



## Review Microbial Fuel Cell United with Other Existing Technologies for Enhanced Power Generation and Efficient Wastewater Treatment

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**Abstract:** Nowadays, the world is experiencing an energy crisis due to extensive globalization and industrialization. Most of the sources of renewable energy are getting depleted, and thus, there is an urge to locate alternative routes to produce energy efficiently. Microbial fuel cell (MFC) is a favorable technology that utilizes electroactive microorganisms acting as a biocatalyst at the anode compartment converting organic matter present in sewage water for bioelectricity production and simultaneously treating wastewater. However, there are certain limitations with a typical standalone MFC for efficient energy recovery and its practical implementation, including low power output and high cost associated with treatment. There are various modifications carried out on MFC for eliminating the limitations of a stand-alone MFC. Examples of such modification include integration of microbial fuel cell with capacitive deionization technology, forward osmosis technology, anaerobic digester, and constructed wetland technology. This review describes various integrated MFC systems along with their potential application on an industrial scale for wastewater treatment, biofuel generation, and energy production. As a result, such integration of MFCs with existing systems is urgently needed to address the cost, fouling, durability, and sustainability-related issues of MFCs while also improving the grade of treatment received by effluent.

**Keywords:** microbial fuel cell; microbial electrochemical system; bioelectricity; power density; wastewater treatment; anaerobic digester

### 1. Introduction

Microbial fuel cell (MFC) can be operated based on the oxidation of biodegradable waste at an anode and oxygen reduction at a cathode [1,2]. Over the last few years, MFCs



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**Copyright:** © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). have received great interest in treating contaminated water; however, most of them are only used in the laboratory. An MFC technique provides treatment of contaminated water and electricity generation simultaneously [3]. The issues related to the power output of an MFC have been improved through a number of research studies. Nevertheless, at the pilot scale, the application of an MFC appears to be restricted, and power outputs appear to have plateaued; consequently, it failed to replace the existing wastewater treatment tools as a stand-alone technology. As a result, some MFC-based integrated techniques have evolved in recent years [4]. Upscaling studies of an MFC as a single unit have not yet been completed, although it has been achieved by many studies using stacked MFCs. Upscaling and practical applicability of MFCs are difficult due to their numerous disadvantages, as mentioned in further sections. The main disadvantage of an MFC is at the cathode region, where there is a shortage of terminal electron acceptor (TEA), which is considered a rate-limiting factor [5-7]. The majority of an MFC's efficiency depends on oxygen available at the cathode as a terminal electron acceptor and its efficient reduction on the cathode surface. Several studies have utilized a platinum catalyst at the cathode to improve efficiency and eliminate cathode issues. However, employing Pt is expensive and, in most situations, unsuitable for use in MFCs due to possibilities of poisoning. The size of an MFC is the second issue. The internal resistance of systems develops as the size of MFCs rises, which in turn reduces energy density. The other issue is maintaining electrode fouling in MFCs when they are applied on an industrial scale [8]. MFCs have been amalgamated with other existing technologies to overcome their limitations, as mentioned above [9]. In this context, numerous interesting hybrid MFC-based sewage treatment methods have been developed in recent decades, some of which show tremendous potential for future applications [10,11]. Hybrid approaches (i.e., combining two or more processes to establish a novel method that integrates the particular qualities of the original techniques) have been emerging. These integrated techniques provide a versatile environment for tackling some difficult challenges, and they often outperform a stand-alone technology [12]. This paper discusses various categories of integrated MFC systems, including integration with capacitive deionization technology, forward osmosis technology, anaerobic digester, constructed wetland technology, microbial desalination cells, and so forth. The integrated CDI-MFC system starts with the electrostatic segregation of ions from water, which is followed by the adsorption of the ions or other pollutants in the electrode-solution interface. This process is used to overcome the disadvantage of stand-alone MFCs of limited ion removal. On the other hand, an MFC integrated with dark fermentation (DF) is used for extensive H<sub>2</sub> generation via an anaerobic hydrogen production process. An MFCintegrated MBR is used to reduce the cost of energy produced in the process, whereas integration of an MFC with an FO membrane aids in the cost-effective treatment of salty water on an industrial scale with simultaneous production of energy.

Not only would such MFC-integrated systems lower overall costs, but they would also increase the combined system's sewage treatment efficacy and sustainability. Micropollutants and new pollutants, such as dyes, which are often not absorbed or decomposed by traditional procedures, can also be effectively removed by combining MFCs with other technologies. As a result, such integration of BESs with existing technologies is vitally needed to address the cost-related issues of BESs while also improving the degree of treatment received by sewage.

This paper reviews several technologies integrated with an MFC to attain maximum benefit in the form of improved wastewater effluent and energy production. It also highlights the disadvantages of a stand-alone bioelectrochemical system and explains the need for the development of integrated MFC systems. Further, future application of an integrated MFC system on the industrial scale is elaborated.

#### Stand-Alone Bioelectrochemical Systems and Their Limitations

Organic compounds such as acetate and glucose are oxidized anaerobically in an MFC, resulting in the liberation of protons, electrons, and CO<sub>2</sub>. The liberated protons

and electrons pass through a medium, which is an electrical circuit for electrons and a membrane for protons, to enter the cathode. The cathode is where water is generated by the reduction of protons and electrons using oxygen supplied from outside. The formation of water in the MFC is depicted by the equation below:

$$\begin{array}{l} CH_{3}COO^{-} + 4H_{2}O \rightarrow 2HCO_{3}^{-} + 9H^{+} + 8e^{-} \\ \\ 2O_{2} + 8H^{+} + 8e^{-} \rightarrow 4H_{2}O \end{array}$$

In MFC, the anode undergoes oxidation, while the cathode undergoes reduction. This oxidation and reduction create the potential difference between the electrodes, resulting in the generation of bioelectricity (Figure 1).



Figure 1. A typical stand-alone microbial fuel cell.

Considering the significance of electrodes in MFCs, selecting the right electrode material is an important part of the system's design. An effective cathode must have high conductivity and a high surface-area-to-volume (S/V) ratio, be noncorrosive, and have the least fouling. Cathode efficacy is critical in the generation of power in MFCs. The ORR is the most prevalent cathode reaction. One of the most limiting aspects of MFC operation is the steady reduction of oxygen on the interface of carbon or graphite electrodes, which results in a significant overpotential reduction. Hence, selecting the appropriate cathode material is important.

The chemical oxygen demand (COD) varies between 60–220 mg/L during the continuous flow system and 23–164 mg/L during the fed-batch system. The COD is also influenced due to wastewater COD concentrations, system alignments, and hydraulic retention time (HRT) [13]. Ghadge and Ghangrekar [14] constructed a 26 L cheap air cathode MFC employing clayware as a divider, claiming that the efficiency was steady for 14 months. With a CE of 5.1%, the MFC had a maximum power of 17.85 mW and removed 78% of the COD. Feng et al. [15] created a stacked large-scale horizontally constructed MFC (SHMFC) with such an overall capacity of 250 L that employs carbon brush electrodes to extract 116 mW of current (0.435 A) while also treating sewage. These SHMFC components may be used independently to process sewage and generate electricity without interfering with other SHMFC modules. Biofilm-based MFCs are unproductive in the elimination of minute entities and, therefore, lead to such high COD of the effluent [16]. Theoretically, the influents include constituents such as azo dyes, which are lethal to microorganisms and thus hamper the biofilm formation and electron transfer, thereby reducing the MFC's potential substantially [17]. Quite a few methods and approaches were employed and examined to improve the energy generation of MFCs [18]. These methods comprise multifaceted enhancement and adaptation techniques [19], improvement of electrode biocompatibility by surface modification, and assembly of three-dimensional electrodes [20]. Even after all these signs of progress, the usage of cost-effective carbon electrodes deprived of the usage of catalysis was established to cause overpotential of the cathode and restricted oxygen transformation competence [21]. The enhancements performed were quite inferior from the contaminated water upscaling viewpoint. These matters have been a massive hindrance in the industrial employment of an MFC as an impartial contaminated water management podium.

The productivity and consistency of the MFC have improved extraordinarily after long research and development. However, despite all these advancements, there are plenty of issues that need to be addressed and improved before this technique can be elevated and employed on a commercial scale. One of the most significant obstacles to the development of microbial fuel cells is the high cost of electrode and membrane components. Due to its arrangement and treatment, the MFC's capital cost is 30 times greater (on average) than that of conventional activated sludge treatment systems for domestic wastewater. Another major hindrance to the MFC's use in wastewater treatment is fouling at the membrane, which disrupts proton transfer. Other constraints of a stand-alone MFC include low electricity generation, electric current inconsistency, sustainability of electrode materials, cost of operating conditions, and so forth. Due to the versatility of the MFC operation, it is easy to integrate this technique with other processes and operations to increase power output and sludge reduction [9]. Additionally, there is a necessity for the invention and optimization of low-cost electrodes to simplify the scale-up procedure. The progress in the configuration of the MFC, the components engaged in the electrode/separator, and the approach of the process leads to the improvement of the physical and chemical confines of the MFC. Therefore, there is a requirement to discover actual assimilation of the complementary functions in the BES and enhance them for easy scale-up [22]. An MFC could produce sufficient energy and can be a modern-day alternative for wastewater treatment, but there are still limitations, such as cost-effectiveness, biofouling, and the need for treatment of the effluent as it cannot be directly discharged. Hence, MFCs need to be integrated with other technologies for more efficient wastewater treatment that eliminates the requirement for additional effluent treatment while simultaneously gaining extra benefits [23]. MFCs were accordingly integrated with a variety of other technologies for the pretreatment or post-treatment of wastewater. These included anaerobic digesters (AD), capacitive deionization (CDI), forward osmosis (FO), and membrane bioreactors (MBRs).

#### 2. Types of Integrated Microbial Fuel Cell System

Microbial fuel cell is an exclusive treatment for the management of contaminated water and energy production based on microbe–electrode interaction. Although the technology has developed significantly in recent years, it is still far from being employed in the practical field. Scalability problems, including external and internal resistance, size challenges, and cathode problems, are the reasons for its confined efficiency. MFCs appear to have the potential to be integrated into other processes, such as sediment MFCs (SMFCs), desalination cells (DS), membrane biofilters, and constructed wetlands (CWs). Some of these integrated systems are mentioned below.

#### 2.1. Integrated MFC-Capacitive Deionization (MFC-CDI) System

Several technologies were integrated with MFC to attain maximum benefit in the form of improved wastewater effluent and energy production. Capacitive deionization (CDI) was one of the recent technologies assimilated with MFC through diverse assemblies.

The integrated MFC-CDI system was tested to associate deionization through energy production and sewage water treatment [24]. CDI is a potential deionization alternative that can be used to remove salt from an aqueous solution to increase freshwater sources [23].

This process depends on the electrosorption that accompanies the charge separation for accumulation and releases considerable quantities of ions with the help of porous carbon electrodes. The mechanism starts with the electrostatic segregation of the ions from the water. This is followed by the adsorption of the ions in the electrode–solution interface. The initiation of the procedure requires a potential gradient (usually less than 1.2 V) to be applied among the electrodes. As the potential difference required is very low, continuous flow MFCs were employed to improve the function of the CDI for the elimination of electrolytes. This integration of MFC-CDI can be used in the post-treatment of the effluent containing ionic pollutants such as  $PO_4^{3-}$  and  $NO^{3-}$ . Here, the MFC is used as a secondary treatment for  $NH_4^+$  and the removal of organic carbon, along with electricity generation, whereas CDI is used to desalinate and decontaminate wastewater effluent [25], as illustrated in Figure 2.



Figure 2. MFC-capacitive deionization (MFC-CDI) system.

In another study, a maximum output voltage of 0.63 V was achieved using a corresponding assembly of dual MFCs, which, when applied to the CDI, was able to attain potential for the elimination of NaCl (60%) from wastewater [26]. The integration of dual MFCs in the corresponding assembly was used to power CDI in another setup, which resulted in a potential of 0.49 V. This aided in the removal of the multi-ionic species from the polluted water and also helped to gain an improved COD elimination of up to 90% [23]. An H-type MFC was functioned in batch mode and was used to supply exterior power to the CDI for less quantity of salt (60 mg/mL) treatment [27]. A salt elimination rate of 35.6 mg/L h was observed in this setup. The key factors influencing the desalination efficiency were ascertained by examining five different circuit configurations [28]. These factors were the internal resistance of the CDI and MFC, the capacitance of the CDI, and the open-circuit voltage of the MFC.

#### 2.2. Membrane-Based MFCs and Membrane Bioreactor-Microbial Fuel Cells

Several studies were conducted where MFCs were integrated with different types of bioreactors to improve the quality of the wastewater (Figure 3). One such method was the traditional activated sludge process integrated with an MFC [29]. The aeration tank was utilized as the cathodic compartment, and the catalyst was the developing aerobic biofilm. The biofilm is an inexpensive alternative for the catalyst and helps in reducing the overall cost of operating this integrated system. The continuous flow process was maintained by a clarifier, which was tailed to the aeration tank. However, extra costs were incurred due to

the installation of the clarifier and the sludge pumping. As this integrated system faced obstacles during industrial scale-up, another setup integrating the MFCs with a sequencing batch reactor (SBR) was introduced. This approach was explored to estimate COD removal and energy production. This setup consisted of a biocathode-equipped membrane-less MFC combined with an SBR. This helps in the retrieval of energy as electricity from the aeration, hence reducing the operating cost for SBR operation [30].



Figure 3. Representation of a typical MFC-integrated MBR reactor.

The MFC combined systems were used to examine different types of wastewaters. For example, a combined system of an up-flow anaerobic sludge blanket reactor, MFC, and biological aerated filter (UASB-MFC-BAF) was employed to treat molasses water [31]. In this arrangement, all three components complement each other by filling up the gaps where the others lack. Here, the UASB was used primarily for sulfate reduction and removal of COD. The resultant sulfide is oxidized by the use of the MFC, along with the production of energy. The phenolic compounds were decolorized and degraded in the BAF. The azo dye, Acid Orange 7 (AO-7), underwent decolorization in the MFC-integrated setup and was denatured into less poisonous complexes. The AO-7 was degraded by a reduction process in the MFC into the constituent aromatic amines [32]. Another system was devised for the degradation of Congo red, where the integration of the MFC and a catalytic oxygen reactor (COR) was used. In this system, Congo red was degraded with the help of oxidants in the existence of a catalyst [30].

One commercially available technology used for the treatment of wastewater is the MBR (membrane bioreactor), which can be combined with the MFC to reduce the cost of energy and develop the superiority of an effluent [33]. These advantages were the reason that many studies were conducted on the MFC-MBR system to check its efficiency and viability. The system born out of the assimilation of the advantages of the MFC and MBR was named a "bioelectrochemical membrane reactor" [20]. Although the scheme of this bioreactor was quite complicated, it was capable of reaching an extreme power density = of  $4.35 \text{ W/m}^3$ . It has also demonstrated improved contaminant removal, which was credited to solid elimination and significant biomass retention [33]. Another setup was reported which had increased O<sub>2</sub> uptake in the aeration tank of the MFCs with the use of a biocathode. The MBR was mounted on the MFC, where sections of steel mesh membranes acted as both filtration units and cathode. These mesh membranes had the advantage of being simply stripped and washed without affecting the anode chamber [30]. A different

study came up with a similar combination but using commercial cathode membranes (stainless steel net altered using a polypyrrole film treated with 9,10-anthraquinone-2-sulfonic acid (AQS)). This added to fouling alleviation and the electrocatalytic denaturation of the pollutants via the bioelectricity that is generated [34].

Several sorts of MBRs (fluidized bed, hollow fiber, tubular) were combined with MFCs. An MFC-tubular MBR system was assembled by integrating a biocathode-equipped MFC with a tubular membrane. Here, the MFC was functioning as a biosensor to monitor the COD in actual time. The combination of individual modules was used to attain contaminated water treatment and energy retrieval [26]. The MFC-MBR combination also helped in reducing the biofouling. This was achieved by merging a hollow fiber-membranebased bioreactor with an MFC. The anodic chamber was an anaerobic compartment that was absorbed into the MBR, while the aerobic chamber of the MBR was used as the cathodic compartment, and in between these compartments [24], the module of the hollow fiber sheath was mounted. The introduction of an electrical field between both the electrodes improves the bacterial movement, which further increases the efficiency and superiority of the wastewater treatment. The electric field also helps in reducing membrane fouling by changing the properties of the sludge and impedes the adhering of negatively charged foulants to the membrane. A system assembled from the integration of an MFC with an electronic MBR (EMBR) was reported. In this setup, the cost was reduced by replacing the expensive proton exchange membrane (PEM) with a quartz silt compartment [33]. The bioelectricity produced in the EMBR was used to successfully hamper the membrane fouling. The electrolysis procedure was magnificently merged with the MFC for a broad spectrum of applications, such as methyl-red-contaminated water and algal pollution management [35]. The electrolysis pretreatment positively enhanced the MBR degradability and removal of chlorophyll a in these systems.

Another successful integration was achieved, which was composed of an MFC and hydrogen bioreactors. Here, the effluent from the hydrogen bioreactor was used as an influent in the MFC for the production of energy and the treatment of sewage water [36]. It was detected that the H<sub>2</sub> yield was inversely proportional to the organic loading rate (OLR), with the maximum value of hydrogen production reached (2.72 mol H<sub>2</sub>/mol glucose) when the OLR was lowest (4 g/L d). It also gained the highest energy output of 4200 mW/m<sup>3</sup>. By combining this method with solid–liquid separation, it was possible to produce value-added biochemicals [37]. Using this combined system, 214 L of methane and 37.7 L of hydrogen were obtained. An amount of 0.3 kg of solid particles/kg of the effluent was acquired using an efficient solid–liquid fractionation process. The solid mass obtained thus displayed features of fertilizers. The supernatant was utilized by the MFC for generating electricity; this supplied the energy demands of the solid–liquid separation and reduced the organic content of the wastewater.

Along with the improved wastewater treatment, the MFC-MBR-integrated setup also reduced membrane fouling. After studying this system for its efficiency, it was depicted that the fundamental elements that affect the generation of electricity were the features of the sludge and the aeration in the cathodic chamber [30]. The membrane filtration was carried out for 27 days in closed-circuit mode and 13–15 days in open-circuit mode [24]. The MFC-MBR setup used for methylene blue degradation achieved maximum antifouling by utilizing the ARS/PPy-modified cathode membrane. In comparison with the nonaltered steel mesh, the power density was amplified 31–37 times due to this modification. As mentioned in Table 1, a system was assembled to mitigate the membrane fouling while achieving efficient wastewater treatment termed the 'antibiofouling MFC-MBR' [28]. When an exterior resistance of 50  $\Omega$  was applied, the power output obtained was 2.18 W/m<sup>3</sup>. Sludge reformation was another technique used to reduce the membrane fouling in the MFC-MBR setup. A fluid bed membrane was installed in the MFC-MBR-integrated setup used for domestic primary effluent treatment. When this setup constantly functioned for 50 days with domestic contaminated water influent with 210 mg/L COD, it displayed improved effluent quality with relatively low consumption of energy. The COD removal

was 92.5%, lowering it to 16 mg/L. This percentage of COD removal is similar (90%) to that in another study in which the MFC-MBR system was used for COD removal [38]. The sludge was reformed as a result of a decrease in sludge production. The reformed sludge was 5.1% greater than the sludge produced in traditional MBRs. The integrated setup of MFC-MBR was cheaper and more effective than the most available techniques for the management of contaminated water and the production of electricity [39]. The reactor was assembled using low-cost materials and was able to generate 1.9 mA and 6 W/m<sup>3</sup> incessant and steady electricity for over 40 days.

When a tubular membrane was installed in the MFC-MBR-integrated setup [20], a power output of 40 mW/m<sup>2</sup> was attained. The setup ran for 30 days, and it was capable of achieving 94% elimination of organic matter and 80% removal of ammonia nitrogen. Another setup was assembled where an EMBR was integrated with an MFC, and the PEM was substituted with a quartz sand compartment [33]. This system achieved remarkable results with regard to the elimination of ammonia nitrogen (93%), phosphorus (50%), and organic matter (97%). The generation of electricity was also improved simultaneously. The MFC-MBR setup was examined as a sensor for an MBR. The competence of the system as a sensor was determined based on an undeviating relationship with a COD of up to 1 g/L. One more MFC-EMBR integration was used for methyl red decolorization and the simultaneous generation of electricity [17]. This system was able to achieve a COD elimination efficacy of 89.3% and 100% decolorization efficiency.

To explore the influence of implementing a process control on petroleum refinery wastewater treatment, a two-stage MFC-MBR-integrated system was built by Zhao et al. [40]. Chemical oxygen demand (COD), ammonium nitrogen ( $NH_4^+$ -N), and total nitrogen (TN) extraction efficiency in the MFC-MBR system were 96.3%, 92.4%, and 86.6%, respectively, compared with 74.7%, 71.2%, and 64.7% in the control system. Further, the use of this system as a biosensor was examined.

The two-stage combined MFC-aerobic membrane bioreactor (MBR)-based wastewater treatment method along with the use of ruthenium/activated carbon (Ru/AC) as a cathode catalyst was studied in another experiment by Bhowmick et al. [41]. The maximum volumetric power density and coulombic efficiency of the MFC-MBR system with Ru/AC as a cathode catalyst (2.7 Wm<sup>-3</sup> and 12.8  $\pm$  1.2%, respectively) were almost 1.4 and 1.5 times higher than those of the control MFC-MBR system without Ru/AC (2.0 Wm<sup>-3</sup> and 8.2  $\pm$  0.6%). Furthermore, these integrated MFC-MBR systems removed more than 96% of chemical oxygen demand (COD) from synthetic wastewater with an initial COD of roughly 1 g·L<sup>-1</sup>. This integrated MFC-MBR system has a lot of potential for being developed into a full-scale application that can provide efficient wastewater treatment and bio-energy recovery.

#### 2.3. FO-MFC Integration

Osmotic MFCs (OsMFCs) are a result of the integration of the MFC with the FO membrane [42]. This setup has also been examined in numerous research studies. Forward osmosis membranes were employed in the MFC to eliminate salt while simultaneously achieving COD subtraction and generation of bioelectricity [18]. The design of the setup included both internal and external placements of the membrane. When the membrane is placed internally, it is either installed between the electrodes as a separator or submerged in the anode/cathode compartment as a purification unit used to divide the draw and feed solutions [43]. The water passes from the anode to the cathode compartment via the FO membrane because of the osmotic pressure gradient, which leads to the dilution of saltwater. In the externally placed setup, the MFC and the membrane unit can work independently. Several studies were conducted by mounting the forward osmosis membrane as an interior segregator to examine the internal configuration of the setup [44]. The external arrangement of the OsMFCs was used for the conversion of the organic constituents in the influent-polluted water to alcohol and short-chain fatty acids, which were restricted by the forward osmosis membrane [30]. This further elevated the alkalinity and conductivity of the

solution, thereby enhancing the ion transfer and generation of power in the MFC. The membrane fouling of the forward osmosis membrane was also decreased by pretreating

the wastewater with the MFCs [45]. Another experiment reported that the power production of such a system was 43 W/m<sup>3</sup>, which was greater than both AEM and CEM, which had an energy output of 40 W/m<sup>3</sup> and 23 W/m<sup>3</sup>, respectively. Despite having most of the acetate removed with an efficiency of 90% in the OsMFC, some of the acetate leaked into the cathode chamber through the membrane. Another study also obtained the same results but with an additional benefit in the form of desalination. Improved proton shuttling from the anode chamber to the cathode chamber by the FO membrane was achieved in the OsMFC [46]. This helped in improving the production of electricity in the integrated OsMFCs as compared with the stand-alone MFCs operated in either mode, batch (NaCl solution) or continuous (seawater). Unfortunately, when a lower concentration of contaminated water was employed, the high values of power generation were not reached, with the highest power density attained being only 4.38 W/m<sup>3</sup> after a time-consuming procedure lasting 40 days. The decontamination potential of organic matter and phosphorus reached 97%, but owing to the lower elimination of the forward osmosis membrane, the elimination capability of nitrogen was hindered [23]. There was significant amplification of the power density of the MFC due to the studies conducted on the solute buildup, improved buffer capacity, and conductivity. The amplification achieved was an increase from 3 to  $11.5 \text{ W/m}^3$  in power density [45]. The membrane fouling was also alleviated as the MFC acclimatized and lowered the generation of sludge. There were remarkable removal efficiencies recorded with the employment of thin-film composites (TFC) in the OsMFC, with organic removal being 95% and phosphorus removal being 99%. The COD of the effluent was also quite low (less than 20 mg/L).

In another study, an MFC was integrated with anaerobic forward osmosis membrane bioreactors for the generation of biogas and methane and the simultaneous elimination of pollutants such as phosphorus and chemical oxygen demand. Using a SnO<sub>2</sub> nanoparticle electrocatalytic cathode in an electrocatalytic-assisted MEC, undesired carbon dioxide produced from biogas was converted to formate, with the maximum faradic yield of formate being 85% at 1.2 V. In comparison with AnOMBR, when AnOMBR-MEC was employed, the concentration of methane increased from 55% to 90% at the end of the operation, and the production of methane increased by 1.6 times. This study found that integrating an electrocatalytic-assisted MEC into AnOMBR is an excellent method for industrial-scale biofuel generation [47].

In an experiment, it was observed that the benefits of organic elimination, bioenergy production, and high-quality water recovery from wastewater are all favored by the osmotic microbial fuel cell (OsMFC), which integrates forward osmosis into the MFC. Waterflux-aided proton advection and net positive charge (NPC)-flux-promoted countercurrent proton exchange revealed an 18.7% increase in power density over a conventional MFC in this analysis. In a three-chamber reactor with a shared anode chamber, the energy production of the OsMFC and MFC was investigated. The voltage outputs of the OsMFC and MFC were maintained at 527.5  $\pm$  9.4 and 470.6  $\pm$  20.3 mV, respectively, with an external resistance of 3000  $\Omega$ . The highest power density of the OsMFC was found to be 3.42  $\pm$  0.18 W/m<sup>3</sup>, which was 18.7% higher than that of the stand-alone MFC (2.78  $\pm$  0.82 W/m<sup>3</sup>). Under varied operating circumstances, the findings of the experiment revealed that water-flux-facilitated proton advection was more essential than NPC-flux-promoted countercurrent proton exchange in regulating the power production in an OsMFC [48].

Type of Integration	Substrate	TEA	Application	Power Density and Efficiency	in Wastewater Treatment	References
Aerobic Reactor	Molasses	-	Acid orange removal	90% removal	52 mW/m <sup>2</sup>	[28]
AHPB	AHPB sewage	O <sub>2</sub>	$H_2$ generation	$2.72 \text{ mol H}_2/\text{mol glucose}$	$4200 \text{ mW/m}^3$	[49]
Capacitive deionization	Synthetic wastewater + sodium acetate	O <sub>2</sub>	Deionization	0.63 V	60% NaCl removal	[27]
Capacitive deionization	Synthetic wastewater	O <sub>2</sub>	COD removal	97% removal	$9.3 \text{ mW/m}^2$	[27]
Catalytic oxidation reactor	Glucose	PBS + Congo red	Congo red removal	90% Congo red degradation	$808.3 \text{ mW/m}^3$	[50]
Dark fermentation + aerobic digester	Waste mix	O <sub>2</sub>	H <sub>2</sub> + methane production from waste	$H_2 = 37.7 L/kg$ waste	Methane = 214 L/kg waste	[37]
Electric membrane bioreactor	Synthetic wastewater + glucose	O <sub>2</sub>	COD removal	97% removal	111 mW/m <sup>3</sup>	[51]
Electric membrane bioreactor	Wastewater	O <sub>2</sub>	COD removal	95.3% removal	$0.15  W/m^3$	[44]
AD-SCMFC (1 m <sup>3</sup> )	Pretreated pharmaceutical wastewater	O <sub>2</sub>	COD removal	35% removal	1.25 A/m <sup>2</sup>	[52]
CW-DCMFC (30 L)	Dewatered alum sludge	O <sub>2</sub>	COD removal	92% removal	$0.448 \text{ W/m}^3$	[53]
Hybrid AA/O- SCMFC (1 m <sup>3</sup> )	Domestic wastewater	O <sub>2</sub>	COD removal	95% removal	0.0036 W/m <sup>3</sup>	[54]
Septic tank-SCMFC (18 units, 700 L) disinfection	Domestic wastewater	O <sub>2</sub>	COD removal	87% removal	$0.00043 \text{ W/m}^3$	[55]

Table 1. Integrated systems of microbial fuel cells (MFCs) with various techniques.

#### 2.4. Integration of MFCs with Dark Fermentation

A hybrid procedure of dark fermentation, microbial fuel cells, and microbial electrolysis cell could convert crude glycerol, a waste by-product obtained during biodiesel manufacturing, to bioenergy (MEC). This combined technique addresses the thermodynamic constraint of dark fermentation, where complete breakdown of crude glycerol is challenging [56]. Large percentages of unutilized organic metabolites remain in the dark fermenter's effluent, which can be used to produce power in MFCs or hydrogen in microbial electrolysis cells. The greatest H<sub>2</sub> generation of 332 mL/L (output of 0.55 mol H<sub>2</sub>/mol glycerol) was attained when crude glycerol with an initial carbon oxygen demand concentration of 7610 mg/L was used in dark fermentation. After 50% dilution, the sewage was deteriorated in MFCs to reach a power output of 92 mW/m<sup>2</sup> and a carbon oxygen demand elimination of 49% [57].

MFCs were also integrated with dark fermentation (DF) for  $H_2$  generation via an anaerobic hydrogen production process. The waste from the hydrogen fermentation is acidic and was reacted further in the MFCs to increase the entire electricity generation and facilitate COD exclusion efficiency [33]. This system was examined to acquire marketable chemicals and fuels, such as biofertilizers, hydrogen, and methane, for bioelectricity [37].

An innovative integrated single-stage dark-fermentation-MFC method was designed in an experiment to produce biohydrogen and power from wastewater treatment at the same time. The integrated system's biohydrogen gas was converted into energy using a proton-exchange membrane fuel cell (PEMFC). The impact of hydraulic retention time (HRT) on biohydrogen and electricity production was also investigated. At an 8 day HRT, the greatest volumetric biohydrogen production rate (VHPR) was 0.44 L H<sub>2</sub>/L·d (0.66 L H<sub>2</sub>/g COD removed), with an electricity output of 530 mV (100 mW/m<sup>2</sup>). The bacteria discovered on the anode of the integrated system were *Chryseobacterium*, *Azotobacter*, *Bacillus*, *Enterococcus*, *Citrobacter*, and *Methanobacterium*, per an 16S rRNA gene-based investigation. The PEMFC employed to generate voltage from biohydrogen produced by the integrated system was capable of achieving a maximum voltage of 459 mV (367 mW) and a maximum cell efficacy of 44% (fuel consumption of  $1.5 \times 105 \text{ mol/h}$ ) [58].

A hybrid approach of dark fermentation (DF) and MFC known as sDFMFC was examined in the study for simultaneous  $H_2$  and electricity production from *Saccharina japonica* in a single reactor. Due to simultaneous  $H_2$ /carboxylic acid (CA) synthesis by DF and electricity production by MFC, the energy recovery of the sDFMFC was significantly improved. A time course of CA concentration in sDFMFC validated the coproduction of H2 and power. With an  $H_2$  yield of 110 mL/g-VS and a maximum power density of 1.82 W/m<sup>2</sup>, *S. japonica* provided good energy recovery of 17.3%. The sDFMFC demonstrated a diversified microbial population for optimal organic substrate microbial conversion. The findings suggest that the sDFMFC could be a promising single reactor process for producing  $H_2$  and energy from a variety of biomass feedstocks while maintaining the individual DF and MFC processes' efficiency [59].

#### 2.5. Sediment Microbial Fuel Cells

MFCs can be integrated into a variety of technologies, including sediment microbial fuel cells, which have the same arrangement as a conventional MFC. The anode (anaerobic) can be immersed in the sediment, and the cathode (aerobic) can be found at the top part of the SMFC, where oxygen acts as a terminal electron acceptor (Figure 4). More intriguingly, it was not discovered until 2001 that bacteria can donate their electrons exogenously. By embedding an MFC in an oceanic sediment, Reimers et al. [60] investigated the feasibility of extracellular electron transmission. They explored exoelectrogenic electron transport by inserting an electrode in a marine sediment and harvesting energy from it. The anode component of SMFCs was inserted into the sediment (anaerobic) region of the ocean, while the cathode was placed at the water surface. Exoelectrogenic microbes are already active in the sediment, according to Reimers et al., and can give electrons to the electrode in

anaerobic environments. The electrons from the sediment can be transmitted to the cathode, which already contains dissolved oxygen from the surface water and can act as a terminal electron acceptor. From the initial SMFCs, they were able to attain an energy output of  $50 \text{ mW/m}^2$ . The aerobic and anaerobic portions of all categories of SMFCs were configured based on the surface and sediment, respectively. He et al. [61] studied the feasibility of rotating a cathode electrode to generate more oxygen in river sediments and reached an energy output of  $49 \text{ mW/m}^2$ , compared with  $29 \text{ mW/m}^2$  for a nonrotating cathode. The rotating cathode provided more oxygen at the cathode in this example, which improved the efficiency of electron transport from the anode. In the nonrotating cathode, on the other hand, it was discovered that the number of terminal electron acceptors at the cathode was limited.



Figure 4. A typical integrated sediment MFC.

Another experiment investigated three laboratory-scale two-chambered MFCs with sea sand as the anode and saltwater as the cathode, and a mean energy output of 0.016 W was achieved. They discovered that Geobacteraceae uses electrodes as an electron acceptor under anaerobic environments [62].

Different types of sediment MFCs, such as benthic MFCs (BMFCs), floating-macrophytedependent MFCs (FMFCs), and soil-based MFCs (SL-MFCs), were developed with time [30]. BMFCs are also an advanced use of SMFCs in an aquatic environment, especially for bioremediation of contaminants, such as sulfur and organic matter in the sediments. The contaminants in the BMFC sediment aid as a source of energy for bacteria, and the oxidation of contaminants in the sediment and the decrease in oxygen at the surface produce power [63,64]. As previously stated, constant monitoring of produced voltage and retention of power from BMFCs can also be used as a monitoring system in the sea [65].

The system design of FMFCs is also influenced by those of sediment MFCs and plant MFCs (PMFCs). Plants release rhizodeposits (organics and nutrients) in the sediment of PMFCs. The microorganisms use secretory rhizodeposits as a fuel in the sediment and oxygen as a reductant at the surfaces and subsequently generate energy. Mohan et al. [66] introduced FMFCs to remove excessive volatile fatty acids (VFAs) and residual organic matter from an H<sub>2</sub> bioreactor's outflow. COD elimination was around 86.67%, and VFA removal was around 72.32%, respectively. The power production was shown to accelerate as COD increased, reaching 224.93 mA/m<sup>2</sup> in some cases.

Huang et al. [67] estimated the use of an SL-MFC for anaerobic phenol decomposition. In a closed circuit, they removed 90.1% of the phenol with a maximum energy output of 29.45 mW/m<sup>2</sup>. Further research used SL-MFC to remove organic pesticides, including hexachlorobenzene from the environment. This research discovered an elimination rate of 71.15% and an energy yield of 77.5 mW/m<sup>2</sup>. The researchers found that the existence of

electrodes encouraged electrogenic microbes to deliver more electrons, which resulted in an increase in hexachlorobenzene elimination.

Quaglio et al. [68] presented an effective way for improving power extraction using SMFCs by alternating the anodes. The setup of a standard SMFC has been changed to incorporate two anodes functioning with the same cathode in the studies. This arrangement is compared with a traditional setup (control) and a setup that uses intermittent energy harvesting, demonstrating that the anode alternation technique improves energy harvesting. Control SMFCs generated average power densities of 6.3 and 8.1 mW/m<sup>2</sup> when operating intermittently. SMFCs employing the anode alternation technique, on the other hand, achieved an average power density of  $23.5 \text{ mW/m^2}$ . These studies support the proposed anode alternation method's superiority to both control and intermittent energy harvesting methods. Anode alternation can also be considered a step forward in the field of intermittent energy harvesting.

In another study, the power generation capability of mixed-culture algal biomass was investigated using a multianode sediment microbial fuel cell (SMFC). The reactor had a multianode structure with a tin-coated copper mesh (TCCM) anode and a platinum-coated titanium mesh cathode. The SMFC generated a high-power density of 2965 mW/m<sup>2</sup>, which is the highest power density yet reported in SMFC research. The biocompatibility of TCCM favors bacterial adherence and accumulation of electroactive bacterial groups, according to microscopic observations and gene profile research (Gammaproteobacteria, Deltaproteobacteria and Alphaproteobacteria). These findings suggest that algal biomass might be employed as a suitable feedstock in SMFCs to enhance electricity output significantly [69]. Table 2 illustrates different experimental performance of sediment MFC.

Sr. No	Reactor Type	Substrate	COD, Conductivity, pH	Cathode	Anode	Power Density (W/m <sup>3</sup> )	Reference
1.	Sediment MFC, 350 L	Synthetic wastewater	-	Activated carbon	C mesh	0.0064	[70]
2.	Sediment MFC, 195 L	River sediment	13.5 mS/m pH 6.8–7.4	Activated car- bon/stainless steel	C mesh	0.0415	[71]
3.	Self-stratifying SCMFC (38 units), 19.2–57.6 L	Urine	5.6–6.8 g/L pH 8.5–9.2	Microporous carbon	C veil fibers	7.3–9.9	[72]
4	Sediment MFC, 72 units, 72 L	River sediment water	890 g/L 27.206 mS/m pH 8	Zinc	Copper _	0.0019	[73]
	Sediment MFC, 35 units, 35 L					0.0069	[74]

 Table 2. Several studies demonstrating the performance of sediment MFC.

#### 2.6. Integration of MFC with MDC

Desalination is another approach for the treatment of contaminated water and potable water production. Distillation, electrodialysis, and reverse osmosis are the methods used in desalination [75]. It is believed that desalination is not applicable in all circumstances due to the high energy requirements and maintenance cost [42]. MFCs have the potential to be integrated into desalination cells due to their advantages in the synthesis of renewable energy, such as  $CH_4$ ,  $H_2$ , and electric power. In 2009, Cao along with his co-workers [76] described a new type of desalination system known as microbial desalination cell, which is based on the movement of ions from water in accordance to the electron supplied by microbes. A desalination cell MFC with three chambers was constructed (as shown in Figure 5. An anion exchange and cation exchange membrane, as well as a central compartment, were included in the three-chambered DS-MFCs. In the anode chamber,

bacteria decompose contaminants and yield power, while negatively charged ions pass through a membrane from the central chamber to the anodic chamber. The cathode utilizes protons in the same way as that in typical MFCs, and a positive charge from the central compartment is transferred to the cathodic compartment. As a result, water in the central compartment is desalinated, and contaminated water is treated at the same time [77].



Figure 5. General representation of MDC-MFCs.

For synthetic wastewater and saline sewage treatment, Zhang and He [78] devised an osmotic MFC with an FO membrane coupled with a microbial desalination cell (MDC). The research achieved an energy output of 0.160 kW h/m<sup>3</sup> and an electrical conductance decline of 95.9%. Due to dilution and desalination, the system was demonstrated to be an excellent technology for salt elimination. Zhang and He [78] used 105 L MDC to test the scaling up of the system. The research revealed that several feeding inlets could improve the current production. The current production improved from 670 to 2000 mA with a salt elimination of 3.7 to 9.2 kg/h when external power was applied (m<sup>3</sup> day). Sevda and Abu-Reesh [79] employed organics from petroleum effluent for an MFC and used osmotic MFCs with up-flow microbial desalination cell for marine treatment. According to their research, the integrated system can eliminate up to 93% COD from saltwater while also removing 48% salt.

#### 2.7. Integrated Constructed Wetland MFC

Constructed wetland (CW) treatment is a less expensive passive sewage management method. The primary cause for its limited recognition is that it has a low treatment efficacy and requires a considerable amount of land to construct. Various MFCs integrated with constructed wetlands are shown in Table 3.

Yadav (2010) introduced a hybrid method that combines an MFC with a CW to improve the treatment effectiveness of the CW while reducing the required landmass. It has found wide acceptance from scientists all over the world over the last 7–8 years. Because the majority of the component in the CW is anaerobic, it has low treatment efficiency (less electron acceptor). As a result, the primary purpose of integrating the MFC into the CW is to improve the CW's treatment potential (Figure 6). However, power generation has recently been discovered to be another CW resource [80].



Figure 6. A typical representation of constructed wetland MFCs.

The first experimental investigation on a CW-MFC for the elimination of dye in a vertical flow CW was carried out. The experiment found that the highest electricity output was around 69.75 mA/m<sup>2</sup>, that the highest COD was 70%, and that dye degradation rate was 93.15% [80]. In the following year, Villasenor et al. [81] tested a horizontal flow CW on various organic loading rates, achieving a COD elimination of 90–95% with the highest electricity output of 43 mA/m<sup>2</sup> and a coulombic efficiency of 0.45%. Zhao et al. [82] evaluated a batch and continuous vertical CW for swine contaminated water in the same year, reporting that the batch experiment eliminated 71.5% of carbon oxygen demand with an energy density of 12.83 W/m<sup>2</sup>. However, in continuous mode, elimination was 76.5%, with an energy output of 9.4 mW/m<sup>2</sup>. Eventually, Corbella et al. [83] investigated the functioning, configuration, and microbiological aspects of CW-MFCs, finding an energy density of 36 mW/m<sup>2</sup> and a prevalence of 13% to 16% *Geobacter* in a horizontal subsurface CW with MFCs. Furthermore, CW-MFCs' power production is still modest, and CE was shown to be very inadequate [83]. Additionally, Ramrez-Vargas et al. and Doherty et al. [84] estimated the evolution and development of MFC integration into the CW. Research on CW-MFCs is still in development, and many fields need to be researched before they can be used more widely. However, a field-scale effort called MET lands is currently underway, with the goal of incorporating conductive material into wetlands.

Wen et al. [85] investigated the extraction efficiency of sulfamethoxazole (SMX), tetracycline (TC), and their common coexisting contaminants, such as COD and nitrogen, in constructed wetlands integrated with microbial fuel cells (MFC-CWs), as a function of plant, circuit operation mode, and influent antibiotic loads. The research revealed that MFC-CWs with plant and circuit connections had optimum SMX (99.70%) and TC (100%) removal performance. Furthermore, the bioelectricity output of MFC-CWs (planted) was superior to that of unplanted systems. With rising influent antibiotic concentrations, coulombic efficiencies in both planted and unplanted MFC-CWs dropped. In conclusion, MFC-CWs with plant and circuit connections have the ability to treat SMX- and TC-containing wastewater.

The study was conducted by Mittal et al. to design a unique earthen-membrane-based two-chambered wetland cum microbial fuel cell (CW-MFC) for the removal of azo dye and other contaminants, including wastewater detoxification. The current design resembles the core of a shallow unplanted CW-MFC that runs anaerobic and aerobic cycles sequentially without mixing cathodic and anodic effluents. A synthetic effluent containing 550 mg/L initial COD and 50 mg/L methyl orange (MO) azo dye was found to have 94.04  $\pm$  2.87%

COD absorption and  $94.22 \pm 1.33\%$  azo dye absorption, with current density and power density outputs of 544.6 mA/m<sup>3</sup> and 148.29 mW/m<sup>3</sup>, respectively [86].

An up-flow constructed wetland-MFC system with various fillers was designed for the removal of Cr (VI) and simultaneous production of energy. These fillers, such as bio-ceramic (CW-MFC1), zeolite (CW-MFC2), calcite (CW-MFC3), and volcanic rock (CW-MFC4), are able to absorb contaminants over a long period of time. All systems removed over 93% COD, and the rate of Cr (VI) removal was as follows: CW-MFC4 (99.0%) > CW-MFC2 (95.5%) > CW-MFC3 (89.7%) > CW-MFC1 (72.2%). Due to abundance of organic substance absorbed by microbes in the filler layer, which weakens the action of anodic microbes, bio-ceramic is a simple way to immobilize microorganisms, which as a filler (CW-MFC1) showed the lowest removal rate of Cr (VI) in the CW-MFC1 system. Furthermore, calcite, which has a reduced surface area but a less porous structure than volcanic rock and zeolite, was unfavorable to microbial life, resulting in decreased Cr (VI) removal in the bottom layer of the CW-MFC3. The CW-MFC system's output voltage and maximum energy density were in the subsequent order: CW-MFC3 > CW-MFC4 > CW-MFC2 > CW-MFC1. As a result, using volcanic rock as fillers was the greatest option, as it permitted for the most Cr (VI) removal (99.0%) and the ideal power production (0.595-0.019 V output voltage, 0.462 W/m<sup>3</sup> power density). Furthermore, due to the extensive Cr(VI) stress, microbial diversity in the cathode was higher than that in the anode, and Acetoanaerobium and *Exiguobacterium* were the leading genera in the anode and cathode, respectively [87].

Table 3. Microbial-fuel-cell-integrated constructed wetlands.

Type of Integrated MFCs-CW	<b>Power Density</b>	COD Removal Rate	References
Vertical flow MFCs-CW	531.04 mW/m <sup>3</sup>	72.17%	[88]
Up-flow_ downflow CW	$50.268 \text{ mW/m}^3$	81%	[89]
Horizontal subsurface flow, continuous mode-CW	36 mW/m <sup>2</sup>	71%	[83]
Horizontal flow bed-CW	-	60–70%	[90]
Vertical CW	$53,714.08 \text{ mW/m}^2$	82.32%	[91]

#### 2.8. Integration of Microbial Fuel Cells with Microalgae

The concept of microalgae MFC is dependent on substantial MFC and microalgae research. A combination of algae and MFCs will be a unique technology that converts solar energy to electric energy through the metabolic processes of photosynthetic microbes (Figure 7) [92].

Likewise, MFCs are used in algal bioreactors, and the bases of microbial-fuel-cellcombined algal bioreactors are solar energy; hence, the existence of algal biomass turns solar energy into chemical energy. Furthermore, the accessible algae transform chemical energy into electrical energy. Photosynthetic fuel cells or solar cells are MFCs that have been integrated into algal bioreactors. Microbes oxidize organics at the anodic compartment, and algae release oxygen at the cathode to meet the need for aeration in the alternative arrangement of algal MFCs. As a result, it is regarded as passive aeration from algae, which provides an oxidant to the cathode for the anode's reaction accomplishment [12].



Proton Exchange Membrane



In the case of a microalgae photobioreactor (PHB), microbes use incoming light to convert CO<sub>2</sub> to biomass and O<sub>2</sub> [3]. Furthermore, the oxygen generated by *Chlorella vulgaris* during photosynthesis in the cathodic chamber of MFCs could act as the terminal electron acceptor. Therefore, the integrated system can be used to sequester carbon dioxide, produce oxygen, and remove nitrogen from contaminated water [93].

In another technique, dead microalgae were employed as the substrate in the anode compartment of MFCs, while viable algae were introduced in the cathode chamber. When the conductive cathode was fixed with multiple strains of algae, such as *C. vulgaris*, *Dunaliella tertiolecta*, and *Synechocystis* sp., the energy production of single-chambered photosynthetic MFCs was detected. When irradiated with 10 W/m<sup>2</sup> of white light, an MFC containing *Synechococcus* sp. had a maximum energy output of 10.3 mW/m<sup>2</sup>. A serial arrangement of four MFCs can produce enough energy to run a commonly available tiny digital clock (currently around 10 mA and a potential of 2 V) [94,95].

An anoxic MFC was generated using dissolved  $CO_2$  as an electron acceptor and a photobiocathode that was activated. The MFC achieved an energy output of 750 mW/m<sup>2</sup> after this biocathode efficiently fixed  $CO_2$  [96]. The MFC was used in conjunction with a tubular PHB as the cathode chamber, which was colonized with *C. vulgaris*. The amount of oxygen produced by *C. vulgaris* and the amount of power produced by the MFC were both light-dependent. Under inconsistent illumination, an MFC with an algal biocathode attained the highest energy output of 24.4 mW/m<sup>2</sup>, which was 2.8 times greater than the MFC with an abiotic cathode [97].

During the anaerobic digestion of Taihu blue algae, biohydrogen employing migrating ammonia as a nitrogen supply and biogas upgrading using hydrogen collected at the biocathode in an integrated bioelectrochemical system (BES) were examined by Wu et al. [98]. The use of an integrated BES allowed for simultaneous ammonia reduction and biogas upgrading. It was observed that under a 0.4 V applied voltage, ammonia utilization and hydrogen production achieved 73.67% and 202.87 mL, respectively. As a result, performing ammonia mitigation, hydrogen production, and biogas upgrading all at the same time with BES appears to be an effective approach.

#### 2.9. Anaerobic-Anoxic-Oxic (AO/O) Integrated with MFC

An MFC was also integrated into an anaerobic-anoxic-oxic (AO/O) system, resulting in a hybrid system with a total volume of 1 m<sup>3</sup> (Figure 8) for treatment of domestic wastewater and power generation at the same time [54]. During its 1-year operation, a maximum current density of 3.6 mW/m<sup>3</sup> and a COD removal of 95% were reached under an HRT of 18 h, a temperature of 8–23 °C, and a recirculation ratio of 200%. Tang et al. [53] combined an MFC with a constructed wetland (CW), resulting in an energy recovery of 0.448 W/m<sup>3</sup> and a COD reduction of 92% while treating dewatered alum sludge.



Figure 8. (A) Integrated MFC-AA/O system for wastewater treatment; (B) integrated MFC septic tank for wastewater treatment.

Notably, Valladares Linares et al. [55] established the feasibility and long-term viability of a linked septic-tank-MFC-disinfection system for residential wastewater treatment. As shown in Figure 8, raw influent from a five-person residence flowed by gravity to a 1300 L septic tank, then to a 700 L Aquox<sup>®</sup> MFC comprising two stacks of 9 MFC units, and lastly to a sodium hypochlorite disinfection system. An ultralow power consumption unit made up of capacitors and microcontrollers gathered and stored energy from the MFC. Because no external energy was required, the system was declared sustainable and viable, and the treated effluent met the local discharge standard.

#### 2.10. Other Integrated MFCs

Many research studies have used MFCs in a stacked configuration to achieve upscaling and treatment performance. There were attempts to improve the COD removal by testing an MFC (without the membrane) employed with a biocathode submerged in an SBR. This MFC was able to reach 2.34 W/m<sup>3</sup>, with a total of 18.7% elimination of COD from the integrated setup [23]. USB-MFC-BAF was used to attain the concurrent management of molasses wastewater and the production of bioelectricity [26]. In this system, the potential of COD removal was 53.2%, and the supreme energy output reached was 1410.2 mW/m<sup>2</sup>. The absolute denaturation of the azo dye AO-7 was obtained through the integration of an MFC and an anaerobic bioreactor [32]. The dual-staged technique was used to achieve a decrease in ecological toxicity and reach 90% COD removal along with the simultaneous generation of bioelectricity. An MFC-COR setup packed with a granular catalyst was used to obtain degradation of the azo dye, Congo red [30]. An amount of 90% of the dye was observed to have degraded in 3 days in a chargeless mixture, and the maximum power density reached was 808.3 mW/m<sup>3</sup>.

In a plug flow environment, Feng et al. [99] combined a number of MFCs in a stacked configuration. With a total volume of 250 L, the flow was horizontal. While treating residential wastewater, the maximum energy obtained in each module was 0.435–0.010 A, with carbon oxygen demand and total nitrogen (TN) elimination rates of  $79 \pm 67\%$  and  $71 \pm 68\%$ , respectively. Moreover, MFC has been applied to a number of other procedures, such as electro-Fenton reactions, to improve treatment efficiency. The strategy for integrating MFCs to improve electro-Fenton reactions is to add hydroxyl ions to the Fenton reactions to speed up the processes. Many studies have shown the generation of  $H_2O_2$  at the cathode of MFCs. The generation of  $H_2O_2$  at the cathode can be accomplished using two or four electron transmission processes, whereas in electro-Fenton reactions, ferrous ions are oxidized to ferric ions using oxidants (Figure 9).



Figure 9. MFC integrated with bio-electro-Fenton.

In one of the studies, a laboratory-scale three-chambered MFC-dependent latrine was built for organic material treatment in the anode compartment, nitrification in the central compartment, and denitrification with the help of a biocathode in the third compartment [100]. During the operation, the highest COD elimination of 90%, a nitrate reduction

of 76.8%, and an energy output of  $3.4 \text{ mW/m}^2$  were attained. Composting was eventually performed with solid waste [101]. A three-column MFC stacking structure that can be easily plugged in a septic tank to yield an energy output of  $142 \text{ mW/m}^2$  was devised. This system is predicted to have a daily power consumption of 24 W/h, which is enough to run a 6 W LED bulb for 4 h. Similarly, 15 MFCs were stacked in a 2.44 m<sup>3</sup> septic tank and linked to a power management circuit, finally discharging the highest current of 1.98 mA and power of 4.51 mW [102].

# **3. Life-Cycle Assessment (LCA) of Integrated Microbial Electrochemical Systems** *3.1. MFC-FO*

Integration of a conventional MFC with a forward osmotic (FO) membrane results in a more efficient system, in terms of bioelectricity production, called osmotic MFC (Os-MFC). Researchers used the International Life Cycle Data System (ILCDS) to calculate the environmental impacts of the working integrated systems. This was performed to characterize the effect of using the Os-MFC system to treat 1 L of wastewater [46]. The system's life cycle is divided into five stages for analysis: raw material extraction and material processing, system fabrication, pretreatment, operation, post-treatment and end of life (EoL). The exploitation of raw materials has a significant impact on metrics, such as global warming potential (GWP) and abiotic depletion potential. The human toxicity potential (HTP) and ozone depletion potential (ODP) are factors in the EoL stage (ODP). Ultimately, the integrated Os-MFC system has a greater environmental impact than a stand-alone traditional MFC system.

#### 3.2. MDC-FO

Overall, three tasks are performed by an osmotic MDC-integrated system, which are removal of salts, treatment of wastewater, and generation of electricity [46]. Although lab-scale systems showed as good as 90% desalination, removal of salts from natural water sources has shown less efficiency. Additionally, serious environmental issues associated with these integrated systems have raised concerns. The discharged water from MDC-FO comprises high concentrations of nitrates and phosphates, which cause water pollution. Even though the operation stage contributes 57.8% to GWP, lesser contribution to greenhouse gas emissions was found when compared with conventional desalination systems, directing towards a hope for sustainable development of the integrated system [103,104].

#### 4. Challenges Associated with Integrated MFC

#### 4.1. High Operational Cost

Integrations of MFC offer advantages, such as enhancement in COD removal and energy retrieval; however, life-cycle assessments in terms of economics need to be carried out to determine the operational costs, such as supply of electrolytes, catalysts, membranes, electrodes, and pH adjustment chemicals. Moreover, additional components such as glucose and acetate are supplied to enhance the activity of exoelectrogens in certain experiments, which adds up to the MFC cost [105]. MFC frequently creates low operational voltage in relation to the cell's electromotive force, which is commonly referred to as thermodynamically anticipated irreversible potentials. Excess biofilm and the chemical molecules generated by the biofilm may aggravate anodic biofouling, thus further lowering the transfer of electrons from the microbe to the anodic material [106,107].

#### 4.2. Reduced Power Density

The major problem associated with an integrated MFC is the low recovery energy, which usually arises during the enlargement of the MFC reactor since the reactor size is inversely proportional to the power output. Moreover, increasing the reactor size leads to increment of capital costs. Therefore, MFCs are connected in stacks in series and parallel to attain an adequate power output. Additionally, integrated MFC reactors with an AnTP and MBR with slight alterations, such as MFC-A2/0 or MFC-submerged MBRs, are

considered suitable for large-scale applications. High power density can also be achieved through appropriate arrangement of MFC stacks and employment of simple upgradable assemblies [105].

#### 4.3. Maintenance and Optimization of Hybrid Systems

Despite the advantages offered through integration of MFCs, there is an increment in the complexity, maintenance, and optimization of the system. An evaluation should be carried out to list the positive and negative impacts of each method to conclude whether the integration is beneficial or not. For instance, in a coupled MFC-MBR reactor, additional costs are essential for the management and removal of the slush through the MBR. Moreover, inadequate functioning of the MFC leads to a reduction in membrane efficacy and operational reliability, an increment in sludge production, and an increase in fouling. Therefore, each factor should be identified and evaluated before integration.

Optimization of the integrated MFC reactor is an essential task in functioning circumstances of a wastewater treatment plant (WWTP). Therefore, it becomes crucial to determine the interdependent parameters for the reactor to operate in its most effective state. Certain available software, such as artificial neural network (ANN) (e.g., KSOFM, MLP) and Design-Expert, can prove beneficial for optimization for diverse variables and data [105].

#### 4.4. Evaluating Sustainability

Prior to the execution of a technology, it has to undergo technical, economic, and environmental analysis. AnTP-MFC and MBR-MFC hybrid systems have proved to be technologically and economically favorable; however, their environmental impacts have not been examined. Life-cycle assessment of an integrated MFC needs to be carried out to study the environmental impacts, and hybrid systems that offer the least detrimental environmental effects need to be commercialized [105].

#### 4.5. Commercialization and Practicality

Integrated MFCs are costly and produce low power output, which are major setbacks in their commercialization. Further study should be accomplished for the facilitation of integrated MFCs in industrial utilization. Studies need to be carried out to fully understand the metabolism and mechanism of action of exoelectrogens. Moreover, since integrated MFCs employ costly chemicals to enhance efficiency, investigations need to be carried out to find a cost-effective alternative, such as using ozone as the final electron acceptor at large-scale applications [108]. Moreover, appropriate reactor configurations contribute to long-term and efficient operation of the reactor [105].

#### 5. Future Prospects and Conclusions

Integrated MFC systems are a promising technology to explore. This review described various integrated MFC systems along with their advantages and disadvantages over a stand-alone MFC. A large number of literatures describes the performance efficiency of an integrated MFC against a stand-alone MFC system. It was observed that the performance of an integrated MFC was much higher than that of a stand-alone MFC in terms of wastewater treatment and energy production. However, there are some bottlenecks related to integrated systems that need to be addressed. One of the major obstacles to overcome the upscale and commercialization of these integrated bioelectrochemical systems is the equipment and operational cost [109]. For that purpose, the costs of electrodes and membranes need to be reduced. The need for an external energy supply also has to be decreased through efficient cathode oxygenation. In conclusion, several technologies can be integrated with BESs, such as FO, DF, MBR, CDI, and aerobic bioreactors, to add various benefits and enhance the already-existing processes. The combined systems have many benefits, such as increased efficiency, lesser membrane biofouling, desalination, and better-quality effluent, along with the simultaneous generation of electricity. An integrated MFC system also has lower

total capital costs than stand-alone systems, but upscaling increases the maintenance and processing costs. Hence, integration is the first step towards the final viable system. These integrations need to be refined and optimized to become viable. These systems need to be studied extensively, and their processes need to be understood deeply. Their fallacies have to be addressed and rectified. The systems should become economically viable and not complicated to scale up. With these obstacles cleared, the integrated systems of BESs have a promising potential to be commercialized for the simultaneous treatment of contaminated water and the production of electrical current.

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