



## Microbial fuel cell applications for removal of petroleum hydrocarbon pollutants: A review

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### ABSTRACT

Bioelectrochemical systems (BESs) are considered as the potential approaches to remediate the environments contaminated by hydrocarbons. This review addresses the application of BESs particularly microbial fuel cells (MFCs) in degradation of petroleum hydrocarbons, including BTEXs, from soil, water, wastewater and sediments. Details on reactor design and critical issues are discussed. Aspects on electrodes, redox mediators and membranes are evaluated, including economic feasibility. The microbial community is considered in detail. It can be concluded, that comparing to classic configurations, single-chamber air-cathode reactors are more cost-effective. Secondly, systems based on small-scale units are recommended for future developments.

### 1. Introduction

Petroleum hydrocarbons (PHs) is the name given to a wide range of chemical compounds, found in crude oil, diesel and gasoline as their primary constituents. PHs can be divided into 4 groups: (i) aliphatics such as alkanes, alkenes and alkynes; (ii) cycloaliphatics such as cycloalkanes; (iii) aromatics such as monoaromatics and polycyclic aromatic hydrocarbons (PAHs); and (iv) other components such as asphaltenes, waxes, tar and resins as shown in Table 1 [1]. PHs are identified as carcinogenic compounds [2]. The physico-chemical properties of these recalcitrant compounds make them extremely portable in the environment, ubiquitous in air, soil, and water bodies. Due to their certain characteristics such as lipophilicity and electrochemical stability, PHs tend to bioaccumulate in organic tissues, causing harmful and chronic effects [3,4]. There are many sources of these hardly biodegradable substances in the environment. The major one is associated with spills and emissions, including releases of petroleum products such as diesel fuel, gasoline, and lubricants, among others [5]. Furthermore, anthropogenic practices such as municipal and industrial runoffs, coal mining, transportation, storage, and onshore and offshore petroleum industry operations induce PHs pollution [6]. The US

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**Table 1**

Classes and characteristics of petroleum hydrocarbons with proper examples (Reprinted with permission from (Ossai et al., 2019). Copyright © 2019 Elsevier [1]).

Hydrocarbons	Characteristics	Examples
<b>Aliphatics</b>	<ul style="list-style-type: none"> <li>Less dense than Water</li> <li>Molecule size is inversely proportional to its volatility and water solubility</li> </ul>	Methane, propane, butane (gases at room temperature); hexane, octane hexadecane (liquids at room temperature); eicosane, triacontane, pentacotane (solids at room temperature)
<ul style="list-style-type: none"> <li>Alkanes</li> <li>Alkenes</li> <li>Alkynes</li> </ul>		
<b>Cycloaliphatics</b>	<ul style="list-style-type: none"> <li>Contains up to six carbon atoms in a ring</li> <li>Fairly resistant to microbial degradation</li> </ul>	Cyclohexane, methyl cyclohexane, methylcyclopentane, 1,2-dimethylcyclopentane
<ul style="list-style-type: none"> <li>Cycloalkanes</li> </ul>		
<b>Aromatics</b>	<ul style="list-style-type: none"> <li>Very volatile and relatively water-soluble</li> <li>Have benzene ring</li> <li>Some are resistant to microbial degradation</li> </ul>	Benzene, ethylbenzene, naphthalene, toluene, xylene and phenanthrene
<ul style="list-style-type: none"> <li>Monoaromatics</li> <li>BTEX</li> <li>PAHs</li> </ul>		
<b>Other Components</b>	<ul style="list-style-type: none"> <li>soluble in aromatics and non-soluble in light alkanes</li> <li>Contains about 18–65 carbon atoms</li> </ul>	Phenols, fatty acids, ketones, esters, porphyrins, pyridines, quinolones, cadaxoles, sulphonates
<ul style="list-style-type: none"> <li>Asphalthenes</li> <li>Waxes and tar</li> <li>Resins</li> </ul>		

environmental protection agency (USEPA) has categorized these PHs as priority pollutants of natural resources [7]. Therefore, any contamination from PHs turns into a burning issue, both for direct (such as inhalation of their vapors) and indirect (such as the presence of these compounds in water bodies) contact [5].

Till now, different methods have been applied for hydrocarbon remediation, such as biodegradation, membrane processes, electrocoagulation, adsorption and advanced oxidation processes (AOPs) [8–12]. However, each of these methods suffers from some disadvantages. For example, biodegradation is a significantly slow process that requires long reaction times, even lasting for days. Thus, exceedingly large treatment facilities are needed. Membranes encounter fouling problems in full-scale operations and high energy requirements. Adsorption on activated carbon, a phase change technique, is complex and expensive since the saturated activated carbon demands interval regeneration, and sometimes the method suffers from the interference of other pollutants. Similarly, AOPs are costly and complicated to install and operate. They are predominantly adequate for low-pollution medium. Electrocoagulation produces large quantities of sludge, insecure and unrecoverable. Operating costs and high capital on industrial level scaling up have made electrocoagulation a non-appropriate approach [8,13].

In the past decade, bioelectrochemical systems (BESs) have shown promising niches compared with the aforementioned techniques. The BES refers to employing microbial electrochemical technologies (MET) in which microorganisms, in the form of catalysts, decompose organic compounds and generate electrons. The electrons can produce electricity and other value-added compounds [14]. Several applications such as microbial fuel cell (MFC), microbial electrolysis cell (MEC), microbial desalination cell (MDC) and microbial electrosynthesis cell (MES) come under the BESs. Unlike many conventional methods, BES has rather mild and simple operating conditions. It has successfully dealt with a wide variety of stubborn pollutants ranging from highly contaminated wastewater to polluted soil and sediment. Moreover, precious and exhaustible electrodes, as well as expensive chemicals, are rarely used in BES, thereby proposing an inexpensive and reliable method without any negative impact on the environment [15]. BES has also the advantage of integration feasibility with other technologies, for example, BES can be coupled with AOPs, constructed wetlands, membranes, adsorption, electrochemical cells, etc. [16–20]. In addition, the electrochemical setup of BES superbly controls the released electrons from microbial metabolism [21].

MFC is a system that sustainably produces energy from biodegradable compounds and simultaneously treats contaminated media such as soil, sediments, or wastewater. Generally, it consists of two chambers, an anodic and a cathodic, and an ion-exchange membrane (IEM) separating the chambers. On the surface of anode, the substrate (organic matter or biomass) is being consumed and oxidized by microorganisms thereby producing electrons and protons. Electrons, via an external electrical circuit, and protons, through the membrane, are transferred from the anodic compartment to the cathodic one, where the reduction of an oxidant, usually oxygen, takes place. Chemical reactions with the aid of protons and electrons occur on the cathode surface and finally form stable reduction products. In a system, organics follow equations (1)–(3) [22]:

At anode:



At cathode:



In a BES, microbial or biotic degradation is the main mechanism to remediate a contaminated environment. Anode functions as an electron acceptor and promotes oxidation reactions. Complex organic compounds such as PAHs are initially broken down into smaller molecules via degrading bacteria (e.g., hydrocarbon degradation bacteria (HDB)) and then the intermediates are degraded further via exoelectrogenic bacteria (EB) (e.g., *Proteobacteria*) at the surface of anode. Generated electrons are subsequently released to the anode directly or through mediators [23]. The presence of EB not only hampers the accumulation of mid-products but also progresses the efficiency of extracellular electron transfer to complete the biodegradation process. In this framework, Wang et al. [24] discerned an increased number of HDB by nearly two orders of magnitude ( $373 \pm 56 \times 10^3$  colony forming unit per gram of soil (CFU/g-soil)) close to the anode in the soil MFC than that in the open-circuit control ( $8 \pm 2 \times 10^3$  CFU/g-soil), indicating strong evidence for the bio-stimulation of HDB and synergy between HDB and EB to degrade petroleum pollutants. Moreover, according to Kumar et al. [25], degradation of volatile organic compounds such as benzene, ethylbenzene, toluene and xylene in the MFC occurs through three steps: (i) mass transfer of compounds from gaseous phase to liquid phase, (ii) compounds diffusion into the biofilm, and (iii) their degradation by microbes present in the biofilm. In fact, the primary difference between this system and conventional biotechnology comes from dissimilar exploitation of electron acceptors, viz. Electrode and oxygen, respectively. In MFC, the attached biofilm on the electrode enables fast and *in-situ* electron transfer from the microorganisms to the electrode. As a result, it facilitates biodegradation and harnesses the existing chemical energy of the organic load [26]. Fig. 1 represents the structural design, working principle and mechanism of a microbial fuel cell.

It should be noted that MFCs are of interest since they do not require an external power supply while treating organic wastes [15, 27]. MFCs release 50–90% less sludge than that in other bioprocesses; hence, they reduce the ultimate cost of sludge disposal. Furthermore, they benefit from simple structure, operational stability, little impact on the microbes, and negligible impact on the environment. In addition, microorganisms in MFCs are rather resistant to toxicity and pH inconsistency. Compared with other methods, MFCs have become an efficient and eco-friendly candidate for removing PHs [28].

Several excellent review articles have been published about the application of BESs [29–33], yet, to the best of our knowledge, there is a limited discussion to specially refer to the MFCs applied for PHs treatment—a field of great potential and value. In this perspective, this review attempts to compile contemporary information of BES application for degradation of PHs and is dedicated to the most practical type of BES— the MFCs. It includes: (i) design of MFCs; (ii) electrodes; (iii) redox mediators; (iv) membranes; (v) microbial community; (vi) co-substrate; (vii) energy consumption and electricity generation; (viii) optimal process parameters; (ix) MFC applications for the PHs removal; (x) cost analysis; and (xi) future outlook and conclusion. Understanding these issues is of critical importance to stimulate more thinking and exploration regarding future promotion in practical applications.

## 2. Design of microbial fuel cells (MFCs)

Numerous MFC reactor configurations have been developed to degrade hydrocarbons and acquire an acceptable quality of the effluent from a cost-effective situation [34]. The most common configuration for degradation of hydrocarbons is a two-compartment MFC which includes H-type (Fig. 2A), cubic (Fig. 2B), tubular (Fig. 2C), or U-type MFC (Fig. 2D). Generally, these systems consist of a cathodic and an anodic chamber separated by a membrane [35,36]. Researchers have studied these systems at laboratory scale to treat polluted hydrocarbon media [24,37,38]. However, the complicated structure and extra cost of these reactors hinder the system from

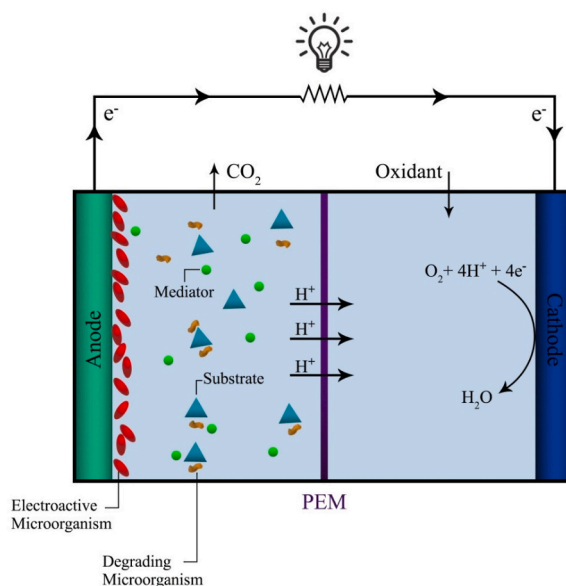
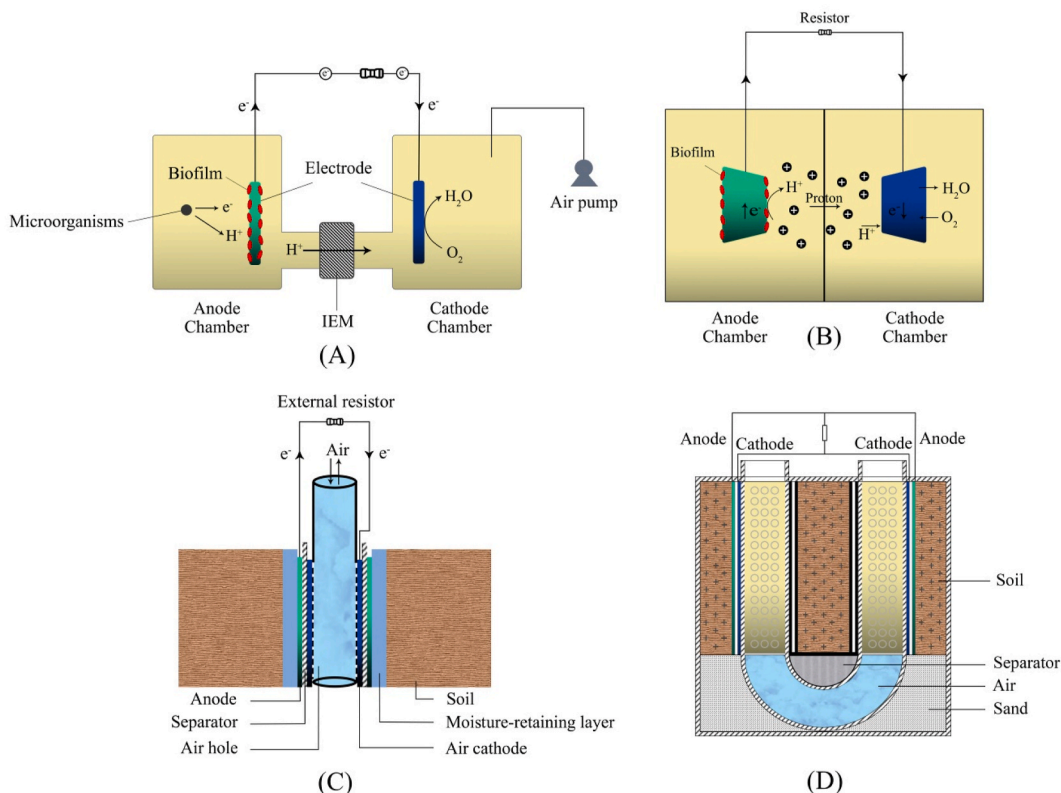


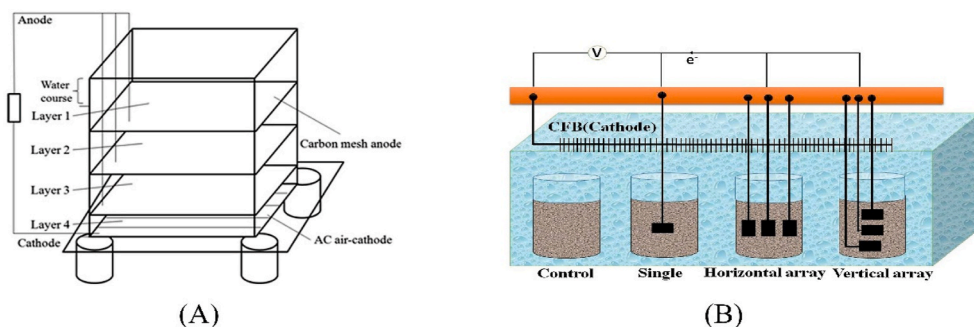
Fig. 1. Schematic design and working principle of a microbial fuel cell [15].



**Fig. 2.** Schematics of dual-chamber MFCs. (A) H-type MFC [28]. (B) Cubic MFC [45]. (C) Tubular MFC (Reprinted with permission from (Wang et al. [37], Copyright © 2020 Elsevier). (D) U-tube MFC (Reprinted with permission from (Wang et al. [24]), Copyright © 1999–2021 John Wiley & Sons).

being scaled up and used in real case scenarios. With the purpose of developing BES/MFC sustainably, several column type single-chamber air-cathode reactors were constructed to remove hydrocarbons from the soil, wastewater, marine sediments, as well as anaerobic sludge stemming from petroleum wastewater [39]. In soil and sediment function, the anode was embedded in the soil or sediment, while the cathode was always suspended in the overlying aqueous part [40,41].

To improve the efficiency of the MFCs in terms of their PHs removal capabilities, a multianode system has been developed [42]. Li et al. [42] have designed a soil/sediment MFC (SMFC) with three parallel anode layers and activated carbon (AC) air-cathode at the bottom. It was observed that the multianode MFC has higher efficiency of PHs removal than a single-anode system. The larger the activated surface area of this system, the easier the transmission of  $H^+$  and  $e^-$  in the process. Liu et al. [43] deployed multiple compacted carbon cloth roll anodes in the ocean water and sediment (Fig. 3A). This system, with three electron collectors, maintained a favorable performance of the single anode but also promoted the effective surface area per sediment footprint and preserved high power density. In an associated study, Prakash et al. [44] constructed three discrete reactors in different configurations by installing



**Fig. 3.** Configuration of MFCs for the application of multilayer anode: (A) A multi-anode system (Reprinted with permission from (Li et al. [42]), Copyright © 2014 RSC Publishing); (B) Vertical- and horizontal-anodes systems (Reprinted with permission from (Prakash et al. [44]), Copyright © 2018 Elsevier).

anode in single, horizontal, and vertical arrangements (Fig. 3B). They observed that the multi-vertical electrode system has higher performance and lower internal resistance than the horizontal one.

The single-chamber systems take advantage of the direct transfer of oxygen from the air to the cathode and thus do not impair the anaerobic environment of the process. In the anaerobic trend, anode microbial communities are not affected by the reductive effect of oxygen, thereby enhancing the microbial operation. However, in a sleeve-type single column reactor (Fig. 4A), where cathodic biofilms act as a separator, the biofilm overgrowth at the cathode would likely diminish the electron transfer and inhibit the degradation process. Furthermore, hydrocarbon compounds, which are required to be reduced by the cathode, are simultaneously influenced by the anode that induces disruption in the system performance. Hollow-type single chambers (Fig. 4B) solved this issue and provided an oxygen source for the cathode at the same time. Hence, it would be an acceptable option for an efficient degradation [46].

Although the lab-scale MFCs successfully treated PHs from polluted media, the produced current in comparison to the chemical energy contained in the wastewater is limited. Hence, the Coulombic efficiency obtained through laboratory MFC is poor. Applying the approach of using a plurality of units, on the other hand, has demonstrated optimal power output. A number of significant issues such as the MFC size, optimized anode surface, the hydraulic retention time (HRT), cascade length, and series or parallel connections should be considered to attain the highest economically feasible power output [48].

In general, desirable surface area to volume (SA:V) ratios in small units maintain diffusion distances near the electrodes and facilitate the achievement of electrical power from organic substances. Small-scale MFC also has more capability for wastewater treatment due to the fact that higher power brings about faster metabolic rates and organic transformation. For a determined anode surface area and length, the power density increases in accordance with the higher (SA:V) ratio. In a comparison of cascade MFCs in a steady diameter tube, Walter et al. [49] revealed that increasing lengths (from 2-fold to 8-fold) result in equal power density and raise absolute power.

In the stacking arrangement of several small reactors (allometric scaling), the outlet of one unit is the following unit's inlet. Power generation is expected to decline more quickly at final units in terms of cascade length or number of MFCs inside a cascade. The feedstock units that have already been treated provide less power output. In this regard, researchers demonstrated that providing a uniform substrate utilization along with the cells can address this issue [48,50]. The uniformity conditions in the reactor are controlled by supplying external mixing, preserving equal flow profile, and increasing the flow rate that leads to the decrease of HRT [51].

Based on the desired purpose, MFCs can be connected either in series or parallel to increase the total voltage or current, respectively. As predicted, setting the MFCs connected electrically demonstrated lower current and greater operating voltage. Hence, while cascade structure makes it possible to set a number of extended MFCs in parallel or series to raise current or voltage, it hardly has an effect on power density [52]. Liu et al. [53] evaluated the performance of benzene degradation from groundwater using vertically three air cathodes in both series-connection and parallel-connection tubular stack MFC. The system reduced benzene degradation time by half compared to a single tubular cell with the same anode capacity. While series connection revealed the highest open-circuit voltage and had showed efficient pollutant degradation, parallel mode demonstrated increased power density and thus consumed less energy [53].

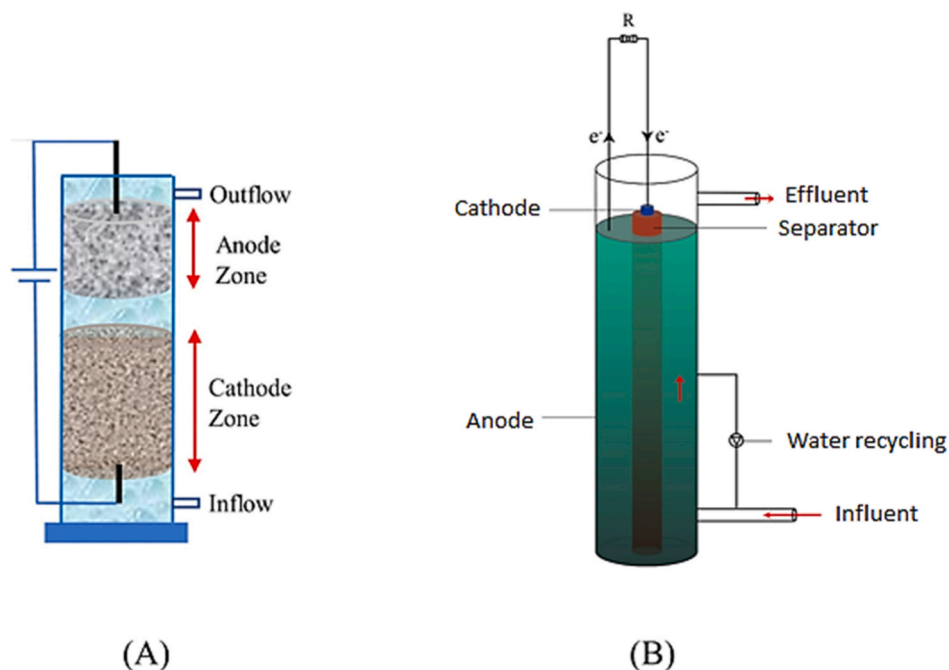


Fig. 4. Schematics of single-chamber MFC. (A) Sleeve-type (Reprinted with permission from (Yang et al., 2020). Copyright © 2020 Elsevier [34]). (B) Hollow type [47].



### 3. Electrodes for hydrocarbon degradation

The performance of MFCs is strongly influenced by the selection of anode and cathode materials in addition to the effectiveness of the catalyst. An appropriate electrode material generally possess: (i) high electrical conductivity to accelerate the electron transfer, and (ii) a large surface area to enhance the sorption effect. As the effective surface area of the biological electrode increases, the mass transfer capacity of the system and thus the microbial activity increase [54]. Furthermore, electrode materials have an impact on the ultimate products obtained during the degradation process. Hence, the selection of environmentally friendly electrodes makes them more useful for large-scale applications.

Properties of the anode affect not only the activity rate of microbial adhesion but also the rate of electron transmission in the cell. Among various anode materials that have been proposed for constructing efficient MFCs, carbonaceous materials are superb due to their beneficial properties such as good biocompatibility, chemical stability, and conductivity. Carbon-based materials consist of carbon cloth (CC), carbon felt (CF), carbon paper (CP), carbon brush (CB), granule graphite (GG), graphite plate, graphite sheet, and graphite rod. The deployment of carbon materials as electrodes has revealed a satisfactory degradation of total petroleum hydrocarbons (TPHs) from contaminated sites [36,55,56]. Venkidusamy et al. [57] demonstrated that the carbon anode/cathode presents 83.4% elimination of diesel with power generation of 90.81 mW/m<sup>2</sup> in a single cell MFC. The results of Adelaja et al. [58] also accounted for more than 90% of PHs degradation for both *in situ* and *ex situ* MFC utilization with CF electrodes. The obtained results confirmed that this system is an advantageous over other complicated and expensive technologies even in anoxic environments under harsh conditions.

Regarding graphite material for the anode, graphite felt (GF) obtained more outstanding performance than other types. This outstanding performance of GF anode was attributed to its larger surface area and greater adsorption potential. Yu et al. [59] constructed soil/sediment MFC (SMFC) in a plexiglass reactor equipped with alternative electrodes made of five different materials: GF, CC, activated carbon fiber felt (ACFF), graphite paper, and aluminum sheet but carbon veil was not tested. The authors revealed that GF anode provided the best result of 59.14% TPHs removal in petroleum polluted soil after 115 days of operation [59]. This relatively higher removal TPHs by GF anode was attributed to the larger surface area of GF, which resulted in greater attachment of organics on the surface of anode, greater mass transfer of substrate, and less internal resistance. The internal resistance owing to the cell components causes interference to the electron and proton transfer [60]. Moreover, metals have been used to overcome the electron transfer barrier. Yan et al. [41] constructed bioreactors in which anode was made up of two stainless steel cylinders (80 meshes with 1 mm thickness) and cathode was prepared from a stainless steel cylinder (4 cm height and 9.6 cm diameter) and obtained more than 90% degradation of phenanthrene and pyrene.

Carbon veil electrodes have already been proved for maximized electricity production and benefits in large-scale MFCs. Veil microchannels, due to their high porosity, facilitate nutrition transport via perfusion. In addition, carbon veil electrodes offer less internal resistance due to their consistent thread structure as compared to other carbon materials with inconsistent strands. Liu et al. [61] evaluated several carbon-based materials as working electrodes in a conventional three-electrode half cell system containing 150 mL reaction solution. Graphite rod and Ag/AgCl electrode were used as counter electrode and reference electrode, respectively. Carbon veil revealed roughly 40% higher current density than graphite and carbon rods. This higher current density of carbon veil electrode was attributed to the formation of microbial biofilms on it. The authors evaluated BET surface area of the materials and found that the real surface area of carbon fiber veil and carbon paper electrodes was about 10 times greater than other tested electrodes. Graphite rod and carbon rod also showed higher BET surface area than expected. The BET surface area measurement may only reflect the fine interior pores of the materials, which are not available to the microbe. As a result, the values are unreliable, and material structure properties played a more important role. Using electron microscopy images, the authors demonstrated that despite the thick and dense biofilms on graphite rod, carbon fiber veil creates a porous structure which colonizes the biofilm on carbon fibers having more than 10 μm thickness. The porous structure may also offer improved substrate access thereby achieving higher current density [61].

Apart from the basic materials described above, both cathode and anode structures can be modified to increase the removal efficiency. Catalysts have been used in several works to accelerate the reduction rate. Liang et al. [60] used porous and conductive GF as the anode while the GF cathode was painted with Pt catalyst (0.5 mg Pt/cm<sup>2</sup>). Electrode modification by catalysts could compensate for the poor kinetics of oxygen reduction in the cathodic compartment which led to higher removal rate of benzo [a]pyrene from polluted soil [60]. Carbon materials can be amalgamated with metals such as Mn<sup>2+/3+</sup> [62,63]. In a relevant study, Nandy et al. [62] compared MnO<sub>2</sub> coated anode with pristine carbon anode in an H-type dual-chamber system to investigate the degradation of hydrocarbons-crude oil components. They found that coated anode, compared to pristine anode, generated higher power density (47 mW/m<sup>2</sup> vs. 38 mW/m<sup>2</sup>) and showed higher alkane removal (36% vs. 25.5%) due to the better adhesion of microorganisms on the coated anode surface [62]. Nevertheless, use of Pt loading was not an economical manner. The integration of nanocomposites such as nanotube/polyaniline on anodic carbon was discovered as a promising material in MFCs [64,65]. In this regard, Liang et al. modified the GF anode applying three kinds of carbon nanomaterials (graphene, graphene oxide, and carbon nanotubes) [60]. The economic modification enormously enhanced the extracellular electron transfer rate, contaminant removal and power output because of high conductivity, high surface area, increased biomass content and excellent electrochemical characteristics of nanomaterials [66]. Potentiality of the modified electrodes due to their better conductivities, biocompatibilities, and optimal/reasonable costs make this technology suitable for large-scale operations [67].

### 4. Redox mediators

The existence of both endogenous (self-production) and exogenous (externally supplied) mediators accelerate mobile electrons'

transfer between microbes and electron acceptors. The redox mediators were applied in several MFCs to speed up the microbial activity of the system. However, external mediators are not commonly used in wastewater treatment due to the complicated and costly separation procedure of mediators from treated solution [68,69]. Recently, exoelectrogens (electricity generating microbes), which are able to transfer intracellular electrons without external assistance to terminal electron acceptors, have been identified as a driving force for electron transmission via electroactive molecules and membranes. Similarly, ferricyanide (FeCN) could also be used as a soluble and stable redox mediator in MFCs. In a related study, neutral red (NR) and FeCN have been used as redox mediators for toluene degradation in a waste remediation cell [70].

The desirable characteristics of effective mediators have been reviewed in detail by Evelyn et al. [71]. These desirable characteristics of mediators include the capability to form redox couples, ability to pass the membrane, stability in both oxidized and reduced form, a nontoxic character in respect to biological species, low cost, non-biodegradability, and sufficient capacity for electron acceptor role [70,72]. An appropriate range of added mediators enables microorganisms to use electrical power for growth as well as metabolite generation. The commonly used redox mediators are compiled in Table 2.

Adelaja et al. [38] investigated the impact of introducing 30  $\mu\text{M}$  of riboflavin into the anolyte medium as a redox mediator. The riboflavin has significantly increased the power generation and Coulombic efficiencies. In comparison to mediator-free MFC, the maximum power density and % Coulombic efficiency were increased by 30-times and 3-times, respectively when riboflavin was added to anolyte medium.

Nature-derived quinone redox mediators (NDQRM)- natural products-have proved significant potential in the electron exchange and formation of biofilms. Biochar -a representative of NDQRM-has been tested for TPHs removal. It was found that biochar has increased the removal rate of TPHs by 17% in comparison to non-biochar SMFC [76,77]. Under this operation, the physicochemical properties of biochar and the formation of different microbial communities of bacteria established an appropriate environment for soil remediation.

Several mediators such as FeCN and NR have been reported to be toxic to the enzymes and thus, hinder cell growth [75]. Therefore, favorable temperature and control pH should be used to achieve efficient degradation of target pollutants. While using 500  $\mu\text{M}$  FeCN and 200  $\mu\text{M}$  NR as redox mediators in MFC fed with 11.09 mg/L toluene, the output voltage was increased from 53.5 mV to 88.2 mV and 109.2 mV, respectively, i.e., an increase of 39% and 51% was observed [75]. However, due to the interference of NR and FeCN in cellular respiration, the degradation time of toluene was 1.56–2.15 times and 2.1 times lengthier for the FeCN and NR containing MFC, respectively, as compared to unmediated MFC [75].

In summary, due to the adverse effects of redox mediators and the possibility of washing away soluble redox mediators in high amount, they are not suitable for industrial applications.

## 5. Membranes

In a MFC system, generally, a proton exchange membrane (PEM) separates anodic and cathodic zones and is responsible for the diffusion of protons from the anode to the cathode. Mass transfer, ohmic losses, power generation, and pollutant removal are greatly affected by the performance of membranes as they can inhibit electrode contact and merging of both compartments solutions. In case of lack of membrane, the solution from the anode chamber could move to the cathode side. It could cause serious biofouling on the cathode surface, thereby deteriorating system's performance. On the other side,  $\text{O}_2$  from the cathode chamber could reach the anaerobic anode chamber, causing inhibition of the anaerobic fermentation process at the anode. Durability, low cost and resistance to fouling are the most significant requirements for a suitable membrane. Lower amount of fuel crossover, oxygen diffusion, and ionic resistance are always favorable parameters for attaining higher power generation [78].

Mostly, Nafion membranes are used in MFCs because of their adequate ion exchange capacity, durability, and high proton conductivity. Nafion is a perfluorosulfonic acid membrane composed of hydrophobic fluorocarbon backbone ( $-\text{CF}_2-\text{CF}_2-$ ) to which hydrophilic sulfonate groups ( $\text{SO}_3^-$ ) are linked. They have been introduced in different thicknesses. Thin membranes encounter higher reactant crossover (e.g., substrate crossover, substrate loss, and oxygen leakage) and lower mechanical strength. Although thick membranes surmount these problems, they have high resistance and thus low current density. However, Nafions experience some problems. They are not suitable under neutral pH conditions and preferentially transport cation species such as  $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  rather than  $\text{H}^+$ , which are typically  $10^5$  times higher than protons in concentration in wastewater [79]. This cationic species transport process leads to increase in pH of the cathodic compartment, salt precipitation on cathode and therefore diminishes the catalytic performance of the cathode.

Several studies have explored the use of alternative membranes such as anion exchange membrane (AEM), cation exchange

**Table 2**  
Common redox mediators used in MFCs.

Mediator	Redox potential	Solubility in media	Toxicants	Reference
FeCN	–	Soluble	Phenols, Nitrophenols	[73]
Riboflavin	(-208 mV vs. NHE)	–	Phenanthrene, Benzene	[38]
Anthraquinone-2-Sulfonate (AQS)	(-225 mV vs. NHE)	–	Phenanthrene, Benzene	[38]
Methylene blue	–	1.34 mM	Volatile Fatty acid (VFA) (acetate)	[74]
FeCN	–	50 mM	Toluene	[75]
Neutral red	–	300 $\mu\text{M}$	Toluene	[75]

membrane (CEM) and bipolar membrane (BPM) (Fig. 5). Some of these materials are generally cheaper than Nafion but are still quite expensive, difficult to handle, and sometimes produce lower power generation owing to high internal resistance. The difference between AEM and CEM is in positively-charged and negatively-charged functional groups which are attached to the membrane polymeric backbone and accomplish the selective transfer of anions or cations, respectively. CEM, which allows the transfer of cations from one chamber to the other, is made of weak or strong acids in a polymeric sheet. AEM is selectively permeable to anions and likewise made from weak or strong bases. Structure of the sheets is inversely correlated with the transferred ion type. For example, negative charges in the case of CEM form a matrix of covalent linkage to a polymer and thus exclude anions. In the same way, AEM carries positively charged fixed groups and permits the passage of anions only. The basic polymer matrix and the fixed ionic charges are the two main parameters that define the characteristics of ion-exchange membrane. Chemical, mechanical and thermal stability of the membrane are determined by the basic polymer matrix. Whereas, the type and concentration of the fixed ionic group in the polymer matrix specify the electric qualities and selectivity of the membrane.

Carboxylic acid and sulfonic acid groups are weak and strong acids, respectively, which are frequently used for CEMs preparation. In the case of AEMs, quaternary ammonium (QA) is used as a strong basic group. Tertiary, secondary, and primary amines as well as phosphonium and sulfonium groups form less strongly functional bases. Despite better buffering, less corrosive environment, lower resistance, and restricted pH drop, AEMs are rarely used in MFCs due to their poorer chemical stability and susceptibility to substrate losses, especially in alkaline conditions, compared to CEMs.

BPM combines both anion-exchange and cation-exchange layers in a laminated structure and dissociates water molecules to  $H^+$  and  $OH^-$ . Afterward, protons through CEM and hydroxide ions through AEM transfer to cathode and anode, respectively [15,27]. This membrane can resolve pH or ionic imbalance, which usually takes place between the cells in MFCs. Even though BPM holds a relatively high polarization resistance, it offers a technology for recovery of acids and bases from salt solutions. BPMs are specially developed for saltwater treatment and desalination. Due to their easy fabrication, high conductivity, low density and chemical stability, stainless steel and graphite have been widely employed for this type of bipolar plate membranes [78,80].

Looking for an affordable and natural alternative to commercial membranes, porous ceramic materials have gained great interest in recent years. Earthenware, terracotta, and clayware membranes facilitate practical application by dropping the cost of the overall system. Interestingly, the internal structure in ceramic membranes can be customized by changing both the raw materials and firing cycles. In fact, modification in the composition of raw materials including alumina, zirconia, mullite, pyrophyllite, and other oxides as well as variation in the kilning method such as curing time and temperature yield comparable or even better performance to those of the polymeric separators [81]. This modification in the composition of raw materials and carefully controlling the kilning process tunes the ceramic properties such as porosity, permeability, internal resistance, water absorption, and cationic transfer selectivity which finally affect the flux of ions and solvent through it [82]. As an important feature, the porous material can be made in a uniform structure which could show resistant to chemical and microbiological degradation. Aside from exclusive chemical, mechanical, and thermal stabilities, ceramics are operative for long-time processes and even reusable after cleansing with harsh agents [83]. However, despite their multiple benefits, employing ceramic membranes is accompanied by low ion conductivity which could fade its privileges. In addition, oxygen diffusion through the membrane could negatively affect anodic respiration, where oxygen incorrectly acts as the terminal electron acceptor [84]. This may lead to a future development trend to find novel materials and/or new surface modification techniques. Accordingly, published articles on the usage of ceramic membranes in PHs treatment are still limited.

It has to be taken into account that deformation and biofilm development on the membrane is almost an inevitable process during

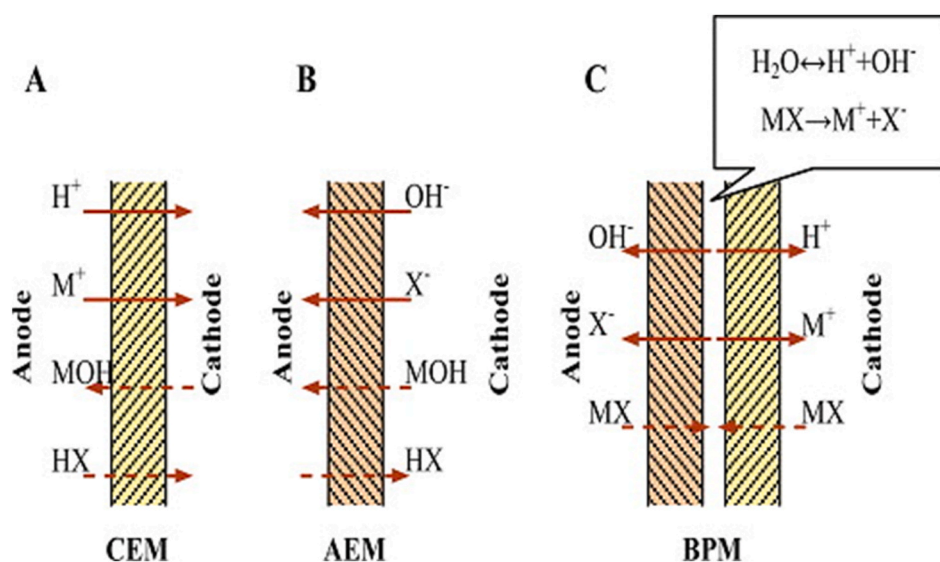


Fig. 5. Schematic diagram of ion and mass transfer across: (A) cation; (B) anion; and (C) bipolar exchange membranes (Reprinted with permission from (Li et al., 2010). Copyright © 2010 Elsevier [79]).



long-term operations. Chemical cleansing agents may help the removal of these formed biofilms from the surface or inside the membrane. Resistance of membrane structure to the agents is a required feature to recover conductivity, ion-exchange capacity, and performance loss [78]. However, the perfect type and material of a separator have not been found yet. Each one has its own advantages and constraints. Considering the operating conditions, a suitable membrane should be selected for practical applications [79].

It is noteworthy that SMFCs do not usually need CEM or PEM, which noticeably decrease the construction/overall cost of the systems [85]. Zhou et al. [86], however, exploited a CEM in their dual-chamber MFC design for hydrocarbon-contaminated soil to produce electric current. Morris and Jin [87] did the same with a Nafion perfluorinated resin in a sediment based MFC.

In water and wastewater PHs treatment, Adelaja et al. [88] presented a robust design of the system with a CEM. Other researchers employed PEM [89], CEM [88] and Nafion proton-exchange membranes [90–93]. Morris and Jin [94] constructed vinyl tube proton bridges having various lengths and diameters to link anode and cathode compartments in two-cell MFC composed of potassium chloride, copper sulfate and ultrapure agar mixed in deionized water which was boiled and then solidified in tubes. They concluded that the largest diameter (1.9 cm) and the shortest length (0.7 m) provided highest cell potential.

## 6. Microbial community

The removal efficiency of hydrocarbons in a MFC depends on two factors -adsorption and microbial degradation. The activity of microorganisms has a significant impact on the degradation rate of recalcitrant compounds. Biache et al. [95] found that the type of microbial communities was the major factor for the polycyclic aromatic compound (PAC) degradation in soil and slurry. It should be pointed out that the biofilms which grow on the anode surface contain degradative and fermentative microorganisms to begin the degradation process. Organic pollutants such as phenolic compounds, toluene, or benzoic acid are subjected to anaerobic reactions resulting in the formation of organic acids; then electroactive microorganisms breakdown the acids into carbon dioxide and electrons [75]. The biodegradability potential of PAHs depends on their molecular weight. The reports indicated that the microorganisms found It has been reported that low molecular weight (LMW) compounds are more appropriate as a sole source of carbon for microorganisms than high molecular weight (HMW) compounds. As result, LMW compounds were found to be mineralized/degraded faster than HMW compounds [39,95].

The performance of MFCs is significantly influenced by the interactions of different microbial species [96]. Both synergistic and antagonistic interactions are possible among different microbial species leading to the enhancement and reduction, respectively, of the power density output and removal rate of hydrocarbons. In this aspect, Guo et al. [96] reported that the best MFC performance in terms of electricity generation and oil removal rate was observed for the combined application of *Deinococcus* sp. and *Paenibacillus* sp. due to their synergistic interaction. For this MFC, the power density output and the oil removal rate were calculated to be 102.93 mW/m<sup>3</sup> and 85.56%, respectively. *Deinococcus* sp. converted PHs to simple fatty acids and esters. These simple fatty acids and esters were further degraded by *Paenibacillus* sp. Thereby both species worked in cooperation and thus, improved the electricity generation and wastewater treatment performances. Similarly, Zhou et al. [39] and Qin et al. [97] enhanced the removal rate of PAHs, including benzo(a)-pyrene, with the addition of LMW naphthalene and phenanthrene respectively. On contrary, the antagonistic effects of microbial strains can adversely affect the process. For example, Guo et al. [96] observed that the *Microbacterium* sp. Interaction with *Paenibacillus* sp. and *Deinococcus* sp. Limited the microbial electron transfer to the anode surface. As a result, the internal resistance was increased and the system operation/output was decreased. Yu et al. [59] and Liang et al. [60] studied the richness/abundance of a microbial community at the phylum and genus levels. The possible paramount phylum in PAH-polluted soils was found to be *Proteobacteria* [59]. Moreover, the process of benzo(a)-pyrene degradation was promoted by the existence of *Gammaproteobacteria* and *Clostridia*, which had significant abundance in the graphite felt anode [60]. Exoelectrogen microorganisms or anode-respiring bacteria, which transfer electrons to extracellular acceptors, can apply organic and inorganic substances as carbon sources and inoculate for the degradation and fermentation of products in the polluted hydrocarbon sites in parallel. Zhou et al. [86] directly adopted PHs contaminated soil to enrich exoelectrogenic bacteria in the MFC system. Meanwhile, they successfully identified and isolated two species of exoelectrogenic strains belonging to the *Geobacter* sp. and *Ochrobactrum* sp. [86]. Wang et al. [24], Venkidusamy et al. [57], Zhou et al. [86] and Zafar et al. [98] found the *Geobacter*, *Flavobacterium*, *Rhodopseudomonas*, *Ochrobactrum* and *Stenotrophomonas* as major exoelectrogens in the anode of MFCs.

Microbial immobilization is a reusable technology that encapsulates microorganisms in a fixed area to control microbial activity and community precisely. Confinement of cells and enzymes inside or on the appropriate carrier plays a significant role in the performance of microbial immobilization [99,100]. An immobilized nontoxic carrier not only increases the attachment area with degrading bacteria but also provides a harmless environment for the growth of exogenous bacteria. The carriers that are used for the microbial immobilization process are categorized into natural, organic, and inorganic materials [101]. Annie et al. [102] indicated that the bio-carrier coconut coir pith is regarded as a suitable amendment for the remediation of crude oil in polluted soil. Partovinia et al. [103] modified polyvinyl alcohol and alginate with halloysite nanotubes (HNTs) to improve the surface area and provide a harmless physical method, resulting in a reliable and low-cost matrix for cell stabilization.

Researchers suggested that usage of immobilized bacteria increased acclimatization of the bacteria with the environment and reduced lag phases and reaction time. In this regard, the start-up time, which is the main issue of MFCs, could be resolved and enable this technology to achieve the highest removal rate [104,105].

It is critical to explore the mechanisms of anodic biofilm communities, develop a better way to facilitate the electrochemical activities of the anodic microorganisms, and improve the reactor performance. It has been reported that the electrical power output is directly related to the biofilm population number of adherent cells in a steady state anodic biofilm system. However, the cell population/quantity is not the only controlling factor for power output. It is axiomatic that there would be no power generation if all cells

were dead with zero metabolic rate. Hence, the metabolic rate of the cells plays a more critical role in power output [106]. In a steady state continuous flow, the metabolic rate is proportional to the growth rate of the cells which, in turn, is related to the electrons' source, particularly carbon-energy substrate.

Microorganisms act as biocatalysts that influence power output by overcoming total internal resistance, degrading the feed, and transporting electrons to the anode [107,108]. Hou et al. [108] utilized electrochemical impedance spectroscopy (EIS) and cyclic voltammetry (CV) to investigate the anodic biofilm growth effect on power generation. The experiments were conducted on the abiotic and open-circuit conditions for 30 days. The authors found that the growth of biofilm greatly enhanced the power production through increasing the rate of electrochemical reactions via enhancing the extracellular electron transfer from microorganism to anode. Biofilm growth has also reduced the anodic polarization impedance and activation losses.

The biofilm structure inside an MFC relies on numerous factors resulting in either a thick (diffusion limiting) or thin (perfusion limiting) structure. The thick biofilm matrix does not contain the impervious electrode substratum, so the formation of a thick biofilm leads to matrix heterogeneity. Under such conditions, there is very low nutrients diffusion rate leading to the starvation of the inner cells which will then fight for their survival rather than growth. On contrary, a thin perfusable biofilm made up of permanently adherent cells have access to flowing nutrients. As a result, there is greater chances for cells to mutate. However, these mutant progeny will have minimal benefit in microbial cells since they are washed away by shear stress or natural cycles and are unable to aggregate [106].

Regarding the microbial community's abundance, the Simpson index measures the community diversity while the Shannon index represents both evenness and richness of the community. Thus the great Shannon or low Simpson index illustrates the less microbial diversity. Yu et al. [59] categorized the microbial communities of different anode materials into two groups: (i) less richness and higher evenness, including aluminum sheet (AS) and graphite plate (GP), and (ii) higher richness and less evenness containing graphite felt (GF), activated carbon fibre felt (ACFF) and carbon cloth (CC) anode materials [59].

Jiang et al. [109] compared the microbial community of the anode biofilms in the dual-chamber and single chamber MFCs. The target DNA extracted from the microbes on the anode carbon brushes was analyzed using the Pyrosequencing technique. The total estimated number of Operational Taxonomic Units (OTU) were 1117 and 843 for single and dual-chamber MFC, respectively. The results indicated that the single MFC was richer with a higher Shannon index (3.94) than dual-chamber MFC (3.65). On the other hand, the Simpson index was greater in dual-chamber (0.0844) than single MFC (0.0476), representing better and more even distribution of microbes. Different electron acceptors, which impacted the extracellular electron transfer speed, were apparently the source of the difference in the microbial community of the anode biofilm. Hence, there is a need to screen and monitor the microbial biofilm to determine the best microbial strains for specific wastewater. Considering the operating reliability and reduced cost of single MFC, it has a brighter future in real-world applications [109].

## 7. Co-substrate

Co-metabolism is regarded as one of the efficient methods to enhance the degradation rate of refractory compounds in MFCs. Co-substrate has the potential to contribute to the proliferation of bacteria and induction of oxidizing enzymes providing biodegradable carbon sources. Indeed, the addition of a co-substrate establishes beneficial synergisms in the digestive media and provides deficient nutrients by generating more electrons, thus improving biogas production [110]. Using biodegradable organic compounds as co-substrates led to the increase in bacterial metabolism and accelerated the degradation procedure. It has been shown that the use of Labaneh whey (LW) wastewater as a co-substrate, due to its high organic and biodegradable contents, boosted the electrons generated in the dual-chambered MFC contained petroleum refinery wastewater. The extra organic matters provided by LW, along with greater amounts of total dissolved solids (TDS), increased electron transfer in the anode chamber and thus have achieved enhanced power generation and higher substrate degradation [110,111].

The previous studies revealed that the use of acetate as a co-substrate in a single-chamber MFC based on microbial communities of *Pseudomonas*, *Geobacter*, and *Shewanella* resulted in electricity generation and phenol removal simultaneously [XY]. In this regard, the promising ability of methanol and acetate as co-substrates in anaerobic biodegradation of petroleum hydrocarbons in marine sediments was confirmed [112]. The results demonstrated that various co-substrates led to distinct structures of the microbial community which subsequently impacted the TPH degradation. The impact of acetate on the degradation of TPH was more promising than that of methanol.

Zhang and Lo indicated above 95% phenol degradation within 60 h in an MFC with an aqueous air cathode, using glucose as co-substrate that incited the growth of whole populations of bacteria in the anode chamber. In addition, glucose-phenol mixture minimized potential phenol toxicity during shock load or temperature fluctuations [112].

## 8. Energy consumption and electricity generation

In an MFC system, the catalytic activity of microbes converts organic matters (chemical energy present in C-C bonds) into electricity [113]. From this perspective, MFCs are theoretically considered energy profitable because: (i) they can be operated under ambient environmental conditions; (ii) they directly produce electricity from substrate energy with a high conversion efficiency; and (iii) they produce less amount of sludge [113]. It is estimated that the system consumes only about 0.024 kW or 0.076 kWh/kg-COD on average (primarily for reactor feeding and mixing) [29]. On the other hand, the activated sludge-based aerobic process consumes about 0.3 kW or 0.6 kWh/kg-COD [29]. In addition, MFCs can be operated at low (<15 °C), ambient (15–35 °C) and high (50–60 °C) temperature ranges without aeration [29]. Moreover, MFCs produce low fossil-related CO<sub>2</sub> because of their low energy consumption.

Interestingly, conversion of biogas (e.g., H<sub>2</sub> or CH<sub>4</sub>) into electricity results in a considerable energy loss (more than 60%) whereas MFCs could produce electricity directly from organic matter present in wastewater with a high energy conversion rate [29].

The term “exoelectrogens” refers to microorganisms with the capability of transferring electrons from outside of the cell to a solid/insoluble extracellular electron acceptor. In MFCs, only exoelectrogens are responsible for the electricity generation. Currently,  $\alpha$ -,  $\beta$ -,  $\gamma$ -, and  $\delta$ -proteobacteria (e.g., *Geobacter sulfurreducens*, *Shewanella oneidensis*, *Escherichia coli*, *Geobacter metallireducens*, *Rhodospseudomonas palustris*) are the most studied exoelectrogens [29]. Previous studies have enriched MFCs with a wide range of feedstocks (electron donors) and innovations to produce electricity such as palm oil mill effluent (1648.7 mW/m<sup>3</sup> and 1280.56 mW/m<sup>3</sup> from pretreated and untreated effluent, respectively), glucose (1499 mW/m<sup>3</sup>), rice mill wastewater (656.10 mW/m<sup>3</sup>), sewage (13.65 mW/m<sup>3</sup>), potato chips processing waste water (612.5 mW/m<sup>3</sup>), municipal wastewater (41.4 mW/m<sup>3</sup>), young leachate (6817.4 mW/m<sup>3</sup>), aged leachate (78.1 mW/m<sup>3</sup>), cassava mill wastewater (18.2 W/m<sup>3</sup>), brewery wastewater (24.1 W/m<sup>3</sup>), fresh human urine (6.93 W/m<sup>3</sup>), sewage sludge (8.5 W/m<sup>3</sup>), and domestic wastewater (12.8 W/m<sup>3</sup>) [113–125]. Yet, the amounts are too weak to be used except for some small devices or ingenious sensors [126]. It is proposed that MFCs should have an output of at least 400 W/m<sup>3</sup> in order to compete with traditional anaerobic digestion as a substitute (at an organic loading rate of 5–25 kg-COD/(m<sup>3</sup> day)) [127] and be able to produce 1 kW/m<sup>3</sup> to fulfill energy self-sufficiency [128].

Abbasi et al. [129] observed a positive correlation between chemical oxygen demand (COD) concentration and generated voltage, possibly due to the increase in organic substrates and higher solution conductivity. At low COD concentration, anode restricted electricity generation due to kinetic limitations, whereas at high COD concentration, cathode demonstrated a more significant impact than the anode on electricity generation [113]. Mohanakrishna et al. [130] measured 2150 mg/L for COD value in petroleum refinery wastewater. On the other hand, the formation of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) during the cathodic reduction of oxygen decreases the attainable power of MFCs, especially with carbon electrodes. It will be better if H<sub>2</sub>O<sub>2</sub> formation is avoid during MFC operation as it could deteriorate the reactor internally owing to its high oxidizing power [131].

Some researchers found that a larger current in SMFCs accelerated the degradation rate. This may be because of: (i) the metabolism rate of anaerobic bacteria could be enhanced by the electrons generated by electrogenic bacteria; (ii) the electric field which possibly affects certain enzymes in electrogenic bacteria to enhance the removal efficiency of organic compounds; and (iii) the electric field influencing the permeability of cell membranes, thus resulting in increased absorption of extracellular substances and thereby promoting the metabolism of microorganisms [28].

Higher generation of power density is subject to several important considerations including: configuration development; anode and cathode function modification with material, biological factors, and redox potential; optimization of microbial colonization; decrease in internal cell resistance; and proper selection of the size, number of units, temperature, retention time, and fuel quality. As an effective solution, incorporating current collectors into electrodes necessarily increases electron transfer and power generation in MFCs [132]. Studies have reported maximum current/power/voltage in PHs treatment in a variety of setups which is mentioned throughout the article.

Solution conditions, high internal resistance, low degradability of substrate, and slow biofilm kinetics mainly account for low power density [133]. Generally, small reactors achieve high currents/voltages. For example, decreasing the size of a fuel cell from liter- or cubic meter-scale to milliliter-scale substantially increases the volumetric power density by 2–4 orders of magnitude [29]. First, “inhomogeneity” or unbalanced substrate distribution is evident in large-sized MFCs which are identified by different hydraulic pressures and substrate concentrations in different cell sections. It would suppress both the energy output and the system’s efficiency. Additionally, extending the volume of MFC increases the internal resistance. Hence, innovative designs are introduced to optimize power output levels for MFC scale-up. In this context, stacking/cascading of small-scale MFC units is being pursued. Reducing the size of consecutive downstream MFCs while avoiding supply recirculation within a cascade is one of the effective strategies for enhancing the power generation. Walter et al. [49] employed two cascade stacks consisting of constant size units (control cascade, CC) and

**Table 3**  
Effects of different parameters on hydrocarbon degradation in MFCs.

Factor Process	Condition	Effect	References
pH	Neutral pH	More current generation, less required current density	[134]
	<6-7	Favorable limits for alkane removal	[39]
	= 7	Favorable limits for PAHs removal	[136]
	Anode = 7.5 Cathode = 6.6	Favorable limits for chlorophenol removal	[137]
Temperature	Increase from neutral	Reduction in the propelling force for proton distribution in the catholyte and protons accumulation in the anolyte	[134]
	Increase to 35–40 °C	Enhancement in electricity production and hydrocarbon degradation	[140,142, 143]
	Increase to 50 °C Decrease from 35 °C to ≤ 20 °C	Inactivation of some microbial species, decrease in substrate consumption Less hydrocarbon degradation	[139] [38]
Conductivity	Sufficient/medium range	Improvement in microbial activity and electricity production, increase the density of biofilm layer on electrodes	[38,146]
	Higher than optimal range	Less hydrocarbon degradation	[149]
Redox mediator	Presence	Developing driving force for electron transfer	[68,69]
		Increase the captured moving electron and electricity generation	[75]

**Table 4**  
Summary of reported studies on MFC for PHs removal.

Pollutant	Reactor type	Initial concentration	Electrode material	Removal efficiency (%)	Treatment time (d)	Max. current/power/voltage	pH	Temperature (°C)	Redox mediator	Predominant species of bacteria	References
Phenanthrene	SMFC	1.23 mg/kg	Anode: GF modified by carbon nanomaterials Cathode: GF	78.1	110	48.61 mV	Anode chamber: 7.2		Self redox potential (Cathode: +220 ± 235 mV vs. Ag/AgCl)	<i>Proteobacteria</i> phylum and <i>Betaproteobacteria</i> class	[165]
Pyrene	SMFC	1.15 mg/kg	Anode: GF modified by carbon nanomaterials Cathode: GF	69.6	110	48.61 mV	Anode chamber: 7.2		Self redox potential (Cathode: +220 ± 235 mV vs. Ag/AgCl)	<i>Proteobacteria</i> phylum and <i>Betaproteobacteria</i> class	[165]
Naphthalene	SMFC		Graphite	76.9	45	17.20 mA/m <sup>2</sup>		27	Self redox potential	Mixed culture	[164]
Phenanthrene	SMFC	0.0954 mg/kg	Stainless steel	99.47	240	17.1 mV	Sediment: 7.8 Medium: 7.0	25	Not detected	<i>Proteobacteria Nitrospira</i> and <i>Chloroflexi</i>	[41]
Pyrene	SMFC	0.0316 mg/kg	Stainless steel	94.79	240	17.1 mV	Sediment: 7.8 Medium: 7.0	25	Not detected	<i>Proteobacteria Nitrospira</i> and <i>Chloroflexi</i>	[41]
TPHs	Single chamber	26 mg/kg	Graphite modified by carbon	46.15	20	179.59 mA/m <sup>2</sup>			Not detected	Mixed culture	[66]
Light PAHs	SMFC	49.73 µg/g	Anode: CC Cathode: Graphite	34.18	28		Medium: 8.1 ± 0.3	20	Not detected	Mixed culture	[40]
Heavy PAHs	SMFC	199.84 µg/g	Anode: CC Cathode: Graphite	4.9	28		Medium: 8.1 ± 0.3	20	Not detected	Mixed culture	[40]
TPHs	SMFC	16,000 mg/kg	Anode: CC Cathode: CP modified by Pt	24	66	2162 mW/m <sup>3</sup>	Sediment 7.2	30	Not detected	Mixed culture	[87]
Diesel	Single chamber	800 mg/L	Anode: Carbon fiber Cathode: CC	100	30	90.81 mW/m <sup>2</sup>	Medium: 7.2		Not detected	<i>P.aeruginosa</i> bacteria	[57]
Diesel	Dual chamber	176 mg/L	Anode: Stainless Steel	82	21	31 mW/m <sup>2</sup>	Medium: 7.0		Not detected	Exoelectrogenic bacteria, genera of sulfate reducers, and electrochemically bacteria	[89]
Phenol	Dual chamber	1000 mg/L	Anode: Carbon Cathode: Carbon modified by Pt	100	2.5	28.3 mW/m <sup>3</sup>	Medium 7.0	30.0 ± 0.1	Not detected	Mixed culture	[90]
Phenanthrene	Dual chamber	20 mg/L		98	5	1.25 mW/m <sup>2</sup>	Medium 7.0	25	Self redox potential (+0.2 V)	<i>P.anguilliseptica</i>	[88]

(continued on next page)



Table 4 (continued)

Pollutant	Reactor type	Initial concentration	Electrode material	Removal efficiency (%)	Treatment time (d)	Max. current/power/voltage	pH	Temperature (°C)	Redox mediator	Predominant species of bacteria	References
PAHs	Single chamber		Anode: CF Cathode: CF modified by Pt Anode: CF & Graphite Cathode: Graphite	89.4	35		Medium: 7.2	25	Vs AG/AgCl reference electrode) Self redox potential (Anode+375 ± 15 Mv cathode: +353 ± 10 Mv Vs Ag/AgCl)	Mixed culture	[162]
TPHs	Dual chamber	130 mg/L	Anode: Graphite Cathode: Carbon modified by Pt	51.16	12	1027 mW/m <sup>2</sup>	Medium: 7.0	22 ± 3	Not detected	Mixed culture	[91]
Benzene	Dual chamber GAC-packed	17 ± 1 mg/L Oil 4.40%	Anode: Carbon Cathode: Graphite	84 ± 3	24	330.4 mW/cm <sup>3</sup>	Medium: 7.2	30	Self redox potential (Anode: 402.60, 266.94 and 146.66 mv (VS-SCE)	Mixed culture	[92]
Volatile phenol	Dual chamber GAC-packed	41.01%	Anode: Carbon Cathode: Graphite	84 ± 3	24	330.4 mW/cm <sup>3</sup>	Medium: 7.2	30	Not detected	Mixed culture	[92]
Aliphatic Hydrocarbons	Dual chamber GAC-packed	54.59%	Anode: Carbon Cathode: Graphite	84 ± 3	24	330.4 mW/cm <sup>3</sup>	Medium: 7.2	30	Not detected	Mixed culture	[92]
DRO (Diesel range organics)	Single chamber	36.3 mg/L	Anode: CF Cathode: CC modified by Pt	89.5	6	132 mW/m <sup>2</sup>	Medium: 7.2	22 ± 3	Not detected	Mixed culture	[130]
Phenol	Single chamber	60 ppm	Anode: CC Cathode: CC modified by Pt	Batch mode: 85 continuous mode: 80	15 d batch mode + 16 h continuous	225 mW/m <sup>2</sup>	Medium: 8.07	29 ± 2	Not detected	Mixed culture	[93]
TPHs	SMFC	840 mg/L	Carbon	40.5	7	286.7 mW/m <sup>2</sup>	Soil: 7.45		Self redox potential (Bioanode: +178 mV)	Mixed culture	[55]
TPHs	SMFC		Anode: Carbon Cathode: AC	77	182	153.8 mA/m <sup>2</sup>			Not detected	Mixed culture	[155]
TPHs	SMFC		Anode: Carbon Cathode: AC	22	135	0.28 mA/m <sup>2</sup>		30	Not detected	Mixed culture	[138]
PAHs (phenanthrene and pyrene)	Plant-MFC	1000 mg/kg	CF	54.2	35	184.9 mV		25	Not detected	<i>Geobacter</i> spp.	[154]

(continued on next page)





Table 4 (continued)

Pollutant	Reactor type	Initial concentration	Electrode material	Removal efficiency (%)	Treatment time (d)	Max. current/power/voltage	pH	Temperature (°C)	Redox mediator	Predominant species of bacteria	References
TPHs	SMFC		Anode: Carbon Cathode: AC	21	135	457 mV	Soil: 8.26	8.26	Not detected	Mixed culture	[153]
TPHs	SMFC		Anode: Carbon Cathode: Stainless steel	12.5	135	0.282 V	Soil: 8.6	23 ± 3	Not detected	Mixed culture	[157]
PAHs	SMFC		Anode: Carbon Cathode: Stainless steel	14	135	0.282 V	Soil: 8.6	23 ± 3	Not detected	Mixed culture	[157]
TPHs	SMFC			12	180	37 mA/m <sup>2</sup>	Soil: 8.92 ± 0.07	30	Not detected	<i>Geobacteraceae</i> sp. and <i>Escherichia</i> sp.	[42]
TPHs	SMFC		Anode: Carbon Cathode: Stainless steel/AC	12	180	37 mA/m <sup>2</sup>	Soil: 9.02 ± 0.04	30	Not detected	Mixed culture	[42]
TPHs	SMFC		Anode: Carbon Cathode: Stainless steel/AC	12	180	37 mA/m <sup>2</sup>	Soil: 9.02 ± 0	30	Not detected	Mixed culture	[42]
TPHs	SMFC		Anode: Carbon Cathode: Stainless steel/AC	12	180	37 mA/m <sup>2</sup>	Soil: 9.28 ± 0.01	30	Not detected	Mixed culture	[42]
PAHs	SMFC		Anode: Carbon Cathode: Stainless steel/AC	27	180	37 mA/m <sup>2</sup>	Soil: 8.92 ± 0.07	30	Not detected	Mixed culture	[42]
PAHs	SMFC		Anode: Carbon Cathode: Stainless steel/AC	27	180	37 mA/m <sup>2</sup>	Soil: 9.02 ± 0.04	30	Not detected	Mixed culture	[42]
PAHs	SMFC		Anode: Carbon Cathode: Stainless steel/AC	27	180	37 mA/m <sup>2</sup>	Soil: 9.02 ± 0	30	Not detected	Mixed culture	[42]
PAHs	SMFC		Anode: Carbon Cathode: Stainless steel/AC	27	180	37 mA/m <sup>2</sup>	Soil: 9.28 ± 0.01	30	Not detected	Mixed culture	[42]
Anthracene	SMFC	103 mg/kg	ACFF	54.2	175	12.1 mA/m <sup>2</sup>	Soil: 7.92	27–30	Not detected	<i>Proteobacteria</i> as dominant phylum	[158]



variable size units with decreasing width (test cascade, TC) with the same volumetric flow rate. The results showed that a cascade comprising MFCs of TC optimizes the received power for a given volume of feed. 400 h power monitoring of the mentioned units showed that the absolute power of the CC stack ( $5.75 \pm 0.08$  mW) was more than that of TC stack ( $3.50 \pm 0.05$  mW). However, The power density of the CC stack ( $8.00 \text{ Wm}^{-3}$ ) was lower than that of TC stack ( $10.00 \text{ Wm}^{-3}$ ), i.e., a 25% increase for TC stack. This 25% rise in the power density of TC was attributed to an increase in power density of each row in the TC stack. However, the power density in a CC stack dropped as every row moved through the cascade [49].

## 9. Optimal process parameters

### 9.1. pH

In MFC systems, the substrates are oxidized to release protons and electrons, so they combine with oxygen to generate water in the cathodic section. The reactor media such as soil, sediment, or wastewater might have an internal resistance that impedes the protons transfer, resulting in a pH gradient from anode to the cathode. Same time, determining appropriate pH values for the effective microbial activity of both degrading and electrogenic microorganisms is essential [134]. The anodic pH affects both anodic and cathodic compartments since it directly controls the rate of the microorganisms' transfer, thereby the proton and electron concentration.

The optimal pH range has a great impact on the bioavailability of substrates and thus hydrocarbon elimination and electricity production [135]. In this regard, the pH spectrum for electricity generation is usually neutral, maximizing electrogenic bacteria's operation. For instance, pH = 6.80 is ideal for the *Geobacter* sp. Moreover, pH = 6–7 and pH = 7 are the most propitious limits for alkanes and PAHs biodegradation, respectively [39,136]. Hassan et al. indicated that with the potential of electroactive dechlorinative bacteria such as *B. subtilis*, the optimized values for chlorophenols degradation was achieved at anode pH = 7.5 and cathode pH = 6.6 [137]. When pH is too low in the catholyte, the propelling force for proton distribution through the membrane will be reduced, resulting in protons accumulation in the anolyte. It should be noted that due to sufficient release of protons in the moderate level of pH, more current generation occurred, and required current density diminished (see Table 3). Briefly, while the pH of the anode zone increased up to the optimal amounts, the current density decreased, and in reverse [134].

Variation between pH values of the anode and the cathode is a dominant factor affecting the activity of microorganisms and thus the internal resistance of the system. To tackle this issue in systems with acid-base differentiation, a relatively acidic catholyte can be added to the cathode, and similarly, alkaline substrates can be added to the anode. The results of monitoring the pH alteration during the process revealed that after a slight pH reduction at the beginning of the process, and ultimate marginal pH increase in the anodic chamber was observed. It might be derived from balanced ion transfer other than  $\text{H}^+$  and  $\text{OH}^-$  during the operation. Li et al. proved that the slow transfer of anodic proton resulted in hydroxyl anions accumulation close to air-cathode and ended up with a pH increase in the soil [138].

### 9.2. Temperature

Temperature is a significant parameter affecting microbial kinetics, mass transfer capacity, and oxygen reaction in the reactor [139, 140]. Different MFC performances have been observed in relation to various starting up temperatures. A decrease in current density was clearly detected with a reduction in operating temperature [141]. According to related studies, lower initial temperatures would slow or impede the system from being properly initiated. Lower operating temperature influenced the microbial communities, which rely on increasing start-up time and MFC efficiency. A power production enhancement took place when the MFC was run at a higher temperature (about 40 °C), and then it was adjusted to a lower one [142,143].

Results of temperature changes during the process in the anodic compartment are more noticeable than the cathodic one. In the anode biofilm, the temperature rise is correlated with the growth of microbial communities. Studies have shown that the cathodic biofilms are more stable even at low temperatures and are also capable of providing great redox activities involved in electron transmission. This adaptation in low temperature is generally attributed to the high metabolic activity of cathodic microbial communities, which are different from those that functioned at ambient temperature [144].

Specific ranges of temperature have been defined for the efficient operation of different microorganisms. Thus, a suitable temperature range enhances system performance in terms of substrate degradation rate and electricity generation. As demonstrated in many studies, 35–40 °C was recognized as an optimal temperature for hydrocarbon degradation and electricity generation [140]. Relatively high temperature facilitates biofilm formation, electron transfer, therefore, current generation. An investigation about the influence of temperature for PHs degradation, ranging from 20 to 50 °C, confirmed a doubled rate of degradation at 40 °C in comparison with 20 °C. However, an evident decrease in substrate consumption rate and performance was observed at 50 °C (see Table 3). This decrease in high temperature resulted from the inactivation of some microbial communities [38,139]. The results correlated to the distinct phases of microbial growth, which determines the microorganisms' growth rate.

Temperature is also regarded as a major environmental factor that has an obvious impact on the archaeal distribution present in petroleum reservoirs, and thus the hydrocarbon degradation. In this regard, Liang et al. [145] investigated the archaeal community of high-temperature petroleum reservoirs and revealed that a wide variety of archaeal was active in high-temperature environments. In addition, they posed that temperature rise was directly related to the presence of more archaea in petroleum reservoirs, and thereby it was more beneficial for hydrocarbon degradation.

### 9.3. Conductivity of media

Conductivity acts efficiently in a BES to improve microorganisms' activity and generate electricity. For sediment and soil as significant sites of hydrocarbon pollution, enhancing porosity, moisture, and salinity are the "available strategies" that can increase the electron transfer rate of MFCs.

Deploying materials including AC and carbon fiber increases the conductivity of media that promotes the function of electrogenic microorganisms resulting in greater performance of the process [146]. Guo et al. reported that the cell equipped with GG had better microbial activity and thus pollutant removal than the one with granule-activated carbon (GAC) anode [92]. It follows from the higher conductivity of G compared to GAC. Furthermore, when MFCs were applied in groundwater or sediment with contaminants such as nitrates, sulfates, or metals oxide, the ions present played the role of electron acceptors, thereby increasing the conductivity of the system [38,147].

Rising ion concentration in both aqueous media such as polluted wastewater and solid media such as sediment and soil is a common way to directly decrease the internal resistance of the BES. In this regard, adding appropriate type and concentration of ions to biodegradation cells of PHs or saline environments facilitates electron transfer and thus the production of electricity. Related studies demonstrated a direct relation between salinity and charge transfer within MFCs [38,139,148]. However, it is worth mentioning that further increase of salinity might have a detrimental effect on microbial metabolism and thereby result in lower performance. Results of Tremouli et al. indicated that although the system took advantage of utilizing optimal salinity (4.1 gKCl/L), the higher salt concentration (5.4 and 6.7 g/L) had an adverse impact on the operation (Table 3) [139]. Similarly, Askari et al. revealed that applying a medium range concentration of NaCl (165 g/L) improved the density of the biofilm layer on the CF electrode and thus enhanced the MFC performance for the treatment of saline sediment [149]. This result is in line with Luqueño et al. Daghigho et al., and Adeleja et al. studies [38,150,151]. Moreover, raising the water content of saline soil or sediment diminishes salinity stress and increases mass transfer.

## 10. MFC applications for the PHs removal

### 10.1. Soil

Hydrocarbons mainly damage the soil environment by changing the composition and structure of the soil, poisoning the medium, disturbing the plants' growth, and altering the soil microbial communities. MFCs have been applied in recent years for remediation to resolve serious soil PHs pollution. The main degradation mechanism is based on the redox gradient between the contaminants and the electrodes [15].

Wang et al. applied for the first time a U-tube soil MFC and obtained TPHs removal enhancement by 120% adjacent to the anode (<1 cm) in 25 days of operation. However, lower soil moisture or larger distances from the anode decreased the performance [24]. Lu et al. attempted to extend the radius of influence by new configurations in a pilot-scale installation. Column-type design worked well beyond the area close to the electrode, enhanced to the edge of the reactor and shortened the remediation period. They indicated that the radius of influence probably reaches 90–300 cm, which means 11–12 times the column radius [152]. With the aim of testing the practicability of long range bioremediation, Li et al. applied extended SMFC in PHs removal by utilizing multilayer anodes in saline soils. The three anodes system with single cathode was identified as efficient in increasing the degradation rates in each layer and the bacterial communities, which played a key role in the process [42].

Mohanakrishna et al. studied soil microenvironment where PHs and carbon electrodes performed electron donors and electron acceptors roles, respectively. The results demonstrated that the average TPHs degradation reached 46% in three cycles of operation. This showed an increase of 40% in comparison with the reactor without electrodes [55]. Li et al. investigated the effect of the addition of carbon source supplementation, glucose, for MFC biodegradation of aged PHs in saline soils. Aged PHs are confronted with a deficiency in electron acceptors, weak electron transfer, and low activity of microbes. The research indicated promising results, especially for soils in extreme environments or barren areas [153]. Zhao et al. introduced plants and surfactants into the MFC system. Therefore, the plants directly acted as electron donors for the microorganisms, absorbed organic contaminants, and helped the transformation and fixation of contaminants. In addition, surfactants were used to pass the electron transfer barrier and desorb contaminants from soil by increasing the solubility of hydrophobic pollutants. The results suggested a plant-surfactant combination in order to boost the MFC performance [154]. Another research by Li et al. was carried out examining the addition of five kinds of surfactants, anionic, cationic, nonionic, ampholytic and biosurfactant, to the aged PHs, which are depleted of easily biodegradable hydrocarbons. They found the most effective one, the ampholytic surfactant, for hydrocarbon degradation and bioelectricity generation [155].

Li et al. promoted mass transport in soil MFCs by the addition of sand which could enlarge soil porosity (45–51%), reduce Ohmic resistance (by 46%), and increase charge output (2.5–3.5C/g soil). This significantly accelerated the degradation rate of hydrocarbons [138]. Similarly, Li et al. overcame the difficulty of mass transfer by amendment of contaminated soil and compared three different biochars (with a mass ratio of 2%) derived from poultry, agriculture, and forestry industries. With the aim of improving soil properties, they deduced variant removal efficiencies related to diverse fractions of PHs. Agriculture and poultry wastes were suited for high removal of aromatics and alkanes, respectively, due to their physicochemical properties [76]. In another research, Li et al. assessed the effectiveness of MFC with 1% conductive carbon fiber mixed with the contaminated soil to reduce the tremendous internal resistance and assist the anode in gathering electrons. This could meaningfully lead to 15-fold bioelectricity generation and improve the effective range of hydrocarbon remediation from 6 to 20 cm [156].

The MFC for in situ soil bioremediation using two different arrangements of anodes (horizontal or vertical) was evaluated by Zhang et al. The superior performance for hydrocarbon removal was obtained using a configuration with anodes horizontally arranged, which removed TPHs about 50% more than the vertically arranged reactor [157]. Yu et al. tried to discover the underlying mechanism of electricity generation and the PAHs removal emphasizing the electrodes interval and the 'anode's microbial community structure and composition. Testing from 4 cm to 10 cm, they achieved more favorable results when electrodes were positioned closer because of smaller internal resistances. Besides, according to their analysis, the system enriched and promoted growth of microbes at the anode surface owing to electric stimulation [158]. Li et al. inquired into electrode aging in MFCs arising from long-term remediation period through examining the surface profile changes of electrode, element accumulation on its surface, and element migration between MFC electrodes in soil. The aging phenomenon undoubtedly reduces self-generated biocurrent and disturbs the operational process. After 300 days, they observed accumulation of cations on cathode (52% of increment) and anode (35% of increment). The stacking of Fe and Ca played a key role in soil hardening, besides the accumulation of Na resulted in electrode corrosion [159].

Several studies developed the remediation system to PHs contaminated saline-alkali soil since salt-influence soils constitute approximately 40% of the 'earth's lands. This common polluted environment typically occurs in coastal areas because big oilfields are often located there. The salts impose unpleasant high osmotic potential and subsequently circumscribe the metabolic activity [160]. Furthermore, they are exposed to high internal resistance. In this sample, Li et al. found the suppressing effect of extreme salinity with soil blended with carbon fiber. The system filled with soil via rinsed salt and mixed with carbon fiber expedited the degradation rate of TPHs, PAHs, and n-alkanes [161]. Accordingly, MFCs have shown satisfactory application prospects to treat PHs polluted soil.

### 10.2. Water/wastewater

To address serious water pollution by hydrocarbons usually caused by spillages and leakages from storage tanks, researchers struggled to develop strategies and scopes of MFC systems. Morris and Jin conducted a study with both single-cell and two-cell MFC design harvesting electricity by utilizing hydrocarbons as the sole substrate in the groundwater, which is depleted of oxygen except at or near the surface. Their preliminary data held the view that the MFC technology made groundwater remediation possible under anaerobic conditions and eliminated conventional in situ aeration [94]. In 2009, the authors implemented a double-cell design for the biodegradation of diesel compounds (C8–C25 hydrocarbons) in water. The results suggested increasing biodegradation by 165% over 21 days as compared to an open circuit control [89]. Gambino et al. explored single-chamber, air-cathode MFC for PAHs degradation and detoxification in the water environment resulting in 90% decrease in PAHs concentration [162]. Venkidusamy et al. documented the influence of pre-enrichment of the anode community in diesel fed single chamber mediator less MFC. The pre-cultured anode, through former long-term culture, in comparison with freshly inoculated new one confirmed excellent performance in terms of the current generation and hydrocarbon contaminant degradation, more precisely 6 times power generation and complete removal vs. 83% removal of diesel [57]. Adelaja et al. published a report on a range of inocula that degrade phenanthrene, a model hydrocarbon, through co-metabolism. All the inocula revealed high degradation potentials (higher than 97%) mentioned in Table 4 [88].

The use of wastewater as a low-grade substrate and an electron source in MFC method, likewise, has attracted attention during years. For this purpose, Mohanakrishna et al. ascertained the decontamination ability of single and dual chamber MFC for significant amounts of residual PHs in produced water. With respect to COD degradation, bioelectrogenesis and TPHs removal, dual-chamber MFC evidenced superior function [91]. Luo et al. determined the contribution of MFCs to phenol degradation in the presence of a co-substrate, glucose, in order to reduce toxicity of phenol during temperature changes or shock load. A distinctive twin voltage peaks pattern, at phenol degradation rates of 20% and 90%, was observed in glucose-phenol mixture as the fuel. Besides, all the experiments concluded above 95% degradation efficiency within 60 h [90]. Guo et al. inspected the influence of packing material characteristics on the system efficiency utilizing petroleum refinery wastewater in the role of fuel. In the anode chamber, packing materials would expand the superficial anode area and accordingly raise microbial enrichment as well as power density output. GG with excellent electrical conductivity improved petroleum pollutants decomposition, electricity generation and microbial community diversity; meanwhile, GAC promoted the stability of operation and preferential removal of volatile benzene and phenols due to strong adsorption ability [92]. Mohanakrishna et al. aimed to boost petroleum refinery wastewater treatment with the aid of externally applied voltage (in the range 100–500 mV) for the initial 24 h. The supplemented potential was identified as effective on bioelectrogenesis and substrate degradation. The 500 mV voltage exhibited the maximum power density, substrate and diesel range organics/straight chain-alkanes removal efficiency much higher than 300 mV, 100 mV, and control MFCs [130].

### 10.3. Sediment

Recently, sediment MFCs have become competitive technology for removing hydrocarbons. Many researchers have reported higher removal efficiencies of the technology compared to in situ natural biodegradation in contaminated sediments. 'Sediment's complex composition of various organic matters, microbial populations, and mineral particles beside water and air, affects the internal resistance and, therefore the performance of MFC systems. In the process, there is no need for mobilization or transport of sediment, thus reducing CO<sub>2</sub> emissions, and low adverse effect on the environment has been observed [85]. It had been reported that MFCs arouse aromatic hydrocarbons degradation with one or two aromatic rings (e.g. benzene, toluene, and naphthalene) in sediments [163]. Morris and Jin tested a modified single-cell MFC setup, appearing to enhance the degradation rate of TPHs by nearly 12 fold over natural biodegradation [87]. Yan et al. proposed construction of plexiglass columns as sediment bioreactors that are greatly capable of PAHs biodegradation in a combined approach (SMFC and Fe (III) addition) with about 95% removal efficiencies of phenanthrene and pyrene. They affirmed the opinion that in such treatment, organic matters present in freshwater sediments could adsorb remaining

PAHs with less bioavailability [41]. Nastro et al. proved the high potential of MFCs for in-situ remediation of marine sediments. Among all, according to the complexity of PAH types, degradation rates ranged from 10% to 86% after four weeks for Indenol [1,2,3-c,d] pyrene and naphthalene, respectively. In terms of power density, the systems reached their highest performance after two weeks [40]. Sherafatmand and Ng did detailed research into the capability of SMFC for bioremediation of PAHs under both aerobic and anaerobic conditions in the cathodic compartment. The results demonstrated more than 3 times removal rate compared with those in the SMFCs without electric current and better performance with 67% TOC removal in the anaerobic reactor as well [164]. Permana et al. executed a comparative study between single-chamber and dual-chamber MFC in sediment remediation. They realized that the single-chamber system can not only degrade more PHs than the dual-chamber system but also produce higher electrical energy on average to the other one [66]. Therefore, MFCs have been successfully investigated in sediment remediation. Fig. 6 exhibits schematic diagrams of MFCs in environmental applications. Moreover, Table 4 represents more accurate information of the reported studies on MFCs in different applications.

## 11. Cost analysis

Worldwide, an enormous amount of research effort is allocated to treatment processes to certify the economic viability of each method. In this regard, economic analysis of MFCs has been done since 2005 [166]. Many considerations have to be taken into account about the evaluation of MFCs cost, including capital and operating costs which are two main areas of focus for any development of this technology into real-world implementation. MFCs operating costs, including tax rates, electricity, labor services, and membrane replacement, are incurred during the process and are estimated to be 4% of the capital cost. The large capital costs of MFC modules mainly appear from the high-priced construction materials associated with the electricity, membrane, and electrodes. Some researchers inferred that the anode accounts for further electrode cost [167,168]; however, Rozendal et al. presumed up to 47% of capital costs of BESs related to the price of cathode materials [169]. Abourached et al. also dedicated 84.5% of the capital cost to the electrodes for a single chamber laboratory size MFC [168]. It is clear that catalyst and current collector which is made of metal materials invariably cause extra cost. Besides the electrode, the high configuration cost is attributed to the overpriced separator material. The result of Ge and He demonstrated that the membrane and electrodes accounted for more than 60% and 19% of material cost for a dual-chamber MFC, respectively [170].

If the technology is to be commercially viable, all changes in the design of the electrode or the reactor structure described above must be aligned with the low-cost goal. Hence, researchers have made strides to reduce the system's overall expenses while

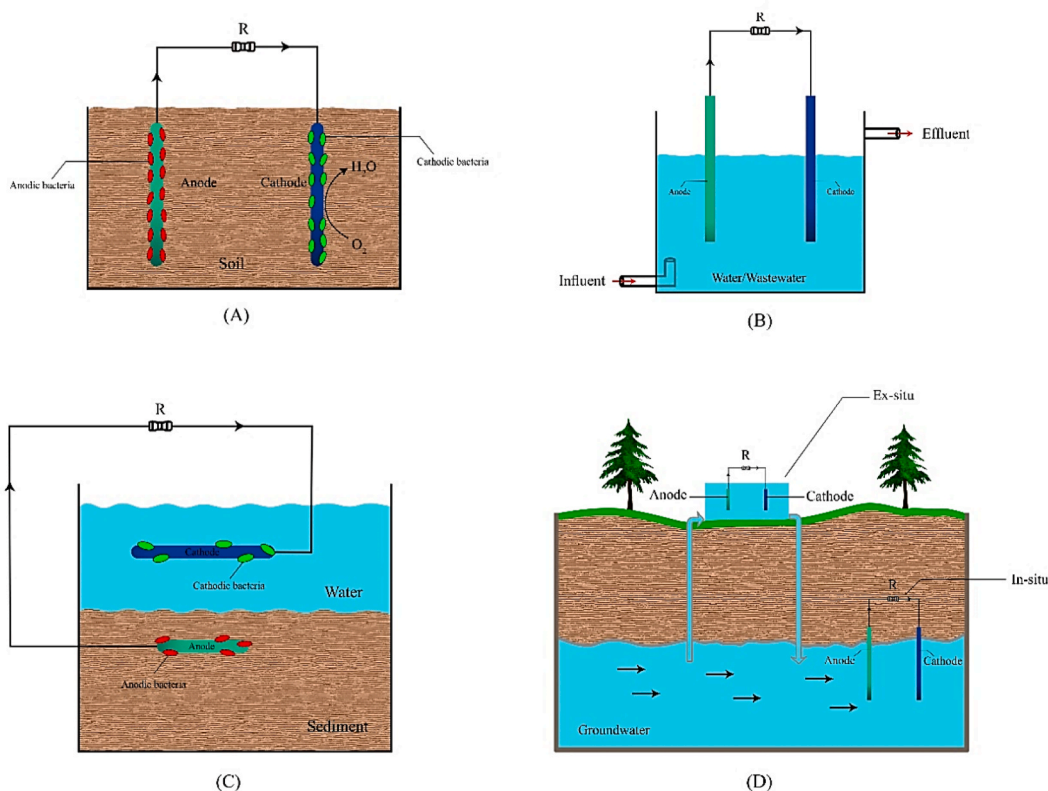


Fig. 6. Application of MFCs for the remediation of environments [28]. (A) Soil remediation. (B) Wastewater treatment. (C) Natural water remediation. (D) Groundwater remediation.



maintaining the same outputs of electricity. Ceramic and different fibers as a low-cost and scalable IEM, highly efficient and less-expensive electrode nanomaterials, and catalyzed biocathodes can considerably increase reaction effectiveness at lower operating costs [171]. Ghasemi et al. acknowledged sulfonated polyether ketone (SPEEK) as a great substitute for Nafion membranes. Results of cost estimation revealed that although MFC with SPEEK produced lower power than Nafion 117, SPEEK (595 \$/W) is about twice as cost-efficient as Nafion 117 (1134 \$/W) [172]. Similarly, Yousefi et al. compared the cost of ceramic membranes (2.5–3.5 \$/m<sup>2</sup>) with Nafion 117 (2300 \$/m<sup>2</sup>) [173].

In the matter of revenue, it is said that an MFC could return about 0.0005 \$/kg-COD assuming a net energy recovery rate of 0.004 kWh/kg-COD and based on an average electricity price of 0.12 \$/kWh [29]. It should be considered whether improving power output or applying the least expensive electrode materials is a cost-attractive approach. Stoll et al. reported that an overall income of 0.08 \$/m<sup>3</sup> for power generation is not economical according to the average cost of electricity in the U.S. (0.12 \$/kWh). Supposing an MFC reactor with about 2 m<sup>3</sup> volume, the generated power from hydrocarbons degradation is below 30 W/m<sup>3</sup>, and thus, the revenue of the system is very low. Furthermore, while applying inexpensive anode materials decreases power generation, the cost of hydrocarbon elimination in an MFC will be significantly reduced, so reducing expenses is more economical [174].

In recent decades, several pilot-scale studies of the stacked MFCs have been conducted and tried to optimize the technical performance related to pollutant removal, Coulombic efficiency, power output, and HRT [53,175]. Overall, it has been said that the capital cost of the MFC system is 30 times higher on average in comparison with the traditional activated sludge process [133]. It is estimated at \$ 6064, \$ 3300, and \$ 36,000 for 96 tubular modules, 18 stacked, and 50 modules, respectively [170, 176, 177]. Jadhav et al. reported a capital cost of \$ 4386 for electrode material and toilet assembly construction to treat 1.5 m<sup>3</sup> capacity of septage wastewater and generate electricity [169]. However, considering the savings related to water consumption, reuse of treated water, and generation of electricity revealed a payback period of 10 years for this system.

## 12. Future outlook and conclusion

Former studies of MFCs for the degradation of hydrocarbons from polluted environments can be applied as references for the real usage of this technology. Previous successful MET cases demonstrated that a single-chamber reactor with modified carbon-based electrode materials has great potential in the remediation of recalcitrant compounds. The data derived from laboratory and in-situ tests reported more than 97% hydrocarbon elimination in 60 days from a polluted sewage treatment plant. In marine sediments, 86% removal efficiency was declared after four weeks. In the same way, MFCs achieved about 90% efficiency in hydrocarbon-contaminated soils in 120 days.

Laboratory-scale MFC successfully provoked anaerobic microbial activity and improved degradation efficiency in comparison with open circuit MFC in environmentally benign conditions. Microbial BES has also enhanced the function of hydrocarbon-degrading bacteria by increasing the adsorption of pollutants on the surface of the electrodes and raising the microbial community. *Proteobacteria* are a common kind of bacteria that affect both power generation and activity of the microbial community in polluted soil.

Scalability requires to be backed by cost-effectiveness, chemical stability against material deterioration and biofouling, and excellent long-term performance. To accomplish commercialization of the technology, studies have mainly focused on continuous-flow MFCs in three categories, including large-size units, stackable individual cells, and/or multi-anode/cathode systems. Common configurations embrace both single- and dual-chamber reactors, and the majority of studies agree that stack modules potentially perform better than other configurations. Additionally, small-scale and package wastewater treatment plants are likely the most suitable options to initially install full-scale units.

One should take into account that operational factors (e.g., feeding substrate, temperature, hydraulic retention time, and organic loading rate) as well as specific design and materials can change the optimal large-scale application. For instance, the stacked assembly may hold an optimum number of units in one stack. Beyond that, there is not a significant improvement. Further, some aspects should be noticed to scale up the optimal cascade systems. Continuous flow mode in MFCs may overcome the development of harmful byproducts, prevent the reduction of electricity generation over time, and enable high productivity per unit volume with less labor. Altogether, it is more practical than batch culture systems. Generally, the selection of overall setup is a trade-off between a cost-effective design and enhanced efficiency in terms of removal and power generation.

However, the real scale-up application of this method is confronted with many challenges that need to be addressed; thereby, more attention should be paid to the following cases to surmount shortcomings.

- (i) In terms of system cost, it must be considered that the main expense is related to reactor electrodes and its income is covered by the energy and power production. Hence, managing a subset of the reactor (cathode, anode, redox mediator, and membrane) can provide a great chance for wide-scale use of MFC. Moreover, in order to attain economic justification, we propose to concentrate on reducing capital expenses instead of increasing the income of power generation [174].
- (ii) Arrangement and conductivity amendment of electrode materials can promote effective areas of remediation. Utilizing low-cost and great performance electrodes and redox mediators ensures the real use of MFCs.
- (iii) Microorganisms play a crucial role in the enhancement of hydrocarbon degradation. When functional microbes are applied at the appropriate temperature, resulting biofilms guarantee their profitable lifetime. Hence, these stable bacteria are more practical and economical than chemical catalysts [38,145].
- (iv) Synergic systems by combining MFCs with other technologies, particularly as a pretreatment, are a priority for future research. It would inhibit the transfer of complex compositions to the cell and shorten the execution time of pollutant elimination. Depending on the purpose, one may attain high effluent quality or more commercial value from MFC-based synergic modes.

- (v) Membranes should minimize or stop the transport of oxygen into the anode because of the anaerobic process of an anodic cell. This requirement would be difficult to control as to when the membrane gets wet. Water through the membrane structure would diffuse oxygen. Imposing a differential pressure (e.g., a small flow of liquid) from the anode to the cathode can prevent oxygen transfer [78].
- (vi) External energy needed to launch the circuit of MFC is one of the main challenges in system scale-up. Energy loss can be improved by designing integrated, highly efficient circuits, and also by collecting electrical energy produced from the system [143].

In summary, this technology, same as other prosperous methods, needs to be further investigated to realize fundamentals and functional barriers in the development of the process.

### 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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