Advances in the development of electrode materials for improving the reactor kinetics in microbial fuel cells

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Abstract

Microbial fuel cells (MFCs) are an emerging technology for converting organic waste into electricity, thus providing potential solution to energy crises along with eco-friendly wastewater treatment. The electrode properties and biocatalysts are the major factors affecting electricity production in MFC. The electrons generated during microbial metabolism are captured by the anode and transferred towards the cathode via an external circuit, causing the flow of electricity. This flow of electrons is greatly influenced by the electrode properties and thus, much effort has been made towards electrode modification to improve the MFC performance. Different semiconductors, nanostructured metal oxides and their composite materials have been used to modify the anode as they possess high specific surface area, good biocompatibility, chemical stability and conductive properties. The cathode materials have also been modified using metals like platinum and nano-composites for increasing the redox potential, electrical conductivity and surface area. Therefore, this paper reviews the recent developments in the modification of electrodes towards improving the power generation capacity of MFCs.

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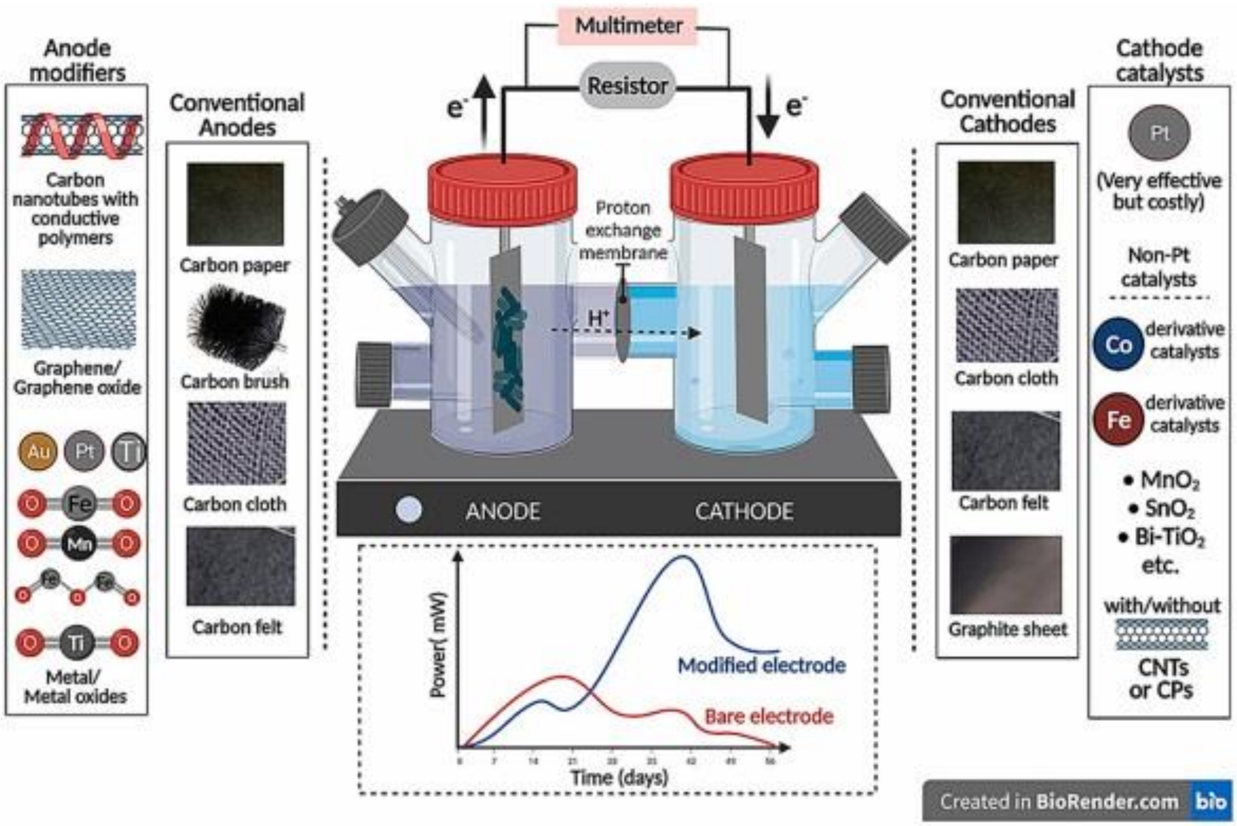
Graphical abstract



Keywords

Microbial fuel cell, Conductive polymers, Carbon nanotubes, Metal oxides, Power density

Abbreviations

CC: Carbon Cloth
CNT: Carbon Nanotube
Co_3O_4: Cobalt oxide
Cu_2O: Copper oxide
DET: Direct Electron Transfer
FTIR: Fourier-transform infrared spectroscopy
GS: Graphene Sheet
MFCs: Microbial Fuel Cells
MWCNT: Multiwalled carbon nanotubes
ORR: Oxygen Reduction Reaction
PDHC: Polyaniline dopamine hybrid composite
CF: Carbon Felt
COD: Chemical oxygen demand
$CoMnO_4$: Cobalt Manganese oxide
DCMFC: Dual Chamber MFC
Fe_2O_3: Iron (III) oxide
GO: Graphene Oxide
MET: Mediated Electron Transfer
MnO_2: Manganese oxide
$NiWO_4$: Nickel/tungsten oxide
PANI: Polyaniline
PEM: Proton exchange membrane

PTFE: Polytetrafluoroethylene
rGO: reduced Graphene Oxide
SCMFC: Single Chambered Microbial Fuel Cell
SMFC: Sediment Microbial Fuel Cell
SSFF: Stainless Steel Fiber Felt
SSWM: Stainless Steel Wire Mesh
XPS: X-ray photoelectron spectroscopy
XRD: X-ray Diffraction

1. Introduction

The global energy crisis has been exacerbated by the world's ever-increasing demand for fossil fuels. According to the “Energy Statistics 2019” by the Ministry of Statistics and Program Implementation, Government of India, the total electricity generation in India from conventional and non-conventional energy sources during 2017-2018 was ~ 1,486,493 GWh. The total output from non-conventional sources was 183,000 GWh (Irfan et al., 2019). The estimated consumption of electricity has increased from 553,995 GWh during 2008-2009 to 1,130,244 GWh during 2017-2018, accounting for a 7.4% annual growth rate (Shetty et al., 2020). Eco-friendly methods should be used to meet this increased demand for energy, which will not further harm the environment.

Along with the energy crisis, the world's lack of clean water is becoming a major concern as the world's population grows and technology and the economy advance. According to the United Nations' World Water Development Report (Rajesh et al., 2020), the global demand for water is increasing at a 1% annual rate and is expected to continue growing in the future. Water is a valuable resource and it is subjected to a variety of forms of contamination and degradation as a result of human interference. It has a significant impact on human health and the environment, and wastewater treatment has become a major concern for everyone. The currently used wastewater treatment processes include physical, chemical and biological treatment processes (Hreiz et al., 2015). However, some of the technologies such as the membrane (bio)technology, ion exchange, and electrolytic reduction are costly due to the extensive infrastructure and operation required (Feng et al., 2020). Some of the technologies such as chemical precipitation, coagulation or flocculation produce secondary pollutants and are therefore unfriendly to the ecosystem.

To address both of these issues, the application of advanced technology that can use the organic component of waste materials to generate energy is required. The problem of energy and water crises, as well as the depletion of fossil fuels, drives intense efforts to develop more sustainable technology, and microbial fuel cells (MFCs) are one of them (Logan, 2010). Biocatalysts are used in MFC to generate electricity while degrading the organic components or wastes present in wastewater (Logan et al., 2019). Several researchers have reported the incorporation of MFCs into wastewater treatment plants in order to generate bioelectricity while also treating contaminated wastewater in an environmentally friendly manner (Dong et al., 2015; Kumar et al., 2019). However, due to economic constraints and its low power output, the feasibility of applying MFC at the field scale is limited (Jadhav et al., 2021). As a result, more research is being conducted to develop low-cost electrode materials and catalysts in order to enable the use of MFC in full-scale applications (Mustakeem, 2015).

Advanced research in MFC design, modification of electrode/substrate material and biocatalyst have been made in recent years (Khudzari et al., 2018). In recent reviews, Sonawane et al. (2017), Hindatu et al. (2017) and Cai et al. (2020) have highlighted the advantages of anode material modifications to improve MFCs performance. Palanisamy et al. (2019) reviewed the electrode (anode and cathode) materials with varying dimensional and structural compositions and integrations that can be used in MFCs. The authors concluded that different categories of electrode materials improved the long-term operation of MFC due to its cost-effectiveness, biocompatibility and high stability. Besides, carbon-based, metal-based materials, and plant-derived materials have also been applied in the field of MFC research (Jadhav et al., 2021). For practical applications, the economic viability, ease of availability, high electrical conductivity, biocompatibility (anode), and enhanced oxidation reduction reaction (ORR) rate potential are all desirable properties of a material to be used as an electrode material (cathode). To achieve high power generation, electrodes are frequently modified with a suitable catalyst. Carbon-based nanomaterials (graphene, carbon nanotubes), metals (platinum, iron), metal oxides (manganese oxide, nickel oxide), and conductive polymers (polyaniline) are among the potential catalysts (Mustakeem, 2015). This review focuses primarily on the various materials used as electrodes and surface modifications for improving the electrode performance and power generation in MFCs.

2. Microbial fuel cell (MFC)

MFC is a bio-electrochemical system based on redox reaction that may be configured as a single (SCMFC) or dual chambered system (DCMFC). In a MFC, the organic matter present in either wastewater or substrates added externally, is oxidized by microbial activity to generate electrons and protons in absence of oxygen (anaerobic condition in anodic chamber), CO_2 is produced as an oxidation product. Unlike in a direct combustion process, the electrons are transferred to anode and then transported to the cathode through an external circuit (Choudhury et al., 2020). In DCMFC, the protons generated, travel through proton exchange membrane (PEM) or a salt bridge and enter the cathodic compartment (aerobic) where they combine with oxygen to form water as shown in equations (1) and (2) , respectively (Fig. 1).

Anode reaction: $C_{12}H_{22}O_{11} + 13H_2O \rightarrow 12CO_2 + 48H^+ + 48e^-$ (anaerobic chamber) (1)

Cathode reaction: $4H^+ + O_2 + 4e^- \rightarrow 2H_2O$ (aerobic chamber) (2)

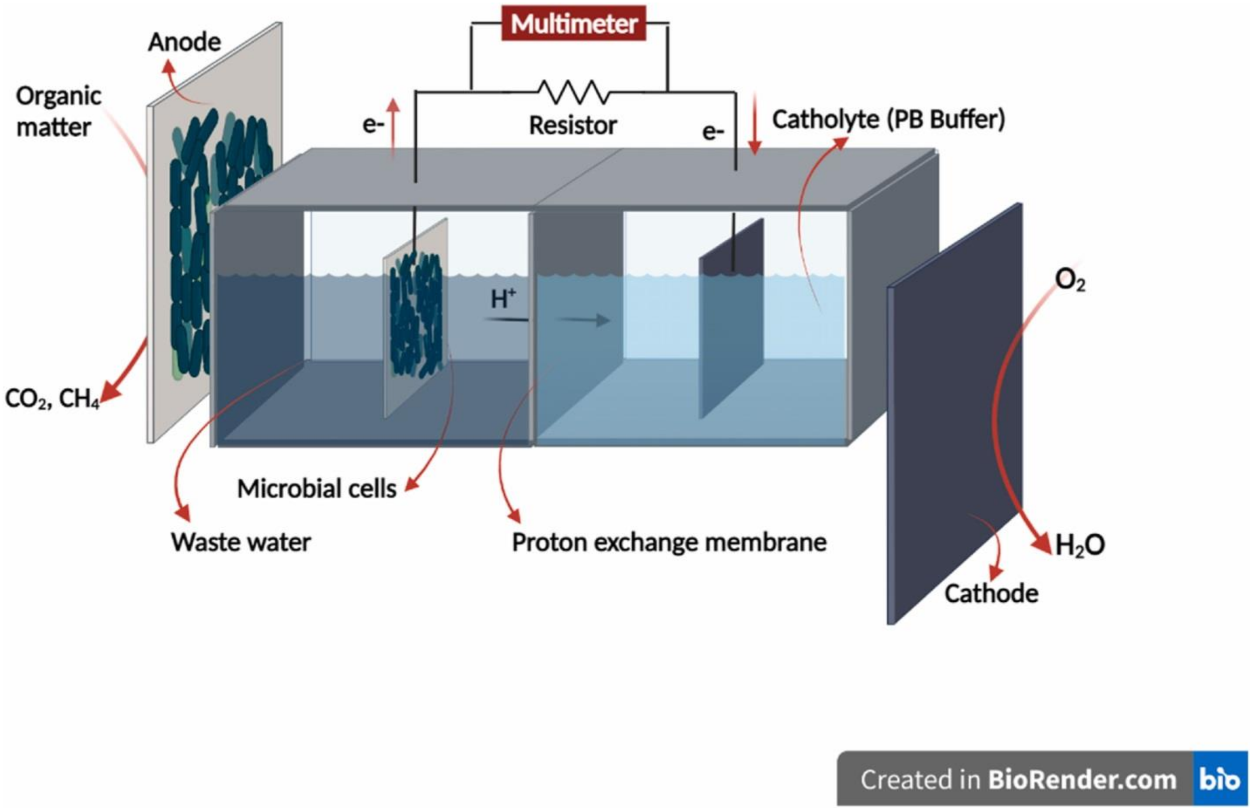


Fig. 1. Schematic of a typical dual-chambered microbial fuel cell (DCMFC).

3. Electrode material

Choice of material to be used as electrode is critical for power generation in a MFC with respect to electron transfer, electrochemical properties and microbial adhesion. Most commonly used electrode materials include different carbon-based materials such as carbon cloth, carbon paper, carbon fiber, carbon felt, carbon nanotube-based composites, and graphene based nano-composites. For the field implementation of MFC technology, material cost must be reduced and power densities must be maximized. Additionally, cathode materials should have high redox potential.

Though the criteria for material selection for anode and cathode differ, in general both should possess the following properties:

Surface area: Surface area and porosity have a significant constrain on electricity generation in MFC. Internal resistance is the major cause of ohmic losses, which reduced overall performance of a MFC. Internal resistance can be minimized by increasing the effective surface area, while keeping the volume constant. Moreover, high surface area enhances electrode kinetics by providing more reactions sites (Di Lorenzo et al., 2010).

*Electrochemical properties***:** Microbial metabolism release electrons in anodic chamber which are taken up by anode and travel along an external circuit, to cathode. High electrical conductivity and low interfacial impedance, facilitates effective flow of electrons through the circuit. Cathode should also possess ionic conductivity to facilitate triple phase boundary reaction (Sun et al., 2015).

*Material stability and cost effectiveness***:** The reducing and oxidizing environment, microbial growth and attachment (on anode) in an MFC often lead to decomposition of the electrode materials. Thus, the electrode material should be stable and durable so that it can provide long term performance with minimum fouling or degradation. Furthermore, electrode material should be low-cost, easily available and sustainable, for economic viability of the technology at large scale (Wang et al., 2013).

*Biocompatibility***:** Anode materials should be highly biocompatible, superior biocompatible material will facilitate effective biofilm formation by bacterial adhesion thereby increasing overall MFC performance (Tao et al., 2016).

4. Anode electrodes

Biocompatibility, convenient microbial adhesion, electrochemical efficiency and electron transfer mechanism from microbial cell to the electron acceptor are key characteristics for the selection anode material (Dumitru and Scott, 2016; Loubna et al., 2019). Hence, the materials from which anode electrodes are constructed should have some unique properties that help to improve the attachment of electro active microorganisms to the electrode surface as well as current collection. Ideal anode materials for MFCs should have good intrinsic biocompatibility for microbial inoculums, growth, attachment, and excellent conductivity for e^- transfer. Other properties, includes large surface area, highly porous structure, long-term stability, and favorable surface modification, are also utmost desirable.

Therefore, one of the major objectives in the MFC research is to design and synthesize low-cost advance electrode materials with increased efficiency, and improved durability. To improve the electron transfer mechanisms in MFC, there are various strategies applied such as, application of bioactive redox compounds, modification of anode etc. (Nosek et al., 2020; Zhao et al., 2010).

4.1. Conventional anode electrodes

Various carbon-based materials, including carbon paper, carbon brush, carbon cloth, graphite brush, have commonly been used as anodes in MFCs. However, due to their restricted biocompatibility, very limited active reaction site, and hydrophobicity, biofilm formation is limited thereby limiting the resulting power density. Apart from carbon based material such as

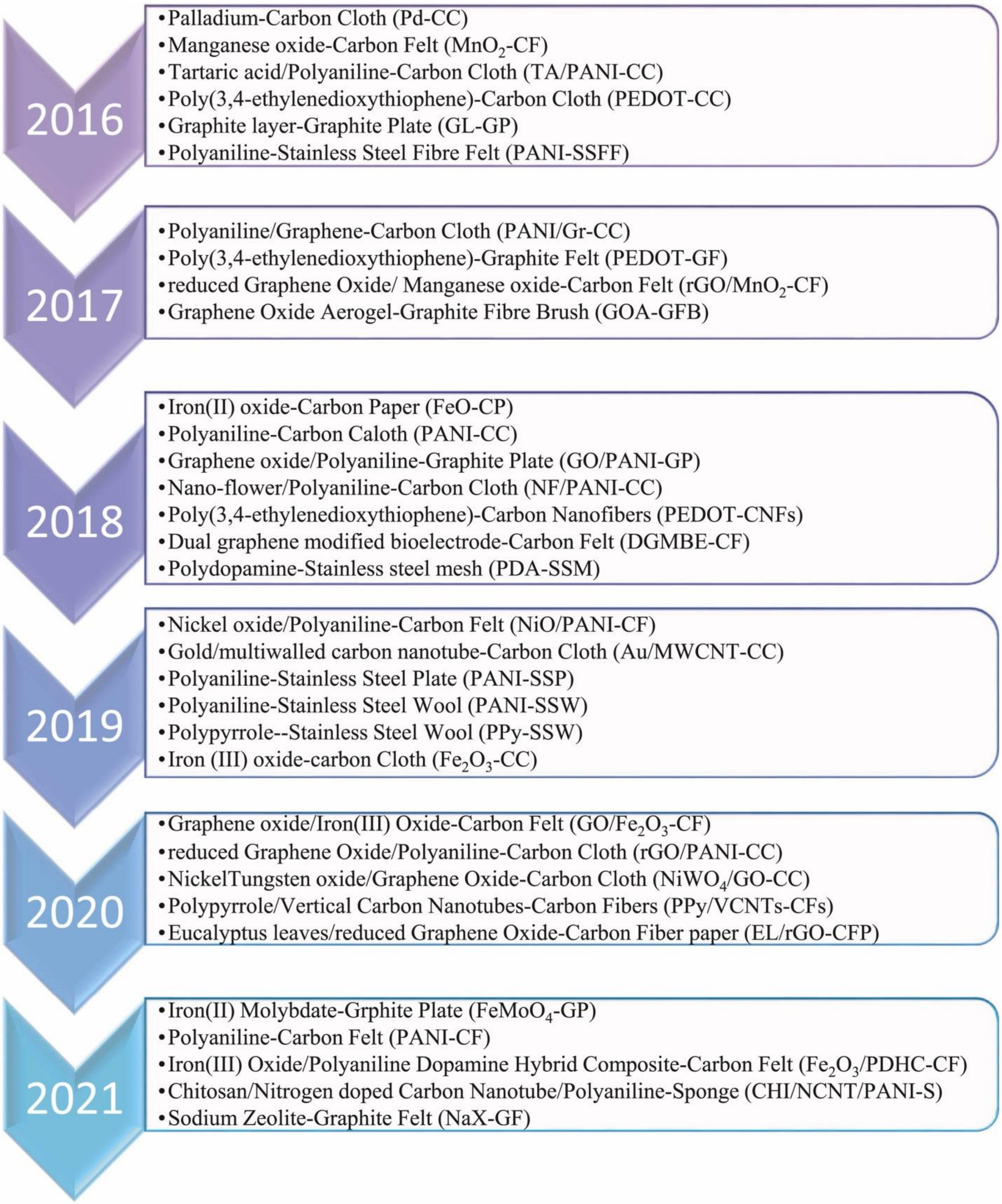


Fig. 2. Timeline depicting the development of anode electrodes.

Table 1. Modification of anode and its performance in term of power density and wastewater treatment.

MFC type	Volume of reactor (ml)	Anode material	Anode modifier	Power density (control)	Power density (modified)	No. of fold increase in power density	COD removal (%)	References
SCMFC	28	Carbon cloth	Graphene + PAni	454 ± 47 mW/m^2	884 ± 96 mW/m^2	1.94	88.6 ± 8.6	Huang et al. (2016)
SCMFC	28	Carbon cloth	PAni	454 ± 47 mW/m^2	589 ± 38 mW/m^2	1.29	81.6 ± 4.9	Huang et al. (2016)
DCMFC	500	Carbon cloth	NCP (Nickel oxide/CNT/PAni)	1.5 mW/m^2	2.5 mW/m^2	1.67	–	Nourbakhsh et al. (2017)
DCMFC	250	Carbon felt	PAni	166 mW/m^2	216 mW/m^2	1.30	67.34 ± 1.89	Rajesh et al. (2020)
SCMFC	288	Carbon cloth	Titanium suboxide/Graphene/PAni	163.2 mW/m^2	2073 mW/m^2	12.7	–	Li et al. (2020b)
DCMFC	250	Carbon felt	Graphene/Fe_2O_3	129 ± 4 mW/m^2	334 ± 4 mW/m^2	2.59	–	Fu et al. (2020)
DCMFC	28	Carbon cloth	PDA-rGO	337 ± 15 mW/m^2	2047 ± 58 mW/m^2	6.1	–	Li et al. (2020a)
DCMFC	28	Carbon cloth	rGO	337 ± 15 mW/m^2	1062 ± 58 mW/m^2	3.15	–	Li et al. (2020a)
DCMFC	28	Carbon cloth	PDA	337 ± 15 mW/m^2	927 ± 45 mW/m^2	2.75	–	Li et al. (2020a)
SCMFC	28	Carbon cloth	Graphene	454 ± 47 mW/m^2	634 ± 78 mW/m^2	1.39	84.5 ± 9.4	Huang et al. (2016)
DCMFC	120	Carbon cloth	PPy-MnO_2	598.4 ± 31.5 mW/m^2	2139.7 ± 67.5 mW/m^2	3.58	–	Zhao et al. (2020)
DCMFC	120	Carbon cloth	MnO_2	598.4 ± 31.5 mW/m^2	1606.7 ± 63.2 mW/m^2	2.69	–	Zhao et al. (2020)
SCMFC	40	Carbon felt	Ru/Fe	0.223 W/m^2	0.6 W/m^2	2.69	–	Qiu et al. (2020)
DCMFC	250	Graphite plate	$FeMoO_4$	75.8 ± 1.5 mW/m^2	106.2 ± 2 mW/m^2	1.40	79.8 ± 1.5	Mohamed et al. (2020)
DCMFC	100	Carbon felt	MnO_2	38 mW/m^2	47 mW/m^2	1.24	–	Nandy et al. (2020)
DCMFC	300	Carbon cloth	Fe_2O_3	0.236 W/m^2	0.285 W/m^2	1.21	69.2	Meicong et al. (2020)
SCMFC	84	Graphite rod	Fe	40 mW/m^2	80 mW/m^2	2	–	Sayed et al. (2020)
DCMFC	250	Carbon paper	Cu doped FeO	123.5 mW/m^2	161.5 mW/m^2	1.31	75	Sekar et al. (2019)
DCMFC	50	Carbon felt	WO_3	0.49 ± 0.11 W/m^3	3.21 ± 0.22 W/m^3	6.55	87 ± 3	Das and Ghangrekar (2020)
Benthic MFC (BMFC)	1000	Carbon felt	Polypyrrole coated Fe_2O_3 (FP)	69.19 mW/m^2	170.45 mW/m^2	2.46	–	Prakash et al. (2020)
Benthic MFC (BMFC)	1000	Carbon felt	Polypyrrole coated MnO_2–Fe_2O_3 (MFP)	69.19 mW/m^2	117.29 mW/m^2	1.69	–	Prakash et al. (2020)
Benthic MFC (BMFC)	1000	Carbon felt	Polypyrrole coated MnO_2 (MP)	69.19 mW/m^2	90.54 mW/m^2	1.31	–	Prakash et al. (2020)

(PAni: Polyaniline, CNT: Carbon nanotube, Fe_2O_3: Ferric oxide, PDA: Polydopamine, rGO: reduced graphene oxide, PPy: Polypyrrole, MnO_2: Manganese oxide, $FeMoO_4$: Iron (II) molybdate, Cu: Copper, Fe: Iron, Ru: Ruthenium, WO_3: Tungsten oxide).

carbon cloth, carbon paper, carbon felt, graphite brush, graphite sheet (GS), graphite rod (GR), granular activated carbon (GAC), reticulated glass carbon (RGC), carbon nanotubes (CNTs), graphene, and stainless steel mesh (SSM), are commonly used anode materials for MFCs (Wang et al., 2009; Wei et al., 2011; Wen et al., 2013). Anode is the conductor of electron and carbon materials which are used as anode has little electro catalytic activity which limits their performance; Thus, modification of carbon materials is the main approach to improve their conductivity (Wang et al., 2015). Semiconductors as well as their composite material have been used to modify the anodes in order to improve their electrochemical properties (Narayanasamy and Jayaprakash, 2020). Nanostructured metal oxides are also commonly used for anode modification as they possess high surface area, good biocompatibility, chemical stability and electro-conductive properties (Liu et al., 2020a).

4.2. Modification of anode electrodes

As discussed previously, modification of anode is primarily done to improve biocompatibility and electrochemical efficiency. Semiconductors, nanomaterials (metal oxides, carbon nanostructures), and conductive polymers have been extensively explored as anode modifiers due to their excellent biocompatibility, chemical stability, good electron conductivity and high specific surface area. Significant developments made towards development of anode materials over the past six years are illustrated in Fig. 2. Table 1 represents the overview of MFC performance using modified anodes.

To simplify the discussion, anode modifications have been classified into three broad categories

I. Modifications using Carbon nanotube (CNTs) and conductive polymer (CPs), **ii.** Graphene based modifications, **iii.** Modifications using metal/metal oxides which are discussed below.

4.2.1. Anode modification using carbon nanotube (CNTs) and conductive polymers (CPs)

CNTs have exhibited very promising properties for uses as electrode material because of their large specific surface area, extraordinarily high conductivity and mechanical flexibility. However, due to their inhibitory or even toxic effect on microorganism, they are rarely used in MFC in unmodified form. Hence, various modifications on CNT are done to make it biocompatible. Qiao et al. (2007) reported that anode modification with CNT and polyaniline composites not only reduces their cellular toxicity but also improves the electrocatalytic activity. They investigated the electrochemical activity of modified electrode through electrochemical impedance spectroscopy as well as charge discharge experiments and reported 42 mW/m^2 of power density which was 1.2-fold higher than that of unmodified electrode. The MWCNT modification via a facile microwave aided unzipping process was used to synthesize core–shell structured multiwalled carbon nanotube–graphene oxide nanoribbons (MWCNT@GONR) with oxygen containing functional groups (Fig. 3) (Sun et al., 2011). The MWCNT@GONR were further modified using ammonia treatment to incorporate the nitrogen containing functional groups (*N*-MWCNT@GONR) (Liu et al., 2020b) and their performance as anode was evaluated in 40 ml-SCMFCs using *Escherichia coli* (Liu et al., 2020b). The power density achieved by using the unmodified MWCNT coated carbon cloth anode was 970 mW/m^2, whereas the power density achieved with MWCNT@GONR and *N*-MWCNT@GONR coated carbon cloth anodes reached 3291 and 3444 mW/m^2, respectively, resulting primarily from the improved biofilm adhesion. The microwave treatment of MWCNT resulted in the formation of a core-shell structure with a nanotube core surrounded by a planar graphene sheet shell, thereby increasing the specific surface area from 18.1 to 49 m^2/g. However, this value was further increased to 72.4 m^2/g by ammonia treatment due to the formation of micropores and macropores structures on the surface of *N*-MWCNT@GONR (Liu et al., 2020b).

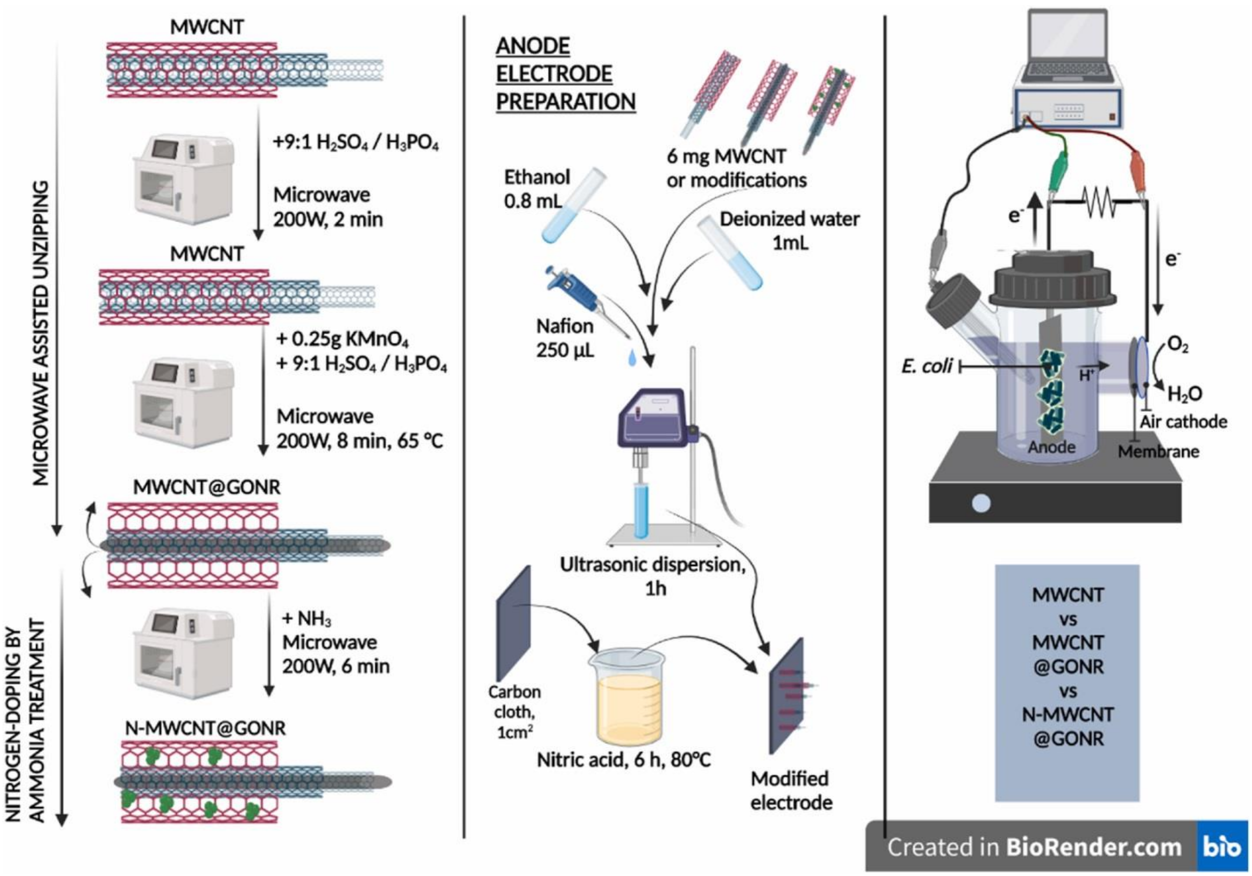


Fig. 3. Multiwalled carbon nanotubes (MWCNT) and its modifications for application in MFCs. Note: MWCNT@GONR: multiwalled carbon nanotube–graphene oxide nanoribbon, *N*-MWCNT@GONR: *N*-doped variant [adapted from the studies by Liu et al., 2020b and Sun et al., 2011].

Conductive polymers are organic polymers that have potential of electricity conduction. They can be metallic conductors or semiconductors. Polyaniline (PANI) is a conductive polymer and has desirable use for MFC anode preparation due to its high electrical conductivity, facile processability, hydrophilicity, good biocompatibility and high stability (Qiao et al., 2007). The positive charge on outer surface of PANI facilitates its interaction with negatively charged bacterial cell membrane that leads to high bacterial density on its surface. Several researchers have combined PANI with metal oxide or graphene or graphene oxide to improve its specified surface area and reluctant conductivity. Metal oxides accelerate the electron transfer between anode and bacterial cells, thus improving the efficiency of MFC. Along with graphene and/or graphene oxide, PANI improves biocompatibility, thus influencing the bacterial adhesion capacity and extracellular electron transfer (EET) efficiency. Khilari et al. (2015) used composite of PANI with bimetal oxides as anode and cathode modifier, they incorporated bi-metal oxide in PANI during its polymerization and found that replacing Vulcan carbon to PANI as catalyst resulted is 1.45 times increase in power density. The maximum power density of 6.1 W/m^3 and 811 mV open circuit potential (OCP) were obtained with MFC-C (in case of bare electrode), which are lower than that of MFC incorporated with $MnFe_2O_4$ nano particles (NPs) (modified electrode). Huang et al. (2016) modified carbon cloth electrode with PANI/graphene and used as anode in a single chambered MFC of volume 28 ml and achieved voltage of 573 ± 37 mV, and power density of 884 ± 96 mW/m^2, which was 1.3 and 1.9 fold higher, respectively, of those of the bare carbon cloth electrode. Fu et al. (2020) combined Fe_2O_3 with graphene and used the composite to coat carbon felt anode and reported significant increase in performance of a dual chambered MFC of volume 250 ml. The maximum power density reported by them was 334 ± 4 mW/m^2 that was 2.59 times more than the unmodified electrode. Jian et al. (2020) prepared a novel anode Fe_2O_3-PDHC/CF (Fe_2O_3-polyaniline-dopamine Hybrid composite) by electrochemical deposition method and reported 90.3% of indole degradation efficiency and reported a maximum power density of 3184.4 mW/m^2 using modified electrode in dual chamber MFC of volume 120 ml (anodic volume). Rajesh et al. (2020) used PANI coated carbon felt as anode and reported a power density of 216 mW/m^2 that was 2 fold higher than bare carbon felt electrode in a dual chamber MFC of volume 250 ml.

These studies concluded that nanostructured PANI is a suitable base for its biocompatibility, hydrophilic nature, positively charged surface and incorporation of metal oxides and other conductive nanostructures like CNTs, graphene further enhances its electrocatalytic activity.

4.2.2. Anode modification using graphene

Graphene is composed of single atom thick sheet of graphite having large specific surface area, high electronic conductivity and good biocompatibility, thus have application as electrode material (ElMekawy et al., 2017; Yuan and He, 2015). Various researchers have used graphene modified electrodes in MFC due to its above-mentioned properties and found improvement in power generation in comparison to respective bare electrode. Liu et al. (2012) modified carbon cloth with graphene oxide and observed a 2.7 fold increased in power generation in MFCs.

Zhang et al. (2011) modified stainless steel mesh (SSM) using porous graphene and PTFE (Polytetrafluoroethylene) paste and used as anode in MFC reactor fabricated using two round polymethylmethacrylate templates and observed improved MFC performance. The maximum power density of 2668 mW/m^2 was reported when modified electrode was used which is 18 times larger than that obtained from the MFC with the SSM anode (without modification). Xiao et al. (2012) tested different shapes of graphene modified anodes and observed that graphene oxide shape which deliver higher surface area also deliver higher power density. Zhao et al. (2013a) have reported that when anode is modified using graphene could achieve 4.2 times higher power density over bare carbon paper anode. Different strategies have been considered to bind graphene oxide on the surface of electrode, these include chemical vapor deposition (CVD) and direct deposition of graphene suspension on the surface of electrode (Zhao et al., 2013a). Kirubaharan et al. (2015) doped nitrogen over graphene nanosheet and used as anode electrode and reported a maximum power density of 1008 mW/m^2 at a current density of 6300 mA/m^2. Nitrogen doped on crumpled graphene sheet induced the structural disorders which facilitated the electrical conductivity. Huang et al. (2016) have designed carbon cloth (CC) with graphene and polyaniline by firstly add oxygen containing group to the CC then combined graphene to the carbon cloth via hydrogen bonds between CC and graphene then PANI added to the anode by soaking into aniline monomer solution and obtain a power density of 884 mW/m^2 which was 1.9 times more to bare CC when used in MFC. Graphene modified anode delivered a maximum power density of 2142 mW/m^2 at a current density of 6.1 A/m^2 in MFC, greatly increase the power density of MFC compared with the bare SSFF-MFC (Hou et al., 2014). The disadvantage of the SSFF is that it has poor biocompatibility as compared to carbon based anode that results in limiting the bacterial colonization on the surface of fibers. To resolve this, carbon nanoparticles were used to modify the SSFF to improve the biocompatibility and decreasing its over-potential as well as increasing the surface area of the electrodes accessible for the bacteria adhesion (Hou et al., 2014). Geetanjali et al. (2019) modified the carbon cloth using $NiWO_4$/GO and used as anode electrode in a single chambered MFC of 0.8 ml volumetric capacity operated using simulated wastewater sample and reported a power density of 1458 mW/m^2 which is 8.5 fold more than bare electrode reported by them.

4.2.3. Anode modification using metal/metal oxides

Semiconducting metal oxide have great potential to work as anode catalyst in MFCs due to its high rate of redox reaction and electrical conductivity. Metal oxide such as TiO_2, MnO_2, FeO_2, Fe_2O_3 have properties such as structural stability, low cost, nontoxicity and good biocompatibility (Priya et al., 2020) and widely used as catalyst to improve the performance of MFC.

Precious metals such as Pt or Au or Ag are widely employed in industries dealing with electrochemistry processes owing to their catalytic activity and high conductivity (HaoYu et al., 2007). However, the high cost of these metals makes them unsuitable to be used as anodic material in MFCs. The catalytic activity of semiconducting metals (PAni or graphene-based catalyst) or non-noble metals (such as Fe-based or Co-based catalysts) and their oxide nanoparticles is almost comparable to that of the precious metals, which can explicitly reduce the ohmic resistance and enhance the attachment of exo-electrogenic bacteria on the surface of the anode (Hindatu et al., 2017). Therefore, the topic needs to be widely explored in terms of the use of such semi conducting materials which have promising catalytic activity, conductivity and are not cost intensive.

One such promising material suitable for modification of anode can be Ti or TiO_2 which has high biocompatibility, anti-corrosion properties, optical and dielectric properties and reduced cost as compared to other metals. Thus, this metal oxide is a prospective material to be used for modifying anode electrode. Some of the metal-based modification along with their power generation and COD removal are tabulated in brief in Table 1.

5. Cathode electrodes

Performance of a MFC depends on various cathode properties, redox potential being the most important one. Other desirable cathode properties include low corrosion, good electrical conductivity, high porosity and high specific surface area. The cathode materials should possess high redox potential so as to capture protons easily, which improves the performances of MFCs (Zhou et al., 2011). In MFC operation, O_2 is an important molecule to act as an electron acceptor in the cathode chamber. Slow oxygen-reduction reaction (ORR) kinetics, an important issue in MFC operation, also directly affects the power generation efficiency. To maintain high rate of ORR, platinum (Pt) based catalysts are preferably utilized as cathode catalyst in MFC due to their potency in reducing the Ea (activation energy) of cathode reducing reactions (Mohan et al., 2014).

5.1. Conventional cathode electrodes

Generally, carbon materials are used as cathode such as carbon cloth, carbon felt, and carbon paper (Merino-Jimenez et al., 2016). Other material such as graphite sheet, graphite paper steel mesh, or other are preferred less than the carbon based material due to their less efficacy in increasing ORR rate. To improve the performances of MFCs, the cathode can be modified using active catalyst. Pt is most popularly used catalyst and supposed to reduce the activation energy and increase the reaction rate of the cathodic reaction (Li et al., 2017a). Considerably the high cost and very limited availability of platinum hinders its use to large scale MFC. A number of modifications have been used by researchers, in order to develop a cheap and highly active noble catalyst for its potential and specific application as cathode catalyst in MFC operation. A schematic representation of important cathode modifications is given in Fig. 4.

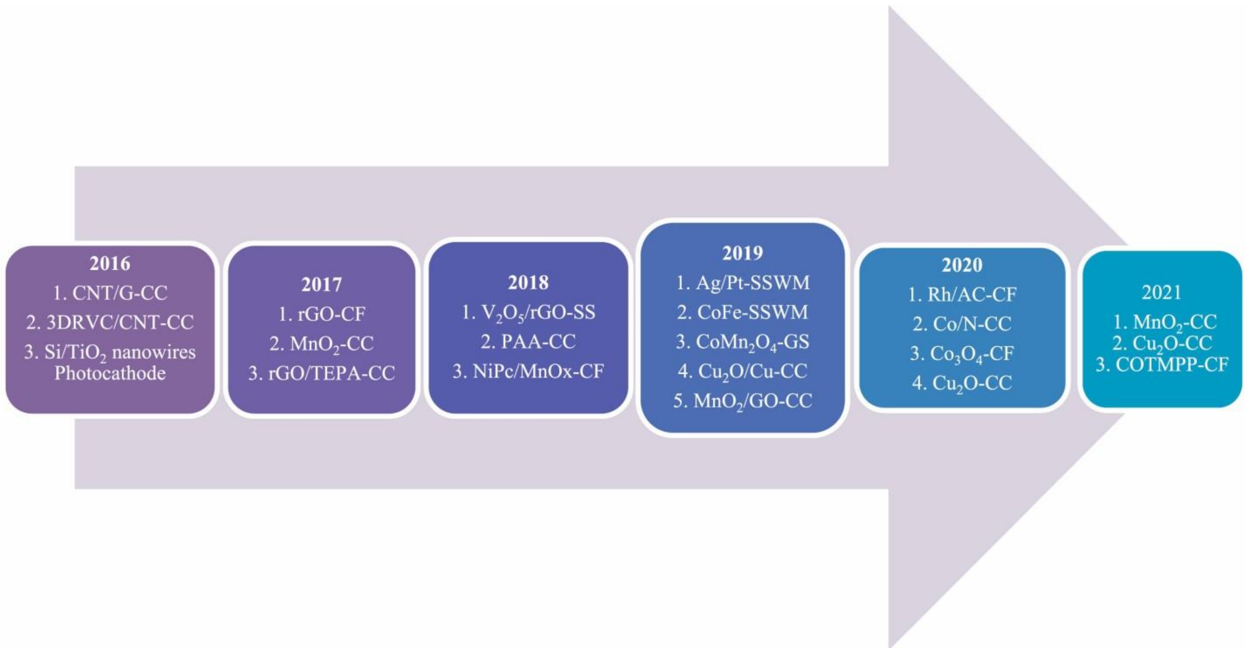


Fig. 4. Timeline depicting the development of Cathode electrode catalyst. Ag/Pt: Silver-platinum, CNT: Carbon nanotube, CC: Carbon cloth, CF: Carbon felt, Co/N: Cobalt with dopped nitrogen, CoFe: Cobalt–Iron, $CoMn_2O_4$: Cobalt Manganese oxide, Co_3O_4: Cobalt oxide, COTMPP: Cobalt tetramethoxyphenylporphyrin, Cu: Copper, Cu_2O: Copper(I) oxide, G: Graphene, GO: Graphene oxide, GS: Graphene sheet, MnO_2: Manganese dioxide, NiPc: Nickel phthalocyanine, PAA: Poly acrylic acid, rGO: Reduced graphene oxide, Rh/AC: Rhodium with activated carbon, Si/TiO_2: Silicon with titanium oxide, SS: Stainless steel, SSWM: Stainless Steel Wire Mesh, TEPA: Tetraethylene pentaamine, V_2O_5: vanadium pentoxide. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

5.2. Modification of cathode electrodes using non-Pt catalysts

Many other cathode catalysts have been researched so far that can manifested similar performances as platinum used as catalyst. Moon et al. (2006) used graphite felt coated with Pt as cathode in their microbial fuel cell and reported a power density of 150 mW/m^2, which was thrice times higher when graphite felt used without modification (Bare graphite felt). As Pt is very expensive metal so this can't be used for realistic application. Cheng et al. (2006) found that the potential did not change noticeably (maximum of 19%) when the Pt loading on the cathode ranged from 0.1 to 2 mg cm^{-2}. This finding resulted in the Pt modified cathode still being competitive and cost-effective. Li et al. (2017b) used $NiCo_2O_4$/carbon composite as catalyst to improve the oxygen reduction rate of cathode (carbon cloth) in MFC and reported a power density of 1249.86 mW/m^3. Sonawane et al. (2019) used PANI/Cu hybrid as catalyst to improve the performance of carbon cloth electrode and used as cathode in MFC and obtained a power density of 0.101 ± 0.01 mW/m^2.

After being doped with Cu and P heteroatoms, freeze-dried, and subjected to high-temperature pyrolysis, bacterial cellulose was used as a cathode catalyst (Fig. 5) (Li et al., 2019a). Doping resulted in changes in functional group and an increase in the number of active sites, thereby increasing the oxygen reduction reaction catalytic activity. The bacterial cellulose-based catalyst was coated on to an air diffusion layer (Ma et al., 2018) and utilized as the cathode in SCMFCs with activated sludge as the inoculum. The results of this study were compared with another similar work that employed a platinum-based catalyst (Li et al., 2019a). A maximum power density of 1177.31 mW/m^2 was obtained for the Cu-P-Co doped bacterial cellulose-based catalyst, while the Pt-based catalyst achieved a power density of 1044.93 mW/m^2. The recent modifications of different cathode electrode are elaborated in Table 2 with the power generation and COD removal values reported in the literatures.

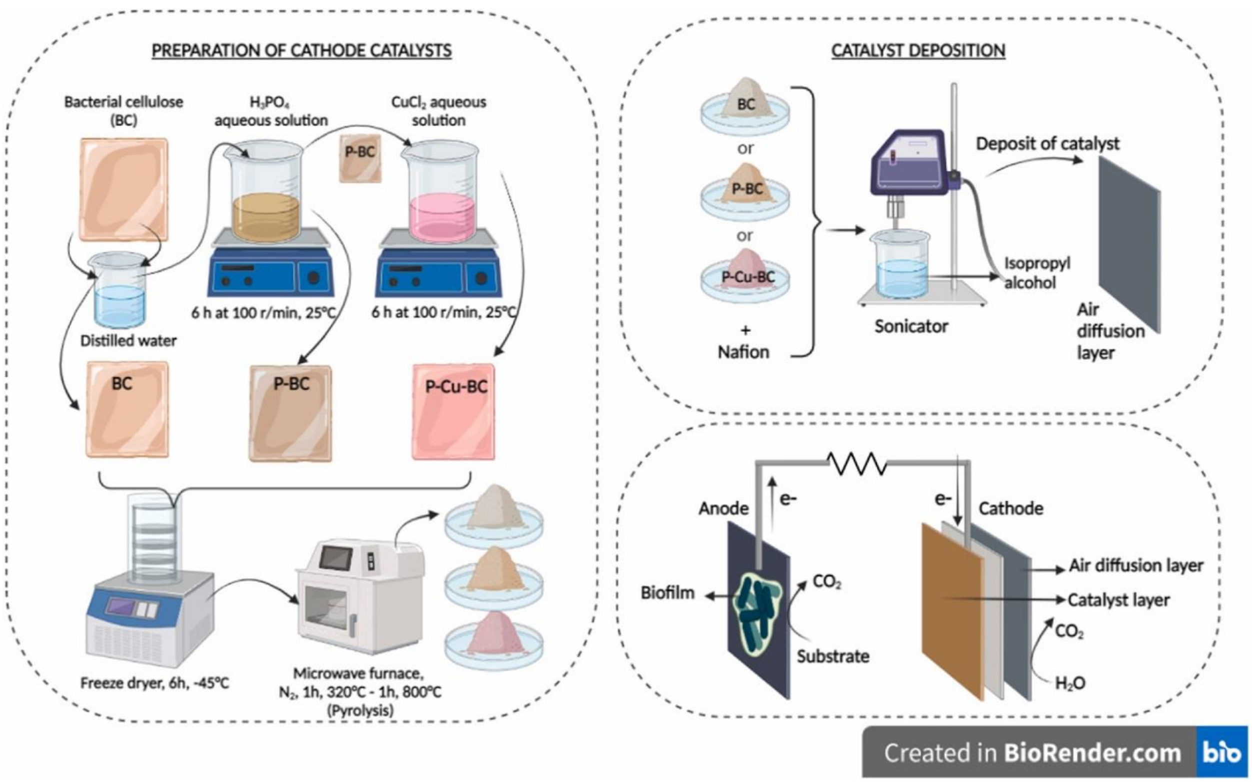


Fig. 5. Bacterial cellulose and its modification as catalyst for cathode in MFC. Note: BC: bacterial cellulose, P-BC: P-doped bacterial cellulose, P-Cu-BC: P, Cu-co doped bacterial cellulose [adapted from the studies by Ma et al., 2018 and Li et al., 2019a].

Table 2. Modification of cathode and its performance in term of power density and wastewater treatment.

MFC type	Volume of reactor (ml)	Cathode material	Catalyst	Power density (control)	Power density (modified)	No. of fold increase in power density	COD removal (%)	References
DCMFC	80	Carbon felt	Co–*N*-CNT	2.9 W/m^3	5.1 W/m^3	1.7	85.0 ± 4.1	Türk et al. (2018)
SCMFC	350	Carbon cloth	Co/N/C	–	931.1 mW/m^2	–	83.6 ± 1.2	Li et al. (2019b)
Cylindrical DCMFC	80 (A) & 250 (C)	Carbon felt	Co_3O_4	2.96 ± 0.20 W/m^3	6.62 ± 0.33 W/m^3	2.2	92 ± 5	Bhowmick et al. (2019b)
SCMFC	150	Carbon cloth	Ni–Co/SPAni	483.48 mW/m^2	659.79 ± 20 mW/m^2	1.4	91.5	Papiya et al. (2018)
SCMFC	25	Carbon cloth	Co/NCNT	74 ± 8 mW/m^2	469 ± 17 mW/m^2	6.3	–	Song et al. (2015)
DCMFC	80	Carbon felt	Fe–*N*-CNT	2.9 W/m^3	6.0 W/m^3	2.3	87.4 ± 3.3	Türk et al. (2018)
Cylindrical DCMFC	80 (A) & 250 (C)	Carbon felt	Fe_3O_4	2.96 ± 0.20 W/m^3	5.07 ± 0.25 W/m^3	1.7	91 ± 4	Bhowmick et al. (2019b)
SCMFC	100	Carbon cloth	Co/FeTMPP	–	33.4 mW/m^3	–	–	Zhao et al. (2013b)
DCMFC	80	Graphite felt	Bi–TiO_2	60 mW/m^2	224 mW/m^2	3.7	89.0 ± 1.9	Bhowmick et al. (2018)
SCMFC	26	Stainless steel mesh	Fe–*N*–C/AC	1.8 ± 0.03 W/m^2	2.4 ± 0.1 W/m^2	1.3	–	Yang et al. (2020)
SCMFC	40	Carbon paper	Fe–*N*-G	561.1 mW/m^2	1149.8 mW/m^2	2.0	–	Li et al. (2012)
SCMFC	1800	Carbon paper	Fe-NCB	0.9 W/m^2	1.85 W/m^2	2.0	–	Erable et al. (2018)
SCMFC	150	Carbon cloth	SGO-TiO_{2}-PAni	483.5 ± 16 mW/m^2	904.18 ± 25 mW/m^2	1.9	–	Papiya et al. (2020)
SCMFC	–	Carbon cloth	$MnCo_2O_4$/PPy	1.77 W/m^3	6.11 W/m^3	3.4	89.8	Khilari et al. (2014)
SCMFC	70	Carbon felt	Rh/AC	3.65 W/m^3	9.36 W/m^3	2.6	93 ± 2.2	Bhowmick et al. (2019a)
SCMFC	900	–	MnO_2	0.491 W/m^3	1.64 W/m^3	3.3	84.4 ± 3.4	Noori et al. (2016)
SCMFC	100	Carbon felt	SnO_2: PAni (4:6)	14 mW/m^2	65 mW/m^2	4.6	–	Tiwari et al. (2020)

(CNT: Carbon nanotube, Co: Cobalt, N: Nitrogen, Co_3O_4: Cobalt oxide, G: Graphene, GO: Graphene oxide, MnO_2: Manganese dioxide, Fe: Iron, NCB: Nicarbazin, Fe_3O_4: Iron (II, III) oxide, Ni: Nickel, Rh/AC: Rhodium with activated carbon, PPy: Polypyrrole, Bi: Bismuth, TiO_2: Titanium oxide, SnO_2: Tin (IV) oxide, PAni: Polyaniline, $MnCo_2O_4$: Manganese cobaltite, TMPP: tetramethoxyphenylporphyrin).

6. Conclusions

Microbial fuel cell is an innovative and sustainable technology for bioelectricity generation and wastewater treatment. Many researchers have focused on the development of electrode material, membranes and construction material to prove this technology as cost effective and ecofriendly with a very low carbon footprint. Carbon-based electrode materials are widely employed in the field of environmental engineering due to their widespread availability, high conductivity, chemical stability, biocompatibility, and low cost, among other characteristics. The surface modification of the anode material also leads to a higher power output for the MFC as a result of its use. In contrast, because of the low rate kinetics of the oxygen reduction reaction (ORR), the cathode materials require an improved catalytic capability in order to function properly. However, despite being the most effective catalyst, platinum has become prohibitively expensive to use in commercial applications due to its high cost. In recent years, transition metal oxides, such as manganese oxide, nickel oxide, and cobalt oxide, as well as nitrogen-doped carbon-based materials, have emerged as potential materials for low-cost cathode catalysts and increased oxygen reduction reaction (ORR) rate. The various anode and cathode modification strategies was reviewed and it was observed that, better power density and higher oxygen reduction reaction rate (ORR) could be easily achieved in MFCs under optimized operational conditions.

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Dedication

The authors dedicate this article in fond memory of our colleague and dear friend, late Ms. Sanchita Gupta, who was a moral support system while inscribing this article.

Declaration of competing interest

All the author and co-authors declares that this article has no conflict of interest.

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