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Engineered nanomaterials in microbial fuel cells – Recent developments, sustainability aspects, and future outlook

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ABSTRACT

Microbial fuel cells (MFCs) have recently emerged as green technology for the direct electricity generation from polluted (waste)water loaded with organic and inorganic contaminants. Despite the remarkable progress in applying MFCs to deal with different types of (waste)water, several issues, including the power density, durability, and costs of the electrode materials, are still to be tackled towards the commercialization of these technologies. The present manuscript provides a critical review of the recent advances and existing challenges in applying engineered nanomaterials (ENMs) to optimize the properties and performance of MFCs. The main advantages of the application of ENMs in the structure of MFCs are to provide a high specific surface area (SSA) for the electrodes, promote the electron transfer and oxygen reduction reactions, thereby representing a high level of biocompatibility for the adhesion of microbial communities, and being durable and cost-effective, especially when fabricated from natural resources. The sustainability aspects of ENMs-based MFC technologies and recommendations for future studies towards the development of sustainable nanomaterials-enabled developments of MFCs are discussed.

Nomenclature

AC	Activated Carbon
AC-MFCs	Air Cathode MFCs
AD	Anaerobic Digestion
AOPs	Advanced Oxidation Processes
CC	Carbon Cloth
CEs	Coulombic Efficiencies
CFF	Carbon Fiber Felt
CNTs	Carbon Nanotubes
CNMs	Carbonaceous Nanomaterials
CNPs	Carbon Nanoparticles
CTR	Charge Transfer Resistance
EAB	Electrochemically Active Bacteria
EPD	Electrophoretic Deposition
EET	Extracellular Electron Transfer
ENMs	Engineered Nanomaterials
GAC	Granular Activated Carbon Anode
LDDT	Long Distance Direct Transfer
MFCs	Microbial Fuel Cells
NPs	Nanoparticles
NOVs	Native Oxygen Vacancies
ORR	Oxygen Reduction Reaction
PAOP	Plasma Advanced Oxidation Process
PEM	Proton Exchange Membrane
PES	Polyethersulfone
PGMF	Pt group Metal-Free
PMPS	Polymethylphenyl Siloxane
PPy	Polypyrrole
rGO	Reduced Graphene Oxide
ROS	Reactive Oxygen Species
SCMFC	Single Chamber Microbial Fuel Cell
SDDT	Short Distance Direct Transfer
SSA	Specific Surface Area
SSFF	Stainless-Steel Fiber Felt
WoS	Web of Science

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1. Introduction

Rapid industrialization has resulted in an over-consumption of fossil fuels, which has caused several environmental crises such as global warming and climate change, mainly through releasing greenhouse gases (e.g., carbon dioxide) into the atmosphere [1,2]. Hence, research on the development of sustainable energy generation and storage strategies to meet the ongoing global needs for clean sources of energy is of high importance [3]. In this regard, various energy harvesting technologies from renewable sources of energy such as solar [4], wind [5], and marine [6] have been developed and employed in recent decades in many regions all over the world. The predictions demonstrate the capability of such technologies to diminish the reliance on fossil fuels in the mid-term future [7]. However, the basis of these technologies is to harvest the energy directly and transform and store it for further use. In this regard, microbial fuel cells have emerged as a sustainable technology for the simultaneous elimination of pollutants from (waste)water and generating electricity through redox reactions of organic substances [8,9]. There are also reports on bioelectricity generation from (waste)water in MFCs by reducing heavy metals such as hexavalent chromium [10]. Hence, MFCs can be used efficiently to deal with industrial (waste)water contaminated by various organic and inorganic compounds such as pulp and paper [11], textile [12], and pharmaceutical [13] effluents.

Principally, MFC can be divided into single- and dual-chamber configurations. A single-chambered MFC is generally comprised of an air cathode as well as an anode. A proton exchange membrane (PEM) may be used to separate the cathode and anode (Fig. 1, a). In the anode area, electrons and protons are produced by the oxidative biodegradation of organic compounds that are transferred to the cathode via an external circuit (for electrons) and a PEM (for protons) [14]. The dual-chambered reactors are the most widely used MFCs, including an anaerobic anode coupled with an aerated cathode (Fig. 1, b). The chambers are generally connected using a close-fitting PEM. The microbial growth forms a biofilm on the anode [15,16]. The specific surface area of ENMs (to provide enough area for the adhesion of microorganisms), and toxicity (which can prevent microbial growth [17]) are considered as the main factors influencing the formation of the biofilm onto the ENMs. It has been also observed that some types of ENMs (such as Mo_2C/CNTS [18]) can promote processes (such as oxidation of hydrogen, as the common metabolite of *E. coli*) that can make the medium suitable for microbial growth. In addition, the surface charge of the microorganisms and ENMs can lead to adsorption or repulsion forces between the ENMs and the microorganisms [19]. The cathode chamber

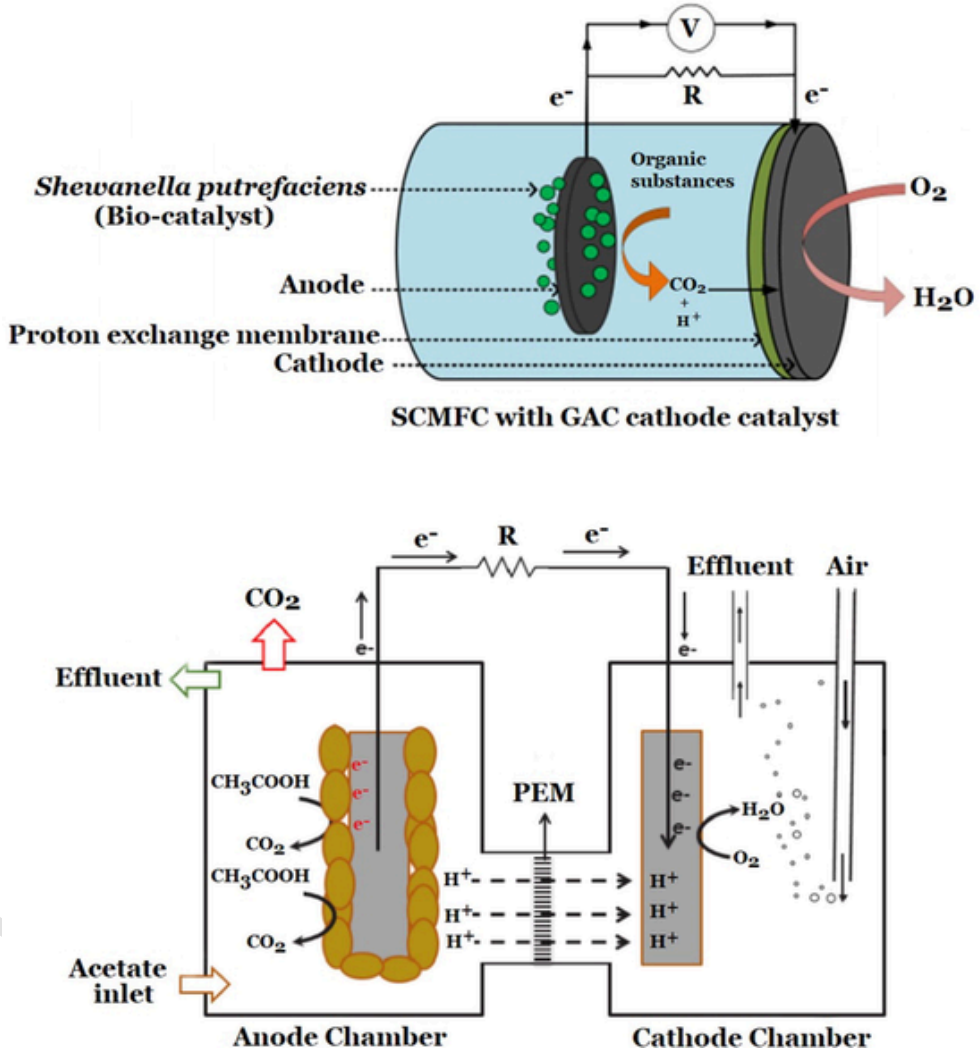

Fig. 1. A single-chamber microbial fuel cell (SCMFC) in which activated carbon (AC) derived from ground nutshell utilized as a metal-free oxygen reduction catalyst for the aerated cathode (up), and a dual-chamber MFC with a PEM (down), the organic compounds are degraded by the microorganisms in the anode to produce electrons, which are transferred to the cathode and participate in oxygen reduction reactions, adopted from Karthick et al., [33], Abu-Reesh, [34].

normally performs under aerobic conditions by continuous supplementation of air and water [20].

The specific interactions between the bacteria and electrode materials govern the MFC performance and density of the generated power [21]. Electrochemically active bacteria (EAB) play an important role in decomposing the organic compounds into carbon dioxide, protons and electrons [22–25]. Transferring the generated electrons is the critical step of the MFC performance, reflecting the importance of the anode that needs to satisfy specific requirements such as being biocompatible, low-cost, and of high electron transfer capacity [22–26]. The cathode receives the electrons generated by EAB from the anode by an external load. In addition, the protons pass through a PEM to reach the cathode area [27,28]. Hence, the cathode material is considered another important factor for the performance of MFCs since the cathode reduction can negatively affect its durability and the overall performance of the cathode [29]. Furthermore, the PEM should support transferring the protons and preventing O_2 and the compounds present in the inlet effluents [30,31]. Antifouling properties are also essential for the PEM to make MFCs more economically and technically viable [32]. To address the abovementioned needs, studies have recently been oriented towards the utilization of ENMs in the structure of MFCs.

The present review aims at exploring the applicability and sustainability considerations of ENMs to enhance the efficiency of MFCs. The current trends and research hotspots in this scientific area have been identified (section 2), followed by discussing the electron transfer mechanisms using the ENMs in MFCs (section 3). Then, the applicability of ENMs in various compartments of MFCs including anode (section 4), cathode (section 5), and proton exchange membrane (section 6) have been assessed and discussed. The sustainability aspects of ENMs in MFCs as well as future perspectives are also discussed (section 7) to push the commercialization of these technologies for real applications.

2. Trends and research hotspots

A systematic approach has been adopted in this review to identify the trends and research hotspots in the application of ENMs to promote the application of MFCs. A specific combination of keywords presented in Table 1 was used for an advanced search to extract all the relevant documents published in the Web of Science (WoS) Core Collection database, which includes all the citations indexed in Science Citation Index Expanded (SCI-EXPANDED). A "Marked List" was then prepared by saving the WoS outputs and then was saved as "text" to visualize the trends in the keywords evolution in the literature using CiteSpace software (version 5.5.R2) according to the existing manual [35]. Fig. 2 represents the output of the keywords analysis and their evolution (2004–2021). In addition to the trends of the keywords, highly-cited documents in this area were retrieved and reviewed to determine the research hotspots, as summarized in Table 2.

According to Fig. 2, studies in this scientific area have been initiated since 2004. The first set of keywords, including "microbial fuel cell" (2006), "performance" (2009), and "electricity generation" (2007) are the most frequently used keywords, demonstrating the starting trend of research in the application of nanomaterials to enhance the performance and electricity generation with MFCs between 2004 and 2010. Besides, "carbon nanotubes" (2004), "oxygen reduction reaction"

Table 1a
Relevant keywords and their combinations used to identify the documents indexed in WoS on the application of engineered nanomaterials in MFCs as well as the WoS highly-cited documents in this field.

Set	Combination of keywords	Results
# 1	TS = (microbial fuel cell*)	11,813
# 2	TI = *nano*	1,235,678
# 3 (Original search)	#1 AND #2	809
WoS highly cited	#1 AND #2	11

Table 1b
Remarks of the highly-cited documents concerning the application of ENMs in MFCs.

Hotspot	Citing documents	Remarks	Ref.
Commercialization aspects	122	The most important challenges for the commercialization of MFCs include insufficient, electricity output, associated costs, and durability.	Review/ Anjum et al., [40]
	120	For promoting MFC commercialization, there is a need to develop electrodes with high conductivity, durability and stability, bio-compatibility, as well as high SSA and porosity (to allow microbial colonization).	Review/ Zhao et al., [41]
	297		Original/Hu and Cui [42]
MFC cathode modification with NMs	41	Modification of the cathode in MFCs with carbon nanotubes increases the electronic conductivity and the power output of the system.	Review/ Gong et al., [43]
	25	Cu_2O/reduced graphene oxide (rGO) represents an excellent catalytic performance and promoted the diffusion of O_2 to the cathode. The catalyst with its high antibacterial activity can also inhibit microbial growth in the cathode.	Original/ Xin et al., [44]
	130	The prepared low-cost and resistant cathode made of N-doped carbon nanosheets can be employed as a strong electrocatalyst for promoting ORRs and hence they can replace the high-cost Pt-based electrocatalysts in MFCs for large scale applications.	Original/Liu et al., [45]
	427	Nitrogen-enriched (core-shell)Fe/Fe_3C-C nano-rod electro-catalyst significantly is capable to enhance the oxygen reduction reactions in an MFC.	Original/ Wen et al., [46]
	277	N-doped carbon nanotubes can be considered as highly efficient and durable cathodes for ORRs in MFCs.	Original/ Feng et al., [47]
MFC anode modification with ENMs	238	Functionalized CNMs can significantly promote the colonization of microorganisms by providing appropriate conditions for the growth of microorganisms.	Original/ Cheng et al., [48]
	177	*In situ* nitrogen and Molybdenum dual doping (N-MoO_{3-x}) can considerably enhance the conductivity, accessible surface active sites, as well as electrochemical stability of MoO_3, which can significantly boost its electrochemical characteristics as MFC anodes.	Original/Yu et al., [49]
	322	The prepared porous CNT-textile anode is characterized by high conductivity and biocompatibility which can enhance the performance of MFCs.	Original/Xie et al., [50]
MFC membrane modifications with ENMs	122	MFC membrane maintenance costs can be considerably reduced by the incorporation of nanomaterials.	Review/ Anjumetal., [40]

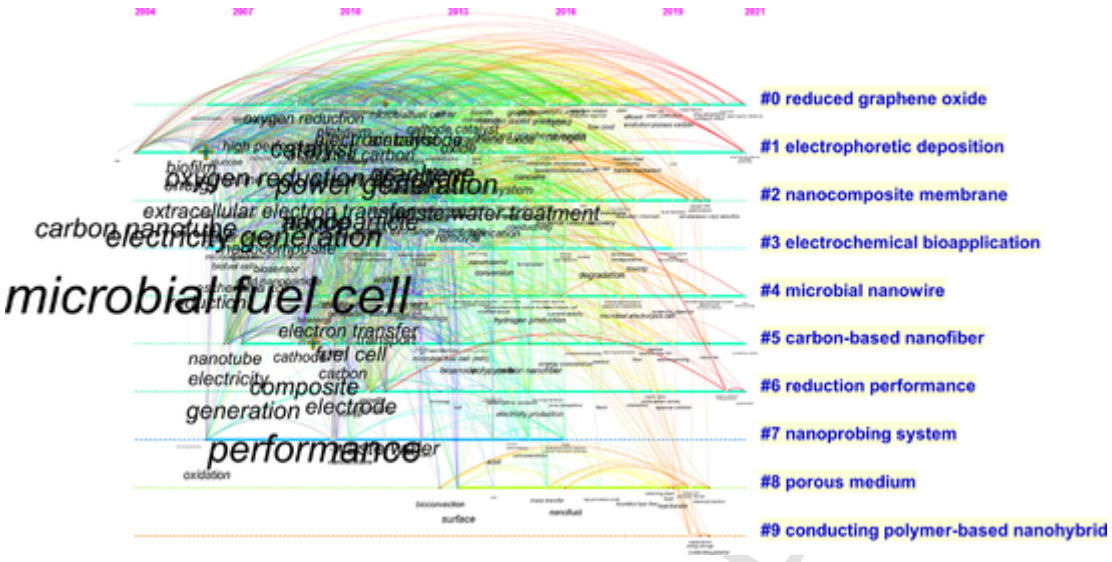

Fig. 2. The pattern of keywords evolution (2004–2021). The size of keywords reflects the keyword frequency in the studied documents. The keywords "microbial fuel cell", "performance", and "electricity generation" display the highest frequencies. The figure has been produced originally by the authors using the CiteSpace.

(2009), and "anode" (2009), and "composite" (2009) have appeared in the relevant scientific documents but with less frequencies. "graphene", and "graphene oxide" appeared in 2012 and 2014, respectively, which highlights the existing trend for the implementation of carbonaceous nanomaterials (CNMs) as sustainable alternatives to enhance the performance of MFCs. Another trend in the period of 2010–2015 was initiated with the keywords such as "membrane", and "proton exchange membrane", both appeared since 2012, which can be in line with the need to overcome the technical issues associated with the membrane performance in MFCs, such as fouling [36,37], which can also bring considerable economic issues. The appearance of "nitrogen" in 2016 indicates the initiation of the next research trend to apply this element mainly as a dopant to improve the electrical properties of nanomaterials used in MFCs [38,39]. Very recently, keywords such as "functionalization" (2020) have started appearing to further improve the performance of MFCs and their generated power density.

Table 2 summarizes the remarks of the highly-cited documents in this scientific area, which is mainly concerned about the MFC commercialization barriers, modifications of the electrodes (i.e., cathode and anode) as well as modifications in the structure of the MFC membrane to overcome the existing before-mentioned technical and economic challenges.

3. ENMs-assisted electron transfer in MFCs

Extracellular electron transfer (EET) is generally known as a process in which electrons are shuttled between the microbes and the anode [51]. EET normally occurs in MFCs via direct electron transfer (DIET) and indirect electron transfer (IET) routes. Three main mechanisms have been discussed so far on the DIET in biological systems including: a) electrically conductive pili, b) extracellular substances, and c) conductive materials (Fig. 3) [52,53]. Biofilm formed on the surface of the anode establishes a direct physical contact and plays a crucial role in short-distance direct transfer (SDDT). In this process, a so-called membrane redox multi-heme protein is the essential element. This protein contains multiple units of heme, especially cytochrome-c protein. The iron-protoporphyrin IX vinyl groups are linked to the two cysteine side chains of a -CX_nCH- motif through the enzymatic machinery [54,55]. On the contrary, multilayer biofilm is responsible for the electron transfer in long-distance direct transfer (LDDT) through the activity of the special pili-like electron carriers, which are produced by *exo*-electrogens such as *Shewanella* and *Geobacter* spp. [56–58]. According to the literature, mixed cultures of *exo*-electrogenic microorganisms can represent a promoted efficiency for current density and power generation compared to pure cultures [59]. Electron transport to the anode in MFCs can also occur via a microbially produced endogenous (e.g., pyocyanin, riboflavin) or exogenous mediator (e.g., 2,2′-azino-bis(3-ethyl- benzothiazoline-6-sulphonic acid) [60].

ENMs can provide a high specific surface area for the adhesion and growth of the microorganisms around the anode, thereby facilitating the DIET process. As indicated in Fig. 3, b, conductive ENMs play the role of a bridge between the microorganisms and the anode to promote the DIET in MFCs. Conductive materials can also result in closely connected microbial communities [61]. Secretion of extracellular polymeric substance (EPS) is considered as the main reason for the attachment of microbial species forming the compact communities [62]. EPS compounds are normally secreted as the protection mechanism when a microorganism is exposed to a new condition including the potentially toxic element [63]. It has been also reported that EPS can also contribute to the DIET process [64].

Indirect electron transfer processes may also occur via so-called mediators [65]. It has been well-documented that some bacterial species such as *Pseudomonas* and *Shewanella* can secrete shuttle molecules such as flavins for transferring the outer membrane of the microbes to the anode [66,67]. Another identified pathway for the EET is through the nanowires mechanism in which microorganisms such as *Geobacter* genera (e.g., *G. sulfurreducens* and *Shewanella oneidensis)* have been employed in MFCs. It was revealed that *Shewanella* uses conductive appendages to transfer the electrons outside of the cell membrane [68].

The application of ENMs to promote the EET process can also be considered as an attractive alternative to enhance the efficiency of MFCs. In this regard, iron-based nanomaterials can represent high magnetic properties [69,70], which can make them appropriate and inexpensive candidates for such an application. However, there are limited reports for the application of iron-based nanomaterials to promote EET in MFCs. According to Jiang et al. [71], the presence of iron sulfide nanomaterials in MFCs results in the formation of *Shewanella* PV-4 cell/nanoparticle aggregates (Fig. 4), promoting the electron transfer efficiency in the network (only in the presence of live *Shewanella)*.

4. ENMs in MFC anodes

The composition of the electrode is a very important parameter for the efficiency of MFCs, which can determine the growth and develop-

Table 2
Modifications in the anodes and their impacts on power density of MFCs.

Modification	Featured properties	Current density (mA/m^2)	Power density (mW/m^2)	Ref.
CFF anodes with vertically aligned TiO_2 and Fe_2O_3 nanolayers	- Increase in microbial density and biodiversity. - Enhanced sulfide removal (over 90 %)	1591 (TiO_2), and 1296 (Fe_2O_3).	608 (TiO_2) and 537 (Fe_2O_3); 1.53 and 1.36 folds for TiO_2 and Fe_2O_3 compared with non-modified electrodes	[85]
Decoration of the anode surface made of graphene oxide with a bimetal oxide ($NiWO_4$)	enhancing the abundance of *γ-proteobacteria* (42.37 %).	3807, much higher than non-modified electrodes (1141)	1458, versus 212 for non-modified electrodes.	[84]
Coating of the anode with Cu-doped FeO nanoparticles synthesized with *A. blitum*	- Increase in hydrophilicity of the electrode with negligible antibacterial behavior. - Reduction in ohmic and CTR	270	161.5	[90]
Fabrication of bacteria-derived iron oxide/carbon nanocomposite	Increase in the roughness, SSA and catalytic activity of the electrode	1600, 3.5 times higher than unmodified CF electrode.	797.0, compared to 226.1 for unmodified CF electrode.	[91]
GL-MoS_2 modified CC and SSFF anodes	- Cost-effectiveness, providing high SSA, biocompatibility	2830 versus 1400, for GL-MoS_2 CC, and SSFF modified anodes, respectively.	960.4 and 713.6 for GL-MoS_2 CC and SSFF modified anodes, respectively.	[98]
Photo-assisted bio-anodes (E. coli@Au_2@CdS_1)	Enhanced electron transfer inside the bacterial biofilms	Up to a 2.5-fold increase in the current density compared to non-modified bio-anodes.	2300 versus 215 for non-modified electrodes	[99]
Cellulosic derived graphene-polyaniline composite anode	Improvement in the electron transfer	1.1	87.71	[94]
cotton textile-based porous structure anode modified by molybdenum carbide nanoparticles	A very high SSA (832.17 m2/g) for bacterial adhesion.		1120	[75]
Carbon-coated TiO_2 nanotube array, as the anode.	Biocompatibility, high electro-active surface, low electric resistance and Tafel slope, resulted in improved bacterial charging capacity and electron transfer.	2500	880 compared to 610 for the MFC with the commercial carbon cloth	[87]
α-MnO_2 nanowires/carbon Vulcan composite	Low cost, capable of providing a high SSA and active sites for redox reactions.	4.0 versus 2.7, for α-MnO_2/carbon Vulcan and pristine α-MnO_2 nanowires, respectively.	180 and versus 111 for α-MnO_2/carbon Vulcan and pristine α-MnO_2 nanowires, respectively	[100]

ment of the bacteria as well as the transport of the nutrients required for feeding the microbial communities [72]. The current trend in the scientific community is to develop appropriate anode materials, which can provide high SSA and porosity as well as high conductivity to enhance the efficiency of MFCs. Cost-effectiveness is another challenge to be addressed for further commercialization of MFCs for real applications. Various types of ENMs have been considered in recent years to overcome such research requirements. Also, sustainability criteria dictate the methods selected for the synthesis of nanomaterials to be simple and eco-friendly developed according to the green chemistry principles [73].

As stated before, the anode is the most important element of the MFCs, where electrons are collected and transferred through an external load to the cathode to generate electricity. Modifications of the MFC anodes with nanomaterials were initiated in 2007 [74] with the application of CNMs because they had been already recognized by the potential to exhibit promising electrical and structural properties. Attempts by L. Zeng et al. [75] resulted in the fabrication of a cotton textile-based porous structure anode modified by molybdenum carbide nanoparticles through a facile two-step method to enhance the performance MFCs. The composite offered a large SSA of 832.17 m^2/g, which can be ideal for bacterial adhesion. The MCF equipped with the prepared electrode represented up to 116% higher power density than the pristine carbon fiber felt (CFF) anodes. The enhancement in the performance of MFC is directly related to high biocompatibility and superior conductivity of the modified electrode.

Carbon nanotubes (CNTs) are of special interest for such applications because they can create 3-D spaces around the anode for the internal colonization of diverse microbial communities and to accelerate the EET from *exo*-electrogens to the anode (Fig. 5). CNTs intertwined with textile fiber to create porous structures to enhance the substrate transport and internal colonization are examples of the successful incorporation of CNTs in MFCs for enhanced current density (157%) higher than that of the MFCs equipped with the traditional carbon cloth anodes [50].

Coating of the anodes with ENMs can provide more active sites for the adhesion of the microbial communities and hence increasing the biomass density. In this regard, the type and the characteristics of the nanomaterials (e.g., SSA and porosity) are of high importance. Various studies have emphasized that nanostructures with porous structures can host microbial communities and can act as efficient garden compost bio-anodes [77], thus enhancing the diversity and performance of microorganisms in the anode region of MFCs [78–80]. In this regard, porous CNMs such as graphene oxide and biochar have represented high efficiencies enhanced power density in the MFCs [81]. Decoration of such materials with ENMs has also been indicated as an effective way to decrease the charge transfer resistance (CTR) and to increase the power density of the modified anodes [82,83]. As evidenced by Geetanjali et al. [84], modification of the anode surface made of graphene oxide with a bimetal oxide ($NiWO_4$) could enhance the abundance of *γ-proteobacteria* (42.37 %), leading to a 6.9-fold higher power density compared to the non-modified anodes. In another relevant study, modi-

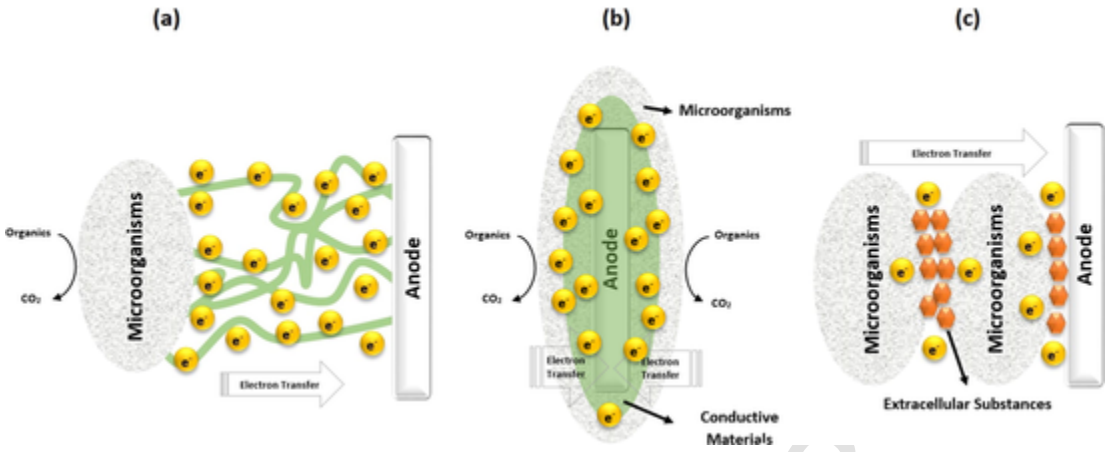

Fig. 3. The main mechanisms involved in DIET in the MFCs. Electrically conductive pili can play a role in the DIET between microorganisms and the anode; (a) direct transfer of the electrons can also occur by the conductive nanomaterials and (b) extracellular substances can also transfer the electrons to the anode (c).

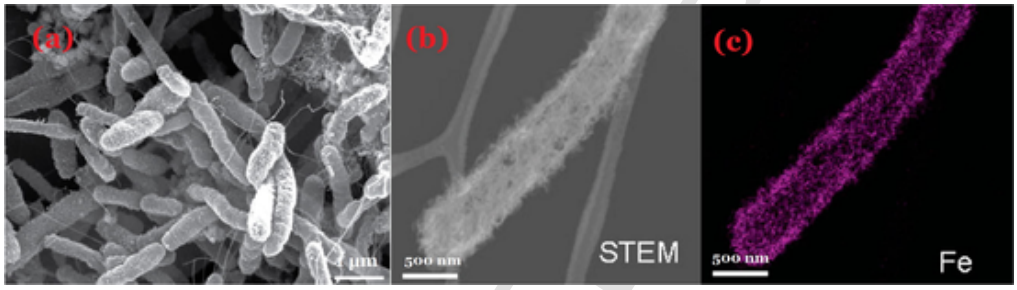

Fig. 4. Structure (a) and composition (b, c) of the cell/iron sulfide nanoparticles. Crystalline nanoparticles in intimate cover the surface of the *Shewanella* PV-4 cells resulted in the enhanced EET in a microbial fuel cell, adopted from Jiang et al., [71].

fication of CFF anodes with vertically aligned TiO_2 and Fe_2O_3 nanolayers increased the microbial density (demonstrated by high-throughput 16S rRNA gene sequencing analysis) as well as microbial diversities and improving the current density (1.53 and 1.36-folds for TiO_2 and Fe_2O_3 compared with non-modified electrodes). This modification resulted in a better sulfide removal (>90%after a 48-h) [85].

As a well-known semiconductor, titanium dioxide can inactivate the bacterial community (such as *Escherichia coli*) under light irradiation [86]. This can potentially limit the performance of MFCs under natural light. A limited number of studies address this, such as that performed by Deng et al. [87] that employed anodization and thermal treatment for the synthesis of a carbon-coated TiO_2 nanotube array as anodes in MFCs. Through a short time (30 min) soaking of the prepared TiO_2-nanotube arrays in Lysine, the authors demonstrated the increase in the bacterial loading capacity when the MFC was exposed to natural light.

Bio-mediated synthesis methods are considered as sustainable routes for the fabrication of nanomaterials due to multiple reasons such as being free of toxic chemicals and enhanced properties of the produced nanomaterials due to the existence of organic compounds in their structure, which act as the capping agents for the stabilization, which can increase the stability properties of ENMs (such as small size and high SSA) by avoiding the aggregation of the nanomaterials [88,89]. Coating the anode with bio-synthesized nanomaterials is hence considered a candidate for large-scale applications. The existing literature supports this idea. Coating of the anode with Cu-doped FeO nanoparticles synthesized with *Amaranthus blitum* [90] is an example that resulted in a hydrophilic electrode with negligible antibacterial behavior. In addition, the Ohmic resistance and CTR of the coated electrode was reduced compared to an uncoated electrode (due to the lower anode activation loss) capable with the delivery of 161.5 mW/m^2 and 270 mA/m^2 as the peak power density and corresponding current density, respectively, in the designed MFC. Such an approach has been adopted to address other issues related to the application of nanomaterials for anode fabrication. Conventional methods for the anode modification principally result in the aggregation of nanomaterials and fabrication of anode materials with low quality. In a recent study [91], bacteria-derived iron oxide/carbon nanocomposite (Bio-FeOx/C) catalyst prepared through the direct carbonization of *Shewanella* on a carbon electrode resulted in well-dispersion of the composite in the N-doped graphitic carbon. Such an approach caused the increase in the roughness, SSA and catalytic performance of the electrode. In addition, the CTR was reduced significantly.

Biogenic synthesis of nanomaterials on the anode surface can also bring the advantage of removing the pollutants from the inlet effluents through the catalytic activity of the ENMs. As an example, modification of the anode surface with Pd can promote the electrooxidation of organic compounds (such as formate and ethanol as the metabolites from the metabolic processes of electrogens) in the anode due to its excellent electro-catalytic oxidation potential [92,93]. A limited number of studies are available on applying anode-coated nanomaterials to remove heavy metals that are discharged from various anthropogenic activities [94]. Matsena et al., [95] co-deposited biogenic zero-valent palladium (Bio-Pd NPs) on the surface of a granular activated carbon anode (GAC) for Cr(VI) removal. According to the achieved results, the Pd-modified GAC can promote the Cr(VI) reduction efficiently. Loading of 6 mg/g of the nanomaterials onto the GAC surface has resulted in an optimum power density of 1965.4 mW/m^3 and a complete removal of 100 mg/L Cr(VI) in 25 h.

Carbon-based materials have also been studied in recent years to modify anodes, which can considerably improve the current density of the MFCs. Despite the progress made in developing such modification materials, especially 2D carbon-based nanosheets such as graphene and graphene oxide, high production and processing costs have been a barrier to the rapid commercialization of MFCs modified with these mate-

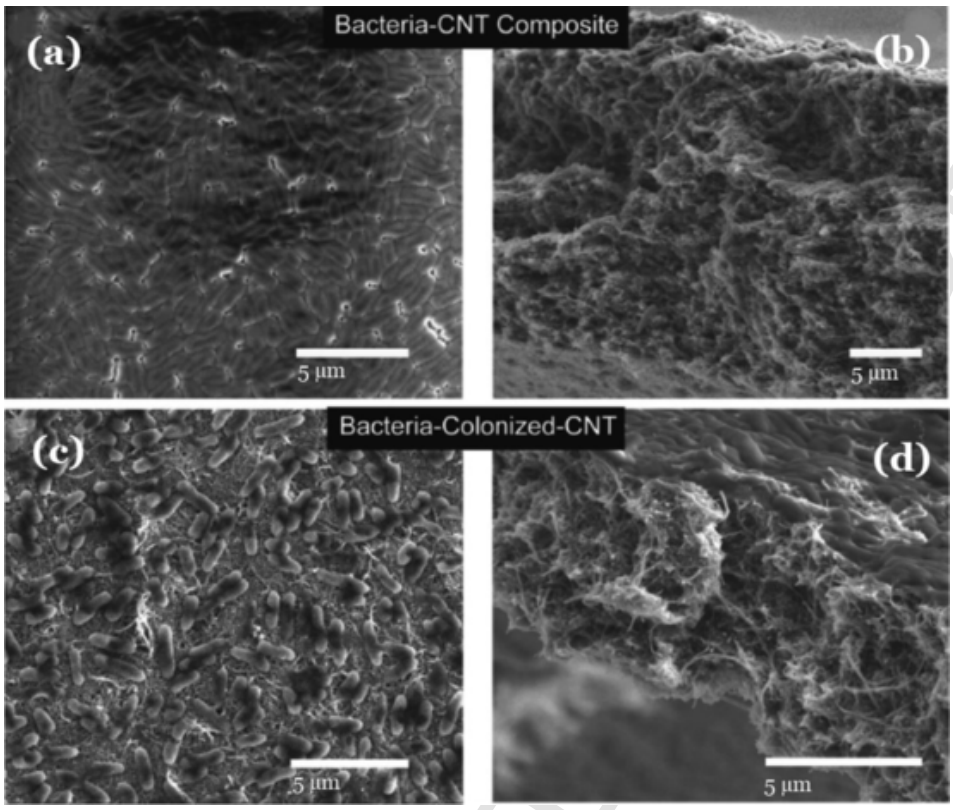

Fig. 5. Scanning electron microscopy of the colonization of bacterial communities on CNTs in an MFC, indicating the top (a) and the cross-sectional view (c) of the bacteria-CNT composite film. The top (b) and cross-sectional (d) images of a bacteria-colonized-CNT anode after 2o hrs of inoculation in bacterial anolyte, adopted from Kou et al. [76]. The figure indicates promotion of the colonization in 3-D space around the CNT anode in MFC.

rials [96]. Emerging of novel nanomaterials with the potential to replace expensive anode modification materials is considered an important milestone in the development of MFCs. Low-cost 2-D nanomaterials such as graphene-like MoS_2 (GL-MoS_2) have attracted attention very recently [97]. For instance, Lou et al., [98] developed a flower-like layered nanomaterial using a facile hydrothermal method for the modification of carbon cloth (CC) and stainless-steel fiber felt (SSFF) anodes and reached a high power density of 960.4 mW/m^2 and 713.6 mW/m^2 for CC and SSFF modified anodes, 1.7 and 3.6 times of their unmodified versions. Providing very high SSA, and biocompatibility that induces the colonization of microorganisms are among the most important reasons for such improvements in the performance of MFCs.

Nanomaterials have also been applied in recent years to develop photo-assisted MFCs. They can provide high conductivity, which promotes the electron transfer inside the bacterial biofilms. A promising approach in this regard is the application of ENMs for the modification of the *exo*-electrogens surface. Coupling the ENMs with high conductivity (such as Au) and photo-responsive materials such as CdS nanoparticles to prepare photo-assisted bio-anodes is an example in this regard [99], resulting in a 2.5-fold increase of current density compared to non-modified bio-anodes. Table 2 represents a summary of the findings on the modification of MFC anodes with ENMs.

5. Nanomaterials for oxygen reduction reactions

The cathode plays a significant role in the power generation in MFCs. EET from the anode to the cathode happens by the basic potential difference created by the cathode [101]. MFCs are principally operating in open-air configurations, and oxygen (with high reduction potential) is used as an electron acceptor. As a result, ORRs occur in the cathode region. However, dissolved oxygen content in most conditions is relatively low (~3–5 mg/L) to support high OPR levels. Hence, there is a need to adopt the strategies to maximize the OPRs. Coating the cathode surface with oxygen-reducing nanomaterials has been examined in recent studies to enhance the power generation by MFCs. Metal oxide such as cerium oxide has shown an acceptable performance in this regard [102,103]. An example is the coating of the cathode of a benthic microbial fuel cell (which is used to support low-power devices) with cerium oxide (CeO_2) nanoparticles synthesized through a hydrothermal synthesis route [103], which resulted in a maximum power density of 60 mW/m^3 versus 14 mW/m^3 produced from non-coated cathodes. The study also found that covering the cathode with cerium oxide nanomaterials performs better regarding the power output than the anode. From a mechanistic point of view, oxygen is adsorbed by the nanomaterials at the cathode region. Then, oxygen dissociation happens by the electrons in the medium in the presence of hydrogen ions, resulting in the production of water molecules [104].

Effective oxygen reduction efficiencies have also been addressed through the application of CNMs. For instance, carbon nanotubes decorated co-doped with cobalt and nitrogen (CuCo@NCNTs) prepared from a straightforward immersion and pyrolysis process represented high EOR capability as well as antibacterial performance, which prevents the biofilm formation on the cathode [105]. Such an approach was adopted by Yang et al., [106] for the synthesis of cobalt oxide NPs grown onto N-doped carbon nanotubes through controlled pyrolysis of the precursors such as graphitic carbon nitride and cobalt acetate. The prepared nanocomposites represented a high ORR performance via a four-electron reduction pathway. The authors argued that the high efficiency of the system is attributed to the growth of Co-N active sites, which can potentially promote the ORR processes. The modified cathode resulted in a 16.6% higher power density generation than the Pt/C catalyst.

The OPR can cause activation losses at the cathode and may interfere with the efficiency of MFC [107]. ENMs have been employed in re-

cent studies as catalysts to avoid destructive OPRs [107]. The 2-electron reduction reactions principally resulted in the generation of hydrogen peroxide, which is toxic for the microorganisms through the reactive oxygen species (ROS) generation in living cells [108]. Application of a catalyst in which the number of electrons produced by the catalyst is close to 4 leads to the direct oxygen reduction reaction, preventing the generation of ROSs. The literature has concluded that noble metals such as platinum can be considered suitable candidates in this regard. However, the application of such materials has been limited because they are expensive and not widely available. Carbonaceous materials have been introduced in recent years as acceptable alternatives for such materials. In a recent study [109], nitrogen-doped carbon-based nanofibers (N-CNFs), prepared under an inert atmosphere pyrolysis process at 900 °C, were used as the catalyst layer developed at the cathode. In this condition, the fine-tuning of CNFs electrochemical properties can play an essential role in promoting the OPRs. The product contained an optimum ratio of pyridinic and graphitic nitrogen, promoting a 4-electron pathway towards ORRs.

One of the other weaknesses in the conventional cathode materials in MFCs is the low electrical conductivity, which can potentially limit the performance of such technologies. For instance, MnO_2 is among the most promising materials for cathode applications because of its abundant, high theoretical capacitance, and relatively low cost [110]. However, it represents a moderate electrical conductivity of $10^{-2} \sim 10^{-3}$ mS $cm^{-1,}$ which is inadequate to lead to fast kinetic electrochemical reactions [111]. Published reports address such a weakness by introducing modifications in the structure of the nanomaterials. Creating oxygen vacancies in the structure of nanomaterials has been considered as an efficient way to enhance the electrical properties and conductivity of the nanomaterials. In this regard, native oxygen vacancies (NOVs) play the vital role of shallow donors in the respective nanomaterials, which can potentially promote electronic conductivity. In addition, more active sites are provided, and the surface reactions were accelerated [112]. Qiu et al., [113] introduced NOVs into the composition of MnO_2 nanorods through a hydrogenation treatment process. As a result, electrochemical performance of the nanomaterial was improved because of the fast charge transfer in the MFC. An energy density of over 50 Wh/kg was achieved that was much higher than the non-modified electrode, which has 2.86 50 Wh/kg, reaching a very high power density of 1639 mW/m^2 with ultra-long durability.

Table 3 represents a summary of the progress in the modification of MFC cathodes to enhance their power density.

cathode.

There is a trend for the use of industrial wastes for the generation of cathode materials. Especially, it is of high interest when the applied material has a porous structure and contains functional groups. For example, coal tar soot resulted from the incomplete combustion of coal tars or heavy oil used by Zhang et al., [120] as an efficient ORR catalyst due to the abundance of oxygen-containing groups. Such a strategy can considerably satisfy the economic considerations by the reduction of synthesis costs of the materials. Biochar has been considered as a low-cost product of biomass waste pyrolysis process, which has been used for various applications including (waste)water treatment and soil applications. Fabrication of cathode materials from biochar is another possibility to reduce the overall costs of MFCs, thus making them more sustainable for real applications. Graphitic BC from woody biomass, prepared under high gasification temperature followed by an alkaline post-treatment, was proposed by Huggins et al., [124] to promote the performance of air cathode MFCs (AC-MFCs). The authors indicated the generation of a relatively high power density of 146.7 mW/m^2, especially when MnO was stabilized in the structure of BC (187.8 mW/m^2). This value is higher than that of vulcan carbon electrode (156.8 mW/m^2). They also indicated that the electrodes made of BC are of very low-cost (0.02 $) compared to vulcan carbon electrode (94.80 $. This has resulted in a substantial reduction of the electricity generated using the prepared biochars.

Biochar can also be used as a low-cost precursor for the fabrication of other carbonaceous nanostructures, which can be even more effective in various processes involved in the performance of MFCs. For instance, the preparation of carbon nanotubes using agro-industrial waste including wheat straw, oat hulls, rapeseed cake, and hazelnut hulls under the pyrolysis temperatures of 400 °C and 600 °C was reported by Hildago-Oporto et al., [125]. It was demonstrated that biochars prepared at 600 °C yielded higher CNT concentrations.

Table 3
Modifications in the cathodes to enhance ORRs in MFCs.

Modification	Featured properties	Power density (mW/m^2)	Ref.
Coating of a benthic microbial fuel cell cathode with cerium oxide (CeO_2) NPs	Enhancing the OPRs and power generation by MFCs. Fast transformation in oxidation states in cerium oxide allowed promotion in the charge transfer rate between the electrode and the microorganism.	60	[103]
Polypyrrole (PPy) membrane anode	Low-cost and highly conductive	612	[114]
Production of electrodes based on the candle soot generated CNPs.	Strong mechanical stability and adhesive properties due to the strong particle diffusive bonding, and biocompatibility.	1650	[115]
Synthesis of a composite of cobalt oxide NPs on N-doped carbon nanotubes	Promoted the ORR electro-catalytic activity.	1260[a]	[106]
Fabrication of Pt free catalysts (Ni-based).		1630[b]	[116]
Polymethylphenyl siloxane (PMPS) coated-stainless steel mesh cathode	High performance, low cost, easy to handle and low water loss capability.	2676	[117]
Iron-nitrogen-carbon nanorod network-anchored graphene nanohybrid	Low-cost and efficient ORRs	1601[c]	[118]
α-MnO_2 nanowires-carbon Vulcan composite	A cost-effective alternative for Pt free catalysts with high stability for long-term operations.	180	[100]
Iron oxide-embedded N-doped biocarbon	The porous structure of biocarbon offered rich active sites and rapid mass transfer capability. Iron oxide nanocrystals enhanced the stability of the cathode.	2740	[119]
Fe-N-doped CNPs prepared using coal tar soot as a highly efficient air–cathode catalyst	Abundant ORR active sites, as well as the presence of interconnected macroporous structure	1337[e]	[120]
Graphene supported V_2O_5-nanorod	Superior electrocatalytic activity	533	[121]
TiO_2-MoS_2 nanosheets based on molybdenite exfoliation	High photocatalytic performance (due to the presence of TiO_2, as photocathode to reduce hexavalent chromium (Cr (VI))	147	[122]
Cobalt/nitrogen co-doped porous carbon embedded with carbon nanotubes (Co-N–PC@ CNTs)	A leaf structure of carbon frameworks, with high contents of carbon nanotubes and cobalt nanoparticles accelerated ORRs through a four-electron pathway similar to Pt/C.	2479	[123]

a. 16.6% higher than that of the Pt/C catalyst.
b. 400% higher than that of commercial Pt catalysts.
c Compared to 1468 for the state-of-the-art Pt/C catalyst.
d. Compared to 14 for non-coated anodes.
e. Much higher than that of a cell with Pt/C.

6. Nanomaterials-based membranes for MFCs

Membranes are essential compartments of MFCs. Electrodes (i.e., anode and cathode) are generally separated using PEM. It is vital for the membrane structures in MFCs to support transferring the protons from anode to cathode and to prevent penetrating other elements (such as O_2 and the effluent compartments) [126]. In addition, the membrane should have a high ionic exchange capacity to reach the desired current density. Coupling these two requirements in the MFC membranes has been a technical challenge for the wider application of MFCs [127]. Among the proton exchange membranes, Nafion, and more recently cost-effective polymeric membranes [128] have received the most attention for application in MFCs.

The current trend among the scientific community is to enhance the separation performance of the membrane structures as well as to enhance their mechanical and thermal characteristics. The incorporation of the nanomaterials into the membrane structures has been investigated in recent years to achieve the desired properties and to enhance the performance of MFCs. Iron-based nanomaterials have been considered as good candidates to be combined with membranes for MFC applications due to their non-toxic structure, cost-effectiveness, and relatively ease to produce and scale-up [70,129,130]. Di Palma et al. [131] employed a melt extrusion process for the fabrication of Fe_3O_4/polyethersulfone (PES) nanocomposite membranes, and its efficiency was compared to commercially available membranes (i.e., Nafion 117 and CMI 7000). They used a synthetic effluent prepared from sodium acetate as a carbon source. The prepared membrane with 20 wt% of nanoparticles demonstrated higher performance compared to the commercially available ones and reached a maximum power and current density of 9.59 mW/m^2, and 38.38 mA/m^2, respectively.

The incorporation of nanomaterials into the structure of the membranes is of special importance to adopt the strategies for homogeneous dispersion of the nanomaterials avoiding their aggregation, which can negatively affect the performance of the membrane [132]. However, some strategies have been adopted in the literature to improve the dispersion of nanoparticles. In a recent study [133], it was demonstrated that the presence of sulfonic groups (-SO_3H) within the polymer matrix of the polyethersulfone (PES) allowed a better dispersion of Fe_3O_4 and, as a result, prevented the oxygen permeation from the cathodic compartment.

Self-assembly of chitosan/montmorillonite (1/1, 1/2, 1/4 %w/w) on the MFC ceramic membrane has been presented in a recent study [134] as an efficient way to diminish the oxygen diffusion coefficient of the ceramic membrane (about a hundred times). This can potentially lead to the better growth of exoelectrogenic anodic bacteria. In addition, the electrical double layer capacitance is boosted (4 folds in the mentioned study) and the anode and cathode electrodes charge transfer impedances decrease by 96.44% and 66.14%, respectively. Finally, the Ohmic resistance showed a drop of 73.2%, leading to improved proton conductivity of the modified ceramic membranes.

The CNMs can be considered another alternative to modify the MFC membranes. The limited number of relevant studies has confirmed the effectiveness of membrane-incorporated CNMs to lower the oxygen crossover and to enhance the chemical, tensile, and thermal stability of the membrane. For instance, an aniline-treated polysulfone containing 1% (w/w) of single-walled carbon nanotubes was able to provide a high power density of 304.2 mW/m^2 and columbic efficiency (17%) [135]. Recently, there is a trend in the literature to prepare CNMs from natural resources such as camphor oil [136] and herbaceous biomass [137] to reduce the membrane fabrication costs and avoid releasing the chemicals used as the conventional precursors. Such an approach can be used for further studies towards the fabrication of sustainable membrane structures for MFCs. Some other inexpensive materials have been tested successfully, such as traditional cotton fabric to prepare flexible and stretchable three-dimensional nanocomposite membrane [138], which can be considered as promising and sustainable alternatives for proton exchange membranes due to its ability to transfer the protons.

Membrane fouling is another drawback of the utilization of membrane structures for (waste)water treatment. This may cause considerable sustainability issues such as impeding the efficiency of the membrane and the need for periodically replacing the membrane, which may bring additional treatment costs. From a mechanistic point of view, there are two main phenomena responsible for this process. Internal concentration polarization generally occurs on the surface of the membrane as a result of the alteration in the polarization happening by the changes in the electrolyte concentration. This can potentially lead to the deposition of molecules present in the effluent on the membrane surface [139]. Also, fouling may happen when the concentration of the solutes exceeds their solubility values [140]. Nanomaterials with known antibacterial activities such as Ag [141,142] can aid considerably in this condition to remove the cake layer. For instance, the application of silver nanoparticles for the modification of thin-film composite successfully mitigated the membrane biofouling in a forward osmosis microbial fuel cell [143].

Nanomaterials can be incorporated into the structure of the membranes to improve the conductivity (to facilitate the proton exchange) and anti-fouling properties of PEMs in MFCs. As an example, including the sulfonated graphene oxide (SGO)@SiO_2 into the homopolymer of poly(-vinylidene fluoride) grafted sodium styrene sulfonate (PVDF-g-PSSA) improved the proton exchange capacity (due to the increase in the -SO_3H content of the membrane and minimized the fouling behavior because of the lower roughness of the modified membrane). Table 4 represents the most important finding in the literature about incorporating the nanomaterials in the composition of PEMs of MFCs and the properties that have resulted from such combinations.

7. Sustainability aspects and future outlook

Several biological and physico-chemical treatment technologies have been developed and implemented to treat (waste)water from various municipal and industrial sources [149,150]. As a general statement, the treatment technologies are materials and energy-intensive, bringing additional production costs for the industries [151]. Industries have widely used biological treatment technologies such as activated sludge to deal with low and medium-strength effluents [152,153]. Despite being less expensive and easy to implement, most of these treatment systems represent limited efficiencies in dealing with high-strength effluents containing recalcitrant and non-biodegradable organic compounds [154]. Hence, this is required to apply efficient methods (such as advanced oxidation processes) as the pre-treatment for the biological treatment systems [155,156], which can make the combined technologies less commercially competitive for large-scale power generation [66,157,158]. Under these conditions, the idea of harvesting the energy from the decomposition of organic compounds has attracted attention for industrial effluents with different degrees of strengths and to push them for commercialization [130].

Anaerobic digestion (AD) [159] and more recently, MFCs [10] are the main technologies developed in this regard. AD-based technologies are more mature, and there is evidence of their successful commercialization. MFCs are less mature still suffering from technical and economic issues. Such technologies represent the high theoretical efficiency of electricity generation (≥80%), making them attractive for large-scale applications. However, the actual energy efficiency of MFCs is hindered due to the existing technical barriers. As an inherent issue, the majority of the organisms tend to decompose low-molecular-weight organic acids released into the medium by the activity of fermenting bacteria, which results in the relatively low treatment efficiency for complex organic compounds [66]. There is evidence for applying chemical-based pre-treatment, such as plasma advanced oxidation process

Table 4
Application of ENMs to improve the properties of MFCs membranes.

Membrane type	Membrane properties	Power density (mA/m^2)	Ref.
Nafion membrane	Rapid membrane fouling, caused by a substantial accumulation of bacteria and their end-products forming a thick biofilm layer on PEM.	119.35, and 152.55, for rice straw and potato peels, respectively.	[144]
Fe_3O_4 (20%)/PES	Improved electrochemical, thermal and mechanical stability	38.38 versus 1.12 for PES alone.	[131]
Chitosan/ montmorillonite incorporated ceramic membrane	The improved oxygen diffusion coefficient, better growth of *exo*-electrogenic anodic bacteria, boosting the electrical double layer capacitance, decreasing the electrode charge transfer impedances, dropping the Ohmic resistance, and promoting proton conductivity of the modified ceramic membranes.	1422, during the start-up operation.	[134]
Cationic aniline-treated polysulfone/ single-walled carbon nanotubes (1% w/ w)	Enhancing the mechanical properties, power density as well as columbic efficiency.	304.2	[135]
PVAc-g-PVDF-coated cotton fabric	Inexpensive with enhanced proton exchange properties	400, versus 300 for Nafion-117.	[138]
SGO@SiO_2/PVDF-g-PSSA	Enhanced proton exchange capacity and mitigation of fouling properties of MFCs	185	
Sulphonated titanium nanotubes	SO3H groups of the modified membrane resulted in the creation of ionic channels, minimizing the membrane fouling.	121	[145]
Sil-ver graphene oxide/graphene oxide/sulfonated polyether ether ketone (AgGO-GO-SPEEK)	54% Improvement in proton conductivity than Nafion® 117 membrane.	1134	[146]
Modified Nafion with silver nanoparticles (AgNPs) immobilized on graphene oxide (GO)	Enhanced hydrophilicity, ionic exchange capacity, and of the modified membrane	400 versus for non-modified membranes	[147]
Nitrogen-doped carbon nanotube	Enhanced morphology and presence of N-functional groups.	408, higher than Pt-coated CNT and pristine CNT membrane	[148]
Nafion and activated carbon nanofiber	Higher production power and coulombic efficiency than conventional membranes (Nafion 117 and Nafion 112)	58 compared to 14 for the conventional membranes	[126]

(PAOP), to break complex organic matter into more simple molecular compounds [160].

There are also technical and economic issues related to the design and configuration of MFC, which need to be addressed to push them for commercialization. Bacterial colonization and attachment of bacterial nanowires to the anode (and hence the electron transfer process) is very limited in conventional carbon cloth anodes used in MFCs, leading to the relatively low power density generation [157,158]. Modification of the anode with appropriate ENMs is an effective and efficient way to overcome such limitations. ENMs provide high electrode SSA and more active sites to host the redox enzymes and microbial communities. They can promote the EET by providing direct contact between the electrodes and the microorganisms. Besides, nanomaterials with higher electrical conductivity can enable faster electron transport in MFCs. Hence, the current trend in the literature is to develop sustainable ENMs with specific properties such as cost-effectiveness, advanced electrical and mechanical characteristics, and high resistance to harsh environmental conditions such as low pH and salinities, which can be expected in highly polluted industrial effluents such as brine and pulp and paper mill effluents [161–163]. Recyclability is also of high importance to satisfy the environmental considerations by preventing the generation of wastes after being replaced.

Economic considerations are also of high importance for the development of sustainable MFCs. Conventional electrodes used so far, such as Pt-based electrodes, are generally expensive and limit the commercialization of MFCs. Also, carbon cloth electrodes are fragile, leading to time-by-time failures, especially in large-scale applications [164]. To address such weaknesses, various types of electrodes have been developed and examined very recently. For instance, stainless steel (AISI 304) has represented acceptable performance with no significant corrosion occurring by bacterial activity [165]. Copper is another element that can be used efficiently as anode materials or as the coating of conventional anodes as a cheap alternative [166]. There is also evidence in the literature that copper can efficiently induce colonization of the biofilm with negligible antimicrobial activity.

The origin and composition of the influents can be considered as one of the most important factors to select the most sustainable type of electrodes (especially in terms of durability and operating costs) for MFCs. For instance, hypersaline effluents, such as brine wastewater [167, 168], can cause corrosion for most of the metallic electrodes [164,165, 169,170]. In these situations, there is a need for sustainable materials to produce electrodes with a high degree of stability to resist these conditions. In this regard, recyclable polymeric substrates [171,172], especially when covered with high specific surface area CNTs [173] can be considered sustainable options. Carbonaceous nanomaterials (such as CNTs) can provide a porous structure to host microbial communities and promote the electron transfer process, enhancing the energy output of the MFCs [50]. Novel strategies such as surface doping of the electrodes, especially those prepared using carbonaceous materials, can maintain the lattice integrity. In this regard, the application of excellent p-type nanomaterials for surface doping such as Au_2Cl_6 [174] can be highly recommended. To benefit from the high conductivity of the conventional metallic compounds such as Cr/Ni [175,176], they can be covered with CNTs to make them more resistant to corrosion under extreme operating conditions. Another issue in this regard to be addressed by future studies is the relatively high production costs of the CNTs with the desired properties (e.g., high SSA and high mechanical properties to tolerate harsh environmental conditions). In this regard, seeking cheap precursor materials such as biochar [177] to synthesize carbonaceous nanomaterials, coupled with sustainable production technologies such as ultrasonic irradiation [166,167], can assist the technology to satisfy the sustainability considerations for large scale applications. Novel materials-based carbon quantum dots developed from natural resources can also be considered for the synthesis of highly efficient cathodes to perform ORRs [178].

The trend for applying inexpensive carbonaceous nanomaterials has been initiated very recently, for instance, by applying candle soot for the production of carbon nanoparticles (CNPs) with strong mechanical stability, which is crucial for MFC anodes and cathodes [115]. It is of high importance for the selected synthesis method of carbonaceous nanomaterials to be facile and easy to implement. It would be evident that synthesis routes that can be implemented without the application of costly and complicated infrastructures can considerably reduce the overall costs of the synthesis process [179–181]. The development of such sustainable technologies is highly important, especially in arid regions of the world with limited access to freshwater resources. It is worth mentioning that the majority of conventional technologies for water desalination, such as forward osmosis is still energy-intensive and expensive [182,183]. In this regard, some attempts have been initiated in the literature, such as the application of MFCs to generate power

from the Barnett Shale-produced water hypersaline autochthonous bacteria and providing the energy to enable a desalination unit to reuse the hypersaline effluents [184].

In addition to electrode materials, the output power of an MFC is influenced by the properties of the membrane for the efficient transferring the hydrogen ions. As stated before, among the various developed configurations for MFCs, H-type is the most widely studied and implemented configuration, which normally uses proton exchange membranes. However, conventional PEMs are not cost-effective and efficient enough for large-scale applications [128,185]. For instance, rapid fouling is highly expected when Nafion membranes are used in MFCs, which can drastically affect the Coulombic efficiencies (CEs), and the power densities generated [144]. There are some reports in the literature for the modification of membrane structures with specific types of nanomaterials. Polymeric membranes can be considered sustainable alternatives in this regard with high efficiency, low fouling and low production costs. Further improvement of such membranes towards satisfying the sustainability considerations such as the inclusion of charged species as well as the incorporation of ENMs (e.g., TiO_2 and ZrO_2) have been adopted for the efficient prevention of fouling [186,187].

Economic considerations are also of extreme importance as one of the main pillars of sustainability when selecting the most appropriate ENMs enabled MCF technologies for real (waste) water treatment applications. In this regard, the costs associated with the electrode materials have been considered as significant challenges for the large-scale application of these technologies. According to a rough estimation, about 50% of the total capital costs of the MFCs is associated with the cathode materials [188].

There have been some attempts to reduce the overall costs of the cathode materials in MFCs. Choi et al., [116] argued that the electrophoretic deposition (EPD) method could be used as a facile and cost-effective technique to produce Pt group metal-free (PGMF) cathode nano-catalyst to promote large-scale applications of MFCs. In this sense, a significant cost reduction in cathode materials has been achieved by Chen et al. (2012) [117]. They developed a low-cost (6$/kg) polymethylphenyl siloxane (PMPS) coated stainless steel mesh cathode with acceptable performance for the ORRs.

Considering that the membranes and electrodes account for over 80% of the total MFC costs, a considerable reduction up to 30–40% can make these technologies more competitive to other (waste)water treatment methods. Utilization of inexpensive raw materials (including recycled or naturally occurring materials) as well as the development of facile and cost-effective methods can be considered a potential approach to reduce the costs of MFC fabrication [189]. For instance, Feng et al. [190] concluded that among the high-performance materials, SS electrodes (SUS 304) are the cheapest option (21 k$/$m^3$), followed by titanium plate and graphite felt (30 k$/$m^3$, and 25–75 k$/m^3, respectively). Grattieri et al., [173] stated that preparation of the anode using a recyclable polymeric substrate with a conductive pain based on CNTs depends on the thickness of the coating layer and varies from 10 $/$m^2$ to 50 $/$m^2$. Estimations are also available for MFC membranes. As an example, successful application of novel nanocomposite membranes (e.g., SGO@SiO_2/PVDF-g-PSSA) to enhance proton exchange capacity and lower membrane fouling has recently been reported in the literature, which can overcome the existing technical issues of MFCs for real applications. Table 5 summarizes the cost-estimations presented by the recent studies for the inexpensive nanostructured materials developed for MFCs. However, there is still a need for more studies on the cost-effectiveness of major research findings in this regard, which can pose difficulties for their sustainability assessments. It is worth mentioning here that most of the reports on the development of novel materials for MFCs lack such economic analysis. More research efforts are highly welcome on the cost-effectiveness assessment of the novel promising technologies discussed in the present review to assist in the commercialization of the most efficient, environmentally friendly, and economic MFC-based technologies.

Table 5
Cost estimations/considerations of the materials recently developed for MFCs.

Component	Composition	Cost evaluation/ consideration	Ref.
Anode	A recyclable polymeric substrate with a conductive CNT	The thickness of coating layer varies from 10$/$m^2$ to 50$/$m^2$	[173]
Anode	Self-assembly of 3D CNT sponges	The costs of raw materials for producing CNT sponge was estimated to be ~ 0.1$/$g_{CNT}$, much less than conventional technologies	[191]
Anode	Corncob-derived 3D N-doped macroporous carbon foam	2–4$/$m^3$, much less than the commercial carbon brush electrode (~270 $/$m^3$)	[192]
Anode	Breadderived carbon anode	~5–8$/$m^3$	[193]
Anode	Graphene-like molybdenum disulfide (GL-MoS_2) nanoflowers	Not given	[98]
Cathode	Polymethylphenyl siloxane (PMPS) coated stainless steel mesh	A significant cost reduction in cathode materials (0.006$/g) was achieved	[117]
Cathode	MnO_2-NTs/graphene composite (with 70% MnO_2-NTs loading)	3.51$/g, much lower than the Pt/C benchmark (with 10% Pt loading, 26$/g)	[194]
Cathode	MnO2/polypyrrole/MnO2 nanotubes (NT-MPMs)	Raw materials for NT-MPMs cost less than 3% compared to the Pt/C (Pt 20%) benchmark	[46]
Cathode	Nanorod β-Ga_2O_3 modified activated carbon	Ga_2O_3 costs 2.3$/g, much less than Pt (66 $/g), as the benchmark	[195]
Cathode	Functionalized carbon nanotubes (CNTs) with poly (diallyldimethylammonium chloride) loaded $CoMn_2O_4$ nanoparticles	0.2 $/g of the prepared electrode	[196]
Cathode	Candle soot-derived carbon nanoparticles	Negligible, compared to the 10% Pt/C benchmark (42$/g)	[115]
Cathode	Duckweed derived nitrogen self-doped porouscarbon materials	~1.2 $/g, much cheaper than commercial Pt/C electrocatalyst ($61.8/g)	[197]
Cathode	Nitrogenous porous carbon embedded cobalt nanoparticles treated at 800 °C	0.12 $/g	[198]
Cathode	Highly graphitic nitrogen-doped carbon nano-onions	0.29 $/g, about 520 times lower than that of commercial Pt/C.	[38]
Cathode	V2O5/rGO composite	4.8 $/g	[121]
Cathode	Porous iron–nitrogen-doped carbon nanospheres	1.16 $/g	[199]

8. Conclusions

This review addresses the applicability of ENMs to overcome the existing issues for the commercialization of MFCs, such as power density, stability, and cost-effectiveness of the electrode materials. Direct electron transfer in MFCs can be promoted by the colonization of microbial communities mediated by ENMs. Conductive nanomaterials can also directly enhance the electron transfer rate in MFCs. The increase in microbial density and diversity can also provide large SSA and active sites for redox reactions to occur. Modified MFCs represented various power densities (from less than 100 to around 2700 mW/m^2), and in most of the cases, modification with nanomaterials increased the power density

of the MFCs. In addition, the incorporation of ENMs in the structure of MFC membranes has led to an increase in the proton exchange and preventing membrane fouling.

Economic analysis performed in the present manuscript can also indicate a considerable cost reduction in the production of electrode materials in MFCs. It is also worth concluding that despite the potential of ENMs-based microbial fuel cells to satisfy the technical and economic considerations, there is not any report available yet for the large-scale application of such technologies. In this regard, low-cost ENMs such as carbon nanotubes prepared using novel technologies can replace the conventional electrode materials such as Pt/C which can facilitate the utilization of such sustainable technologies in large-scale wastewater treatment processes.

CRediT authorship contribution statement

Mohammadreza Kamali: Methodology, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Tejraj M. Aminabhavi:** Conceptualization, Validation, Writing – review & editing, Visualization, Supervision. **Rouzbeh Abbassi:** Writing – review & editing. **Raf Dewil:** Conceptualization, Validation, Writing – review & editing, Supervision, Funding acquisition. **Lise Appels:** Conceptualization, Validation, Resources, Writing – review & editing, Supervision, Funding acquisition.

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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