Chapter 11
Advances in Concurrent Bioelectricity Generation and Bioremediation Through Microbial Fuel Cells

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11.1 Introduction

Industrialization and economic development of different countries can be measured in term of available energy source. For the past 200 years, fossil fuels were pillar of growth supporting the demand for energy, but each coin has two phases; therefore, along with advantages comes disadvantages. It damaged the environment leading to pollution, over-exploitation of natural resources and damaging the flora and fauna. These factors acted as promoter for different stakeholders to search for an affordable and environmental friendly alternative, such as biofuels and bioenergy. One such alternative was MFC; the concept came into existence in year 1910, when Michael C. Potter at University of Durham, UK, observed the ability of *Escherichia coli* to generate electricity (Potter 1911). It has emerged as a promising tool for bioenergy generation. MFC is a device that converts the chemical energy to electricity via biological pathways (Santoro et al. 2017; Zhang et al. 2016). The conventional chemical approaches are costly and require sophisticated infrastructure (Zaffar et al. 2016). MFC is a green tool for the treatment of different pollutants and simultaneous generation of electricity to meet the growing energy need of increasing human population. General schematic MFC found its application in wastewater treatment plants along with electricity and hydrogen generation (Wang et al. 2015), sediment bioremediation (Li and Yu 2015) and detoxification of polluted soil from toxic xenobiotic compounds (Rodrigo et al. 2014). The growing interests of scientific community in application of MFC and its further improvement can be easily observed

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Fig. 11.1 Articles published on MFCs with major focus on bioremediation. The data is based on the article mentioning MFC and bioremediation in the SCOPUS citation database in November 2017

through growing number of publication in this area (Zhang et al. 2016) which has gained impetus in the past 10 years (Fig. 11.1). This chapter gives an insight into the recent development in MFC technology as pollutant treatment units besides generating electricity, its limitations and economic feasibility for commercialization.

11.2 Improvement in the Microbial Fuel Cell Technology for Bioremediation

The development of MFC technology has come a long way since the discovery "animal electricity" by Luigi Galvani in year 1780. In the early eighteenth century, Volta's experiment led to the invention of early battery, and in 1859 lead acid battery was invented by French physicist Gaston Plante. William Grove is considered as the father of fuel cell technology, and the concept acted as the theoretical base for the discovery of fuel cells in the future. Various vehicle manufacturers have shown keen interests in fuel cell technology which led to the discovery of different fuel cells such as soft oxide fuel cells (SOFCs), molten carbonate fuel cells (MCFCs) and proton exchange membrane fuel cells (PEMFCs). However these technologies had their own limitations such as high operating temperatures, slow start times, need of precious metals as catalysts, high temperature, high cost involved and highly corrosive media in some cases. The alternative to these technologies was MFC, an efficient alternative to the costly abiotic fuel cells, as it can be operated under ambient temperature and pressure. The MFCs technology also attracted the attention of NASA scientists in the year 1960s when they showed interests in turning of organic wastes into electricity during the space missions; however, that was a short-lived project. The MFC technology went on back seat until the work by Bennetto et al.

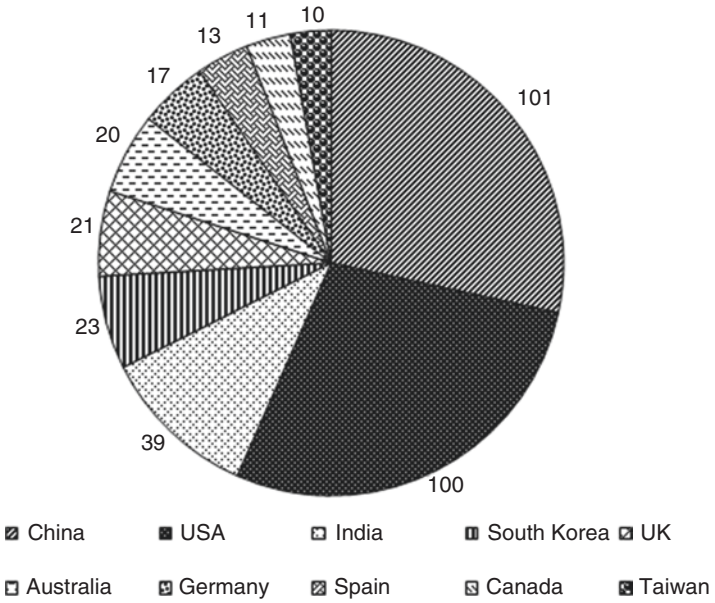

Fig. 11.2 Country-wise distribution in MFC research, number of articles on MFCs with focus on bioremediation. The data is based on the article mentioning MFC and bioremediation in the SCOPUS citation database in November 2017

(1983), where they reported the functioning of MFC with special focus on the use of mediators for electron transfer during electricity generation.

Since the advent of the twenty-first century, MFC attracted attentions of research groups around the world which is clearly visible from Fig. 11.1. Increase in number of publications of MFC with emphasis on bioremediation increased nearly sixty (60) times in year 2015–2016 as compared to year 2002–2003. It attracted attention of both developed and developing nation; China and the USA are in first place, and India is in third regarding MFC–bioremediation publications (Fig. 11.2).

In order to improve MFC, different strategies are used such as selection of electrodes, membranes materials, use of pollutants as substrates and designing new type of MFCs for specific applications which is explained later in the chapter.

11.3 Design of Microbial Fuel Cell

MFC can be divided into mainly types on the basis of design: single chambered and dual chambered. General schematic representation of MFC is described in Fig. 11.3 which contains separate cathodic and anodic chambers called as dual-chambered MFC whereas the single-chambered MFC contains both cathode and anode in a single chamber. Major components of MFC are cathode, anode and membrane.

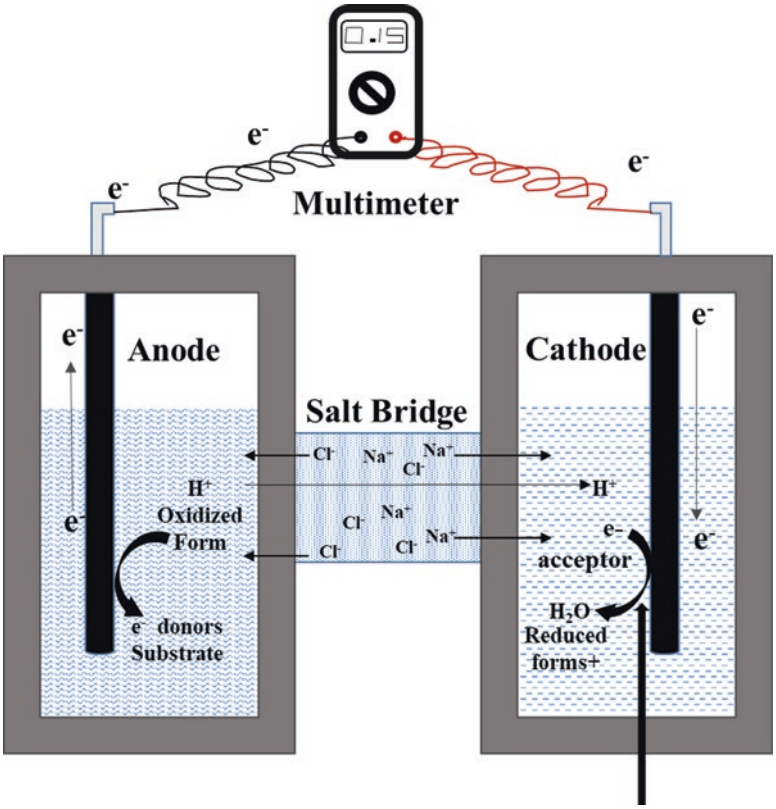

Fig. 11.3 Schematic representation of two-chambered microbial fuel cell and mechanism of electron and proton transfer in MFC (Based on Chaturvedi and Verma 2016; Zhang et al. 2016)

The choice of material for each component is an important factor in energy output and economics of MFC as technology. Each component and advancement in the selection of materials for the cathode anode and membrane is described in the next section.

11.4 Electrode Materials

The performance of MFC for electricity generation and bioremediation is based on selection of proper anode and cathode materials. The basis for electrode selection usually depends on bacterial adhesion, electron transfer and electrochemical efficiency (Mustakeem 2015). In general practice the cost of materials used for electrode must be low and power densities maximized (Mustakeem 2015). The criteria for selecting the anodic and cathodic material are as follow:

A. **Electrical Conductivity:** The electricity generation through MFC, an electron released from microbes, has to travel from the anode to the external circuit. The

electrode material having less resistance will have higher conductivity for the electric current as it allow effortless flow of electron. The lower the resistance, the higher is the conductivity for the flow of electron and result generation of electricity. The low interfacial impedance also plays a key role in facilitating the electron transfer. In case of cathode, the higher ionic conductivity is required for facilitating the triple-phase boundary reaction (Mustakeem 2015; Natarajan and Van Nguyen 2004).

B. **Surface Area and Porosity:** The surface area of electrode affects the final output power of MFCs. The loss of current is directly proportional to the electrode resistance. The efficiency of the MFC can be enhanced by decreasing the resistance of the electrode material that can be done by increasing the effective surface area whilst keeping the volume same. The larger surface area will provide large area for reactions that will enhance electrode kinetics (Wang et al. 2011; Rismani-Yazdi et al. 2008). Higher porosity allows bacteria to access and colonize more that help in biofilm formation.
C. **Stability and Durability:** The material used for electrode must be capable of withstanding highly reducing and oxidizing environment in MFC that may lead to the swelling and decomposition of the materials. The electrode material must be highly durable which may be provided by increasing the surface roughness. However, excessive roughness may increase the probabilities of fouling that may decrease overall performance of the MFC in longer term. Therefore, a highly stable and durable electrode are required for overall enhanced performance of MFC (Santoro et al. 2017; Mustakeem 2015).
D. **Cost and Accessibility:** It is necessary to use such material which is easily accessible to develop at lab scale and will help in impletion at large scale. The cost of the electrode material will influence the capital involved in construction of the MFC to a large extent. Thus, for implementation of the MFC at commercial scale, the material should be cheap, sustainable and easily available. For example, platinum is widely used; however, it is expensive. Carbonaceous and non-precious metal materials such as composites might be a substitute to precious metals in electrodes in the future (Santoro et al. 2017; Mustakeem 2015).
E. **Biocompatibility:** It can be seen in term of compatibility of electrode surface for proper adhesion of the microbial biofilm. A highly biocompatible material will increase the bacterial adhesion and hence the life of the MFC and ease of current flow (Santoro et al. 2017; Mustakeem 2015).

11.4.1 Anode Materials

Anode is the electrode used in MFCs and is the site where electron donors undergo oxidization reactions (Zhang et al. 2016). The material used in anode preparation plays a significant role in the biofilm formation and the electron transfer between the microorganism and the electron acceptor. Various carbonaceous, metallic and composite materials are used for the construction of anode for increased power density and better energy output (Mustakeem 2015). List of different anode along with properties are tabulated in Table 11.1. The interaction between biofilm and

Table 11.1 List of anodic materials and their properties used in MFC

Anode materials	Properties	References
Carbon cloth	High surface area, porosity, electrical conductivity, flexibility and mechanical strength in forming complex 3D structure. But expensive	Guerrini et al. (2014); Santoro et al. (2013)
Carbon brush	Expensive material consists of titanium core with twisted carbon fibres, High surface area that provides an optimal area to volume ratio. The central titanium core guarantee the electrical conductivity	Liao et al. (2015)
Carbon rod	Most affordable anode used in MFC. Surface areas for carbon rod are low. Thus they are more preferred as current collectors rather being used as anode	Jiang and Li (2009); Liu et al. (2004)
Carbon mesh	Commercially available at low cost. Carbon mesh can be folded efficiently to prepare a 3D electrode, low electrical porosity conductivity, mechanical strength and poor durability	Wu et al. (2017); Wang et al. (2009)
Carbon veil	Very cheap carbonaceous material with high electrical conductivity and porosity. It can be easily folded to form a robust and porous 3D electrode. Carbon veil is fragile	Artyushkova et al. (2016); Boghani et al. (2014); Winfield et al. (2014)
Carbon paper	Planar and relatively porous carbonaceous material. Expensive and fragile with its application limited to lab-scale batch system.	Santoro et al. (2014); Srikanth et al. (2008)
Carbon felt	High electrical conductivity. Carbon felt has high porosity that allows the microbes to colonize the biofilm by penetrating through the large pores. High mechanical strength owing to thickness of the material and its cost is relatively low	Seviour et al. (2015); Roy et al. (2014); Calignano et al. (2015)
Granular activated carbon (GAC)	The granular activated carbon is used as packaging material rather than using GAC as stand-alone anode. Due to GAC's intrinsic property of very high surface area, it can help in the adsorption of organics pollutants or heavy metals. Low cost and electrical conductivity, high porosity	Yasri and Nakhla (2017); Zhao et al. (2016); Jiang et al. (2011)
Granular graphite	It has properties similar to GAC except the granular graphite has much lower surface area because of lack of activation. It is also used as packing material rather than using a stand-alone anode. High electrical conductivity	Feng et al. (2010)
Carbonized cardboard	3D materials consist of single wall corrugated cardboard from recycled paper. The material is very low cost and has high electrical conductivity and porosity	Kretzschmar et al. (2017); Chen et al. (2012b)
Graphite plate	It has relatively lower surface area, surface to volume ratio and electric output. Due to its high mechanical strength, it is often used as support for modified structures. Graphite electrode is simple anode with high electrical conductivity and relative low cost	Dewan et al. (2008); Heijne et al. (2008)

(continued)

Table 11.1 (continued)

Anode materials	Properties	References
Reticulated vitreous carbon	Reticulated vitreous carbon is highly conductive. The unique property of high porosity allows the biofilm to penetrate through the entire structure and colonize the entire electrode. The major disadvantage is the material is quite fragile and very expensive	Lepage et al. (2012)
Electrospun carbon fibres	Electrospun carbon fibres are prepared by layer-by-layer (LBL) electrospinning of polyacrylonitrile onto thin natural cellulose paper followed by carbonization. High-density layered biofilm propagation and high bioelectrocatalytic anodic current density	Chen et al. (2011); He et al. (2011)
Activated carbon nanofibres	Activated carbon nanofibres have an ultra-thin, porous interconnected structure with high bioaccessible surface area that promotes well-supported biofilm growth. Application in simultaneous power generation and removal of organics from wastewater	Delord et al. (2017); Karra et al. (2012)
Carbonized plant materials	Several plant materials (e.g. corn stem, different mushrooms) are carbonized and tested as anode. The carbonized plant materials have highly porous architecture support, high electron transfer rate and electroactive biofilm growth. Low-cost alternative to costly anodes. Very conductive, robust and cheap and easily accessible. Major drawbacks are low surface area, biocompatibility issues and corrosion	Karthikeyan et al. (2015); Chen et al. (2012a)
Metallic anode materials	Several metallic materials such as stainless steel (plate, mesh, foam or scrubber) have been used as anodic material. Copper, nickel, silver, gold and titanium have been also successfully tested as anodic material. However, copper and nickel ions can be poisonous for microbes forming biofilm due to its metal toxicity but high and stable performance	Baudler et al. (2017); Guo et al. (2016)
Composite material	Carbon nanotubes–polyaniline composite enhanced the electrocatalytic property and adhesion to the bacterial cell. Polypyrole-coated carbon nanotubes have enhanced power density by six times as compared to graphite electrode. PPy-coated CNT showed high electron transfer	Mustakeem (2015); Sharma et al. (2008)
Material treatments	Several surface treatments such as ammonia-treated electrode and graphene over stainless steel mesh. Acid treatment of electrode and electrochemical oxidation treatment helps in protonation of functional group and addition of functional group, respectively	Lim and Wilcox (2012); Cheng and Logan (2007); Lowy et al. (2006)

anode electrode is affected by surface morphology and chemistry. The bacterial attachment with the electrode can be regulated by controlling the surface chemistry properties such as surface charge (Santoro et al. 2015), hydrophilicity/hydrophobicity (Du et al. 2017), oxygen/nitrogen functional groups and immobilized mediators (Santoro et al. 2017; Li et al. 2014). Further attachments can be regulated by surface morphology and can be controlled at nano- and micro-scale level using chemical

treatments, surface coatings and electrochemical and thermal treatments. Recently lot of surface modifications such as 3D electrode are preferred over 2D electrode, as theoretically it is suggested that surface area is directly proportional to electricity generation. However, several studies have suggested that there are limitations for 3D surface such as pH gradients and diffusion transport phenomenon associated with product and reactant (Blanchet et al. 2016). Whilst designing a MFC, anode electrodes must be selected such as to avoid clogging and dead zone, as it will help in long-term operation (Santoro et al. 2017).

11.4.1.1 Role of Anode in Bioremediation

Most of the organic pollutants present in waste and wastewaters are in reduced form, and anode acts as a site for oxidation of electron donors (Zhang et al. 2016). The chemical oxygen demand (COD) is adopted as mean to quantify the oxidation power of the pollutants. In the anodic chamber, COD is converted to carbon dioxide and water (Zhang et al. 2016; Kim et al. 2015a, b). The recalcitrant pollutants such as azo dyes (Thung et al. 2015), polycyclic aromatic hydrocarbons (PAHs) (Sherafatmand and Ng 2015), benzene derivatives (Zhang et al. 2015), inorganic wastewaters containing sulphide (Raschitor et al. 2015), industrial wastewaters (Abbasi et al. 2016) and several organic wastes such as chicken feather (Chaturvedi and Verma 2014), poultry droppings and human excreta (Kretzschmar et al. 2017; Ieropoulos et al. 2013) are used. It has also been used for the treatment of various waste treatments such dairy manure, polluted soil sample and landfill leachate. However, solid waste digestion efficiency of the anode is low, but several modifications have been carried out to improve its efficiency, e.g. polluted sediments/soil/groundwater can also be remediated by embedded anodes in the polluted matrix with an external cathode exposed to the air.

11.4.2 Cathode Materials

The oxygen reduction reaction (ORR) takes place at the surface of cathode under three phasic interfaces, i.e. electrode (solid), electrolyte (liquid), air (gas), to form water. The ORR is the limiting reaction of the MFCs, and typical MFC cathode can be divided into three layers, i.e. diffusion layer, conducting support layer and catalyst. The materials used for the cathode (Table 11.2) must bear certain properties to act as robust cathode such as (a) high mechanical strength, (b) catalytic property and (c) high electronic and ionic conductivity (Santoro et al. 2017; Mustakeem 2015). Most of the materials used for anode can be used as cathode; however, carbon-based materials have poor catalytic activity, and an additional catalyst material is required to boost the reduction process. Different types of material used as cathode are tabulated in Table 11.2:

(a) Cathode with Pt-based catalyst
(b) Cathode with non-Pt-based catalyst

Table 11.2 List of cathodic materials and their properties used in MFC

Cathode material	Cathode catalyst	Properties	References
Cathode with Pt-based catalyst/ platinum metal based	Carbon-Pt	Platinum is the most successful catalyst used for carbon cathode coating for oxygen reduction reaction because of its high surface area and low over potential for ORR. Limitations such as high cost, pH sensitivity, sulphide poisoning and non-sustainable	Watanabe (2008)
	Double-layer polydimethylsiloxane (PDMS)/carbon loaded with Pt	High-power density and hydrophobic materials like PDMS decrease the water diffusion into a single-chambered MFC	Zhang et al. (2016); Mustakeem (2015)
	Carbon nanotubes (CNTs) loaded with Pt	CNTs loaded with Pt by depositing it over CNTs surface. ORR catalytic activity was improved for CNT–Pt composite. The ORR was not affected considerably by decreasing Pt loading by 20-fold	Ghasemi et al. (2011); Cheng et al. (2006a)
Cathode with non-Pt-based catalyst/ platinum group metal-free	Iron phthalocyanine	Rapid electron transfer takes advantage of π–π interaction of metal and carbon	Yuan et al. (2011)
	Iron phthalocyanine-amino-functionalized multiwalled CNTs	High-power density	Yuan et al. (2011)
	Cobalt tetramethoxy-phenylporphyrin (CoTMPP)	Coulombic efficiency is comparable to Pt-based catalyst	Cheng et al. (2006b)
	Lead dioxide–carbon	Alternative to Pt as a catalyst, lead oxide has low cost and shows four times higher-power densities as compared to conventional Pt electrodes	Morris et al. (2007)
	Transition metal-based oxide	The cathode current with MnO_2-catalysed cathode was found to be much larger than graphite. β-MnO_2 deposited over glassy carbon using polyvinylidene fluorine (PVDF) as binder showed high catalytic efficiency and power densities as close to Pt based. CNTs covered with MnO_2 as cathode material showed high-power density increased by two order of magnitude as compared to plain stainless steel cathode	Amade et al. (2015); Cao et al. (2003)
	Palladium	Palladium, a Pt-like transition metal, has excellent catalytic. High stability and low cost as compared to Pt	Huang et al. (2011)
	Lead oxide	Four time higher-power density and eight time less cost compared to commercial Pt/C	Morris et al. (2007)
	Activated carbon	Large surface area, synthesized by electrospinning and pyrolysis of polyacrylonitrile precursor. Activated carbon fibre-treated electrodes was increased by more than 75%	Ghasemi et al. (2011)

(continued)

Table 11.2 (continued)

Cathode material	Cathode catalyst	Properties	References
Cathode with metal-free catalyst/ carbonaceous materials	Chemically modified carbon-based materials	Carbon nanotubes, graphene, and graphite foam can be used as catalyst for ORR	Su et al. (2010); Matter et al. (2006)
		Low cost, relatively high and stable performance	
	Nitrogen-doped graphene nanosheets	High catalytic activity, onset potential, higher electrocatalytic activity, lower over potential and long-term stability than commercially available Pt (10 or 20 wt%, E-TEX) for ORR	Lu et al. (2013); Ci et al. (2012); Gong et al. (2009)
Biocathode	Aerobic biocathode	In aerobic biocathode, oxygen acts as terminal electron acceptor, hydrogen peroxide acts as intermediate and transition metals such as iron and manganese act as electron mediators between the electrode and oxygen	Park and Zeikus (2003)
	Anaerobic type biocathode	In anaerobic type biocathode, oxygen is not present, whereas nitrates and sulphates act as terminal electron acceptors. Advantage by preventing the loss of electrons through oxygen also microorganisms at the cathode can act as biosensor for detection of biological oxygen demand (BOD) in water. Further microorganisms at biocathode can be used to produce as methane, ethanol and formic acid	Rabaey and Rozendal (2010); Kim et al. (2003)

(c) Cathode with metal-free catalyst
(d) Biocathode

List of cathodic materials used in MFC along with their properties are tabulated in Table 11.2

11.4.2.1 Role of Cathode in Bioremediation

In MFC the electron acceptors undergo reduction reactions at cathode. The oxidized substrates are reduced at the cathode if the available electric potential exceeds the threshold of oxidized substances (Zhang et al. 2016; Nancharaiah et al. 2015). The environment of cathodic chamber is highly reducing; therefore it is widely used in waste (landfill leachate) and wastewater treatment (liquid fraction of pig slurry, swine wastewater), organic substances (chlorobenzene and trichloroethylene) heavy metals like (Cr6+, V5+) and inorganic substances like ammonia (Sotres et al. 2015), xenobiotic compounds, etc.

11.4.3 Membrane Material

The architecture, choice of material and overall arrangement of membrane in MFC affects the performance, cost and multi-level applications. A large array of materials have been tested for its application as membrane such as natural rubber (Rajan et al. 2006), laboratory gloves rubber (Winfield and Chambers 2014), j-cloth, nylon fibres (Zhang et al. 2010), glass fibres, biodegradable shopping bags and ceramics (Winfield et al. 2013). The cation-exchange membranes (CEM), e.g. Naflon, is the most commonly used membrane system (Santoro et al. 2017). Dual-purpose ion-exchange bridge, monolithic 3D printed materials and porous materials (sufficient strength, chemical inertness and longevity) can be employed as the membrane materials (Santoro et al. 2017; Philamore et al. 2015). Several microporous filtration membranes (Zhuang et al. 2009), nylon infused membrane (Hernández-Fernández et al. 2015), photocopy paper (Winfield et al. 2015), ceramics and terracotta materials (Winfield et al. 2016) have also been tested as membrane**.**

11.5 Types of Waste Materials Used as Substrates in MFC

MFCs are considered as an efficient technology which effectively utilizes wastewater for energy generation (Winfield et al. 2016), Various waste materials used as substrates in MFC are as follows:

1. **Lignocellulosic Biomass:** Lignocellulosic materials are abundant and renewable natural resource. However it cannot be directly utilized as it has to be first converted to monosaccharides or low-molecular-weight compounds for the utili-

zation by the microorganism (Huang et al. 2011; Ren et al. 2007) for electricity generation. Cellulose and chitin are cheap, renewable and readily available biopolymeric materials which can be used for electricity generation (Rezaei et al. 2009). For direct conversion of cellulose, the microorganism(s) must be able to hydrolyse cellulose anaerobically, utilizing anode as an electron acceptor as well as oxidizing metabolites obtained after cellulose hydrolysis. Thus, using a solid substrate such as cellulose or chitin, the power production is limited due to a low rate of hydrolysis of the particulate material. However few studies have been carried out using particulate substrates in MFC.

2. **Synthetic Wastewater:** It has also been observed that synthetic or chemical wastewater having precise composition have also been used (Pant et al. 2010). However synthetic wastewater may contain redox mediators, such as cysteine and sulfur species (Aldrovandi et al. 2009). These redox mediators can act as abiotic electron donor and help in enhancing the production of electricity for a short while but it would not represent the true performance of the system (Aldrovandi et al. 2009) resulting in the ambiguity being generated in the results. Thus, this can be overcome by the use of minimal salt media with a single electron donor such as glucose or acetate.
3. **Brewery Wastewater:** Breweries wastewater has been used in MFCs, primarily because of its low strength, suitable for electricity generation due to food-derived nature of organic matter and lack of high concentrations of inhibitory substances, e.g. ammonia in animal wastewaters (Feng et al. 2008). It can be an ideal substrate for MFCs due to its nature of high carbohydrate content and low ammonium nitrogen concentration (Pant et al. 2010).
4. **Starch Processing Wastewater:** Starch processing wastewater (SPW) contains a relatively high content of carbohydrates sugars protein and starch which can be potentially converted to a wide variety of useful products (Jin et al. 1998). SPW was used as a fuel to enrich a microbial consortium generating electricity and current generation.
5. **Dye Wastewater:** Azo dyes constitute the largest class of synthetic dyes and are extensively present in the effluent of dye and textile industries. The removal of toxic substances present in the effluents before discharge is of paramount importance as they are effecting the environment adversely. In aquatic system intense colour of dyes has led to obstruction of light and transfer of oxygen into the water bodies which is having detrimental effect on the aquatic flora and fauna (Pant et al. 2007). Very recently, efforts were made to utilize these dyes as substrate in MFC leading to detoxification and generating electricity. The concentration of glucose and dyes plays an important role in the current generation; however, the microorganism are unable to decolorize high concentrations of dye (Sun et al. 2009). Another method which can be used includes simultaneous treatment of azo dye-containing wastewater and readily biodegradable organic matter-containing wastewater. The addition of later could help in improving the dye degradation efficiency due to microbial consortium associated with it along with the presence of nutrient material, thus saving both cost and energy. However, the system still requires considerable improvements in terms of finding appropri-

ate bacterial community that is capable of utilizing a mixture of dyes and other simple carbon sources in order to make MFCs a realistic solution for its extensive utility at large scale.

6. **Landfill Leachates:** The use of landfill effluent in a biological fuel cell for COD removal was first reported by Habermann and Pommer (1991). Landfill leachates are heavily polluted landfill effluents with a complex composition containing four major groups of pollutants: dissolved organic matter, inorganic macro-components, heavy metals and xenobiotic organic compounds (Kjeldsen et al. 2002). Recently, Greenman et al. (2009) demonstrated that it is possible to generate electricity and simultaneously treat landfill leachate in MFC columns. Gálvez et al. (2009) operated three MFCs fluidically connected in series for simultaneous leachate treatment and electricity generation.
7. **Inorganic and Other Substrates:** There are various substrates which have been used in the MFC as substrates such as paper-recycling plant wastewater (Huang and Logan 2008), unamended wastewater, phenol (Luo et al. 2009), carbon monoxide (Kim and Chang 2009), 1,2-dichloroethane (Pham et al. 2009), sulphate and thiosulphate (Zhao et al. 2009). MFC was used to remove the fermentation inhibitors in cellulosic biorefineries which included furfural, 5-hydroxymethylfurfural, vanillic acid, 4-hydroxybenzaldehyde and 4-hydroxyacetophenone with simultaneously generating electricity (Borole et al. 2009).
8. **Heavy Metals**

Hexavalent Chromium: Chromium has various industrial applications such as leather tanning, metallurgy, electroplating and wood preservatives (Chaturvedi and Verma 2016). Cr(VI) is more hazardous due to its mutagenic and carcinogenic properties (Humphries et al. 2004); therefore, there is a need for detoxification of hexavalent chromium [Cr(VI)]. Chemical or electrochemical reduction into nontoxic chromium is the most preferred method; however, other approaches such as ion-exchange resins, filtration and direct chemical reduction have also been employed (Kurniawan et al. 2006). These technologies require high-energy inputs and produces by-products which itself are pollutants. Therefore, reduction of chromium coupled with electricity generation using MFC and can be applied for Cr(VI) treatment (Tandukar et al. 2009) at the cathode. Various microorganism used in the process include *Trichococcus pasteurii* and *Pseudomonas aeruginosa*.

Selenite: Selenium and its derivatives such as selenite (SeO_3^{-2}) and selenate (SeO_4^{2}) are widely used in industries, e.g. glass manufacturing and electronic industries. Selenium can enter in the environment through sewage sludge, fly ash from coal-fired power plants, oil refineries and mining of metal ores (Tandukar et al. 2009). Selenite is more toxic than selenate to aquatic invertebrates and fishes (Hamilton 2004; Lemly 1997), and it accumulates in aquatic plants thus causing bioaccumulation in higher organisms which can cause both acute and chronic toxicity in aquatic organisms (Rovira et al. 2008). The application of MFC technology in reduction of selenium and production of electricity by using selenium-containing waste was investigated by Catal et al. (2009).

9. **Nitrate:** The use of nitrate-based fertilizers and animal waste has increased the presence of nitrate in water. Nitrate (nontoxic) can be transformed to nitrite (NO^{2-}) after entering human body, which can cause "blue baby syndrome" normally observed in infants, or it can be converted to N-nitroso compounds which are carcinogenic into humans. Some of the treatment methods employed include electrochemical treatment, ion exchange (IE), reverse osmosis (RO), electrodialysis (ED) and heterogeneous catalysis (HC) (Park and Yoo 2009). As the methods employed are expensive, the use of MFC for removal of nitrate has gained importance due to the feasibility of the process. Few workers have employed a metal catalyst (Polatides and Kyriacou 2005) or microorganisms as catalysts on cathode electrode (He and Angenent 2006). In few studies, electrochemical denitrification process to remove nitrate ions was employed at cathode chamber of bio-electrochemical denitrification system (Kondaveeti and Min 2013).
10. **Marine Sediments Rich in Acetate:** Marine sediments rich in acetate have been used as a substrate in MFC. Acetate presence helps in providing inertness towards alternative microbial conversions, fermentations and methanogenesis at room temperature (Sun et al. 2009; Aelterman 2009). Acetate can act as a carbon source to induce electroactive bacteria, and it is an end product in several metabolic pathways (including the Entner–Doudoroff pathway for glucose metabolism) for higher-order carbon sources (Biffinger et al. 2008; Bond and Lovley 2005). When Chae et al. 2009 compared the performance of four different substrates in terms of CE and power output, acetate-fed MFC showed the highest CE, followed by butyrate, propionate and glucose. According to Liu et al. (2009), acetate-based MFC achieved more electric power and external load resistance compared to those based on consortia induced by a protein-rich wastewater.
11. **Animal Wastes:** Animal wastes such as poultry dropping, cow dung, human faeces and urine are reportedly used as substrate for the bioelectricity generation. Barbosa et al. (2017) and Jimenez et al. (2016) suggested that animal waste can be considered as potential substrate for generation of electricity due its high daily production, high COD, high nutrient (N and P) concentrations and high solution conductivity. Based on these observation, Ieropoulos et al. (2016) suggested that the treatment of urine can be performed in MFCs whilst generating electricity for low-power devices, such as light-emitting diode (LED) lights or sensors. Ieropoulos et al. (2016) ran field trials by connecting a stack of MFC to public urinals and used it for lighting a room. This was an attempt for taking the technology beyond laboratory, and it helped understand how fast the technology evolves and is being able to address the issue of public hygiene, sanitation, direct treatment of human waste and energy generation. Melhuish et al. (2006) designed a robot "EcoBot-II", which was powered by on-board MFC with oxygen cathodes, it simultaneously utilized unrefined insect biomass and converted into useful energy. Kretzschmar et al. (2017) provided an experimental proof of concept using human faeces as substrate in MFC for power generation.
12. **Petroleum Hydrocarbons:** MFC can be a cost-effective and eco-friendly approach for bioremediation of petroleum hydrocarbons as these hydrocarbons

act as substrate for microbes. The MFC technique was used for detoxification of soil decontaminated with petroleum at an increased rate and is energy sufficient. A U-tube MFC designed by where they observed enhanced degradation rate of petroleum hydrocarbons by 120% high charge output. Morris et al. (2009) used MFC for enhancing biodegradation of diesel. They demonstrated that in MFC diesel removal rate was increased by four times using MFC because the electrode served as an electron acceptor. MFC utilizing diesel (v/v 1%) as sole carbon source also resulted in high-power density and current density (Cheng et al. 2017). Therefore, MFC technology can be effectively used for detoxification and enhancing biodegradation of polluted soil/wastewater containing petroleum contaminants in anoxic environments, thus, eradicating the need to adjust terminal electron acceptors such as oxygen.

11.6 Types of Microbial Fuel Cell for Bioremediation of Pollutants

11.6.1 Anaerobic Microbial Fuel Cell (ANMFC)

ANMFC require oxygen that may cause loss of electrons and lead to increase in energy demand to carry out the process (Abbasi et al. 2016). Therefore, anaerobic MFC can be an environmental friendly and cost-effective alternative for simultaneous electricity generation and bioremediation. Wastewaters generated from brewery industries (Akman et al. 2013; Pant et al. 2010); distillery and domestic wastewater (Jiang et al. 2011); pharmaceutical industry (Velvizhi and Venkata Mohan 2012), petrochemical, vegetable oil, food industry and animal carcass wastewater (Li et al. 2013); textile (Solanki et al. 2013) swine wastewater (Zhuang et al. 2012); and municipal wastewater (Zhang et al. 2013) could be treated using anaerobic MFC. Abbasi et al. (2016) designed an anaerobic MFC where they utilized wastewater samples from vegetable oil industries, metal works, glass and marble industries as substrate. This process has significant effect on wastewater treatment efficiency for COD in range of 85–90% at 96 h of hydraulic retention time (HRT). The coulombic efficiency of 5184.7 C with maximum voltage of 890 mV was generated when vegetable oil industries discharge was treated in MFC. A positive significant co-relation was observed between COD concentration and voltage generated.

11.6.2 Sediment Microbial Fuel Cell (SMFC)

SMFC comprises of an anode buried in sediment and a cathode in an oxygen-rich water (Rezaei et al. 2007; Xia et al. 2015). The SMFC have been successfully tested in removal of persistent organic compounds such as polycyclic aromatic hydrocarbons (PAHs), xenobiotic compounds and pesticides, etc. along with electricity

generation (Sherafatmand and Ng 2015). Similarly Yu et al. (2017) reported maximum power density together with removal of anthracene, phenanthrene and pyrene by a closed SMFC utilizing PAH-polluted soils. They further studied the influence of electrode interval and role of microbial community in electricity generation and removal of PAHs. The decrease in electrode interval resulted in efficient electricity generation and removal of PAHs. The SMFC was dominated by the genus of *Geobacter* and enriched in electrogenic bacteria at the anode surface. The growth of certain microbes (except electrogenic bacteria in the soil) was improved by electrical stimulation. Similarly Xu et al. (2017) found that genus *Geobacter* are predominately found in SMFC and more electrogenic bacteria are found attached in biofilm of anode. They also demonstrated that addition of Fe (III) oxide in SMFC enhances the removal efficiencies for organic pollutants. Xia et al. (2015) reported enhanced biodegradation of organic chemicals of high polarity by operating a SMFC in heavily contaminated sediment and analysing its global organic chemical degradation profile. The study showed that SMFC prefers to stimulate the degradation of organic chemicals with higher polarity. Cao et al. (2015) constructed a SMFC in the top soil contaminated with hexachlorobenzene (HCB), a toxic refractory organic pesticide. Under anaerobic condition in the soil MFC, HCB was degraded via the reductive dechlorination pathway and an existence of the anode promoted electrogenic bacteria provided more electrons that subsequently improved electricity generation.

11.6.3 Benthic Microbial Fuel Cells (BMFC)

BMFC consists of an anode present in anoxic benthic sediment and a cathode in oxic overlying water which is further connected using an external electric circuit. BMFC are different from SMFCs as the latter does not require in situ deployed in real water bodies (Li and Yu 2015; Holmes et al. 2004). BMFC has emerged as sustainable and efficient technology for cleaning up of contaminated sediments and simultaneous energy generation. The bioremediation of the contaminated sediments is performed by utilizing the natural metabolic activities of microbes in detoxification, decomposition or immobilization of environmental contaminants present in the sediments. BMFC is at initial stage but have shown many potential benefits such as accelerated decontamination, relatively easy deployment, self-sustained operation and control and environmental benignity. The relatively lower efficiency, limitations in respect of system design, electrode selection, microbial control and selection of deployment environment severely limit its application (Li and Yu 2015).

11.6.4 Enzyme-Based Microbial Fuel Cells (EBC)

The fuel cells are of two types first employing living cells known as microbial biofuel cells and the other utilizing enzymes and referred as enzymatic biofuel cells (EBC). The microbial biofuel cells have long lifetimes ~5 years (Moon et al. 2006;

Kim et al. 2003) and can completely oxidize simple sugars to carbon dioxide (Bond et al. 2002). However they have low-power densities in W/cm^2 per unit electrode surface area which is due to slow transport across cellular membranes (Palmore and Whitesides 1994). By contrast, enzymatic biofuel cells typically possess higher-power densities (although still lower than conventional fuel cells) and can partially oxidize the fuel and have limited lifetimes (typically 7–10 days) owing to the sensitive nature of the enzyme (Kim et al. 2006; Barton et al. 2004) and to eliminate the need for a membrane separator. One of the most significant advances in enzymatic biofuel cells is the development of biocathodes and bioanodes that employ direct electron transfer (DET) instead of mediated electron transfer (MET). The DET is more preferred over the MET (Moore et al. 2004). The second method is the immobilization of the enzyme which has helped enzyme increase in the active lifetime of the enzymes (Topcagic et al. 2004). Recently, the active lifetimes have been extended beyond 1 year by encapsulation in micellar polymers (Akers et al. 2005; Moore et al. 2004; Topcagic et al. 2004). The EBC can be used extensively in biodegradation of toxic organic pollutant such as azo dyes, polyaromatic hydrocarbons, etc. The EBC have many disadvantages over traditional fuel cells and primary batteries; they remain limited by short lifetimes, catalytic inefficiencies, low fuel utilization and low-power densities (Minteer et al. 2007). However working solutions to short lifetimes and catalytic inefficiencies have been introduced, but advances in improved fuel utilization and power density are required.

11.6.5 Air-Breathing Cathode-Based Microbial Fuel Cells (ABC-MFC)

An air-breathing cathode consists of electrode substrate, catalyst layer and air diffusion layer (Wang et al. 2017). Single-chamber, air-breathing MFC with a flexible graphite sheet as the anode was designed by Sonawane et al. (2017), in which they used landfill leachate as substrate. They obtained open-circuit voltage (OCV) of 1.29 V, which is the highest reported OCV in the literature till date by utilizing landfill leachate as substrate. The reactor also resulted in generation of maximum cathode area-specific power density of 1513 mW m^{-2}. Jimenez et al. (2016) demonstrated the use of gas-diffusion air-breathing cathode-based MFC for generation of electricity utilizing urine as substrate. As discussed earlier cathode is site of ORR, and it is one of the limiting factor for MFC performance; therefore Kodali et al. (2017) used Mn-, Fe-, Co- and Ni-containing platinum group metal-free catalysts along with aminoantipyrine, AAPyr precursor for enhanced electricity generation. With increase in solution conductivity, it was observed that Fe-AAPyr was found to be the most suitable catalyst–precursor combination in air-breathing cathode-based MFC for enhanced electricity with high-power density.

11.6.6 Constructed Wetland Microbial Fuel Cells (CW-MFC)

Constructed wetlands (CWs) and MFCs are two different technologies, nevertheless very compatible technologies (Liu et al. 2013). Both the techniques are dependent on the actions of bacteria to remove pollutants from wastewater, but MFC has added advantage being energy generator. Therefore, the two techniques are combined in such a way that anode is buried in the anaerobic condition of constructed wetland and the cathode exposed to oxygen in the plant rhizosphere and collectively called as CW-MFC. The low oxygen availability at anode and higher redox gradient are an essential feature for generating electric current in CW-MFCs. The upflow regime of CW-MFC is such that it reduces the availability of dissolved oxygen (DO) at the anode whilst ensuring its maximum availability in the cathode region and also provide sufficient redox profile for MFC integration (Corbella et al. 2014; Fang et al. 2013). This natural redox gradient comes at cost of ohmic resistance of 120–500 (Doherty et al. 2015b; Villaseñor et al. 2013). A multi-electrode MFC with a separator electrode assembly was suggested by Ahn and Logan (2012) that help in reducing the ohmic resistance to 33 Ω. Glass wool separators can be used in construction of CW-MFC in order to minimize electrode spacing. This technology is excessively used in wastewater treatment with the aim of improving the wastewater treatment capacity of wetlands whilst simultaneously producing electrical power (Doherty et al. 2015a).

11.6.7 Thermophilic Microbial Fuel Cells (TMFC)

MFC used for the generation of electricity usually operated at ambient or mesophilic temperatures. Carver et al. (2011) suggested that thermophilic systems have potential for increased microbial activity rates on the anode that can subsequently enhance electricity generation. Air-cathode single chambers and two-chamber designs are usually used as for electricity generation, but the real role is played by thermophilic microbes associated with anode under anaerobic condition. Carver et al. (2011) described a thermophilic MFC design maintained at 57 °C with an anaerobic and thermophilic consortium. This suggested design minimized evaporation and associated microbes respired with glucose to generate a power density of 375 mW m^{-2} after 590 h. Voltage data and polarization showed that the design can work in both batch and continuous mode. Dai et al. (2017) constructed a two-chamber TMFC and utilized ethanol as an electron donor. They obtained an open-circuit potential of approximately 650 mV, maximum voltage of 550 mV and maximum power density of 437 mW m^{-2}, and the coulombic efficiency was 20.5 ± 6.0. They also analysed the microbial dynamics by high-throughput sequencing and 16S rRNA clone library sequencing; *Firmicutes* bacteria accounted for 90.9% of all bacteria associated with TMFC biofilm. The development of TMFC-involved biotechnologies will be beneficial for the wastewater treatment, production of valuable chemicals and generation of energy at the same time (Dai et al. 2017).

11.7 Commercial Application of MFC and Economic Feasibility

MFC technology has come a long way, and over the last decade, significant scientific and technological development had brought the MFC to the point of becoming commercialized technology. During commercialization of a technology, various market forces decide its success such as cheap, cost-effective, environmental friendly, larger consumer base and above all profitability. For assessing the profitability of the MFC and checking whether the technology is ready to enter market for commercial energy generation and waste treatment, Trapero et al. (2017) used classical evaluation criteria for investment decisions such as the net present value (NPV) and the internal rate of return (IRR). They have presented an economic assessment of a MFC in a juice processing plant where maximum power density of the cell using two different MFC cases, i.e. cathodes with and without Pt, was studied. The performance was then compared to the conventional activated sludge process. Three different scenarios, optimistic, pessimistic and most likely scenarios, were analysed. They also performed study to find the important factors and design influencing MFC performance. By a sensitivity analysis of the electrode area, and the annual growth rate of the electricity pricing, it was revealed that the electrode area parameter is the most influential factor. The results of the study clearly showed that the implementation of MFC is a favourable substitute to the use of classical aerated activated sludge, and it has potential economic benefits. Whilst designing for a current state-of-the-art MFC keeping in view the future challenges, research directions may be focused on selection of microbial consortium, along with operational consistency and stability of the developed system.

11.8 Future Prospects and Directions

MFC is a promising technique with great potential as it is an environment-friendly tool which does not involve the use of renewable natural resource and emission of pollution. In recent times substantial technological advancement and improvement are observed for waste/wastewater treatments in MFCs. MFCs have multiple applications which include energy generation, waste treatment and production of chemicals, sensors for pollution level in water/soil, etc. It can generate energy out of waste without any external/additional energy. This property will enable MFC technology to be used in remote areas for energy generation and waste treatment e.g., maintaining sanitation and hygiene, catering the need of poor people for affordable electricity which is of particular interest for countries and regions of the developing world. However, several aspects need to be addressed in order to improve the MFCs performance such as power output and efficiency, material costs for electrodes and separators, microbial consortium, suitable MFC design for treatment of specific waste and the optimization of the bio-electrochemical reactions. Keeping in mind

the funding/development perspective, MFCs can be considered as nascent technology, but recent involvement of government laboratories, NGOs, philanthropist and business houses has provided helps and support for continuing research into MFC technology with a hope to find solutions to global environmental problems.

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