

Review

Progress in microbial fuel cells for sustainable management of industrial effluents



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ABSTRACT

Microbial fuel cell (MFC) technology is a promising solution for both organic and inorganic effluent treatment, as it is capable of handling a variety of complex contaminants with simultaneous energy and resource recovery. Unlike conventional treatment technologies, MFCs can achieve removal of organic matter, persistent organic compounds, heavy metals and nutrients from categorically different waste effluents, while recovering energy and valuable substances. This makes the technology economically attractive, and viable for a wide range of industries for their waste treatment applications. However, in order to implement this technology as an on-site treatment unit, limitations pertaining to costs, scale-up and performance have to be addressed, with due emphasis on developing strategies that can be easily applied in industrial settings. This review summarizes the recent progress in the field of waste treatment and environmental remediation using MFCs, and examines these operational challenges. Cost-effective materials that have been implemented on the field and at large scales have also been highlighted. In addition, the review highlights the fact that there is a need to optimize the technology depending on energy or resource recovery. The MFC-based technology utilizes the synergy of bioremediation and bioelectricity production from wastes, offering a sustainable approach to industrial waste management.

1. Introduction

The industrial economies of today have a major undertaking and that is, environmental protection. The proliferation of factories without particular regard for effluent treatment has led to large-scale contamination of water and land. These effluents pose severe health risks, because of the bioaccumulation potential of their contaminants that include persistent organic compounds and heavy metals [1,2]. It is apparent that wastewater and toxic sludge volumes will only rise, and wastes are also becoming more difficult to treat. Conventional treatment technologies are energy-intensive and cannot achieve the stringent limits of discharge without requiring additional operating expenses and costly retrofitting to the existing systems [3]. For developing countries undergoing rapid industrialization, improper treatment of wastes is a significant problem, exacerbated by the use of inefficient technologies. Effluent treatment is expensive, complex and therefore, does not receive the adequate attention that is required [4,5]. Improper management and disposal of solid wastes is a persistent problem plaguing industrial economies across the world [6,7].

Microbial fuel cell (MFC) is a rapidly progressing technology that has

garnered an interest among the scientific community for its dual ability to treat wastes and recover energy in the form of electricity. With the ability to degrade a variety of organic compounds, MFCs offer a sustainable approach to environmental remediation, achieving treatment with resource recovery. At its core, an MFC is a bioreactor where a special group of microorganisms, having the ability to transfer electrons to an external anode (called electroactive microorganisms), oxidizes organic substrates. The current then travels to the cathode, to reduce oxygen or other ions. Over the years, extensive research has been conducted on MFC substrates, electrode materials and device configurations to improve power generation and degradation efficiency. In addition to having other advantages, MFCs combine the major concepts of sustainable waste management *i.e.*, almost energy-neutral operation, low investment and environmental impact, energy and material recovery and robust performance [8].

The components necessary to operate an MFC are the anode, cathode, ion-exchange membrane, an external electrical circuit with resistors and organic matter for the bacteria. The anode compartment is inoculated with bacteria that can be collected from wastewater, sewage sludge or compost. The bacteria feed on substrate solutions prepared

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using simple sugars, such as, glucose, acetate and butyrate. However, complex organic matter in the form of wastewater, sludge, sediment and soil have successfully served as both substrate and the source of microorganisms [9–11]. The mixed microbial consortia present in these substrate sources, along with the higher organic content, make these substrates capable of yielding greater power output when used in MFCs [12]. This is promising, because such substrates tend to contain biodegradable wastes. The oxidative breakdown by bacteria can provide the benefit of energy recovery alongside treatment. In fact, the more immediate application of MFC technology is in the field of treating wastes, by harnessing the power of the indigenous electroactive microorganisms present within the waste substrate itself [13].

A variety of configurations have been tested, the most common being the dual-chambered MFCs (DCMFC) where the anode and cathode chambers are separated by a proton exchange membrane (PEM) or a salt bridge. Single-chambered MFCs (SCMFCs) are cheaper, as all the electrodes of the MFC are placed in the same compartment. In some cases, the PEM can also be eliminated, resulting in membrane-less MFCs [14]. However, the stacked MFCs consisting of several MFC modules connected in series or parallel can generate greater power output. Upflow MFCs are continuously fed with substrate solution from the bottom. Other designs of MFCs utilize a soil or sediment matrix as both the substrate and source of bacteria, and have been used for bioremediation purposes (Fig. 1). Constructed wetlands-coupled MFCs (CW-MFCs) [15], microbial desalination cells (MDCs) [16], microbial electrolysis cells (MEC) [17] and membrane bioreactor MFCs [18] are some of the more advanced and integrated configurations.

For industries in developing countries, use of this technology could be readily beneficial, due to the comparative advantages over other costly, energy-intensive treatment technologies and its flexibility to treat effluents of varying characteristics [19]. In the developing world, lack of technical expertise regarding control and operation of effluent treatment

plants, and cost overruns are usual problems in factories, especially mid-scale ones that are expanding at a rapid pace [20]. MFCs treat the waste without generating secondary pollution, recovering energy whilst operating under mild, easy-to-control conditions [21]. The technology not only has the ability to reduce the organic load in wastewater [22, 23], it can effectively remove nitrogen and phosphorus from waste streams. Removal of these nutrients from the wastewater is essential for complete treatment, as these nutrients can contribute to eutrophication (due to nitrogen and phosphorus), global warming and ozone depletion (due to reactive nitrogen) and pose other risks to human health [24–26]. Instead of removal of these nutrients, recovery is the more sustainable and valuable approach, because a major percentage of these valuable nutrients are lost to the environment. MFCs have been successfully applied to achieve both these strategies.

Undoubtedly, the discharge of these effluent streams into soil and water bodies has led to widespread contamination of these natural systems. Even if the effluent itself is treated to sufficient levels, the sludge generated during the treatment process contains accumulated pollutants at concentrations far exceeding the levels present in the untreated effluent [27–29]. This sludge becomes a toxic repository of heavy metals, persistent chemicals and recalcitrant organic compounds, and without adequate treatment, the contaminants present in this solid waste leach out, eventually accumulating in soils and river sediments [30–32]. Remediation of these polluted systems requires strategies that are inexpensive, effective, easy to deploy and minimally intrusive to the natural environment [33]. MFCs utilizing soil and sediment as substrates have been shown to be successful for *in situ* treatment of these polluted systems [34–37]. These MFCs are benign to the natural system, triggering the activity of electroactive microorganisms native to the soil or sediment for degradation of the waste [38,39].

In recent years, heavy metal contamination of natural resources has become rampant as toxic metals (Cd, Cr, As, Hg) are being found in

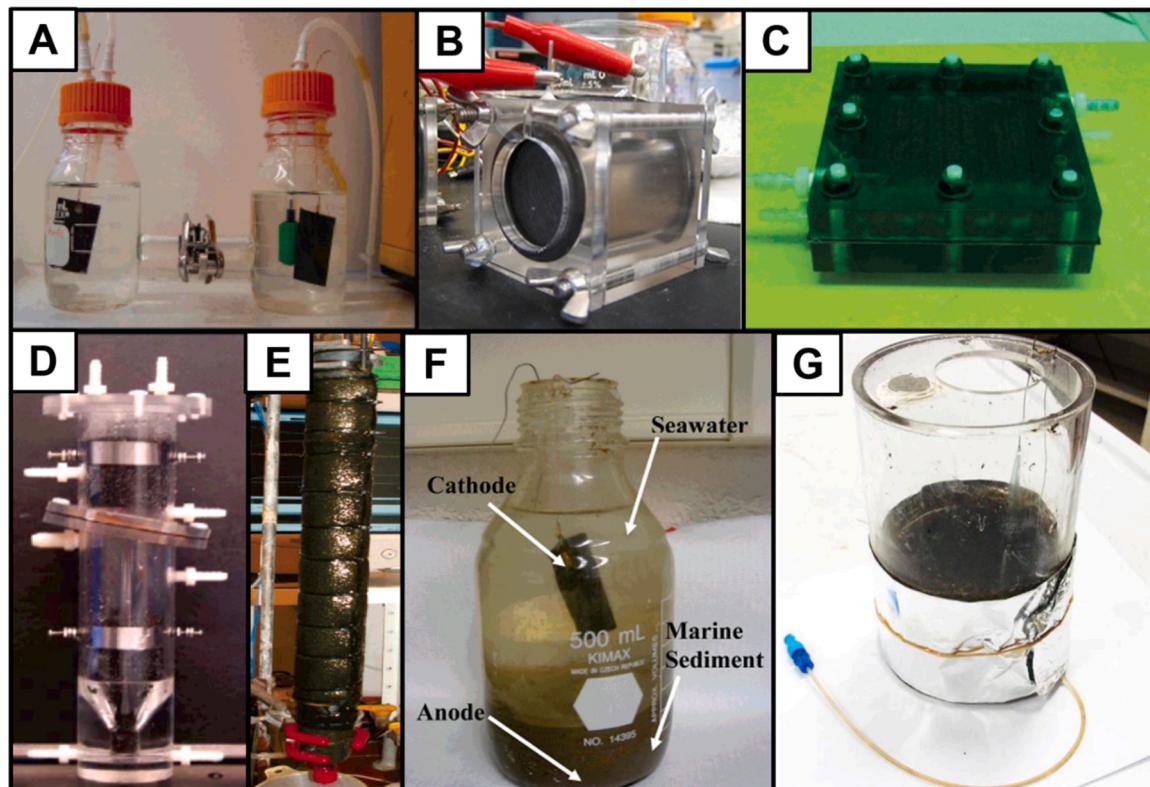


Fig. 1. Design of various types of MFCs. (A) H-type DCMFC [276]; (B) SCMFC air cathode system [277]; (C) Flat plate MFC with PEM [278]; (D) Upflow, tubular type MFC (anode below, cathode above) [279]; (E) Upflow, tubular type MFC (inner anode and outer cathode) [280]; (F) Sediment MFC [281]; (G) Soil MFC [282]. Reprinted with permission from references.

elevated levels in soil and water bodies. The common practice of unchecked dumping of hazardous wastes from metallurgical, mining, electronic and tannery industries have contributed greatly to heavy metal pollution [40–43]. MFC-based systems can either reduce these metal ions at the cathode or initiate electrokinetic migration [44–46]. This makes the technology a successful alternative to other expensive technologies of heavy metal removal [47].

The versatility of the technology lies in the fact that not only is it capable of treating effluents at the source, but it can also be an effective bioremediation solution for natural systems contaminated by these effluents. This review therefore is intended to highlight the current progress of this multifaceted technology for the treatment of a wide variety of industrial wastes. For each category of contaminant, the industries that can adopt MFCs for removal and recovery have been identified. Successful studies conducted in large scales have been discussed, including those where the MFCs were operated efficiently over long periods. Emphasis has been placed on configurations and design parameters that would be more economically attractive to industrial effluent treatment plants. An analysis of the operational challenges that an MFC-based treatment process entails and the corresponding solutions are presented. Future considerations that need to be made in order to employ this technology alongside traditional treatment units have also been discussed. A proper effluent management strategy involves treatment, recovery and remediation to support the efforts of preserving the natural systems [48]. MFCs offer the benefit of energy and resource recovery while being readily adaptable to wastes containing a diverse class of contaminants. Therefore, this technology, if its limitations are addressed, has the potential to serve as a green, cost-effective treatment solution for proper management of industrial wastes.

2. Industrial pollutant removal using MFCs

MFCs have been studied extensively for wastewater treatment, in order to examine the suitability of this technology as a practical treatment option for various types of effluent. Current treatment processes are energy-intensive and the associated costs can be significant. In addition, the generated sludge needs to be properly disposed of. Most importantly though, majority of these processes do not yield any economic benefit as their operation involves a net input of energy and does not recover any of the valuable resources present in the waste effluent. MFCs offer a sustainable approach in wastewater treatment because the degradation of the organic contaminants at an anode by electroactive microorganisms makes the treatment a net energy-generating process.

Untreated sludge from industrial wastewater treatment plants, pesticide residues, excess fertilizer, toxic organics and hazardous heavy metals all eventually find their way into the water bodies or accumulate in the soil. This diverse class of contaminants make remediation a challenging task. Numerous techniques have been developed [49,50], however, not all of them have been equally successful and cost-effective [51]. Excavation of the contaminated site and/or disposal to landfills, incineration and chemical precipitation for the removal of heavy metals still remain the methods of choice [52,53]. Although guidelines exist for proper implementation of these techniques, adhering to these requirements usually proves to be expensive [50,54]. As such, in the developing world, use of these techniques is limited [54]. The solutions themselves give rise to secondary environmental concerns. For instance, landfills bear the risk of contaminants leaching into the ground, incineration can release carcinogenic dioxins, and chemical precipitation simply transfers the pollutants to the solid sludge which itself needs treatment [50,52,53]. Increased focus is now being placed on developing sustainable solutions that have minimum environmental impact, and bioremediation techniques therefore seem favorable for decontamination [55].

Bioremediation technologies all use indigenous population of microbes in wastewater and can be based on a range of different techniques: bioaugmentation, biofiltration, biostimulation, bioreactors and

bioventing [50]. Among these, in biostimulation, the indigenous population of microorganisms in the contaminated region are already adapted to degrade the contaminants. This technique could be the most effective solution for a wide range of industrial effluents [56], as it does not bear the risk of mutating the microbial species [57]. However, without the presence of suitable electron acceptors and due to the inefficient rates of electron transfer, the process can be slow to deliver reasonable results [35]. Use of a bioelectrochemical system solves these issues, as the anode acts as an electron acceptor to which the bacteria can easily transfer their electrons, and the current generated enhances the growth of the microbes [39]. The directed flow of electrons causes redox processes to occur without much impediment, and no power input is necessary [35], unlike the more expensive electrokinetic remediation (which has been touted as being better than existing remediation techniques [51]).

An integral mechanism of MFC operation is reduction. In cases where the reduced species is the contaminant, MFCs would be beneficial as a bioremediation tool. MFC technologies have been utilized to remove heavy metals, organic substances and even toxic dyes present in soil, sediments or water [11,36,58,59]. This promising new bioremediation solution can be applied to real-life systems in the immediate future. Operation is effective at moderate temperatures; however, the category of MFC and the configuration can vary, depending on the contaminant type. The various aspects of operation to be taken into consideration to make the MFC a viable solution for industrial waste management is discussed in the following sections.

2.1. Removal of simple organic pollutants from industrial wastewater

Recent developments in MFCs for wastewater treatment have reached a stage where the technology is no longer simply confined to laboratory scale units operating on synthetic waste streams. There are several advantages that MFCs provide over traditional treatment technologies such as a high theoretical energy conversion rate, 50–90 % less sludge generation compared to aerated activated sludge units [9,60] and suppression of any gas production, with concomitant output of power [61]. The amount of energy recovered in the treatment of wastewater is almost half of what is required for aeration in conventional treatment systems [9]. The organic molecules of target are not just limited to hydrocarbons. Sulfides (via *Pseudomonas fluorescens* and *Paracoccus denitrificans*) and sulfates (via *Desulfovibrio adipica*, *Desulfovibrio desulfuricans*, and *Thermodesulfovibrio aggregans*) have also been removed from wastewaters via a variety of MFC configurations. In fact, attaching an MFC unit to treat sulfide-contaminated waste stream emerging from an anaerobic digester could be a solution for sulfide treatment [62]. The pollutant removal rates and MFC configuration for various industrial wastewaters have been summarized in Table 1. These results serve as a strong testament to the versatility of MFC technologies in the treatment of diverse classes of wastewater. The performance of MFCs with food-industry wastes is significant, with more than 90 % COD removal. High-strength industrial wastes are difficult to degrade, which is reflected in the moderate COD removal efficiencies achieved [121,139,140]. It is possible to treat high-strength wastewaters by utilizing longer hydraulic retention times (HRT). Use of two air-cathode MFCs in series having an HRT of 16.7 h could continuously treat swine wastewater having a COD concentration of 8 g/L over a period of 185 days [63].

In order to determine whether MFCs can be implemented as a practical solution to effluent treatment, the performance of scaled-up MFC reactors with real wastewater has to be assessed. Several successful studies on large-scale units have been reported. The basic strategy for scale up involves using modularized units where multiple reactors are stacked in series or parallel, instead of a large single unit where transport limitations impede reasonable performance [64]. Stacked modules generate reasonable power output, while having higher liquid capacities [65,66]. A 1000 L unit consisting of 50 modules operated with municipal wastewater (at two different COD concentrations of 80 mg/L and

Table 1

Results of treatment of various types of wastewater by MFCs with different configurations.

Wastewater	Configuration	COD removal %	Anode material	Power output (mW)	Working volume (mL)	Power density, ^b sed on ^a node chamber (W/m ³)	References
Dairy industry	DCMFC	90	Graphite plate	5.2	2000 ^a	2.6	[283]
Petroleum refinery	DCMFC	64	Carbon rod	0.132	400 ^a	0.33	[284]
Human feces	DCMFC	71	Carbon paper	787	1000 ^a	0.787	[285]
Furfural	SCMFC	68	Carbon fiber brush	0.324	18 ^a	18	[286]
Effluent sludge of palm oil mill	DCMFC	3	Carbon graphite	nr	100 ^a	451.26 [*]	[287]
Distillery	SCMFC	72	Graphite plate	0.87	500	1.74	[288]
Dyeing	SCMFC	71	Graphite rods and granular activated carbon bed	nr	2500	8.0	[158]
Sewage	SCMFC	88	Graphite rods	0.0711	10.6×10^3	0.0067	[289]
Primary clarifier effluent	SCMFC	40–50	Graphite rod	9.26×10^{-4}	388 ^b	2.4×10^{-3}	[290]
Hospital wastewater	SCMFC	–	Graphite granules and graphite rod	1.5	2000	0.75	[291]
Human urine	SCMFC	75	Carbon brush	0.055	130 ^a	0.42	[292]
Swine wastewater	SCMFC with air-cathode	83	Carbon brush	13	340	37.5	[22]
Effluent from fermentation processes differing in their end products	SCMFC with air-cathode	85–90	Graphite fiber brush	nr	28	62–835 [*]	[293]
Polyalcohol water	SCMFC	71–91	Carbon cloth	0.304–0.446	12	25–37	[294]
Industrial	SCMFC	84	Carbon cloth coated with LiNbO ₃	nr	nr	0.131	[295]
Phenol	SCMFC	88	Carbon felt	0.119	330	0.36	[296]
Synthetic penicillin	SCMFC with air-cathode	90	Carbon felt	0.21	100	2.1	[297]
Rice mill	SCMFC with earthen pot	96	Stainless steel	0.108–0.92	400	0.27–2.3	[298]
Tannery	SCMFC	88	nr	nr	nr	7 [*]	[299]
Domestic	Upflow membraneless MFC	78	Carbon fiber brush	nr	nr	20.3	[101]
Animal carcass	UTMFC	51	Graphite felt and granular activated carbon	1.64	750	2.19	[300]

Note: MFC – microbial fuel cell; SCMFC – single-chamber MFC; DCMFC – double chamber MFC; UTMFC – upflow tubular MFC.

nr- not reported.

W/m³- power normalized with respect to net working volume of anode chamber or in case of SCMFC, working volume of reactor.^a Power density: mW/m²- power output normalized with respect to anode area.^a Net anode chamber volume.^b Empty bed volume.

250 mg/L) for over a year had achieved COD removal rates at an average of 70–90 %, whilst ensuring effluent concentrations met the discharge standards [67]. Studies of continuous treatment of brewery wastewater was conducted with a 20 L membrane-less MFC system that removed up to 95 % of the COD [68]. Ge and He studied the performance of a 200 L MFC system (96 modules) at a wastewater treatment facility where almost 75 % COD and 90 % TSS removal percentages were reported. With the use of carbon brush anodes and carbon cloth cathodes, the capital cost of the system was USD 58 per gallon per day of wastewater treated, which is comparable to that of a small wastewater treatment facility [69]. Their setup is shown in Fig. 2. This indicates the potential

of an MFC system to replace septic tanks, or to serve as a wastewater treatment unit in remote areas where a lack of power limits the use of aerated bioreactors for wastewater treatment [13]. To treat the primary clarifier effluent from a domestic wastewater treatment plant, five flat-panel air-cathode MFC units (where the electrode area was 600 cm²) were connected in series. During eight months of operation, the COD removal rate was found to be 85 %, while total nitrogen removal was 94 % [70]. Instead of reducing dissolved oxygen, which is a common feature in most large-scale MFCs [71,72], cathodes that directly reduce the air must be developed. This can significantly reduce the energy expenditure in operation of the MFC as it eliminates the need to dissolve

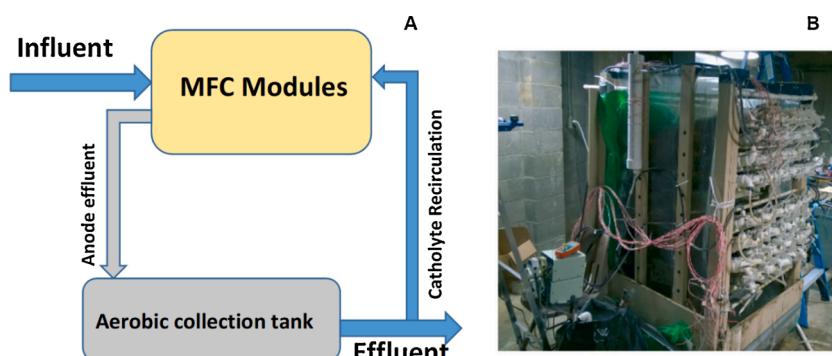


Fig. 2. Modularized MFC system for continuous treatment of wastewater: (A) the schematic of the modules and an aerobic collection tank; (B) the setup in a wastewater treatment plant. Reproduced with permission [69].

the air in the catholyte [73]. Rossi et al. developed a multi-panel air-cathode system, consisting of 15 smaller cathode panes (projected surface area of 324 cm²) welded onto a single stainless-steel frame, forming a cathode panel with an active surface area of 6200 cm² [74]. The maximum power density obtained using this cathode in an 85 L MFC was 0.083 W/m², almost 1.5 times higher than that achieved in a stacked MFC configuration where the total cathodic surface area was around 10,000 cm² [75]. With the use of the metallic frame, the electrical resistance between the frame and the individual cathodes could be maintained below 0.2 ohms. In large systems, resistances associated with the cathode can govern the power production, thus any modifications to reduce Ohmic losses or catholyte resistance can raise the power output [74,76]. To assess the long-term potential of these multi-panel cathode systems to treat raw wastewater, a 255 L MFC module consisting of two of these panels (85 cm by 85 cm), was exposed to municipal wastewater and operated for 98 days. For a flow rate of 144 L/d, the power density reached 317 mW/m³ (with respect to the reactor volume), and 41 %, 36 % and 18 % of the COD, TSS and total nitrogen were removed, respectively [77].

Generally, in effluent treatment plants, several treatment technologies are combined to produce a high-quality effluent. MFCs can be an additional option to incorporate into this treatment chain [8]. For instance, a single-chamber MFC with air cathode was combined with a low-cost flocculation unit to improve effluent quality from raw swine wastewater containing more than 2700 mg/L COD. The system removed 83 % of the COD and 99.1 % of the ammonia (at a rate of 269 g/m³ per day) in the MFC. With a further reduction of the COD in the flocculation process, overall 97 % of the COD was removed, at a net cost benefit of USD 0.026 per m³ of wastewater [22]. MFCs connected to osmotic membrane bioreactors were able to achieve high power densities (11.5 W/m³). Due to the pretreatment in the MFC, the sludge production was lower (0.076 g TSS/g COD and < 350 mg/L TSS), which could have otherwise fouled the thin-film composite polyamide forward osmosis membrane, and the effluent COD was maintained below 20 mg/L while PO₄³⁻-P rejection was more than 99 % [18]. A microbial electrochemical fluidized bed reactor (containing granular activated carbon as the anode bed) treating the outflow from an electrocoagulation unit removed 93 % of the total suspended solids (TSS) and 88 % of the COD from brewery wastewater containing 625 mg/L of TSS and 2888 mg/L COD [78].

For rapidly developing countries, management of effluent, more often than not, is a secondary concern, which may only be realized when the damage is manifest. Small-to-mid-scale factories cannot afford to invest their limited resources in effluent treatment plants [4]. Commonly used biological treatment units in these factories are activated sludge processes, which produce large quantities of sludge, handling of which constitutes almost 50 % of the operating cost [79,80]. Aeration of the units is another major expense, covering from 10 to 30 % of the operating cost [81,82]. For these establishments, in order to reduce the costs of treatment, an MFC system is a highly promising option.

2.2. Removal of inorganic pollutants from wastewater

In conventional effluent treatment facilities, soluble nitrogenous compounds, ammonium and orthophosphate ions generated during anaerobic and aerobic digestion are released into the side streams, causing the problem of nutrient rich effluents. Wastewater containing excess amounts of inorganic nutrients is not suitable for discharge, due to the risk of eutrophication [83]. The usual method of handling these streams with elevated nutrient concentrations is to recycle them to the inlet of the plant, but the non-uniformity in the concentration of the constituents and flow rates impede the performance of the treatment units. A better practice is to treat the ammonium and phosphate-enriched streams in separate units [84]. Although efficient conversion of nitrogen can be carried out using the Annamox process, this requires significant energy input and produces emissions of nitrous

oxide [85,86]. MFCs have presented a sustainable treatment approach for removing nitrogen and phosphorus from wastewaters without the extensive energy input [13,87]. In addition, it was determined that nitrogen compounds, when present in the organic substrate, can interfere with MFC operation if the denitrifying bacteria compete with electroactive microorganisms for the electrons [88,89]. Sukkasem et al. reported that the power production decreased to a certain extent when nitrate ions were present in an SCMFC [88]. This is because typical denitrifying bacteria are heterotrophic in nature, thus they consume organic matter, preventing electrogenic bacteria from utilizing it instead. There is therefore an additional motivation for controlling the nitrogen levels in the MFC's wastewater substrate, which is to limit the negative impact on the performance of electroactive microorganisms and consequently, on energy recovery.

Conventional denitrification involves nitrate ions accepting electrons from the degradation of organic compounds. A bioelectrochemical alternative to this process is autotrophic denitrification, where bacteria capable of carrying this out occupy a cathode where they reduce nitrate ions to nitrogen. This denitrification process is suitable if the target is the removal of nitrogen from the waste substrate. However, typically the wastewater is more enriched in ammonium ions and therefore the strategy should be to oxidize the ammonium ions to nitrate first, which can be accomplished via aerobic nitrification [90]. The resulting nitrate-rich stream is then denitrified using autotrophic denitrifying bacteria in a biocathode MFC [91], as the following studies demonstrated.

A membrane-less MFC achieved a 37 % reduction in total nitrogen content and generated a maximum current density of 0.844 A/m³ using a biocathode that was inoculated with an anoxic denitrifying culture [92]. Zhang and He designed a dual-cathode system, where the inner cathode reduced nitrate ions, removing between 67 % and 90 % of the total nitrogen [93]. Another configuration employed a dual DCMFC system, in which the first MFC oxidized sulfides and reduced oxygen to convert ammonium ions to nitrate. The nitrate-rich effluent from this cathodic chamber was then fed to the second MFC where the anoxic cathode converted the nitrate to nitrogen gas, removing more than 58 % of the total nitrogen in the process [94]. This option is suitable for treating streams that have a high ammonium content [95]. Flat-panel air cathodes connected in series (Fig. 3), having a hydraulic retention time (HRT) of 2.5 h, removed 94 % of the total nitrogen from domestic wastewater. Upon analysis of the bacterial community at the cathode, presence of *Nitrosomonas*, *Nitratreducator* and *Acidovorax* strains were detected, which were determined to be involved in nitrogen removal. The first two units sufficiently reduced the overall load of total nitrogen at a rate of 0.62 kg-N/m³.day [70]. Heterotrophic denitrification also contributes to the nitrate removal at the anode if the organic content is high [96]. Ammonium nitrogen (NH₄⁺-N), toxic to living organisms, is present in elevated levels in coking wastewater, and cannot be treated adequately in systems that are more directed towards the removal of the refractory carbonaceous pollutants. An integrated electrochemical system consisting of three MFCs in addition to a general electrochemical system removed 71 % of the total nitrogen in coking wastewater. The NH₄⁺-N was first converted to NO₃⁻-N, which was then removed in the final denitrifying reactor through microbial action. The effluent, containing only a low quantity of residual hydrocarbons, could then be treated in conventional biological treatment units [97]. The Annamox process can also be integrated into an MFC system by inoculating the anode chamber with anaerobic digestate, in order to treat wastewaters containing a low carbon-nitrogen ratio. Analysis of the anodic bacterial community revealed the coexistence of annamox bacteria with the electrogenic population [98,99]. The Annamox bacteria in the anaerobic anode chamber oxidized the ammonium ions, while the nitrate/nitrite ions produced in the reaction were reduced either in the anodic chamber [98] or by denitrifying bacteria present in the biofilm of the cathode [100].

Phosphorus removal can also be achieved if a photosynthetic process

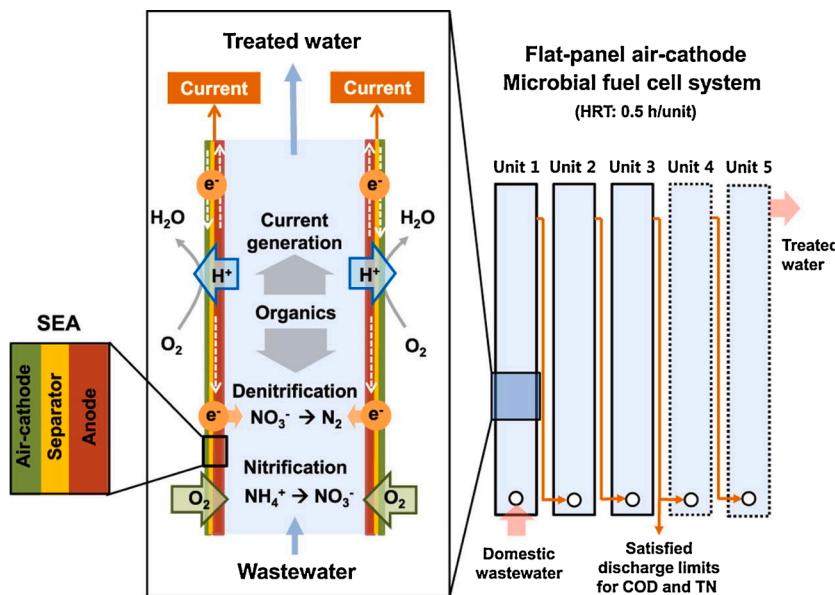


Fig. 3. Flat-panel air-cathode MFC (FA-MFC) with five MFC units connected in series. Reprinted with permission [70].

is incorporated into the MFC. Microalgae grown in MFC systems have reportedly removed between 70 and 92 % of the phosphorus [23, 101–103]. An integrated photo-bioelectrochemical system consisting of an MFC inside an algal bioreactor was operated over a period of 12 months, and removed 98 % nitrogen and 82 % of the phosphate, achieving a power density of 2.2 W m^{-3} . The algae produced the necessary amount of oxygen for reduction at the MFC cathode, which is a cost-effective strategy as it did not necessitate sparging air into the system [102]. The combination of an electrocoagulation unit with a fluidized bed MFC was used to treat brewery wastewater containing more than 66 mg/L total nitrogen and 15 mg/L total phosphorus. Overall 93 % of the nitrogen and 98 % of the phosphorus were removed; less than 5 mg/L of the total nitrogen and less than 0.3 mg/L of the phosphorus were detected in the effluent [78]. However, because phosphorus is a valuable and limited resource, efforts should be directed towards its recovery, not removal [104].

Limiting the concentration of nitrogen in groundwater by employing MFCs has also been analyzed [105,106]. Treatment of actual groundwater contaminated with nitrate ions was carried out in a variation of the MFC, called a microbial electrolysis cell, where a slight external potential was applied and a removal rate of over $130 \text{ g NO}_3\text{-N/m}^3\text{.day}$ was achieved [96]. Zhang and Angelidaki designed a denitrification cell where, during the startup period, the MFC was operated with oxygen as the electron acceptor. The generated electric field caused nitrate ions to migrate towards the anode, followed by autotrophic denitrification at the cathode [107].

MFCs can simultaneously treat the raw wastewater and the nutrient-rich side streams from the other treatment units, with the oxidation of the contaminants in the former supporting the reduction of those in the latter. However, this is not a feasible strategy when the raw wastewater itself contains nutrients, as heterotrophic denitrification will take precedence. Furthermore, a high organic content also stimulates heterotrophic denitrification [108]. In such cases, energy recovery efficiencies would dwindle, and thus, operating the bioreactor as an MFC would not yield any additional returns. Therefore, removal of these nutrients is only suitable when the nutrient load in the waste stream is low and the waste itself is not highly concentrated with organic substrates [87]. In other cases, the more sustainable option is to recover these nutrients.

2.3. Heavy metal remediation

Industrial effluents discharge heavy metals at alarming levels that have the potential to create lasting ecological damage. Oxidation and reduction both occur in an MFC, and the latter process also participates in contaminant degradation. As organic matter is oxidized in the anode compartment, heavy metal ions at higher oxidation numbers (chromium (VI), copper (II), vanadium (V) etc.) can be reduced at the cathode by the electrons that are released by the electroactive microorganisms [46]. Usually these heavy metals accumulate in the sludge generated in conventional treatment units, which is often discharged into the environment without adequate treatment of the sludge. This is the primary cause of soil and sediment pollution. MFCs have been successfully applied for the reduction of several of the common heavy metals found in these polluted soils and sediments.

2.3.1. Chromium removal

Extensive studies have been conducted to test MFC performance in converting toxic Cr (VI) to harmless forms. Wang and coworkers had achieved complete removal of chromium (VI) from 100 mg/L in synthetic wastewater in a DCMFC [109]. Some authors report that specific classes of bacteria are adapted to degrade metal contaminants, including Cr (VI) [11]. On the other hand, mixed cultures derived from anaerobic sludge, or algae biomass were also reported to be able to remove Cr from contaminated sites successfully [110,111]. These studies reported removal efficiencies from 75 to over 90 % [46]. In 26 h, 97 % of the Cr was reduced from Cr (VI) to Cr_2O_3 that precipitated on the surface of a rutile-coated cathode, where 0.80 V was generated under irradiated conditions, indicating that light catalyzed the electron transfer [110]. Cr-contaminated sludge from a leather tanning wastewater treatment facility was used to inoculate a DCMFC operated with synthetic wastewater achieving a 93 % reduction in Cr (VI) from an initial concentration as low as 5 mg/L [113]. Effective soil and sediment remediation has also been achieved in MFCs where Cr (VI)-contaminated soils were taken in the cathode chamber of a DCMFC. The anode chamber contained acetate as the substrate for bacterial consumption, and the electrodes were made of carbon felt. The results showed Cr (VI) removal efficiencies of 99.1 % and 62.7 % from the two types of soil tested. The lower efficiency in the second type of soil was attributed to its higher iron content, which competed with the Cr ions [36].

2.3.2. Arsenic removal

Arsenic contamination of groundwater is of the most concern in South-Asian regions. Rice grains tend to accumulate arsenic (mostly as H_3AsO_3) that the roots draw up along with the porewater. Rice being a semi-aquatic plant compensates for the lack of oxygen in the soil it grows on by releasing residual oxygen through its roots. The oxygen converts ferrous (Fe^{2+}) to ferric ions (Fe^{3+}), forming iron oxides/hydroxides that act as sequestering agents for As (V); this is known as iron plaque. It prevents As(V) from being absorbed through the roots and accumulating in the grains [114,115]. However, flooding during cultivation causes the soil to become anoxic, wherein bacteria oxidize the organic matter, with iron oxides serving as the electron acceptors [116]. The iron ions dissolve, releasing As (V) that is then reduced to As (III). It then binds less readily to the plaque and easily diffuses to the porewater, from where the plants can draw up the arsenic. This problem is especially exacerbated when As-contaminated groundwater is used for irrigation [117]. Current methods of decontamination are costly and require frequent maintenance. Detection and remediation of arsenic ions (As (III)) in water bodies demand cost-effective and convenient solutions. Miller et al. isolated an As (V)-respiring bacterium called *Bacillus sele-nitireducens*, which is also electrogenic in nature. The power production in an MFC inoculated with the bacteria decreased from $60 \mu\text{W}/\text{m}^2$ to $3 \mu\text{W}/\text{m}^2$ in the presence of As (V) in the anode chamber, as the ions compete for the electrons against the species in the cathode chamber. This device can thus function as an easy-to-use detector of arsenate ions. Another species, *Alkalilimnicola ehrlichii*, can oxidize arsenite (As(III)) ions in the anodic chamber, releasing electrons that make the MFC operational [118]. An indirect method of controlling the release of As (III) in groundwater was studied by Gustave et al., who packed paddy soil into an SCMFC and operated the system for a period of 60 days to observe the changes in As and Fe concentrations in the soil porewater. The average Fe and As concentration in the porewater samples collected from the vicinity of the anode were significantly lower (65.0 % and 47.0 % of the control MFCs at day 50) than in the negative control MFC (open-circuit conditions, no power produced). This was attributed to the fact that at such conditions, electroactive microorganisms dominated the environment, preventing iron-reducing bacteria or As (V) reducers from oxidizing the organic matter (OM) instead. Upon consumption of the OM by the electroactive microorganisms, the released electrons were transferred to the anode, which then went on to reduce the oxygen in the cathodic chamber [119].

2.3.3. Copper removal

Wastewater contaminated with copper (II) ions can also be treated in MFCs, with removal efficiencies ranging from 85 to more than 95 % [120,121]. Copper ions can also stimulate bacterial activity in MFCs, resulting in additional benefits of high-power outputs and superior sulfate removal efficiencies. The sulfate-reducing activity of *Desulfovibrio*, on a bioanode enriched with the bacteria, was enhanced by the addition of Cu^{2+} ions (the maximum concentration tested was 20 mg/L), resulting in the copper ions precipitating out as the metal sulfide. As a result, 98 % of the ions were removed. In this study, the power density was $224.1 \text{ mW}/\text{m}^2$, while COD and sulfate removal efficiencies were greater than 85 % and 70 % respectively [122].

2.3.4. Removal of other heavy metals

Cadmium and lead are two other toxic metals that are serious contributors to soil toxicity. In one study, an air-cathode DCMFC was constructed using a graphite anode and a carbon felt cathode, with the electrode chambers separated by a cation exchange membrane. The cathode chamber was filled with soil contaminated with Cd and Pb (98 mg/kg and 910 mg/kg respectively), and it was observed that the electric field generated in the chamber facilitated the migration of Cd and Pb ions from the vicinity of the anode towards the cathodic zone. The homogenous concentration no longer existed; the soil near the anode experienced respectively a 31 % and 44.1 % decrease in Cd and Pb

concentrations [45]. Abourached et al. tested the removal efficiencies of Cd and Zn in the anode chamber of an air-cathode SCMFC (carbon cloth electrodes). 90 % and 97 % removal of Cd and Zn respectively were reported from a substrate medium containing $200 \mu\text{M}$ Cd and $400 \mu\text{M}$ Zn. The power output was also significantly high, reaching $3.6 \text{ W}/\text{m}^2$. The mechanism of removal in this case was biosorption by the cell walls of the bacteria and sulfide precipitation (produced when bacteria reduced sulfate ions present in the medium). These results have strong implications that the removal can be effected with simultaneous power production if the anode chamber contained the heavy metal contaminants [44]. In place of an aqueous substrate medium, studies can be focused on the remediation of contaminated soil/sediment [11]. During treatment of oil sand tailings in a DCMFC, removal efficiencies of 97.8 % for selenium, 96.8 % for barium, 94.7 % for strontium, 81.3 % for zinc, 77.1 % for molybdenum, 66.9 % for Cu, 44.9 % for Cr, and 32.5 % for lead were achieved along with a power density of $392 \text{ mW}/\text{m}^2$ during a 1700-h operation [123]. Membrane-less sediment MFCs were used to treat sediment contaminated with mercury, silver and zinc, where 69.53 %, 66.57 % and 65.33 % of the respective ions were removed [34].

2.3.5. Practical issues with MFC use for heavy metal remediation

From the foregoing examples, it can be inferred that treatment of heavy metals in MFCs can be achieved without requiring any energy input, which establishes the superiority of the MFC technology over conventional heavy metal removal processes. The primary removal mechanism is either absorption into the bacterial cell walls or immobilization on the cathodes [124], and so MFCs minimize chemical sludge production and provide the possibility of recovering these metals [125]. A major shortcoming, nevertheless, is the fact that the technology cannot be universally applied for the removal of all heavy metals. Mercury, for instance, is converted to toxic methyl mercurate [33]. Sulfide generation can poison microorganisms, eventually limiting the power output. The metals that have been removed can, at high concentrations, pose a threat to the viability of the bacteria [44]. In addition, most studies have been conducted using MFCs where the metal ions were in the cathodic chamber and their migration towards the anode was prevented by the use of membranes. This is beneficial because heavy metal ions can be toxic to the microbes at the anode, which degrades the performance of the MFC [126,127]. However, when real wastewater or contaminated soil is treated, it is not possible to prevent the anode from being exposed to the ions because both the organic compounds and the metal ions coexist in the substrate. In these cases it might be necessary to maintain the concentrations well below levels at which the ions begin to exert their inhibitory effect on the bacteria [34,122,128–130]. In fact, Xu and coworkers determined that at trace levels, Cu^{2+} and Cd^{2+} could enhance the power production by improving bacterial attachment on the anode [128]. Table 2 summarizes studies reporting configurations where the entire chamber was exposed to the metal contaminated substrate (MFCs treating metal-contaminated wastewater, soil or sediment MFCs). Although the removal rates are consistent, the power output varies over a range, which emphasizes the need to set the upper limit of the metal ion concentration based on the power requirement.

2.4. Degradation of complex organic compounds

Certain organic compounds present in complex industrial effluents tend to be more persistent, undergoing only limited degradation in nature under normal conditions. These recalcitrant organic compounds have toxic, mutagenic and carcinogenic potential [131]. The use of MFC technology can stimulate the process of degradation, as the current generated is able to accelerate the redox processes [35]. The generation of the current also stimulates the microbial population to consume substrates at a greater efficiency. This is due to the fact that the degradation products are metabolized by the electroactive microorganisms in the final stages and electrons are transferred to terminal electron acceptors [132,133]. Complex organic contaminants that have been

Table 2

Removal of heavy metals from contaminated organic substrate present in the anode chamber.

Heavy metal treated	Concentration	System configuration	Electrode materials	Maximum power output (mW)	Maximum removal efficiency (%)	References
Hg, Zn and Ag	Hg(II): 1.941 mg/kg Zn(II): 512 mg/kg Ag(I): 10.5 mg/kg Cd(II): 200–300 μM	Membrane-less sediment MFC	Graphite felt	0.34	Hg(II): 69.5 Zn(II): 66.57 Ag(I): 65.33 Cd: >89	[34]
Cd and Zn	Zn(II): 400–500 μM	Membrane-less air-cathode MFC	Carbon cloth	0.65	Zn: >94	[44]
Fe and As	Fe(III) and As(V): nr	Soil MFC	Carbon felt	0.06	Fe: 65 As: 47	[119]
Cu ^a	Cu(II): 20 ppm	DCMFC containing metal-contaminated wastewater in anodic chamber	Carbon felt (anode); Pt-coated carbon cloth (cathode)	0.56	>98	[122]
Cu	Cu(II): 12.5 ppm	SCMFC	Carbon brush (anode); carbon cloth (cathode)	0.29	98.3 Cr: 81 Cu: 73	[129]
Cr, Ni and Cu	Cr(VI): 390.3 mg/kg Cu(II): 480.1 mg/kg Ni(II): 180.5 mg/kg	Sediment MFC with aerated cathode	Graphite plate	2.7	Ni: 80	[301]

Note: MFC – microbial fuel cell; DCMFC – double-chamber MFC; SCMFC – single-chamber MFC.

^a Conversion to sulfide at the anode.

effectively degraded include polycyclic aromatic hydrocarbons (PAHs) such as in pesticides [59,134,135], dyes [136,137] and petroleum hydrocarbons [138–140].

2.4.1. Removal of pesticide-based pollutants

Moderate-to-low degradation rates in the soil allow pesticides in the form of atrazine and the bioaccumulative hexachlorobenzene (HCB) to persist in the natural environment. Soil MFCs have been successfully employed to destroy these contaminants at levels exceeding 90 % and 50 % (for atrazine and HCB respectively) [59]. Removal of potential carcinogens, phenanthrene and benzene was tested in a tubular single-chamber MFC containing carbon felt electrodes whose projected surface areas were close to 200 cm². At a hydraulic retention time (HRT) of 10 days, more than 90 % of the hydrocarbons were degraded (at initial concentrations of 30 ppm phenanthrene and 200 ppm benzene), while the peak power density reached 6.75 mW/m². Changing the HRT did not negatively affect the hydrocarbon removal percentages. The system proved robust as it could handle extreme concentrations, as low as 50 ppb for both contaminants; however, the power output in this case was quite low. At higher ends, the system could degrade up to 1500 ppm benzene and 100 ppm phenanthrene at an HRT of 30 days. Operation lasted 155 days, signifying the long-term potential for the treatment of contaminated groundwater and soils. In the same study, a similar set-up was tested for continuous treatment of contaminated wastewater and the effect of high organic shock loading was investigated. When the concentrations of phenanthrene and benzene were raised from 30 and 200 ppm to 100 and 1500 ppm respectively, the COD removal rate dropped from 77 % to 64 %. The maximum power density decreased from 6.75 to 5.74 mW/m². The removal efficiencies of the hydrocarbons continued to be over 80 % when the concentrations were changed. The fact that even at a high organic shock loading, the system maintained its power density, and COD, benzene and phenanthrene removal percentages with only slight decreases, proved that continuous operation was also stable. Additionally, the oxidizing agent at the cathode was bromate ions, which resulted in power densities comparable to when the cathodes were coated with a platinum catalyst; simultaneous removal of the contaminant bromate was achieved. This suggests that the use of expensive platinum cathodes can be avoided in such large-scale treatment applications if bromate contaminated wastewater is used instead [141]. The banned herbicide atrazine, still widely used in certain parts

of the world, was effectively removed from a contaminated soil sample in an MFC consisting of graphite plate anodes and graphite felt cathodes. A maximum current density of 66 mA/m² was obtained, for a removal efficiency of 83 % with complete genotoxicity elimination within 7 days of treatment and complete detoxification of the soil on the 14th day. MFC treatment restored chlorophyll fluorescence in *Sorghum saccharatum* species planted in the treated soil to levels comparable to those in non-polluted soil samples [132]. This is promising, as it indicates that electrically conductive materials can assist electron transfer from microbes in the soil, which has been observed in several other studies, demonstrating the concept of ‘graphite-assisted bioremediation’ [142, 143]. Removal efficiencies of persistent polychlorinated biphenyls (PCBs) reached 38 % after 60 days of operation in a sediment MFC (initial PCB concentration was 62 mg/kg sediment) where the sediment was augmented with nanoscale zero-valent iron (NZVI). The latter has a strong reducibility, therefore being able to oxidize strong organic pollutants and improve the redox potential by enhancing extracellular transfer rates [144]. In fact, microbes cultured from anaerobic sludge in an MFC could utilize N-heterocyclic compounds (y) as the sole carbon source, destroying 90 % of the compounds with power densities ranging from 140–228 mW/m² [145]. Benzo(a)pyrene, Benzo(k)fluoranthene and Benzo(b)fluoranthene present in contaminated river sediment was removed by 74 % over 72 days in the vicinity of the anode where 13 % of the microbial population consisted of *Longilinea*, which is efficient in decomposing refractory organics [37]. This study validated that unlike traditional biological oxidation processes, MFC systems can potentially treat recalcitrant pesticides that had been accumulating over long periods [146]. Agricultural field runoff and even the discharged wastewater from the effluent treatment plants of food packaging and processing industries have been determined to contain pesticide residues at levels above permissible limits [147–149]. Once these chemicals contaminate surface and ground waters, they can contribute to acute ecological toxicity [147]. MFCs can be a suitable treatment option for these contaminants, which require advanced oxidation processes for removal that may not be economically feasible otherwise.

2.4.2. Removal of organic dyes

Azo dyes, ubiquitous in the world of textile manufacturing, contain aromatic amine groups that are strongly suspected to be carcinogenic. Benzidine, a known carcinogen was discovered in effluents containing

disperse orange 37, disperse blue 373 and disperse violet 93 dyes [150]. Studies on textile factory workers revealed the occurrence of kidney, liver and urinary bladder cancers, dermatitis, asthma and rhinitis after prolonged exposure to azo and reactive dyes [151,152]. Conventional aerobic biological processes do not adequately tackle azo dyes, while advanced oxidation processes and ozonation are expensive. Microbial fuel cells have been a useful technology for the decolorization and consequent reduction of azo dyes. The azo dye Congo red was decolorized in a proton exchange membrane-air cathode MFC by 98 % within 36 h, where a co-substrate (glucose, ethanol and acetate in turns) had been used to accelerate the bacteria growth and redox process. The power density was exceptionally high, ranging from 63.2 mW/m² to 103 mW/m² [58]. The reductive decolorization is understood to involve redox mediators such as flavins or certain cell-surface proteins called cytochromes [153,154]. Studies with acid navy blue dye revealed that reduction of the azo group to aromatic amines had primarily taken place [155]. In another study on the degradation of the same dye, peak power density of 2236 mW/m² and an overall removal of 80 % were achieved in a system where glucose was added as the growth substrate [156]. For complete degradation of the reduction products, the system was coupled with a downstream aerobic unit. Degradation in the cathodic chamber is a viable option as well, and has been determined to lead to a more stable and lasting result.

Mani and coworkers compared the degradation of acid orange 7 in the anode with that at the cathode. Power density was higher when the dye was degraded at the cathode by laccase enzymes added to the cathode chamber (50 mW/m² compared to 42.5 mW/m² for anodic degradation). COD reduction exceeded 80 % when the dye was in cathode; and was 69 % with the dye in the anode. During 24 h of operation to remove acid orange 7, decolorization of the dye at the cathode was 80 %, whereas only 20 % decolorization could be achieved in the anodic chamber. Furthermore, toxicity assays revealed anodic degradation had produced the carcinogenic 1-amino-2-naphthalenol, whereas laccase degradation in the cathode had yielded phenol, hexanoic and benzoic acids that are easily removed in conventional treatment processes [157].

Operability at the large-scale without the use of expensive membranes and catalysts has also been successfully tested using dye wastewater. A single chamber MFC, having an active volume of 2.5 L with a granular activated carbon bed (GAC) as anode (100 cm³ packed volume) and GAC-filled stainless steel enclosures as cathodes was operated for 5 months using dye wastewater collected from an actual effluent treatment plant. The MFC achieved 75 % decolorization, 71 % COD removal and almost complete toxicity inhibition at an HRT of 48 h. Degradation of the dye was not attributed to adsorption into the GAC, instead it

occurred almost entirely at the electrodes, first at the anode, followed by aerobic treatment at the cathode. Both processes were influenced by microbial biofilms present on the electrodes [158]. Instead of costly GAC adsorption units that require continuous regeneration and can only be applied for the final polishing stage, the MFC alternative could be used to treat raw wastewater in textile effluent treatment plants, whose primary concern is dye removal. Table 3 lists some MFC configurations that do not use costly catalysts or have been operated continuously for dye treatment. MFCs are therefore promising when it comes to the treatment of industrial wastewaters containing complex dyes, because successful removal rates can be achieved without requiring expensive operating elements.

2.4.3. Hydrocarbon removal

Morris and coworkers reported 82 % removal of diesel from diesel contaminated groundwater collected from a refinery. In this MFC, the waste was removed from the anode chamber over 21 days of operation, and the strains isolated from the anode included *Citrobacter* sp., *Pseudomonas* sp., and *Stenotrophomonas* sp.; 31 mW/m² of cathode was generated in the process [159]. Quite recently, a novel marine microbe of the *Gammaproteobacteria* class, E2, which is highly specific to diesel, was discovered. 50 % of the diesel was utilized as the carbon source, during an operation that lasted 8 days [160]. Phenol removal was achieved with simultaneous current generation and a peak power density of 9.1 W/m³ where phenol was the sole carbon source for the bacteria in an MFC operated using an air-cathode. The phenol was degraded at an efficiency of 95.5 % in 144 h, with 95 % efficiency having reached within first 60 h. [161]. A study conducted in a DCMFC with phenol-contaminated groundwater in the anode chamber examined the degradation products of phenol. Fermentation of phenol was found to have occurred, where the final product was acetate, which was then consumed by the electroactive bacteria. 71.4 % of the phenol had been removed in the process, while the system achieved a power density of 1.8 mW/m² [162].

Soil MFCs spiked with a petroleum hydrocarbon mixture consisting of diesel oil at a concentration of 840 mg/L removed 57 % of the recalcitrant pollutants, while achieving a power density of 287 mW/m². Simultaneously, 12 % TDS and 62 % sulfate removal percentages were observed, proportional to the hydrocarbon loading. This indicated that the oxidation of the hydrocarbon stimulated the activity of the sulfate reducing bacteria in the system, confirming the presence of a synergistic ecosystem inside MFCs [163]. This is another example of the versatility of MFCs as a treatment technology. Enhanced removal (92 % degradation) of hydrocarbons (benzene and phenanthrene) was achieved under saline conditions (NaCl 1%), suggesting the potential that MFCs have for

Table 3
Dye removal in MFCs with readily adoptable design parameters.

Dye treated	System Configuration	Type of degradation	Notable operating parameters	Results	References
Acid orange 7	SCMFC with CEM integrated to an aerobic bioreactor	Reductive degradation at the anode, followed by further degradation in aerobic bioreactor	Continuous mode with HRT of 12 h; mixed microbial culture from MFC treating azo dye wastewater	>90 % dye removal; 51.9 ± 4 mW/m ²	[155]
Reactive blue 221	DCMFC	Oxidative degradation by laccase immobilized at the cathode	Graphite cathode electropolymerized using methylene blue followed by laccase immobilization	74 % dye removal; toxicity reduced up to 5.2 %	[302]
Reactive brilliant red X-3B	Constructed wetland coupled MFC system	Reduction at anode, including mineralization by the rhizosphere microorganisms	Granular activated carbon (GAC) bed as anode, <i>ipomoea aquatica</i> planted onto the GAC air cathode layer	91.24 % decolorization at a maximum rate of 55 %; maximum power density 0.302 W/m ³	[15]
Acid orange 7	MFC with CEM	Reduction at anode	Enzymatic oxidation by laccase at cathode	89 % dye removal; 78 % COD reduction	[303]
Acid orange 7	SCMFC	Reduction at anode, including consumption as substrate by the fungus	Enzymatic oxidation by laccase produced from white-rot fungus <i>Ganoderma lucidum</i> BCRC 36,123 immobilized on the cathode surface	>90 % dye removal; 13.38 mW/m ² maximum power density	[304]
Congo red	MFC with air-cathode	Reduction at anode	Addition of humic acid as mediator to enhance power and decolorization rate	100 % decolorization in 18 h; 76.8 ± 3.31 mW/m ² maximum power density	[305]

Note: MFC – microbial fuel cell; CEM – Cathode exchange membrane; SCMFC – single-chamber MFC; DCMFC – double-chamber MFC.

the treatment of effluents from refineries around coastal areas [140].

3. Resource recovery using MFCs

Metal ions and nutrients are often present in sufficiently high concentrations in industrial waste, and therefore can be recovered from the waste stream. This is the more sustainable approach to waste management. Reactions at the cathode are involved in the recovery of metals and nutrients, which can increase the value of the MFC output and may be more economically viable than energy recovery [164]. Reduction of metal ions leads to the electro-precipitation of the pure metal or one of its oxides that can be readily collected from the electrode surface. In case of nutrients, the design of the MFC itself facilitates recovery. Most of the nitrogen present in the waste is in the form of ammonium ions that migrate under the action of the electric field, diffusing across the cation exchange membrane even against a concentration gradient [165]. The ammonium ions accumulating in the cathode chamber convert to ammonia by combining with OH^- produced during the reduction reaction, thus the nutrient is transformed to an easily recoverable form. Phosphate, on the other hand, is recovered in a mineralized form (struvite, hydroxyapatite, vivianite, etc.), depending on the type of metal ions present in the system.

3.1. Nutrient recovery

Ammonium ions serve as proton shuttles and about 90 % of the ionic flux in an MFC can be attributed to these ions [166]. The ammonia produced in the cathode chamber can easily be separated [167] as shown in Fig. 4.

Desloover and coworkers determined that microbial electrolysis cells, which are different from microbial fuel cells, as the electrolysis cells have to be supplied with an external current and reduce hydrogen ions at the cathode, had the ability to reduce the ammonium content in the waste stream of an anaerobic digester. From 2.1 g N/L, a decrease of 62 % was obtained, with a power input of 13 kW h /kg N. A titanium electrode coated with iridium-ruthenium oxide served as the anode in a membrane-based cell, which was connected to an absorption-stripper unit. Cost-benefit analysis revealed that such a technology becomes profitable at applied current densities above 20 A/m² [168].

Side-streams having high ammonium concentrations could be

treated similarly. The setup by Wu and Modin recovered 79 % of the ammonia from a real reject water containing 1000 mg N/L [167]. Zou and coworkers developed a microbial electrolysis cell-forward osmosis hybrid system to treat high-strength side streams as well, where 99 % of the ammonia-nitrogen was recovered [169]. Integrated MFC-algal systems can also be used for the recovery of ammonia nitrogen. *Spirulina* growing in the cathode chamber of an MFC maintained the oxygen levels above saturation (sufficient to sustain the cathodic reaction), unlike MFCs that were operated under the usual conditions (without the algal culture). 83 % of the $\text{N}-\text{NH}_4^+$, 89 % of the COD and 40–60 % of the micronutrients (Na, K, Ca, Cr and Co) were removed from the anodic chamber. The $\text{N}-\text{NH}_4^+$, Na, K, Ca, Cr, Mn, Co, Fe were partly recovered in the cathodic chamber, whereas P and Mg were retained in the anode compartment. A maximum power density of 0.85 W/m² was obtained [170]. The ammonia can also be recovered as a more valuable product. Christiaens et al. separated the ammonia generated in an MFC where the catholyte was urine via stripping columns and used it as a substrate in the production of microbial protein [171]. In fact, the ammonia can also be incorporated into a mineral called struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$), which is the primary means of recovering the phosphorus present in the waste [172].

In an MFC, reduction of oxygen at the cathode produces hydroxide ions and this develops a high pH zone. This is suitable for the formation of the struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) precipitate, which also requires magnesium and ammonium ions to be present in the wastewater [173]. Although the precipitate slows down the current generation, removal of the precipitate restores the performance almost immediately [174]. Struvite precipitation was able to recover 70–82 % of the phosphate from swine wastewater in an air-cathode MFC [173]. A cost-effective study involving terracotta separators in an air-cathode MFC was found to achieve removal rates of 60–90 % of the soluble COD from nutrient-rich wastewaters, along with significant recovery of several macronutrients (C, N, P, K, Fe, Mn, Ca, Mg). Over a period of 125 days, a total of 10 g of the nutrients was collected for every kg of the terracotta separators, mostly seen to have been deposited on the separator. 0.32 g of the ammonia-nitrogen and 0.20 g of the phosphorus was recovered per kg of the separator, mostly as a struvite precipitate. Exceptionally high recovery was that of calcium, iron and magnesium, which was likely due to the high cation exchange capacity of the terracotta. Such a system truly embodies the concept of recycling, as the nutrient-enriched

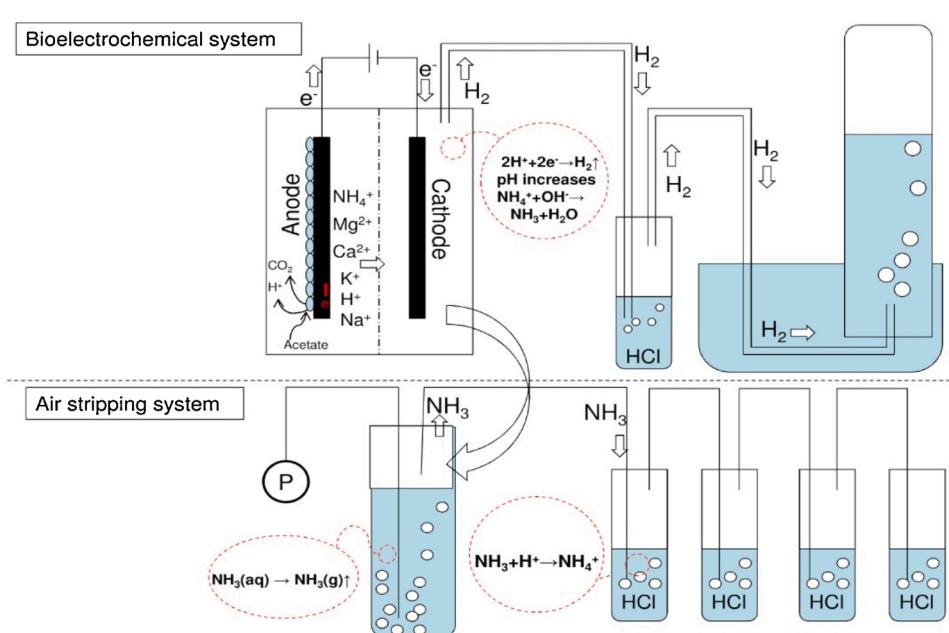


Fig. 4. Ammonia recovery in a microbial fuel cell. Reprinted with permission [167].

terracotta can then be applied to soils as a conditioner, provided the concentration of the heavy metals captured in the separator is within specified limits [175]. The microbial electrolysis cell-forward osmosis (MEC-FO) hybrid system developed by Zou et al. recovered 79.5 % of the phosphorus as struvite [169]. However, struvite recovery is not highly efficient (reported to be below 50 %) when the organic loading is high [173,176,177]. Struvite production also requires Mg^{2+} , which may need to be externally supplied [176]. Phosphorus can also be recovered as calcium phosphate if calcium ions are present in abundance. Lei and coworkers reported a phosphorus removal rate ranging from 20 to 74 %, where part of this phosphorus was recovered as amorphous calcium phosphate at the cathode. The setup was a microbial electrolysis cell, and increase in phosphorus concentration from 0.23 mM to 0.76 mM reduced the energy input from 224 kW h/kg P to 56 kW h/kg P. The recovery cost of phosphorus in this study was determined to be similar to that for mined phosphorus [17].

For recovery of nutrients to be economically viable, the nutrient load in the wastewater has to be sufficiently large. Wastewater from dairy, meat and poultry processing plants can therefore be ideal targets for nutrient recovery [178,179].

3.2. Heavy metal recovery

An MFC coupled with a thermoelectric generator (TEG) was used to sequentially recover copper, cadmium and cobalt present in smelting wastewater. By regulating the applied voltage of the cathode using the TEG (that was operated on waste heat), metallic copper was recovered at a rate of 121 mg/L.d, whereas higher recovery rates were recorded for Cd and Co at 158 and 194 mg/L.d respectively. These metals were precipitated as $Cd(OH)_2$, $CdCO_3$ and $Co(OH)_2$ on the cathode [180]. This showed that a modification of the system could be used to recover a range of heavy metals present in the same system, including those that have negative reduction potentials [181]. Plant-microbial fuel cells, a more recent derivative of MFC-based systems, harvests electricity from the degradation of organic matter by the microbes existing in the rhizosphere of the plant [182]. This region contains one of the most diverse and richest repositories of microorganisms that exist in symbiosis with the plants [183]. Nutrients and water released by the roots sustain this dynamic microbial ecosystem, while the microorganisms influence plant growth and health via complex interactions. Among these, symbiotic interactions between the plant and plant-growth promoting rhizobacteria (PGPR) mitigate toxic effects on plants and enhance the tolerance of plants to heavy metals [184]. Plant MFCs, constructed using ryegrass, achieved 99 % reduction of Cr (VI) from a concentration of 19 mg/L; the ions precipitated out as $Cr(OH)_3$ onto the electrodes. Assessment of long-term operation in field settings was performed by conducting tests without amending the system with additional nutrients. 100 % treatment of 10 mg/L Cr (VI) was observed, at a maximum rate of 1.66 mg/L/day. The accumulated residues could then be easily desorbed from the electrodes, enabling Cr recovery, while ensuring continuous operability [185].

Silver and gold, two valuable metals whose recovery is an attractive venture, have been effectively recovered via MFCs. Recovery of silver at levels above 95 % has been reported [186–188]. Wang and coworkers obtained the highest removal efficiency for silver in a DCMFC, where 99.9 % of the silver ions were reduced from a wastewater containing ammonia-chelated silver alkaline wastewater. 1.6 g Ag was deposited at the cathode for every 1 g of COD removed [188]. In another such configuration containing a carbon brush anode and a carbon cloth cathode, 98.3 % of silver was recovered after only 8 h of reaction from a wastewater system containing 200 ppm $AgNO_3$. It was calculated that 0.0143 kW h of energy output resulted from every kg of silver recovered, which was an economic advantage over conventional electro-deposition method, where an input of 3.81 kW h is required for every kg of silver recovered [186]. Wastewater MFC systems have also been utilized for the electro-deposition of gold on graphite electrodes and the formation

of gold nanoparticles [189,190]. Metal processing and electronics manufacturing industries have to discard large quantities of precious metals with the waste effluents. The recovery strategies are expensive and often involve toxic chemicals such as cyanides. In addition to preventing heavy metal contamination when these effluents are discharged, recovery of these valuable metals would complement resource conservation efforts [191–193]. The value of these metals can offset the cost of this technology.

4. Barriers in implementation of MFCs and potential solutions

4.1. Limitations in scale-up

The recent literature suggests that the low energy recovery of MFCs might make the technology more suited to contaminant removal. As a green solution to tough environmental challenges, MFCs have now been rediscovered, and efforts to solve operational and system architectural issues are being undertaken. For MFCs to compete against existing treatment systems, costs of scale-up, robustness and longevity of the fuel cells, and net positive benefits (in the form of energy and material recovery) are major concerns that need to be addressed. Fig. 5 summarizes the major challenges that need to be tackled to utilize MFCs for real-world applications. The notion that there is no profit to be incurred in running effluent treatment plants (ETP) discourages most factory owners in the developing world from setting up proper ETPs. Because MFCs are capable of recovering resources, it makes the technology more economically attractive, which might persuade these owners to install MFCs to complement conventional treatment units [194]. However, it must be ensured that the benefits outweigh the costs. Therefore, any scale-up efforts must ensure that the energy output increases at a similar proportion, because simply increasing the size of the reactors would not achieve this objective, and would in fact, reduce the power densities [195]. In a scaled-up system, the relative electrode size and spacing between the electrodes must be maintained to avoid mass transfer losses from arising [195,196]. Compartmentalized multiplication, in other words, stacking several smaller individual units (allometric scaling) preserves the diffusion distances near the electrodes, while increasing the overall surface area. In a study by Walter et al., a scaled-up system was designed by vertically stacking 20 individual MFCs (connected in series) inside a 5 L chamber; a high surface area to volume ratio (total anodic surface area was 30,000 cm²) raised the power output whilst maintaining the power density to that of the individual units [197]. However, simply focusing on maximizing the power output may not be suitable; a better target would be to obtain the most economically viable power output. Mehravanfar and coworkers, who conducted an economic analysis on the configuration that maximized the power while keeping the operating costs to a minimum, demonstrated this. Series connection resulted in the lowest ratio of operating costs to maximum power (USD 0.048 /mW.d), whereas for the parallel unit, the costs per unit power was almost two-fold higher [198]. However, series connection of units (where the components within each unit are also placed in series) submerged in a common electrolyte may be subject to ionic short-circuiting, leading to voltage losses [199,200]. A possible solution to this issue is to arrange the individual components of a module in a parallel arrangement while connecting the modules in series as such a setup has not exhibited short-circuit currents [197,200]. Stacked systems also remove COD by considerable percentages; for both vertically and horizontally stacked modules with parallel electrical connections, the range extends from 60 to over 90 %, the value depending directly on the number of units added [64,198,201]. For series electrical connection, similar removal percentages have been reported [198,202]. Modularized systems having treatment capacities over 500 L have been operated successfully over long periods. A 720 L stacked MFC unit containing six modules removed 87 % of the COD from sanitary wastewater at a hydraulic retention time (HRT) of 36 h [203]. Liang et al. operated a 1000 L modularized MFC system for a period of one year, which achieved a

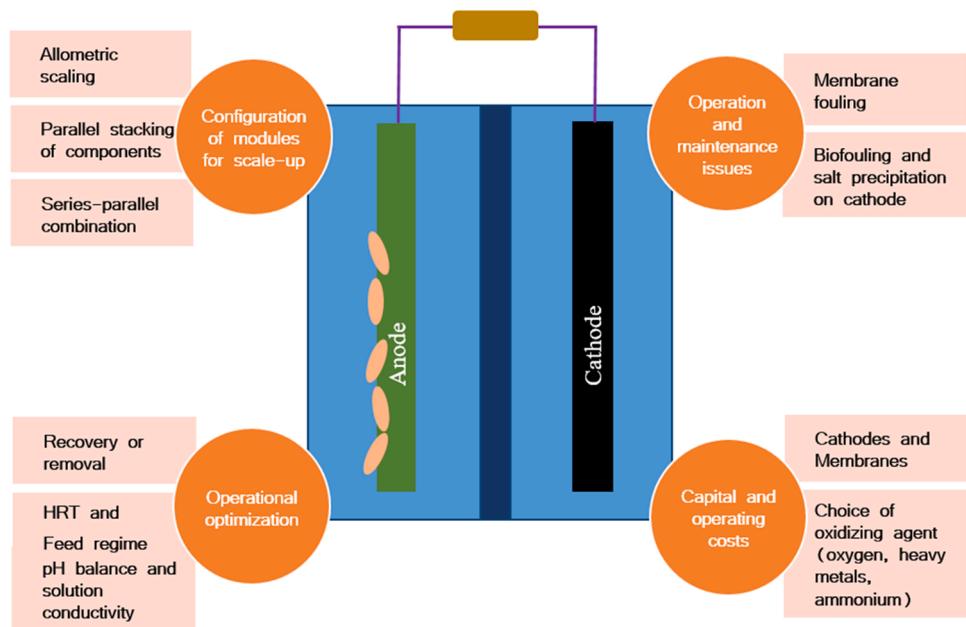


Fig. 5. Major challenges faced in using MFCs for industrial waste management.

COD removal rate of 70–90 % [67].

4.2. Capital costs associated with electrodes

Cell construction, electrode and membrane materials as well as the chemicals consumed during operation contribute to the overall cost of setting up and running an MFC system [69,204]. For long-term operation of a large-scale unit, inexpensive yet durable materials must be developed to a point where these are just as efficient as those used in laboratory-scale systems. Carbon-based bioanodes are biocompatible, durable, have comparatively lower costs and are suitable for a variety of substrates [205,206]. Graphite, despite its excellent performance in the laboratory, has a resistivity whose effect becomes more pronounced in large-scale units, and it is expensive [207]. Carbon cloth and carbon paper are also expensive and lack mechanical strength and durability, making these materials incompatible to large-scale usage. Stainless steel (SS) anodes may be well suited to large-scale units for continuous operation because of the affordability and corrosion resistance, whereas polymer coatings on SS anodes can raise the power output by about two-folds [208,209].

Packed bed designs where granular activated carbon (GAC) functions as the anode could easily be adopted in a treatment plant. In a laboratory-scale system, a membrane-less MFC using GAC as anode exhibited a power density 2.5-times higher than the MFC utilizing carbon cloth [210]. Pilot-scale studies conducted using GAC bed anodes (where graphite rods acted as current collectors) achieved 80 % removal of COD from domestic wastewater when operated at a hydraulic retention time (HRT) of 20 h. This system was estimated to generate a power density of 15 W/m³ [211]. However, it must be ensured that there is adequate electrical conductivity within the beds (requiring a high packing density) and a lack of oxygen access [207,212]. There is however a limitation in the use of GAC beds if waste streams having a significantly high organic load are to be treated and that is fouling, which necessitates replacement [213]. Biochar can be a practical, low-cost alternative to GAC due to its high specific surface area (as high as 774 m²/g has been reported [214]), and recent successes with biochar electrodes in MFC systems have demonstrated its effectiveness [214–217]. Huggins et al. studied the performance of biochar electrodes in MFCs treating industrial wastewater. 95 % of the COD was removed, while nutrient recovery rates were 73 % and 88 % (for NH₄-N and PO₄

respectively). After 20 days of operation, a high amount of nutrient retention (from 0.16 g phosphorus per kg electrode to 1.9 g per kg) was observed on the biochar cathode, which suggested that the spent cathodes could be utilized to amend soils. The use of waste wood as the precursor for the biochar made the process more sustainable [218]. Chakraborty et al. have reviewed extensively the use of biochar in MFC systems [219].

Current collectors in the form of graphite rods, stainless steel or copper are necessary to minimize the flow path of the electrons through the electrodes, to reduce the Ohmic losses. However, copper, despite being an inexpensive and readily available material, is subjected to extensive corrosion that can drastically limit the power output [69,207]. However, in a study conducted over 27 weeks, a low extent of biofouling was observed on copper collectors, which was attributed to copper's antimicrobial activity [220]. A number of losses degrade MFC performance, among which activation losses brought about by the limited rates of the reduction reaction may be the more dominant [221]. For industrial settings, the most cost-effective and sustainable option would be the reduction of oxygen directly from the air [73], but the rate of the reaction is too slow, necessitating the use of catalysts. Rozendal et al. [222] discussed how almost 50 % of the capital costs of an MFC could be associated with the air-cathode materials. There has been significant progress in this domain; however, the challenge is to develop cheaper catalysts or even cathodes that do not require any catalysts.

Activated-carbon cathodes offer high specific surface areas, which has been shown to be effective without catalysts, albeit in laboratory units [223]. Another possible, inexpensive option would be to enhance the activity of the cathodes by combining certain chemicals with a carbon-derived base material. Nitric acid treated carbon powder demonstrated current densities above those achieved by Pt-catalyzed carbon cathodes, and as such, may be an economically attractive option [224]. Low-cost metal-doped catalysts have also performed on par with Pt in pilot-scale studies. Cu-MnO₂ and Co-MnO₂ cathodes respectively reached peak densities of 465 and 500 mW/m² [225]. A bimetallic Cu-Zn catalyst produced a 4-fold improvement in performance of bare carbon cathodes. The power density per unit capital cost was eight times higher than the Pt-based cathode, which justified the use of this cathode in field-scale units, where the COD reduction and the power density far surpassed the units operated using the bare carbon cathodes [226].

Another suitable choice is a biotic cathode, where biofilms

developing on the cathode surface reduce other contaminants, such as heavy metals. Power densities and coulombic efficiencies achieved with biotic cathodes far exceeded traditional carbon-based cathodes (430 W/m³ and 59.6 % compared to 257 W/m³ and 15.6 %) [227]. Wu et al. established a convenient method of obtaining biocathodes, by first enriching the electrode as an anode in an MFC operated using anaerobic digester sludge, and thereafter transferring the electrode to an MFC containing Cr (VI) ions in the catholyte. Higher power densities (9.7 mW/m² against 7.9 mW/m²) and voltages (291 mV against 179 mV) were a direct result of the lower internal resistance of the biocathode. The time to obtain stable current generation for the biocathode that had been acclimatized originally as an anode was 19 days lower than the control MFC operated using a biocathode prepared by the conventional route, which is by the inoculation of the cathode medium [228]. In a similar study by Beretta et al., a polarized biocathode was shown to be populated with Cr(VI)-reducing *Moheibacter*, *Nitrobacter*, *Truepera*, *Flavobacteriales*, *Nitrosomonadales* and *Rhizobiales* families, and achieved almost 90 % removal of Cr(VI) even in the absence of organic carbon. The biocathode had also been acclimatized previously as the anode of an MFC where anaerobic digestate was used as the inoculum [229]. With such acclimatization studies, novel Cr(VI)-reducing strains can be isolated [230]. It has been shown that over 90 % of nitrate ions can also be removed using biotic cathodes [231]. This creates a truly synergistic treatment environment if the biocathodes are used alongside bioanodes that degrade organic contaminants in the anode chamber. However, a better understanding of the biocathodes is necessary to evaluate the adequacy of these cathodes in large-scale systems.

4.3. Cost and performance of membranes

The second largest cost proportion belongs to membranes. In a 200 L MFC system developed by Ge and He, the cost of the cation exchange membrane (CEM) was 60 % of the total capital cost [69]. These membranes are also subject to biofouling, which blocks proton transfer [232]. In addition, CEM tend to selectively transfer other cations (K⁺, Na⁺, Ca²⁺ and Mg²⁺), whose concentrations in the cathode chamber can be 10⁵ times higher than the protons [165,233]. Because of the uneven and slow proton distribution, over time, protons build up in the anode chamber while the pH increases at the cathode due to the oxygen reduction reaction. Sharp pH gradients are thus established across the membrane, leading to voltage losses that can reach significantly high levels in substrates containing a strong concentration of cations (such as wastewaters) [234,235]. Alternative membranes such as anion exchange or ultrafiltration membranes may remedy the problem; however, their costs and fouling potential deter their use in large-scale treatment units. Membrane-less designs therefore seem to be the most cost-effective option, and several pilot-scale units have been effectively operated without a membrane, achieving COD removal rates ranging from 41 to 80 % [77,211,236]. Unfortunately, the energy output from these MFCs dropped as the operation was extended over long periods, primarily due to the precipitation of calcium and magnesium salts on the cathode [77,211].

Membranes and separators also act as a reasonable barrier to the diffusion of oxygen towards the anode that can prevent aerobic degradation of the substrate. If oxygen was allowed to infiltrate the anodic chamber, the Coulombic efficiency of the system would be reduced, which is a major drawback if power generation is the desired output and not the COD removal [236]. Membraneless designs also suffer from an inherent limitation, as the catholyte and anolyte fluids can mix, leading to reactant crossover. This allows electrons to transfer directly between the electrodes, shorting the fuel cell [237]. Studies have shown how reducing the distance between the anode and cathode can lower the internal resistance of an MFC, and improve the power output [238]. In addition, to reduce the space requirement in stacked systems, the MFC units would have to be packed into a chamber where the components would be in close contact [197]. Because of the risk of short-circuiting in

membraneless designs, spacing between the electrodes can only be decreased to a limited extent. With the addition of the membrane or separator, the anode and cathode can be placed relatively close to improve the efficiency of ion transfer between them that would minimize the internal resistance of the cell and prevent internal short-circuiting. Therefore, instead of developing a membrane-less configuration, a more effective approach would be to install inexpensive membranes or separators. Porous ceramic membranes have emerged as viable alternatives [239–241]. A feasibility study determined that ceramic membranes made from red soil and modified with cation exchangers and Nafion solution achieved a power density of 84 mW/m³ [242–244]. Walter and coworkers developed a 5 L stacked-module MFC system consisting of ceramic membranes, which validated the use of these membranes in large-scale units [197]. Low-cost, non-woven cloth separators have demonstrated considerable success in long-term operational studies spanning over more than two months [243,244]. Biochar has also been investigated as another low-cost alternative [245]; Chakraborty et al. determined the power generated from an MFC utilizing a biochar membrane was 26 times higher per unit cost of the membrane than that of the MFC operated using the more common and expensive Nafion membrane [246].

The choice of membranes depends largely on the pollutant being treated and the objective of the operation. Fouling remains a limiting factor, therefore if power output is the major goal, then there is a need to suppress the onset of fouling to reasonable limits. Pretreatment of the waste stream to reduce the concentrations of hardness ions, or controlling the biofilm thickness using ultrasonic treatment may be suitable strategies [247]. Anti-fouling membranes have also been developed, but their use will be determined by their cost [248]. If the goal of the operation is pollutant removal, membrane-less designs are more preferable, because the loss of organic pollutants to aerobic oxidation at the cathode chamber would be a desirable outcome. In fact, this is why MFCs designed for the remediation of polluted soil and sediment do not tend to have separators, even in pilot-scale units [37]. An integrated system combining an MFC with an electric membrane bioreactor (cathode chamber) consisted of a sand chamber to replace the PEM, and removed 97 % of the COD, >93 % of the ammonia nitrogen and 50 % of the phosphorus [249]. The design used an air-contact oxidation bed and an embedded trickling filter to increase the DO levels in the cathodic chamber to reduce the cost of aeration. This can be an attractive alternative for existing treatment plants.

4.4. Other operational issues

There are other operational concerns that need to be addressed if MFCs are to be integrated with existing waste treatment units. Continuous operation of MFCs is a matter of optimization where the change in the hydraulic retention time (HRT) involves tradeoffs. Increase in HRT raises treatment efficiency, but has negative consequences for the power output due to substrate depletion, which can induce a voltage reversal if the levels are too low. Unit size also needs to be increased that adds to the costs. HRT therefore is a critical parameter, whose optimization is governed by the preferred objective of the MFC. Power generation and pollutant treatment can often be competing objectives; in fact, the configuration of stacks in a large-scale unit that yielded the maximum power density achieved the least COD removal [198]. Lu et al. operated a 20 L MFC that achieved almost 95 % COD removal from brewery wastewater, but the power production dropped by three orders of magnitude when the HRT was raised [68]. It is therefore imperative that either power production or the treatment efficiency be targeted, and strategies that improve the results of the desired outcome be implemented.

The properties of the organic waste (in the form of wastewater or solid waste), namely the composition, conductivity and pH, will have direct impact on the microbiology and output of an MFC. When MFC modules are stacked, increasing salt concentrations improved

conductivity, which raised the maximum power density and enhanced the COD removal by an average of 20 % [198]. However, the parameter needs to be optimized, because excessive concentrations of cations can lead to fouling [250] due to the formation of sodium carbonate on the air-face of the cathodes and insoluble calcium carbonate on the interior face [211]. High salt concentrations can also be detrimental to the microbial consortium [251]. Maintaining a neutral pH in the system by the use of buffers is also critical, as the neutral pH level is not only conducive to the growth of microorganisms, but the buffer also prevents a pH gradient from developing across the electrodes. A sharp pH gradient can lead to voltage losses because it causes cell potential to drop. Low pH also limits the activity of the anodic biofilm [252]. Buffers can also enhance solution conductivity. Rossi and coworkers recorded the highest power density in an air-cathode MFC (7.1 W/m^2), obtained upon increasing the concentration of the neutral pH buffer to 100 mM [253]. In wastewater treatment processes, pH adjustment is a common stage; therefore, buffer addition could be achieved up to a certain economically feasible limit. In fact, if the MFC is operated using a substrate containing ammonium ions, it may not even be necessary to add chemicals for pH adjustment, as these ions combine with hydroxide ions at the cathode. This lowers the pH, preventing the onset of a pH imbalance across the two chambers [170,254].

The specific type of pollutants removed and the performance of the MFC is governed by the microbial population, whose demographics can be dramatically changed by the removal mechanism (aerobic or anaerobic oxidation) and the anode potential [255,256]. The community can evolve dynamically even during operation of the MFC, as the operating parameters are changed. Quan et al. determined that switching from aerobic to anaerobic operation reduced the power output and the dominant species in the anodic biofilm. The sparging of air into the anode chamber selectively influenced the growth of aerobic bacteria [255]. Decrease in the anode potential was observed to enhance the growth of the anodic biofilm that could then oxidize more substrates [257,258]. The internal resistance of the anode decreased, causing the power output to rise [258]. Analysis of the pollutant degradation rate revealed that at higher anode potentials, degradation percentages were lower [257]. However, the influence of anodic potential on degradation rate and power output is not a strictly inverse relationship, and studies revealed the existence of an optimum anode potential, where pollutant removal and power output were the highest. Fan et al. analyzed that among 350 mV, 200 mV and -200 mV, the internal resistance of the anode was the lowest when its potential was set to 200 mV [258]. Similarly, the highest quantity of volatile fatty acids was degraded when the anode potential was 550 mV, compared to removal rates at 300 and 800 mV [257].

Fouling of cathodes by salt precipitation can degrade its performance significantly, and along with biofouling, can lead to an average of 20–40 % decrease in the power density over long-term operation [74,250]. Although the biofilm layer on the cathode can be removed by simply scraping it off, in a large-scale unit consisting of several stacked modules, the design must allow easy disassembly of individual modules for cleaning. In this case, parallel arrangement of the components inside a module may be a suitable option. Salt precipitates may need chemical treatment, because simple mechanical cleaning was shown to restore only 22 % of the power output, while nutrient removal did not revert to the original rate [77]. Moreover, for large-scale units, chemical cleaning is a relatively expensive strategy. Increasing the buffer capacity of the wastewater or reversing the pH gradient by the periodic addition of acids and bases has shown promising results in limiting salt precipitation at the cathode [222,234]. Continuous monitoring of the cathode and the solution pH are therefore useful in mitigating fouling. In case of soil and sediment MFCs, the high concentration of salts and humic acids would be particularly detrimental to performance [259]. Periodic replacement of the electrodes may be the only option for these MFCs.

Industrial effluent treatment plants can derive greater benefits from using MFCs because the waste streams are highly concentrated in

contaminants and COD, which raises their energy recovery potential. In addition, the semi-continuous operation of these treatment plants is advantageous to the operation of MFCs, as pulse feed regimes at high flow rates can be maintained, which can replenish the units with fresh feed by continuously maintaining a high substrate concentration [197, 253]. This eliminates the problem of substrate depletion and maintains homogeneous distribution of nutrients, thus preventing voltage reversal in the fuel cells. A pulse-feed strategy also avoids the need to have larger reactors [260]. Energy self-sufficient designs can be adopted to reduce the energy needs of the system. Dong et al. developed a 90 L pilot-scale MFC system containing 5 modules to treat brewery wastewater. Studies conducted over a period of six months revealed that the MFC unit generated enough power ($0.056\text{--}0.097 \text{ kW h/m}^3$) to run the influent pumping system, where 88 % of the COD and 86 % of the TSS were removed. The VSS removal rate was higher than values obtained in upflow anaerobic sludge blanket reactors (90 % compared to 81 %), thereby reducing the sludge load [261]. More units need to be stacked for an energy self-sufficient system if the treatment capacities are raised [262]. In another study, an auto-circulating continuous unit (treatment capacity of 8.4 L/day) consisting of an activated carbon bed as anode was designed to operate without requiring any external agitation for liquid mixing. No energy input was necessary to operate the system, whose maximum energy yield was sufficient to generate around 60 W.h annually, while exhibiting a COD removal efficiency of 90.4 % and generating 83 % less VSS than aerated systems. Most notable however was the life cycle analysis of the system, which showed that long-term operation of the system was feasible because the treatment process has low to moderate environmental impact [263].

4.5. Considerations in process selection and optimization

It is certain from the aforementioned discussion that to use this technology in field-scale treatment facilities, extensive optimization is necessary. As with any optimization strategy, there is a need to set the target objective. In case of MFCs, the choice is between energy and contaminant removal/resource recovery. Only those operational and architectural issues that are relevant to the target need to be addressed, because certain parameters, when altered, may improve power generation but can negatively affect contaminant removal. However, in both cases, cost-effectiveness of the operation must be assessed at every stage of optimization. It can be gleaned from Table 1 that even though power output varies across a wide range and depends on a number of factors (electrode materials, substrate type, configuration, etc.), the contaminant removal efficiencies are consistent. MFCs therefore behave as robust effluent removal systems, and in fact, may be more suitable to contaminant removal and product recovery [264,265]. The operating parameters also depend on the product to be recovered. Both heavy metal ions and nitrates are reduced at the cathode, and a competing effect may arise in systems containing both. Increasing the concentration of lead for example was found to lower the rate of denitrification at the cathode [266]. Although membraneless designs have performed well in MFCs where heavy metal ions were being reduced, use of cation exchange membranes was shown to be beneficial in nutrient recovery. Nutrient removal and recovery was found to be over 95 % in DCMFCs operated using CEM (98 % of NH_4^+ -N and 94.9 % of PO_4^{3-} -P), significantly higher than those using forward osmosis (98.81 % of NH_4^+ -N and 83.18 % of PO_4^{3-} -P) and non-woven membranes (97.2 % of NH_4^+ -N and 90.6 % of PO_4^{3-} -P) [267]. The high pH gradient established by the CEM facilitated the nitrogen recovery, while the increased permeability of Mg^{2+} ions through the membrane might be the reason behind the improved phosphate recovery in the study, since precipitation of phosphate as struvite depends on the concentration of the Mg^{2+} ions in the system [268]. In a study conducted using sediment MFCs, Sajana et al. determined that increase in distance between the electrodes enhanced the removal of organic matter but reduced the power density [238]. These are just a few examples that highlight the importance of setting

the target of the MFC operation. Although primarily the energy and resource recovery potential is what makes MFCs more attractive than other technologies, there is a need to conduct life cycle analyses for this system at reasonably large scales to determine its superiority [8,263]. Additionally, it is necessary to develop robust process monitoring and control systems to ensure stable operation of MFCs when these are used as treatment units. MFCs can be monitored in real-time as the electric current and electrode potential can serve as useful and effective monitoring parameters. Models capturing the complex relationship between the microbiology and the electrochemistry of the MFC would certainly aid in understanding and controlling the system better [8,269]. Research in this direction would undoubtedly advance the current state-of-the-art towards practical implementation in order to achieve effective pollution management.

Despite these challenges, studies with MFCs in practical scales have shown promise. An MFC stack system was designed for secondary treatment of the effluent from a septic tank, employed to treat grey water from a real household. The two-stack system consisting of 9 units per stack had a total capacity of 700 L, and used a granular carbon bed as the anode. Interestingly, the circuit design incorporated capacitors to store the energy from the MFCs. Operation lasted continuously for 60 days, and the HRT was around 3.3 days for a daily treatment capacity of 600 L. Removal efficiencies reached above 80 % (for COD, TSS and BOD) with the addition of the MFC system, whereas with the septic tank, only less than 5% of the TSS, BOD and COD had been removed. Total nitrogen and phosphorus were removed by 30 % and 64 % respectively [270]. It is to be noted however, that domestic wastewater tends to be homogeneous in its contents, unlike industrial effluent streams. Whether such an integrated unit would apply well in industrial settings is a research area gaining increasing importance. In a study by Hiegemann et al., a 45 L MFC unit, where four single chamber membrane-less modules had been stacked, was operated using the effluent from a primary clarifier for nine months. Although the analysis revealed COD, TSS and TN removal was moderate, the net energy benefit was positive: 0.36 kW h was received for every kg of COD removed. The savings were in the operation of the sludge digester and dewatering system as the sludge production had been significantly reduced [271]. Field-scale experiments at even larger scales have been attempted with success. By stacking six MFC units in parallel (each with a working capacity of 120 L), a system for treating sanitary wastewater was set up to operate for 255 days. Low-cost ceramic membranes and cathode catalysts ($\text{Co}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ and $\text{Sn}_5\text{Cu}_{84}$) were used. Treatment efficiencies were over 80 %, increasing with the HRT. However, the power output was seen to fluctuate with the dynamic changes in the loading rate, which is a very likely scenario in industrial ETPs. A possible remedy is to utilize a power management circuit involving capacitors that can store the electricity generated and discharge it in order to power devices when required. The researchers employed 12 supercapacitors of 500 F (F) capacity, which were charged continuously by the MFC system during the day and the energy was used to operate 6 LED bulbs (2 W in total) for a total of 60–90 min at night [203]. Addition of power management circuits (PMS) is an efficient strategy to recover the energy of MFCs, because these may not provide energy in an uninterrupted and stable manner, especially when operated in ETPs where effluent loading is subject to dynamic variations throughout the day. In fact, to boost the output from MFCs, a PMS is invaluable, and studies have shown the voltage output can increase by over 70 % [272]. PMS can also protect against voltage reversal in MFCs modules stacked in parallel [273,274].

The application of MFCs for bioremediation, however, has not seen comparable progress. The main consideration here is the distribution and arrangement of the MFC modules in sufficient quantities for maximum coverage over the area contaminated. Pilot-scale studies for the remediation of contaminated soils introduced the concept of the radius of influence (ROI) for an MFC module, which is defined as the distance from the surface of the anode to the point in the soil/sediment where the contaminant concentrations were the lowest. The parameter

was found to depend on the properties of the contaminated soil (water content, permeability and porosity) and the pollutants, the MFC configuration and the electrode materials. Column-style configurations, where a hollow perforated tube was placed adjacent to the cathode in a horizontally stacked MFC module was determined to have the highest ROI. Such a configuration removed more than 80 % of petroleum hydrocarbons in a 50 L pilot-scale MFC unit. The current density reached a maximum of 70 mA/m^2 , sufficient to power small sensors. The ROI extended up to 3 m from the module, which was promising [275]. The manner in which individual MFCs can be distributed across contaminated soils therefore depends on the ROI, and *in-situ* remediation is more enhanced when ROI is higher. Thus, large-scale remediation studies should look towards optimization of the ROI.

Although field-scale studies are still limited, the practical strategies and limitations addressed here could serve as useful guidelines to design such studies. In addition, the success of integrated MFC units could spark the interest of researchers and practitioners to test MFC systems in existing treatment plants. MFCs obviously cannot completely replace conventional units, but as part of an integrated network, they can be used to derive net positive benefits from wastewater treatment, which current treatment strategies cannot provide.

5. Conclusion

Environmental bioremediation is an expansive domain of MFC applications. In the last decade, significant strides have been made to improve this technology for waste treatment purposes. Currently, inexpensive electrode materials are being developed and designs without expensive membranes are being evaluated, with soil and sediment based MFCs, having eliminated the requirement of membranes altogether. One of the main hurdles in the application of this technology in industrial settings was that of scaling up of the MFCs. This issue has been addressed in many recent studies, by stacking several smaller modules in series or in parallel to raise the output power and increase the treatment capacity. Unlike conventional treatment options, MFCs require no energy input and sludge production is also reduced. Therefore, bioremediation by MFCs is more cost effective than conventional solutions, has minimal environmental footprint and produces benign end-of-chain products. Recent MFC technologies, combined with reactor design and factor optimization, presents new avenues for exploration. Advancement in this field with respect to energy and resource recovery along with waste treatment could make industrial utilization a profitable venture. A sustainable waste management system does not stop with treatment, but it needs to valorize the chemical waste. This is why industries struggling with proper handling of their effluents can benefit from the integration of MFC technology to their treatment plants. MFCs effectively create a closed-loop system, thus complementing the efforts to create a sustainable future. Knowledge of the progress of this technology is imperative for industrial stakeholders striving to find innovative and sustainable waste management solutions for the coming decades.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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