locally isolated microorganisms to produce clean and renewable energy [151]. Liu et al. [153] developed a UASB-MFC dual sensors system to treat brewery wastewater. The MFCs developed in this study have a voltage range of 0.34–0.42 V, a COD removal rate of about 90%, and an $NH4^+$–N concentration of less than 15 mg/L. The long-term performance of MFCs was studied by the authors using electrochemical methods, and it was discovered that biosensor degradation was primarily caused by Ca^{2+} and Mg^{2+} precipitation on the cathode surface, which was influenced by concentration. Hence, cleaning the electrode using a self-enhanced method that does not require external assistance ECS (electrode connection switching) will boost the efficiency of MFCs to 83.2–84.6% [153].

Microbial fuel cells (MFCs) have been demonstrated as an efficient wastewater treatment system. Integrating MFCs into present wastewater treatment plants can lower operating costs while also improving treatment efficiency, and the scaling up of MFCs will be critical. However, only a small number of studies have recorded successful scale-up efforts. Before MFCs can be commercialized, the fabrication costs, treatment efficiency, and operating lifetime must all be optimized. To examine these factors, a 20-L MFC system with two 10-L MFC reactors was operated for almost one year with brewery wastewater by Lu et al. [154]. The highest COD removal efficiency attained by the authors was $94.6 \pm 1.0\%$. Thus, based on this study, MFCs are capable of supporting several rates of treatment over a long time period and are successful enough in maintaining high efficiency treatment processes [154].

6. Nutrient Removal in MFCs

6.1. Nitrogen Removal in MFCs

The removal of nitrogen from wastewater is a critical process that requires large amounts of energy, necessitating the development of more energy-efficient treatment methods. Koffi and Okabe [155], electrically operated a single-chamber microbial electrolysis cell (MEC) using a double-chamber microbial fuel cell (MFC) to investigate the removal efficiency of bioelectrochemical ammonium nitrogen (NH_4^+-N) from real municipal wastewater without requiring aeration with large amounts of energy. At several applied voltages, the total nitrogen (TN) removal rates were obtained by the authors. The results of the authors' study show that, without aeration and at a voltage of 0.8 V, a TN removal rate of 95 ± 42 g-TN m^{-3} d^{-1} was achieved. The nitrogen removal rates obtained in this study are greater than the previously recorded values in past studies. Thus, the MFC-driven single-chamber MEC provided by this study is a successful method for the removal of nitrogen from wastewater [155].

The residual nitrogen present in the effluents of municipal wastewater degrades the aquatic systems, and the addition of an external source of carbon is the most commonly used approach to fix this issue. The external carbon sources that are mostly used by wastewater treatment industries that are limited on carbon are agricultural biomasses, due to their low cost and high availability. Hence, the feasibility of introducing agricultural wastes in a microbial fuel cell-constructed wetland (MFC-CW) to optimize the production of bioelectricity and the removal of nitrogen was estimated, and the results obtained were compared by the authors to those in an MFC study by Tao et al. [156]. Several agricultural wastes, including corncobs, rice husks, and straw, were compared by the authors, and the corncob turned out to release more carbon. The results of the authors' study show that corncob carbon release was a diffusion process, with the highest COD removal of 47.6 mg $(gL)^{-1}$ fitting a second-order kinetics. Corncob addition improved nutrient removal dramatically in MFC-CWs with an original influent COD of 22 mg L^{-1}, with an overall total nitrogen (TN), nitrate nitrogen (NO_3-N), and ammonia nitrogen (NH_4^+–N) removal of $86.6 \pm 1.6\%$, $97.2 \pm 0.3\%$, and $73.1 \pm 2.8\%$, respectively. Furthermore, the production of bioelectricity was improved with a maximum power density of 23.5 mW/m^3, despite a small increase in internal resistance. This study proves that when treating carbon-limited wastewater, MFC-CW can remove nitrogen, recover electricity, and handle agricultural wastes in a cost-effective manner [156].

6.2. Sulphate Removal MFC

In a MFC, the mass balance of organic matter, reaction kinetics, and sulphate transformation are studied by changing the sulphate and the COD concentrations in the substrate. Experiments show that in a MFC with an inlet COD/sulphate ratio of 0.75, a sulphate removal greater that 99% can be reached, yielding about 1.33 kg/m^3 COD removal per day. These fascinating results make MFC an interesting sulphate removal technique [157]. Various research studies have focused their work on the removal of sulphates from wastewater. Chakraborty et al. [158] demonstrated that sodium dodecyl sulphate (SDS) can be biodegraded effectively in MFC after a 12-h retention period. This was the first to quantify the use of MFCs for SDS degradation and its effect on MFC power generation and organic matter removal capability. The SDS-induced microbial diversification of the anodic biofilm was also demonstrated in this study, which correlates with the established SDS degradation pathway. The authors have successfully reached more than 70% SDS removal efficiency in their study [158].

MFCs have been previously applied in the conversion of carbon-based substrates into electricity. Sulfur compounds, on the other hand, are abundant in organic waste and wastewater. Rabaey et al. [159] converted dissolved sulfide to elemental sulfur by using a MFC with a hexacyanoferrate cathodic electrolyte. Two types of MFCs were used by the authors: a square type with a closed cathode compartment, and a tubular type with an open cathode compartment. In addition, MFCs were attached to an anaerobic up-flow anaerobic sludge blanket reactor, resulting in total sulfide and acetate removals of up to 98% and 46%, respectively. The results of the authors' study show that the MFCs were successfully able to simultaneously remove sulfate via sulfide [159]. Furthermore, sulfate-reducing bacteria (SRB) can be used to biologically remove sulfur via reducing sulfate to sulfide, which is then oxidized into elemental sulfur (S(0)) by sulfide-oxidizing bacteria (SOB) for recovery. Lee et al. [160] treated, in their study, sulfate+organic carbon wastewaters using a MFC cultivated with the SRB+SOB anodic biofilm. The results of the authors' study show that the SRB cells in the biofilm have efficiently converted excess sulfate ions to sulfide, and the generated sulfide was then diffused to the neighboring SOB cells, where it was converted to elemental sulfur S(0). The electron flux of the MFC was primarily determined by cell-to-cell sulfide transport [160].

7. Metal Removal in MFCs

In the last 15 years, the bio-electrochemical technology of microbial fuel cells has arisen as a modern and appealing technology that presents a new pathway for the generation of electricity and the removal and recovery of heavy metals from wastewater. The MFC approach precedes the traditional techniques in the following manner: it is energy intensive, produces less amounts of sludge, and has low efficacy at high concentrations. Studies demonstrate that the pH, mixed metal systems, the gas environment in the cathode, the composition of the biofilm, and the metal redox potential all have a major effect on the effectiveness of MFCs in the removal and recovery of metal and the generation of power [161].

The removal and recovery of organic pollutants using MFCs is a clean approach for industries to develop greener technologies. Lim et al. [162] investigated the ability of MFCs to remove zinc from industrial effluents. The results of the authors' study show that, for industrial and synthetic samples and after 22 h of operation, the removal of Zn^{2+} in MFCs was greater 96%. Furthermore, in the industrial samples, the electroprecipitation process was found to be more dominant than the electrodeposition process. According to the findings of this study, MFCs can be used to remove heavy metals in a safe and environmentally friendly manner without the use of electricity or chemical inputs [162]. In a further study, Wang et al. [163] used an up-flow constructed wetlands-microbial fuel cell

(CW-MFC) to characterize a microbial community structure and resistance gene (CzcA) for the treatment of wastewater that is contaminated with Zn (II). The results of the authors' study show that the CW-MFC had a high Zn (II) and COD removal efficiency, in addition to a high power density [163].

Wastewater treatment and precious metals recovery via MFCs offers an appealing solution for a cleaner environment and industrial processes. Recovering silver from wastewater and valorization as silver nanoflakes (AgNFs) aids in the transformation from linear to circular economies by bringing waste material back into the production stream. Ali et al. [164] compared a MFC that is fed with silver-laden artificial wastewater (MFC-Ag) to a MFC fed with potassium ferricyanide (MFC-FC) and a MFC fed with phosphate buffer as catholyte (MFC-blank) in terms of bioelectrochemical performance. The authors' study results demonstrate that the silver removal and recovery efficiencies after 72 h of operation of MFC-Ag has reached 83 $\pm$ 0.7% and 67.8 $\pm$ 1, respectively. Furthermore, the MFC-Ag has appeared to have a greater maximum power density and current density compared to MFC-FC and MFC-blank. Additionally, MFC-Ag has a high coulombic efficiency and a low solution resistance, indicating that silver-laden wastewater has the potential for large-scale applications. This study offers a broad range of applications for scaling up the technology of MFCs with greater sustainability and limited facilities [164].

8. Challenges and Future Perspectives

Based on the published research, MFC systems offer great potential for sustainable wastewater treatment in addition to energy production. Although the studies published in this regard state the possible advantages of the system, a full-scale application is still in the early stages of use, and further optimization and scale-up studies are required.

As the current produced by the MFC cells depends on the ability of the used microorganisms to oxidize the available substrate and the effective transfer of the electrons between the electrodes, the efficiency depends on many variables. The type of waste used and its composition play an important role in influencing the resultant energy produced. In addition, other experimental variables related to the cell design, electrode types, and set-up conditions can easily affect the resultant bioelectricity production and degree of wastewater and pollutant degradation. Energy production is also heavily dependent on the type of substrate used, its initial concentration, and other operational parameters such as pH and temperature. Optimizing all such variables and a lack of consistency remains a main challenge for scaling up long-term industrial operations. Another main hindrance is associated with the costs related to the exchange membranes. Furthermore, regular cleaning to remove any biofouling deposits from the membranes can incur additional costs and provide high and undesirable resistance to electron movement, affecting power generation. The main costs are also associated with the expensive metal catalyst (for example, platinum). Alternative non-platinized cathodes are required and research studies are being conducted to find suitable replacements with similar performance to platinum. Manganese dioxide, stainless steel, and nickel alloys are cathode catalysts that may offer a great alternative to platinized ones. In addition, and as a main potential for the use of MFCs, is the fact that there has been a huge improvement in the power production of the MFC systems since its early reports in 2004. Further optimization in the future is expected to make MFCs a great contender in sustainable wastewater treatment, compared, for example, to anaerobic digestion.

MFCs have been shown in several studies to be an efficient candidate for the removal of various pollutants. Some limitations of MFCs for wastewater treatment should be further explored, including high cost and energy requirements. In order to further enhance the performance of wastewater treatment, more research should be focused on new MFC materials. Finally, a deeper understanding of the nature and role of electrode materials is needed. By advancing MFCs alone or in conjunction with other methods, multiform wastewater can be substantially degraded.

9. Conclusions

Wastewater is widely acknowledged as a significant source of contamination in the world. However, treating and recovering wastewater is hard to attain and maintain because most of the present wastewater treatment systems require large amounts of energy and are very expensive to build and operate. Thus, an alternative approach using microbial fuel cells (MFCs) is greatly investigated currently as a cost-effective and energy efficient pathway in the wastewater treatment field.

This review highlighted the main factors that still hinder the scaling up of this technology. The main issues can be summarized as:

1. Costs associated with platinum electrodes;
2. Biofouling effects on the membranes;
3. Non-consistence power generation depending on the type of substrate and operational conditions;
4. Monitoring and harvesting the power generated in the system.

In this review paper, the applications of MFCs in the removal of nutrients (nitrogen and sulphates) and precious metals from wastewater were also intensively reviewed. As a result, the efficacy of various MFCs in achieving sustainable power generation from wastewater has been critically addressed in this study. MFCs precede several other wastewater treatment methods in the following manner: they produce less sludge, require less energy to operate, and produce large amounts of energy. MFCs are also limited in their practical application due to the high cost of mediators and exchange membranes.

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