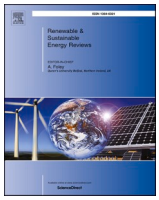
Renewable and Sustainable Energy Reviews 188 (2023) 113813

Contents lists available at ScienceDirect

Renewable and Sustainable Energy Reviews

journal homepage: www.elsevier.com/locate/rser



Recent advancements in the development of Two-Dimensional nanostructured based anode materials for stable power density in microbial fuel cells

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ARTICLE INFO

Keywords:
Microbial fuel cell
2D nanomaterial
Power density
Nanocomposite
Anode

ABSTRACT

The demand for alternative energy sources from non-recyclable waste materials will be a hot research topic in future industries. This interest is primarily due to the ability to harness energy from waste materials, the provision of localized power solutions, and the promotion of efficient power conversation. In this respect, Microbial Fuel Cells (MFC) represent new energy sources possessing unique qualities for many applications. MFC generates power by utilising exoelectrogens forming the biofilm on the surface of the anode. Since in the MFC, wastewater is primarily converted into protons and electrons at the anode surface, where biofilms typically develop, the anode becomes the most vital component. Consequently, significant research has been conducted on anode material to improve MFC performance. The present review focuses on different aspects of the MFC, including a comprehensive summary of the recent developments in the field of MFCs, including a state-of-the-art literature review based on a bibliometric analysis using keywords, a description of the mechanism and operational principle of MFC, applications and a summary of current research perspectives including the use of carbon nanotubes, graphene, graphitic carbon nitride, MXene, and their nanocomposites as anode materials with stable power density performance. Lastly, we present the challenges and future perspectives regarding the continued development of MFC anode materials, culminating in overall conclusions related to MFC research.

1. Introduction

Population growth and increasing urbanization are accelerating the global economy, resulting in greater energy demand. However, fossil fuel resources are non-renewable, and their use results in significant environmental pollution and there is a real need to find sustainable alternatives [1]. One such technology is the microbial fuel cells (MFC) which uses specially adapted microorganisms fed on organic waste to generate electricity [2]. A basic dual-chambered MFC contains a cathodic and an anodic chamber that are segregated by a proton exchange membrane (PEM). Within the cell, anodic exoelectrogens, i.e., microorganisms that can transfer electrons extracellularly, break down organic substrates while at the same time producing electrons and protons [3,4]. The electrons are then transmitted from the microorganisms to the anodic surface and ultimately to the cathode via the external circuit, whereas protons get internally transmitted to the cathodic chamber via the PEM. This process results in a continuous generation of current while simultaneously treating wastewater [5]. Accordingly, oxidation at the anodic region and reduction at the cathodic region enable electron transfer to the cathode from the anode, thus completing the circuit [5].

Although MFC has gained recognition as an environmentally friendly renewable next-generation technology, they still have a long way to go

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https://doi.org/10.1016/j.rser.2023.113813
Received 14 December 2022; Received in revised form 27 July 2023; Accepted 1 October 2023
Available online 12 October 2023

Abbreviations:

MFC	Microbial Fuel cell
2D	Two-dimensional
3D	Three-dimensional
CNTs	Carbon nanotubes
MWCNTs	Multiwalled carbon
nanotubes	
g-C_3N_4	Graphitic carbon nitride
Gr	Graphene
PEM	Proton exchange membrane
ORR	Oxygen reduction reaction
CB	Carbon brush
CC	Carbon cloth
CP	Carbon Paper
CR	Carbon Rod
GF	Graphite felt
GO	Graphene oxide
rGO	Reduced graphene oxide
NCNTs	Nitrogen-doped carbon
nanotubes	
TiO2	Titanium dioxide
EET	Extracellular electron transfer
Iron (III) oxide:	Fe_3O_4
PANI	Polyaniline
PDDA	poly(diallyl dimethylammonium chloride)
GCE	Glassy carbon electrode
PTFE	Polytetrafluoroethylene
WO_3	tungsten trioxide
SnO_2	Tin(IV) oxide
MOFs	Metal-organic Metal-organic framework
MnO_2	Manganese oxide
WC	Tungsten carbide
Au	Gold
CHI	Chitosan
Ti3C2	titanium carbide
CeO2	Cerium(IV) oxide
ZIF	Zeolite imidazolate framework
$NiFe_2O_4$	Nickel ferrite
$NiWO_4$	Nickel tungstate
ZnO	Zinc oxide
Ac	Activated carbon
MoS_2	Molybdenum disulfide
PPy	Polypyrrole
APTES	(3-Aminopropyl)triethoxysilane

before becoming commercially applicable which is mainly due to insufficient current production. This low current is mainly a result of inefficient MFC components such as electrodes and membranes, poor biofilm formation, lack of suitable catalysts, and high operating costs associated with electrode materials [60]. The choice of anode material is also crucial since it can results in weak microbial adhesion on the electrode surface, reducing electron transfer [60]. The main strategies to increase microbial surface attachment are surface modification of anode material and exoelectrogens selection. Amongst all the different components that influence MFC efficiency, the anode is the most crucial, as it in not only responsible for the transfer of electrons upon adhesion of microbes, but also dictates the overall cost of the MFC [61].

The advent of the nanomaterial era and fabrication process with the recent developments in nanotechnology have significantly improved MFC performance [62]. Nanomaterials show different novel properties with respect to their bulk counterparts. For example, a high surface-to-volume ratio provides maximum surface area for biofilm formation, enabling the microbes to accelerated biocatalytic activity. Furthermore, nanoscale materials or metal catalysts can improve the rate of oxygen reduction reaction (ORR) [63]. Moreover, metallic oxide nanomaterials display antibacterial properties, enhancing the electrode's performance and shelf life [64]. Today, 2D planar nanomaterials are extensively utilised for their ability to provide more active reaction sites. Such modifications have helped improve MFC performance for eco-friendly applications such as renewable electricity generation, wastewater treatment and recycling and environmental pollutants biosensing.

Conventional carbon materials, e.g., carbon cloth (CC), paper (CP), rods (CR), and graphite felt (GF)) [65], are suitable anodic materials in MFC fabrication due to properties like highly specific surface area, elevated electric conductivity, and chemical/structural stability [66]. However, an of these materials is their porosity; microbes can get trapped in the pores causing bactericide, thereby reducing the cell's overall performance. Alternatively, graphene-based nanostructures such as carbon nanotubes (CNTs), graphene and its oxides (GO and rGO), graphite, graphitic carbon nitride, and MXene are promising anodic materials having enhanced properties such as mechanical strength, thermal stability, non-corrosiveness, and electric conductance [65]. Nanomaterials such as CNTs, GO and rGO have also been incorporated into polymer/metallic materials to enhance these beneficial characteristics further [67].

The selection of microorganisms is crucial in determining the stable power density. Modern biotechnological tools can help better understand the selection of desired microorganisms for MFC. The microbial selection for the MFC depends on several parameters for power generation, and screening of electroactive species is essential [68]. There are few reports for screening electroactive bacteria using MFC arrays, nano probes, and MFC based on U-type tubes [68]. However, these methods are laborious and provide only semi quantitative and qualitative data. Recently, a novel electron shuttle protocol has been reported to screen potential electroactive bacterial, which can efficiency reduce ferric ions (Fe^{3+}) [69]. In recent years, the research is mainly focused on *Geobacter sulfurreducens Escherichia coli, Achromobacter insuavis, Cronobacter sakazakii, Kocuria flava, Bacillus encimensis, Shewanella algae, Cronobacter sakazakii, Pseudomonas otitidis* and *Pseudomonas otitidis.* [70,71] These microorganisms are used in MFC as a single strain or in a consortium to maximise power density. Recently, *Shewanella algae* which is known for its iron reduction capacity, was proposed as an effective catalyst for MFC [72]. It is also essential that any microorganisms used do not produce harmful by products. In recent years, yeast based MFC has also gained importance because of its simplicity in operation and easy to isolate. *Saccharomyces cerevisiae* is prominently used in MFC can efficiently utilize a variety of substrates, making them versatile for high power density [73–77]. Yeasts tend to metabolize a wide range of organic substrates, which can be advantageous in waste water treatment plants. Electron transfer in yeast are similar like bacteria which is through cellular membrane and involved direct and mediated electron transfer.

Electron transfer at the electrode surface when using microorganisms is vital for the performance of MFCs towards achieving stable power density. Several microorganisms that can exchange electrons with nanomaterials as mediators have been studied in depth. This bioelectrochemical system is a new revolution in MFC research as it generates electrical current using microorganisms acting as biocatalysts in the anode chamber [78,79]. To further elaborate the work in the direction of microbial attachment to solid surfaces, researchers have focused on optimizing the electrode material, which enhances the

attachment and formation of biofilm on the material's surface.

Recombinant strains (RS) of microorganisms are becoming more popular due to their adaptability and high power outputs [80–82]. In the case of RS, biofilm formation involves complex adhesion, growth, maturity and cell-to-cell communications processes. Adhesion is mainly through extracellular DNA, proteins and polysaccharides. Extracellular DNA is a nucleic acid released from the inside to the outer membrane of bacteria that provides rigidity and cell protection from host defence mechanisms response [83]. This extracellular DNA provides structural integrity to the biofilm and promotes bacterial adhesion and structural stability, promoting electron transfer efficiency. In addition, an increase in extracellular DNA makes the cell membrane more hydrophobic, thereby enhancing cell adhesion on the electrode surface [84].

Researchers have engineered the extracellular DNA of *S. oneidensis* via various biology-driven strategies that allow it to attach itself to the electrode surface efficiently. The cell layers can generate a current density of 39 μA/cm^2 [85]. Another study used engineered *S. oneidensis* $G_7\Delta RSL_1$ researchers utilised glucose and lactate to improve cell adhesion. The authors constructed a $G_7\Delta RSL_1$-rGO/CNT biohybrid with a maximum power density of 560.4 mW m^{-2} and 373.7 mW m^{-2}, the highest reported for a genetically engineered *S. oneidensis* [86]. This study provides a new strategy to facilitate practical applications of MFC in wastewater remediation and efficient power recovery. It was also reported that cytochrome-c with extracellular polysaccharides has the properties for cell adhesion and attached cytochrome-c for extracellular electron transfer (EET) [87]. Several researchers have also reported increased power density and promotion of EET utilising RS [88–90]. Ultimately, MFC power performance depends on several parameters, such as the selection of microorganisms (electroactive), biofilm formation, electrode material, and type of MFC [70].

Microbial adhesion properties are depended on adhesion proteins and structural proteins (flagella and pili) which are also responsible for the electron transport [68]. There are few reports on the possible direct electron transfer mechanisms from the substrate to the electrode and biofilm formation [91,92]. Indirect electron transfer is mainly through internal or external mediators (redox active materials), responsible for electron flow between microbial biofilm to the anode surfaces. Direct electron transfer requires either direct attachment of cells or via nanowires. Fig. 1a shows a typical morphology of bacterial cell with different components involve in attachment and electron transfer. A possible mechanism of electron transfer (Fig. 1b) and the interaction of bacteria on surfaces using different parameters (Fig. 1b) [93] are also illustrated.

Carbon-based nanoscale materials with tailored dimensions and functions are being increasingly used depending on their features at the nanoscale level. A promising area of research in MFC that has garnered immense interest is the development of various engineered anode

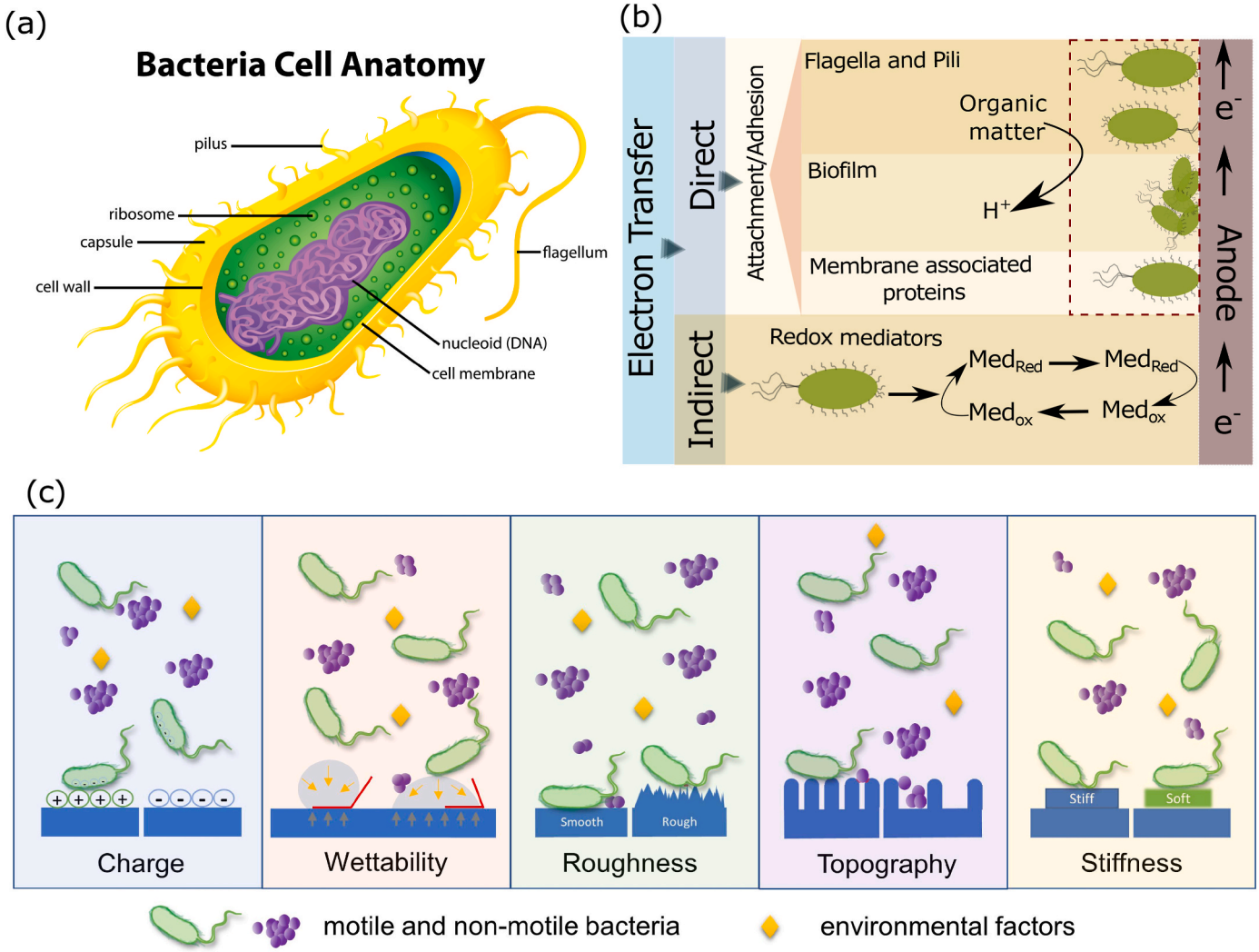

Fig. 1. (a) Morphology of bacteria (source attribute www.vecteezy.com) (b) Schematic illustrations of the mechanism of extracellular electron transfer in electrochemical microorganisms and (c) the influence of different parameters on the adhesion properties of bacteria on different surface properties Adapted with permission from Ref. [93] Copyright 2021, Frontiers in Bioengineering and Biotechnology.

design, modification, and preparation methods [94].

This comprehensive review has been organized to overview current advances in anode materials and their nanocomposites for MFC applications with enhanced power density. Among 2D materials, CNTs and graphene have been widely used for application because of their excellent semiconductor nature and thermal stability. However, several nanostructures have been doped or integrated with graphene/CNTs to boost the desired characteristics for electrode fabrication applications. Few research groups have published comprehensive reviews on applying and using potential nonmaterial-based anode in MFCs and their importance in power generations [66,95,96]. Functional nanomaterials are primarily used to modify the anode surface for better adhesion of bacteria and improved EET. A detailed explanation of exoelectogens, nanomaterials, surface modifications and bacterial surface modification is given by Jiang et al. [97] Together with this, the mechanism will help to design better electrode material for MFC. Similarly, in yet another review, Lu and coworkers provides an interesting discussion on the performance loss of MFC anode towards electrochemical studies [98]. They show how this is possible via anode surface improvement through modifications, biocompatibility, and increased surface area for adhesion, facilitating improved EET.

Conducting polymers, natural anode materials (bamboo charcoal, carbonized form of mushrooms and corn) and composite nanomaterials are ideal for constructing the anode material for MFC [67,99,100]. Slate et al. provides a detailed overview of the bacterial attachment to the surface of the anode, mechanisms of EET via pili and electron shuttles, optimization parameters, MFC configurations, and electrode materials [101]. Recently, the focus has centred on improving the power density of MFC by improving the bacterial strains, incorporating novel 2D nanomaterials, and better 3D nanocomposite for the adhesion of bacteria to form biofilm [5,66,95]. These above reviews presented the interesting and narrative work towards improving MFC, but lacks details of new emerging materials, appropriate power densities and preparation of nanocomposite materials. To identify the importance of new emerging materials and their potential application in MFCs, we have listed the various anodic nanomaterials and well-known modification techniques used to enhance anode performance. The review also includes the importance of g-C_3N_4 and MXene as possible future anode materials and their nanocomposites suitable for enhancing power output for MFC. This review focuses on the recent developments in MFC anode materials using CNTs, graphene, g-CN, and MXene nanocomposites (Fig. 2). Finally, the review ends with the current challenges, and future

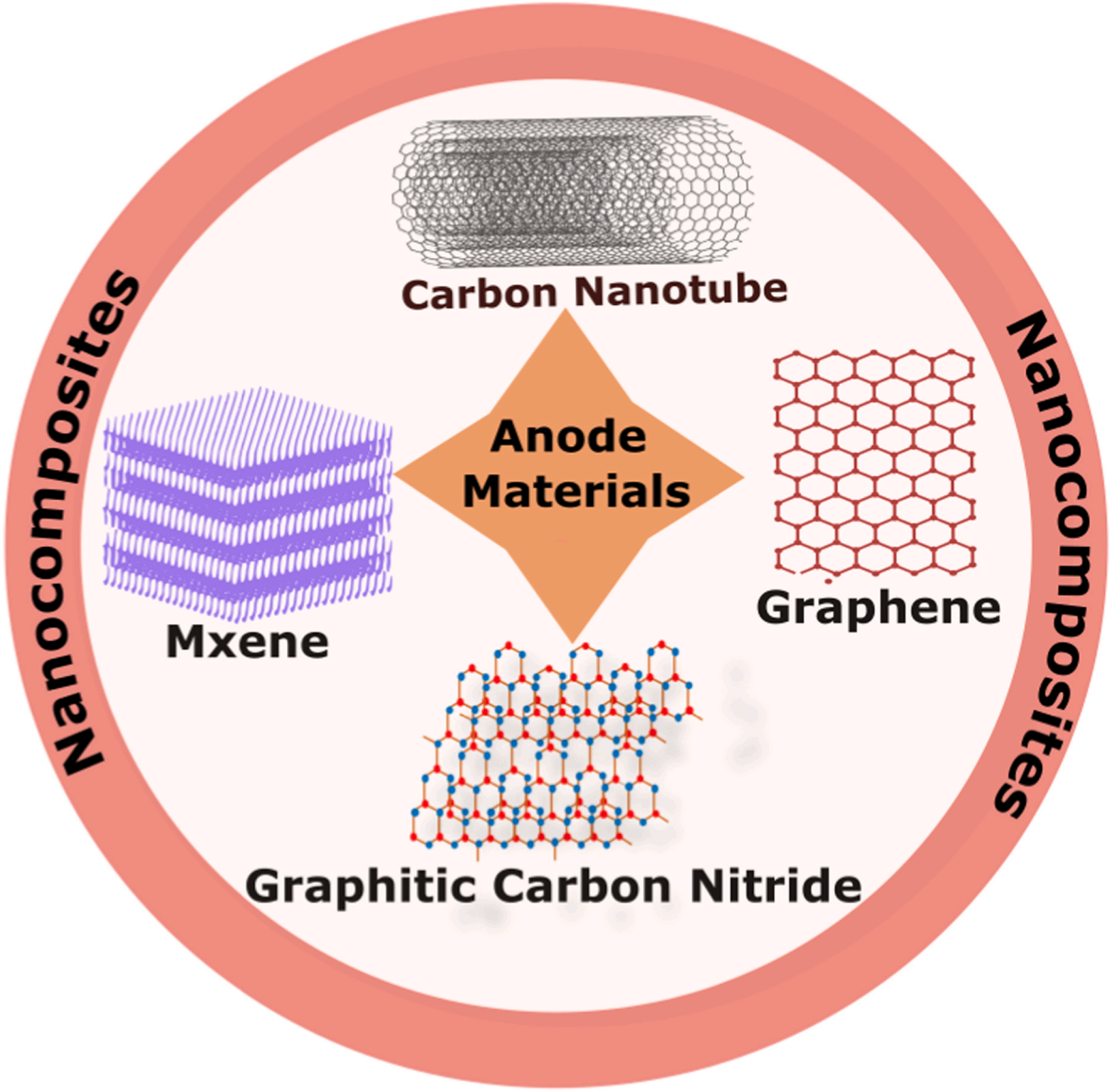

Fig. 2. Overall illustration of the present review comprising of different nanomaterials employed for anode fabrication.

perspectives and overall conclusions.

2. Bibliometric examination

The bibliometric study of MFC was centred on utilising different nanostructures as potential anode modifiers for biofilm formation in waste water treatment. Research professionals in this discipline may use this knowledge as a base to analyse and develop new MFC fabrication strategies. Elsevier's database for abstracts and citations, Scopus, contains relevant data and its linked research articles from many different areas. This platform was utilised to review recent academic and scientific articles on MFC research. Research articles were searched using different key word queries such as ("microbial fuel cell" or "MFC"), ("anode" or "anode modification"), and ("nanomaterials" or "nanocomposites"). The search produced 115 articles that matched the keyword criteria, ranking chemistry, energy, engineering, chemical engineering, and environmental sciences as the top 5 fields of study. The operators "or" and "and" were used to guarantee that the final search result had at least one of the phrases in between the parentheses whereas the quotations ("") were required to scan the publications for the precise terms whilst eliminating synonyms. Eighty-six research papers and 14 literature reviews were found in journals from Jan 2010 to Jan 2023 as a result of this database search. The critical publishing years for this field were found to be from 2020 to 2023, which suggests that MFC is a more recent field of study.

In this study, the occurrence of the search topics was analysed and visualised using VOSviewer, and a map built on the bibliographical information was produced (Fig. 3). The circles indicate where the search words appear most frequently in the chosen database; the wider the circle area, the more frequently the search words appear, demonstrating their similarity. As a result, the association is stronger when the circles are closer together and the lines are shorter. The co-occurrence of keywords analysis and counting method was used to create a map by choosing search words that appeared at least five times. From a total of 1439 search words, 137 met the criterion, and these were then organized into five major groups. Cluster 1 comprised 99 words, most associated with MFC and their uses. Most of the 70 words in Cluster 2 were related to the anodic electrode for bioenergy and bioelectric applications. Cluster 3 contained 55 words related to the use of nanomaterials for anode fabrication. The 36 words in cluster 4 primarily describe the various nano structures used for MFC development. Lastly, there are 12 words in cluster 5 associated with the transport of electrons and the highest power output in MFC. At least five instances of each phrase were stipulated in order to determine the trending regions, yielding 120 words that do not fulfil the requirements. To generate the current hotspots, the timeline was adjusted from 2020 to 2023, and the following major search words were reported: "Microbial fuel cells", "nanocomposites", "biofilm", "power density", "nanomaterials", "anode", "electrode", and "MFC".

3. Operational principle and setup of microbial fuel cell

MFC is a set-up that uses bacteria to transform chemical energy into electricity by degrading organic material present in sewage [66]. The advantages of applying MFC as an alternate energy source include synchronised treatment of sewage and electricity production from

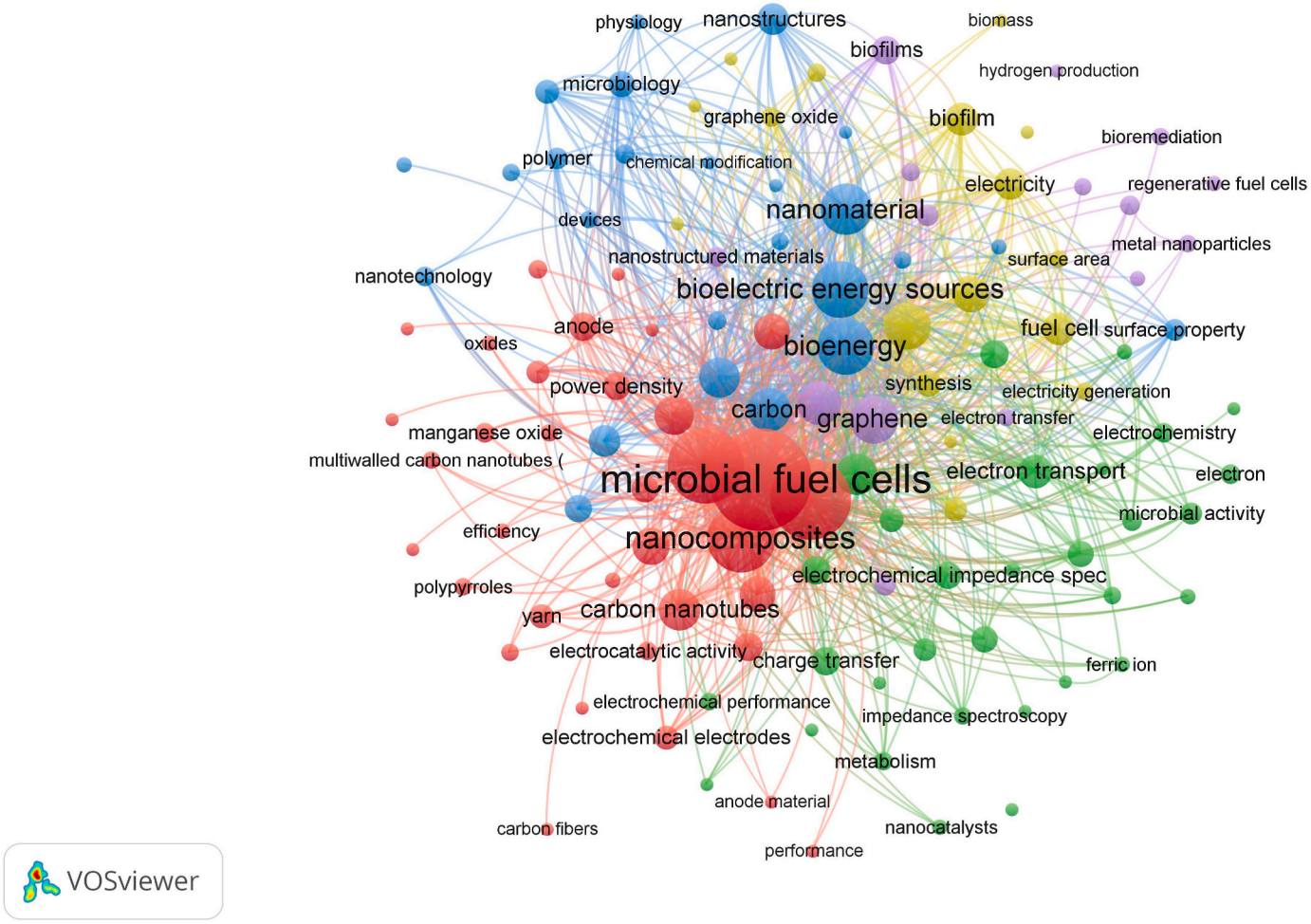

Fig. 3. Network map created using bibliometric data of co-occurrence of keywords (2020–2023) related to a directory of MFC publications.

organic matter, demonstrating MFC as a cutting-edge and proactive research area. In MFC, the electrodes and the bacteria work together to create biofilms on the anodic surface. This kind of MFC simplifies the process for complex organic matter to be transformed into a resource for energy production via microbial metabolism. Potter, in 1911, introduced and brought forth the original idea of MFC [102]. In principle, MFC consists of anodic and cathodic chambers segregated by a PEM (Fig. 4). The anodic compartment consists of a substrate and bacterial solution made up of *Escherichia coli, Shewanella*, *Gammaproteo, Geobacter,* and/or *Deltaproteo*.

Exoelectrogens act on the substrate in the anaerobic compartment to metabolically oxidise it and produce protons and electrons (Fig. 4) [3,103,104]. The electrons travel to the cathode, linked via an externally connected circuit, after directly or indirectly reaching the anode. Parallelly, protons (H^+) enter the cathodic compartment via PEM, wherein they engage in a reducing process with oxygen (O_2) to generate water (H_2O). In this way, a steady current is produced while simultaneously treating sewage. Substrates play a critical role in MFC performance and with proper choice (sugars, glucose, cellulose, and starch), oxidation of the microorganisms has been attained [105]. Selecting appropriate fabrication material for MFC has enabled the generation of sufficient electricity from various feedstocks (industrial, animal, and sewage). Elements that affect the functioning and efficiency of MFC comprise of the source of the substrate, PEM, and type of anode [106]. In three types of ways, microbes transmit electrons to the anodic surface, i.e., direct/mediator-less transmission, indirect/mediated transmission, or via nanowires [107]. Another electron transfer mechanism is to the anodic surface through the cell membrane [107,108].

MFC consist of two types: Single and double chambered cells. The primary purpose of MFC is to converts organic matter into electrical energy via microorganism mediated metabolic events [109,110].

MFCs, either single or dual, have the following specific advantages and limitations.

- The anode and cathode are placed in the same chamber in a single-chamber MFC. Compared to a single MFC, two separate compartments housed individual electrodes with a proton exchange membrane (PEM) or ion exchange membrane separating the two compartments. The design selectively allows ions to flow and maintain the charge at the anode and cathode compartments.

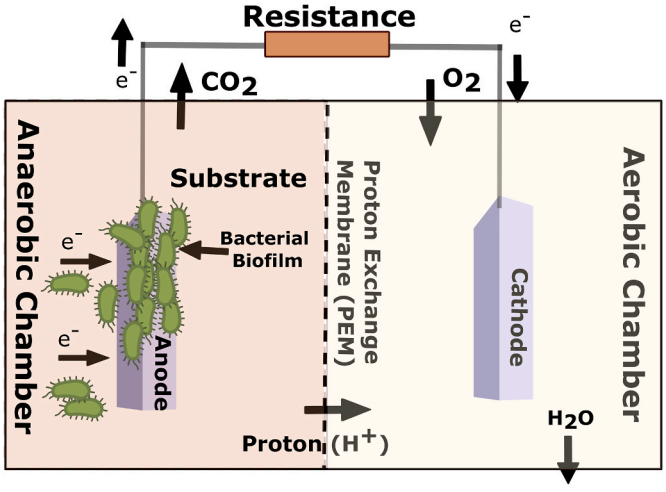

Fig. 4. A schematic showing the different parts of a Dual-chamber MFC. The anaerobic chamber consists of an anode electrode covered with bacterial biofilm and filled with substrate which undergoes metabolic oxidation via exoelectrogens to generate protons and electrons. The electrons produced at the anode further arrive at the cathode in the aerobic chamber through the PEM after reacting with O_2 resulting in reduction to H_2O.

- In single-chambered MFC, both electrodes are close to one another. Whereas in the dual chamber MFC, the electrodes are separated in two compartments which prevents them from direct contact ruling out short-circuiting of electrons with further interacting/mixing of oxygen to the anode.
- Single MFCs are simple to construct and operate, require fewer components and are less sophisticated in design. A dual chamber possesses a complex structure due to the separation of the two electrodes.

The performance of MFC depends on the following factors, including (a) anode and cathode material, (b) the substrate used as the catalyst and (c) PEM. Apart from this, there are other characteristics of the electrode material that contribute towards improving the ability of MFC such as (a) surface area of the material, (b) porosity of different anode materials, (c) electrical conductivity associated with the material, (d) material cost, and (e) Stability over a wide range of environmental conditions.

4. Anode modification

Surface modification is designed to enhance the compatibility of microorganisms to the substrate, such as.

- Better microbial adhesion: Modification of anode creates surfaces more compatible with microbial attachment. It can be achieved by roughness/functionalization of the surface of the MFC anode. This facilitates and provides a large surface area, allowing Pilli, flagella and cellular proteins to attach and a biofilm to form on the surface of the anode [101].
- Improved EET: Modification can also facilitate the improved EET between bacterial community and anode. Conductive nanomaterials enhance the electrochemical activity between bacterial and anode surfaces by promoting direct EET or mediated electron transfer phenomenon, efficiently transferring electrons between the microbial cell and anode [5,66,95].
- Promote microenvironment: Modification can also create a microenvironment suitable for the growth of bacterial community and activity [70,71]. This can provide the anode surface with appropriate pH, availability of nutrients for microorganisms, and redox potentials, which are essential for the activity of cells. By customizing the properties of the anode material surface, it is possible to create a microenvironment that is more conducive to microbial compatibility and improves the performance of the MFC.
- Inhibition and Toxicity: Some anode modification methods can reduce the toxicity/inhibition of the microorganism activity. For example, applying a special coating, polymers, and material on the surface of an anode provides a shield, protecting the microorganism from harmful substances present in wastewater. This shielding allows the microorganism to proliferate, ensuring long-term compactivity with the anode [111].

Anode surface chemistry is essential in microbial adhesion and biofilm formation, improving the overall EET in MFC [112]. Functional groups, surface charges and hydrophilic/hydrophobicity of nanomaterial which render the electrostatic interaction, cell surface interaction and wetting properties. Chemical and thermal treatment of the anode was also reported earlier, facilitating the attachment of microorganisms and biofilm formation, resulting in enhanced EET [113]. Surface roughness, charge, and wettability enhance the cell adhesion and promote biofilm formation (Fig. 1c). To overcome the above issues, different 2D nanomaterials have been used to increase the power density and biocompatibility of anode material. Different nanocomposite-based anode materials are presented in the below sections.

5. Carbon nanotubes-based nanocomposite electrodes

Due to its distinctive physiochemical qualities, for example high conductivity, excellent mechanical strength/flexibility, large surface-to-volume ratio, and suitable biocompatibility, CNTs with CP, CC, and CF, have been utilised as modifiers in anode fabrication for MFC [114]. In this section, a detailed discussion has been carried out on CNTs and nanocomposite-based materials which has been used previously and recently by researchers as potential anode materials in MFC. The anode modification with CNTs and other materials (polymers, metal oxides and 2D materials) has been briefly outlined to examine their effectiveness in improving the power density of MFC.

Modifying a CNTs structure or doping with different metals are ways to increase the power density in MFC. In this regard, a recently developed facile method to synthesize nitrogen-doped CNTs using bamboo-like structures (Bamboo-NCNTs) was developed by catalytically pyrolyzing ethylene diamine as anode material in MFC [115]. The as-prepared material has the defect region of Bamboo-NCNTs-MFC which results in larger surface area and can achieve a very high power density of 1040 mWm^{-2}. The advantages of using Bamboo-NCNTs was their higher specific surface areas (205.3 $m^2\ g^{-1}$) compared to bare CNTs (165.8 $m^2\ g^{-1}$) which was calculated using nitrogen adsorption-desorption measurement. Nitrogen in doped Bamboo-NCNTs may promote desired microorganisms to adhere to and proliferate on the electrode surface, lowering the potential and boosting biocatalytic performance.

Inorganic compounds are also being used alone or in combination with different nanomaterials to improve the performance of MFC, either as a support, carrier, membrane or catalyst itself [116]. Taking the advantages of the above properties, a facile synthesis method was developed to prepare a nanohybrid nanocomposite of CNTs decorated with TiO_2 nanoparticles [117]. The as fabricated CNTs@TiO_2 nanohybrids are very attractive because the CNTs@TiO_2 may be economically feasible and effectively synthesized without requiring any strong surfactant treatment or harsh environmental conditions. Additionally, the resulting CNTs@TiO_2 showed significantly enhanced activity as the anodic electrode material in MFC compared to pure CNTs or TiO_2 nanoparticles alone. The nanocomposites prepared with this method generate a topmost power density of 1120 mWm^{-2}, about 1.5–1.7 times more than those obtained with CNTs-MFC and TiO_2-MFC (730 mWm^{-2} and 670 mWm^{-2} respectively).

Surface modification of the anode with various minerals and their oxides using nanoscale engineering methods has been applied to efficiently facilitate the transmission of electrons in-between the bacterial biofilm and the anode [118]. The effective transmission of electrons relies on the adherence of the microbes and development of biofilms on the anodic surface. Several anode modification techniques, such as 2D nanostructure fabrication, have been devised to enhance power density, EET, and the electrodes' capacity to receive electrons [94]. Iron (III) oxide (Fe_3O_4), occurring in nature as mineral magnetite offers a platform for electroactive microorganisms (EAMs) to transmit electrons to the outer side of the cell's surface to enhance the EET [118]. It is regarded as the optimum substance for altering the MFC anodic surface because of its availability, affordability, superior electric conductivity, and high level of biocompatibility [118]. With the above properties of Fe_3O_4, Fe_3O_4/CNT nanocomposites were produced and utilised as anodic materials in MFC [26]. Using this nanocomposite, an increase in power density was observed [26]. The first mono-dispersed $FeSO_4$ on CNT was prepared using a solvothermal method to achieve this. Afterwards, the Fe_3O_4/CNT nanocomposite was mixed with *Escherichia coli* culture to create an *E. coli* and Fe_3O_4/CNT network. Next, employing magnetic fields, a thick multi-layered network was deposited onto the carbon-based paper electrode and applied as an anode. With a large surface area to enable microbial growth and improve anodic EET, CNTs were bound to the anode surface attributable to the magnetic characteristics of Fe_3O_4. Superior catalytic activity was demonstrated by the Fe_3O_4/CNT enhanced anode, which showed a maximal power density of 830 mWm^{-2} [26].

A critical step in MFC and similar bio-electrochemical networks is the transport of electrons from an exoelectrogen to an electrode. As a result, a highly porous N_2-doped CNT that induced reduced graphene oxide (rGO) and polyaniline (PANI) as an N_2 supplier, was utilised as an anodic material. The nanocomposite as created had a surface doped with nitrogen atoms to improve the flavin redox process and a 3D structure that facilitates microbial growth and transmission of electrons from the surface of the MFC to the anode. It was observed that the performance of *Shewanella oneidensis* used in this study could be enhanced by modifying the surface of the anode N_2-doped CNTs by treating the CNTs with nitric acid/ammonia to raise the N_2 proportion [119]. Additionally, after N+ ion incorporation, the surface roughness, hydrophobicity, and charge transfer resistance, enhanced bacterial adherence and biofilm growth between the bacteria and electrodes [120].

Graphene and CNTs are both exceptional 2D carbonaceous electrode nanomaterials that have been employed in various applications due to superior biocompatibility, high electrochemical stability, and enhanced electrical conductivity [121]. CNTs formed on graphene also function as dependable conducting pathways between electrodes and the corresponding components. The as-developed nanocomposite provides relatively good adhesion towards EET and a maximal power density of 1137 mWm^{-2}, which is 8.9 fold greater than a CC electrode as well as *N*-doped nanocomposites and bare CNTs/rGO nanocomposite [9]. The study focused on the synergistic measurement of surface chemistry design and tailored pore structuring to enhance both bio and electro-catalytic activity in MFC, which offers inputs for designing of bioelectrodes in other bio-electrochemical applications as well (Fig. 5a).

Metallic oxides have been used as anode materials and have capabilities to facilitate EET [10]. Combining metal properties and the mesoporous nature of tungsten trioxide (m-WO_3), which was synthesized using mesoporous silica and incorporated with CNTs, resulted in CNTs/m-WO_3 [10]. In addition to having exceptional characteristics for electrical conductance, the synthesized nanocomposite also produced a delocalization of m-WO_3, which optimized the conjugation of CNTs with m-WO_3. The developed CNTs/m-WO_3 nanocomposites displayed intense electrocatalytic activity and allowed MFC *E. coli* to operate at a maximal power density of 1110 mW m^{-2}, which is noticeably more significant than the MFC utilising only m-WO_3 (760 mW m^{-2}) [10]. Similarly, with other types of CNTs such as multi-walled carbon nanotubes (MWCNTs), the nanocomposites were studied using the incorporation of tin-oxide (SnO_2) [122]. A glassy carbon electrode (GCE) was used as the sensor platform and the nanocomposite (MWCNTs/SnO_2) was prepared with 5 % Polytetrafluoroethylene (PTFE) as a binder and later drop cast onto the electrode surface. According to reports, adding CNTs to a conducting polymer-based anode increased the MFC's power output [122]. Furthermore, MWCNT/SnO_2 nanocomposites showed significantly increased functioning as an anodic material compared to bare GCE and/or pure MWCNTs. The electrochemical efficiency of the nanocomposite may be explained by the synergetic effect of SnO_2 with MWCNTs due to the possible transmission of electrons through the electrostatic linkage of Cytochrome-C, SnO_2 and MWCNTs. The MWCNTs/SnO_2/GCE yielded a maximal power density of 1421 mWm^{-2} with increased charge transfer energy because of the large surface area and conductance that also promoted bacterial biofilm formation on the surface [122]. The power density of the nanocomposite was stable after 2–3 days of operation with three feeding cycles. Another PANI-based MFC anodic fabrication involved the electrophoretic adsorption of CNTs upon PANI- modified graphite felt (GF), which severs as an anodic material in a mediator-free MFC using *Shewanella putrefaciens* as exoelectrogens [25].

In addition, anode surface modification with CNTs enhances the material's surface area and conductance. As more microbes adhered to the CNT/GF/PANI anodic surface, the fabricated MFC achieved a maximal power density of 257 mWm-2. As a result, MFC anodes with

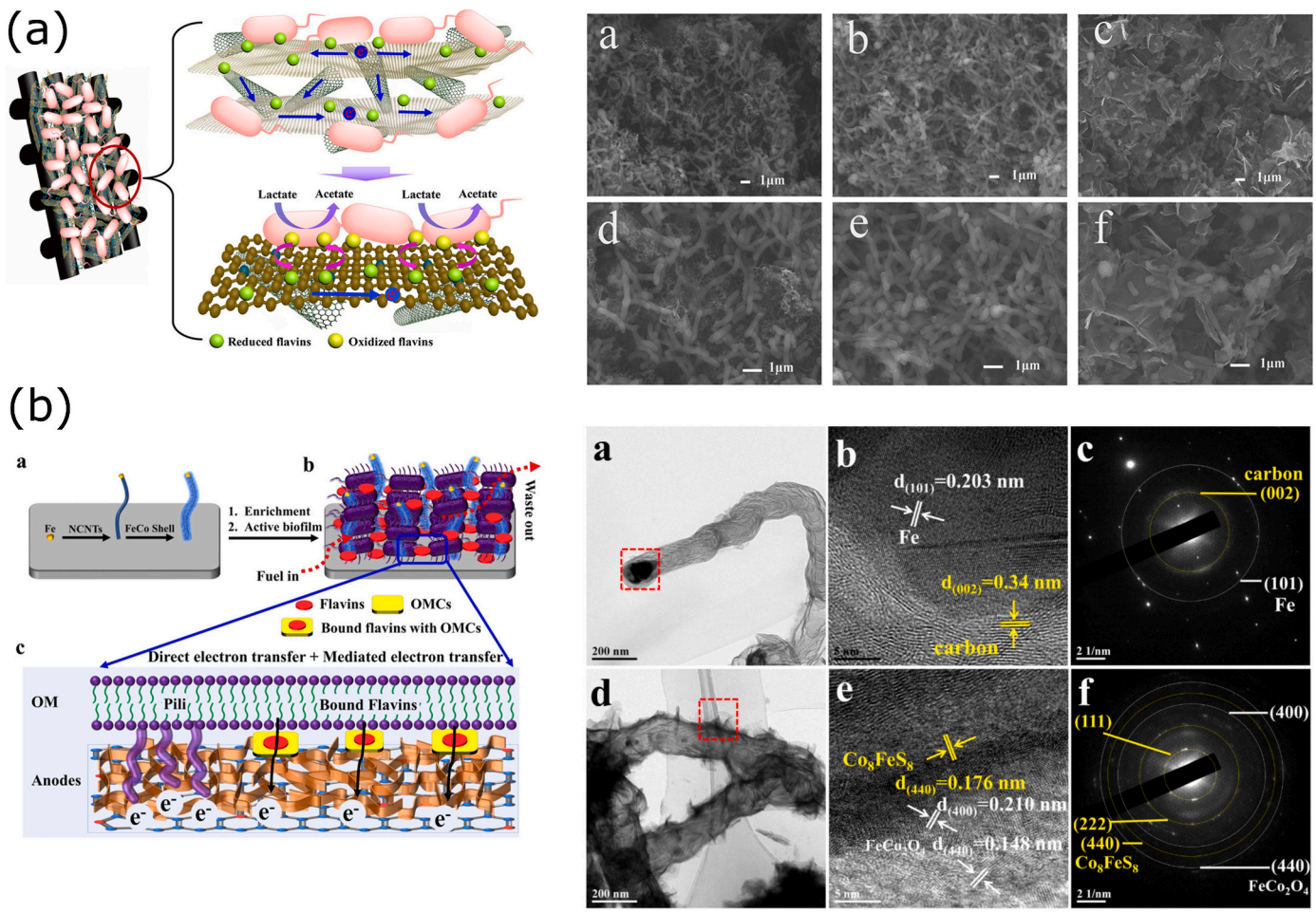

Fig. 5. Carbon nanotube-based nanocomposites in MFC. (a) Electrochemical mediation of the N_2-CNTs/rGO anode and FESEM imaging of microbial cells attached onto the surface of the electrode upon discharge depicting potential working of synchronised increased biofilm extracellular electron transmission ((a,d) N_2-CNTs, (b, e) N_2-CNTs/rGO, and (c,f) N_2-rGO). Adapted with permission from Ref. [9] Copyright 2018, American Chemical Society. (b) Growth procedure of FeCo/NCNTs@CF, active biofilm formation, and proposed EET processes and, (a) TEM, (b) HRTEM imaging of NCNTs@CF, and its (c) SAED pattern, (d) TEM, (e) HRTEM imaging of FeCo/NCNTs@CF, and its (f) SAED pattern. Adapted with permission from Ref. [6] Copyright 2022, American Chemical Society.

excellent efficiency at low cost were made [25].

In search for conductive 2D nanomaterials which can be combined with metal or metal oxide nanoparticles will tailor the integration between exoelectrogens and the anode towards the improvement in power densities of MFC [123,124]. CNTs used to enhance anodes and showcase the potential to increase biocompatibility and EET [125]. Published findings have also proved that the length of the nanotubes, doping, and growth density, all play a key role in bacterial adherence with housing while inoculating, biofilm formation, and EET [126–129]. Combinational approach in mixing CNTs and Fe/Co, improve the power density of bacteria with the anode in certain MFC, enriching more exoelectrogens and promoting EET rate [130]. However, it was observed that most of the CNTs and Fe/Co nanocomposite enhanced anodes could be incorporated with polymers for fixed stabilisation, promoting high internal resistance [130,14]. To overcome these issues, MOFs were directly grown on CNTs and Co/Ni nanocomposites which were aligned vertically on 1D carbon nanofibers and showed enhanced trifunctional electrocatalysis and stability [131]. However, in some MFC, anodes comprised a combination of various metallic oxides/sulfides as an approach to coat N_2-doped CNTs.

In doing so, there will be a substantial decrease in the internal anodic resistance when N_2-doped CNTs are synthesized on carbon-based surfaces with a consistent sp2 carbon interface assembly [132]. Taking the points mentioned above into consideration, the integration of mixed metallic oxides with N_2-doped CNTs would be advantageous in synergistically enhancing the electrochemical performance, conductivity, and surface biocompatibility, which could offer practical alternatives for increasing the maximum power density of the MFC [133]. Combining properties of mixed metal ions, metal-organic framework (MOFs) and N_2-doped CNTs, the NCNTs were synthesized directly onto CF utilising melamine as a monomer and Fe as a catalyst [6]. This 3D structure provides more surface area and robustness to the MFC enabling more microorganisms to grow for better electron transmission to the electrode surface.

N_2-doped CNTs (Fig. 5b), have been employed on carbon fibre (CF) substrates for the synthesis of core -shell (Co_8FeS_8–$FeCo_2O_4$/NCNTs) nanoparticles consisting of (FeCo/NCNTs@CF) [6]. Compared to the bare CF, these nanocomposites exhibit enhanced hydrophilicity, diffusion coefficient, specific capacitance, and reduced charge transfer resistance. The MFC integrated with FeCo/NCNTs@CF demonstrated a maximum power density of 3040 mW m^{-2}, significantly more than bare CF electrode MFC (2060 mW m^{-2}) and other previously fabricated transition metallic electrode MFCs. These studies also detailed a large number of redox species present in the electrochemically active biofilm morphology and studies on the 16s rRNA gene sequence analysis supported this enriched exoelectrogen biofilm on the anode surface. The high capacitance of nanocomposites at the exoelectrogens and anodic interaction surface effectively promoted the EET. These findings provide a new direction in designing and fabricating 3D hierarchical anodic electrodes with ideal biofilm formation and a rapid EET output for

optimum MFC performance [6] (Table 1).

6. Graphene-based nanocomposite as anode material in MFC

Graphene (Gr) is a promising candidate for an active anode-enhancing material due to its acceptable biocompatibility, excellent catalytic activity, and high electron mobility [60]. This includes graphene and its by-products, such as single/multi-layered graphene, graphene oxide (GO), hybrid nanocomposites and reduceed graphene oxide (rGO) [65]. Different graphene-based nanocomposites that showed exceptional anode material in MFC applications have been emphasised and explained in this section. Since the discovery of graphene in 2004 b y Andre Geim and Konstantin Novoselov [134], it has attracted great research attention in several fields (nanoelectronics, energy storage materials, biosensing and catalysis) [133], and provides a new opportunity for improving MFCs [65].

Using Graphene's qualities, rGO and manganese oxide (MnO_2) were coated onto the CF to create a novel anode [44]. This binder-free anodic nanomaterial was shown to boost the enrichment and proliferation of exoelectrogenic microbes and promote the extracellular electron transmission from microbes to the anode due to its exceptionally wide surface area and outstanding conductance. A rGO/MnO_2/CF nanocomposite-modified MFC anode generated a maximal power density of 2065 mW m^{-2}, that is 154% potent than a bare CF anode [44]. The synthesized rGO/MnO_2/CF nanomaterial-modified anode may be a potential anodic material for high-throughput MFC applications. It was prepared using a straightforward dip-coating/electrodeposition procedure. Another study used electrosynthesis to create a polypyrrole and graphene oxide (GO) nanostructure using GF electrodes [135]. In comparison to an anode modified solely by graphene or polypyrrole, coating the GF electrode with a mixture of nanocomposites resulted in a discernible improvement in several attributes, including enhanced surface area, electric conductance, biocompatibility, as well as stability [136]. When employed for MFC applications, the fabricated nanocomposite anode demonstrated a maximal power density of 1326 mW m^{-2} [137]. In addition to enhancing electricity production, these alternative anodic materials also offered the MFC long-term stability [137].

Polyaniline (PANI) is a conductive polymer that is applied as an anode modifier in MFC based on its easy processing ability, biocompatibility, hydrophobicity, electric conductance, and stability [133]. Researchers from different fields have been drawn to PANI's novel characteristics, and have been investigating its potential as a nanomaterial to increase the power output of MFC [66]. Additionally, because PANI is positively charged, it encourages negatively charged microbes to adhere to it [116]. In order to fabricate an anode, carbon cloth (CC) was modified on-site using PANI and Graphene. A power density of 884 96 mW m^{-2}, i.e., 1.3 to 1.9 times greater than that attained with bare CC anode MFC, was developed by modifying the CC anode with PANI and a graphene nanocomposite [138]. In the future, it is anticipated that the mutual alteration of both PANI and Graphene as anodic materials will improve MFC efficiency.

Based on its excellent metallic conductance, biocompatibility, catalytic performance, and endurance, tungsten carbide (WC) as a metal catalysts has caught the interest of scientists as a metal catalyst [139]. However, its usage remains limited due to low surface hydrophilicity and complex fabrication processes. WC and WC + rGO nanolayered nanocomposites exhibit exceptional results as an anodic material in MFC for parallel power generation and sewage treatment. These catalysts were developed by Mohamed et al. by applying a straightforward and inexpensive urea glass method and reduction-carburization strategies [140]. Anodic catalysts were evaluated on-site under actual MFC parameters, and the results demonstrated that the WC + rGO/CF enhanced anode showed exceptional efficiency in terms of power output (1120 mW m^{-2}) and current density (5680 mA m^{-2}). Utilising this approach, WC + rGO finds broader applications in high-performance MFC as cost-effective anode catalysts.

Magnetite (Fe_3O_4), which is earth-abundant, cost-effective and readily available is another ideal material for anode fabrication in MFC. Magnetite nanosphere (*NS*–Fe_3O_4) provide low efficiency of electron transmission which is a significant drawback in MFC applications, even though the affinity of Fe_3O_4 has been reported previously [141]. When Fe_3O_4 nanoparticles and rGO nanosheets are combined to form nanocomposites, the poor electron transmission of Fe_3O_4 can be effectively addressed, and at the same time, the affinity of the nanocomposites for exoelectrogenic microorganisms is increased, making them desirable MFC anodic nanomaterials. As anodes for MFC, the Fe_3O_4-NS/rGO nanocomposite was synthesized using a one-pot solvothermal technique [142]. Wide electroactive surface, low charge-transfer resistance, and high electron transmission ability are all attributes exhibited by the Fe_3O_4-NS/rGO anode. With an enhanced current density of 3719.9 mA m^{-2} and a maximum power density of 1837.4 mW m^{-2} at a scan rate of 0.001 V s^{-1}, the Fe_3O_4-NS/rGO electrode displays remarkable reproducibility and endurance.

Table 1
Details of some of the recently developed single and multi-composite coating anode materials.

Anode material	Method	Inoculum Type	Reactor	Areal/volumetric Power density	Ref.
FeCo/NCNTs/CF	Electrodeposition	Mixed	Dual	3040 mW m^{-2}	[6]
CC/CNT/Co-Pi	Electrodeposition	*Shewanella loihica*	Dual	1200 mW m^{-2}	[7]
CNT/PPy	Chemical Vapour Deposition	Mixture Electrigens	Single	1876.62 mW m^{-2}	[8]
N-CNTs/rGO	Deposition	*Shewanella putrefaciens*	Dual	1137 mW m^{-2}	[9]
CNTs/m-WO_3	Sonication/Deposition	*Escherichia coli* (DH5α)	Single	1110 mW $^{-2}$	[10]
CNT/GF	Dipping/Deposition	Mixed	Dual	2.10 ± 0.05 mW m^{-2}	[11]
CNF/CNT	Electrospinning	Mixed	Dual	1016 mW m^{-2}	[12]
MWCNT/rGO	Sonication/Solvent Method	*Shewanella putrefaciens*	Dual	789 mW m^{-2}	[13]
N-MWCNT/GONR	Microwave energy-assisted unzipping process	*Escherichia coli*	Single	3444 mW m^{-2}	[14]
SC/AC/CNTs	Sonication/Deposition	Mixed	Dual	899.52 mW m^{-2}	[15]
CNTs/PANI*n*/APTES/ITO	Electropolymerization	*Shewanella loihica*	Single	34.51 mW m^{-2}	[16]
GNP/CNT/CPE	Deposition	Mixed	Dual	80 mW m^{-2}	[17]
MoS_2/CNTs/CP	Hydrothermal method	Mixed	Dual	645 ± 32 mW m^{-2}	[18]
MWCNT/MnO_2/PPy	In-situ/Electrochemical	Mixed	Dual	1125.4 mW m^{-2}	[19]
CF/N-CNT/PANI/MnO_2	In-situ polymerization	Mixed	Dual	13,800 mW m^{-3}	[20]
S/N-CNT/PANI/MnO_2	Dip and Pull Method	Mixed	Dual	1019.5 mW m^{-2}	[21]
PANI/PPy/CNTs-Fe_3O_4/Hydrogel	Coating	Mixed	Single	5901.49 mW m^{-3}	[22]
CHI/NCNTs/PANI/NCNTs/sponge	In situ grafting and polymerization	Mixed	Dual	2816.67 mW m^{-3}	[23]
PPy/CMC-CNTs/CB	Polymerization	Mixed	Dual	2970 mW m^{-2}	[24]
CNT/PANI/GF	Electrophoretic Deposition	*Shewanella putrefaciens*	Dual	257 mW m^{-2}	[25]
Fe_3O_4/CNT	Solvothermal	*Escherichia coli*	Dual	830 mW m^{-2}	[26]
CNT/Au/TiO_2	Sol-gel	*Escherichia coli*	Single	2.4 mW m^{-2}	[27]

The graphene electrode with FeS_2 nanoparticle coating was developed in a different work using a straightforward hydrothermal process and utilised as an anode [143]. In comparison to the start-up times of the bare CC and rGO anodes, the FeS_2/rGO anode is 90 and 83.8%, respectively, faster by two days. The FeS_2 nanostructures significantly enhance the efficiency of the MFC by promoting surface contact with the bacterial biofilm and facilitating extracellular electron transmission from the electroactive microbes to the electrode surface. Finally, the graphene anode coated with FeS_2 reached 310 mW m^{-2} of power density while simultaneously removing 1319 ± 28 mg L^{-1} of COD from a brewery's wastewater effluents [143]. Developing nanocomposites by combining graphene with other nanomaterials provides robustness to the MFC, as discussed above. While doing this, the performance of MFC increased by multiple folds than graphene alone. The newly emerging field of multicomponent composites to synthesize nanomaterials often display synergistic characteristics because of the interdependency of the components, providing an efficient solution for enhancing electrode output.

A multicomponent anode nanocomposite using a core/satellite structure of Fe_3O_4/gold nanoparticle-doped 3D graphene foam (Fe_3O_4/Au NCs-3DGF) was developed by combining on-site synthesis methods (both freeze drying and hydrothermal) [144]. The Fe_3O_4/Au nanostructures enhance the adhesion between the anode and microbes based on the high bio-affinity of bacteria towards Fe_3O_4 (Fig. 6a). Moreover, the Au provides a platform for high-speed electron transmission pathways, which help tackle poor conductivity issues of the Fe_3O_4 core. Fe_3O_4/Au NCs-3DGF showcase improved microbial attachment and highly efficient extracellular transfer of electrons, which in combination result in a 71 times enhancement of power density output (2980 ± 54 vs 41 ± 4 mW m^{-2}) in comparison to bare graphite electrode (41 ± 4 mW m^{-2}) (Fig. 6**a**) [144].

A different experiment involved the preparation of a 3D graphene foam/MWCNTs/Fe_3O_4 anode (3D G/MWCNTs/Fe_3O_4) rather than Au nanoparticles, and the ensuing MFC demonstrated a significant power production capacity and stability. MWCNTs can avoid irreversible aggregation in between graphene sheets (Fig. 6b) [133]. The power density and polarisation curves were also analysed by adjusting the external load resistance. The maximal volumetric power density of the MFC built on the 3D G/MWCNTs/Fe_3O_4 anode was 882 mW m^{-2}, that is greater than that of a bare graphite electrode (76 mW m^{-2}) (Table 2).

7. Graphitic carbon nitride as potential anode material

Graphitic carbon nitride (g-C_3N_4) is one of the first synthetic polymers in the line of 2D nanomaterials. In 1834, Liebig gave the g-C_3N_4 precursors their nomenclature after Berzelius synthesized them [65]. Based on its exceptional mechanical, electrical, thermal, and optical characteristics, distinctive structure, lack of metallic materials, increased porosity, high surface to volume ratio, more surface active sites, and lesser surface defects, an allotrope of carbon nitride (C_3N_4) is also making a mark in improving the quality of life [145]. g-C_3N_4 has a layered arrangement made up of weak van der Waals attraction between the neighbouring *C*–N layers, much like graphite. Notably, g-C_3N_4 is affordable, abundantly available, and a more environmentally friendly option (non-toxic) [145].

The tris-*s*-triazine g-C_3N_4 was assumed to be a stable allotrope among the many forms of C_3N_4 (pseudo cubic C_3N_4, α-C_3N_4, β-C_3N_4, cubic C_3N_4, g-*0*-triazine, g-*h*-triazine, and g-*h*-heptazine) [145]. Triazine units or heptazine rings (tri-*s*-triazine) were the two structural models that were proposed for g-C_3N_4, with heptazine rings tending to be the more energetically favourable building blocks of g-C_3N_4 complexes [145]. Since it only consists of carbon and nitrogen, g-C_3N_4 has relatively minimal biological toxicity and is readily metabolised in biological systems. Additionally, g-C_3N_4 shares structural similarities with graphene but has drawn more interest because of its superior characteristics [145].

The electrocatalytic reduction of g-C_3N_4 is inadequate because of its semi-conducting nature (band gap: 2.7 eV). As a result, its potential for use in MFC research and other electrochemical techniques is largely unexplored. However, because of the outstanding electron capturing and transportation characteristics of graphene, electrons may be transmitted from the graphene layer to the g-CN layer via Mesh-on-Mesh g-CN@Graphene. These electrons accumulate in particular on the N atoms in g-C_3N_4, which serve as the composite's catalytic activity sites (Fig. 7a) [146]. Graphene-based carbon nitride nanosheets exhibit superior electrocatalytic activity, long-term endurance, and high selectivity due to its elevated N_2 amount, less thicknesses, large surface area, and improved electric conductance as a metal-free catalyst for reduction reactions [147]. Recently, scientists have focused on integrating mesoporous carbon nitride (mpg) into graphene sheets to enhance the efficiency of electron transmission in MFC. In a recent research on yeast MFC where commercial carbon brush was used as microbial biofilm formation was modified with g-C_3N_4 through a single-step preparation technique [148]. The developed g-C_3N_4@CB as an anode material demonstrated an outstanding performance with a maximal power density of 772 mW m^{-2}, 12 times greater than using bare CB [148]. The efficiency of g-C_3N_4@CB may be attributed to the biocompatibility and roughness, which enhances the yeast biofilm attachment to the anode and decreases the ohmic resistance.

One of the main constraints limiting power generation is electrode resistivity, which significantly impacts on the flexibility and commercial application of MFC. However, coating of nanomaterials on the surface of the electrode results in innovative electrodes with greater electric conductance than conventional electrodes, thereby maximising the power generation from the MFC. In recent studies, Attia et al. used three 2D nanomaterials (graphitic carbon nitride, graphene, and carbon nanotube) to check the power density and coulombic efficiency [150]. The g–C_3N_4–coated electrodes displayed voltage stability for the entire 140-h period with a maximum value of 1.367 V and a maximal power density of 116 mW m^{-2} [150]. With a maximal areal power density of 23.6 mW m^{-2} and a maximal volumetric power density of 3.2 mW m^{-3}, the voltage steadily rose to 0.616 V after 22 h in comparison to the control (no coating), and it remained stable for up to 140 h.

The g-C_3N_4 nanosheets outperformed all other nanomaterials in terms of efficiency. The findings demonstrate that covering MFC electrodes with g-C_3N_4 enhances the electrical power by 4.9-fold compared to the uncoated control [150]. MFC electrodes coated with g-C_3N_4 were superior to graphene, CNTs, and the control in that order. Additionally, it was demonstrated that manufactured electrodes were non-toxic for microbial organisms and highly conducive to bacterial growth. The electrodes coated with g-C_3N_4 made excellent results due to their unique attributes.

It is clear that the 3D structure supports more adhesion of microbial biofilm on the surfaces. You et al. fabricated a facile 3D microporous N_2-enriched g-C_3N_4 (NGC) scaffold utilising melamine foam by applying a pyrolysis technique (Fig. 7b) [149]. The developed anode material promotes EET efficiency by attaining a power density of 750 mW m^{-2} using *Shewanella oneidensis* in MFC. The novel 3D open-cell structure supports electroactive biofilm formation and supplies a porous network for internal mass transfer avoiding any issue of bio-blocking and/or bio-fouling [149].

8. MXene as a promising anodic nanomaterial for high-throughput MFC

A new group of 2D nanostructures called MXenes are a transition metal carbide with a structure resembling the layers of graphene. Due to its high feasibility for ion intercalation, stability, hydrophilicity, relative specific surface area (compared to preceding 2D materials listed above), and environmentally benign attributes, it is an ideal support matrix [54,55,151,152]. By modifying the electrophilicity of catalysts in multiple component systems, MXene promotes quick electron transportation and

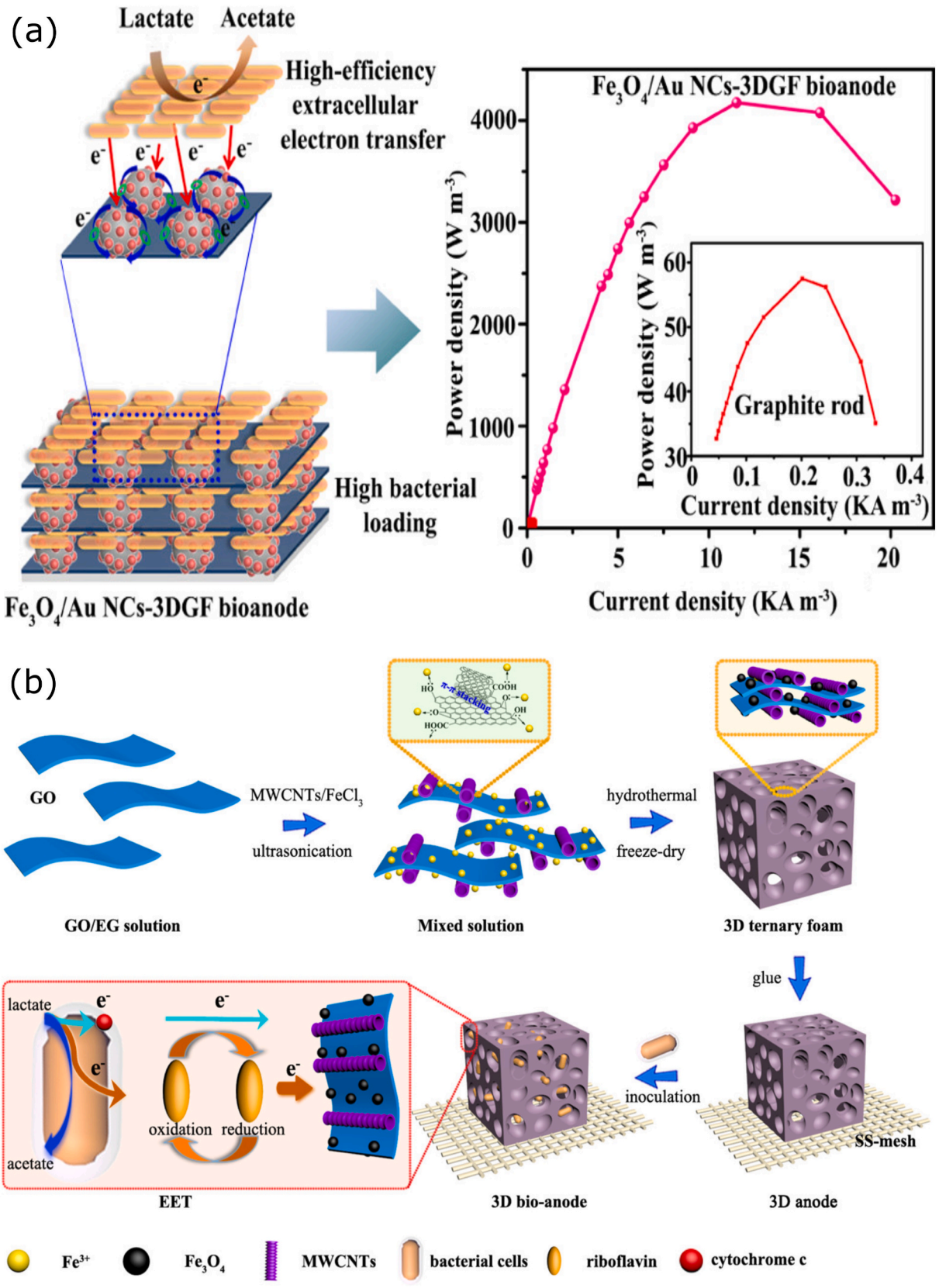

Fig. 6. Schematic representation of the synthesis procedure: (a) Fe_3O_4/Au NCs-3DGF modified bioanodes based on the mechanism of possible electron transfer and current density profile compared to the commercially available graphitic rod. Adapted with permission from Ref. [144] Copyright 2019, American Chemical Society. (b) Schemme 1. Development of the 3D macro-porous G/MWCNTs/Fe3O4 foams and their applications as MFC anodic materials. Adapted with permission from Ref. [133] Copyright 2016, American Chemical Society.

Table 2
Details of recently developed graphene-based anode nanocomposites.

Anode material	Method	Inoculum Type	Reactor	Power density	Ref.
GO/CC	Dip coating	Mixed	Dual	14 mW m^{-2}	[28]
GO/Ni	Electrodeposition	*Shewanella putrefaciens*	Dual	468.16 mW m^{-2}	[29]
Gr/graphite	Exfoliation	Mixed	Dual	1110 mW m^{-2}	[30]
CF/br-GO	Dipping	*Shewanella putrefaciens*	Dual	240.2 mW m^{-2}	[31]
PANI/rGO/CB	In-situ polymerization	Mixed	Single	862 mW m^{-2}	[32]
PANI/rGO/CC	Dipping	Mixed	Dual	306 mW m^{-2}	[33]
rGO/PPy/graphite rod	Electro-polymerization	Mixed	Dual	835.21 ± 1.40 mW m^{-2}	[34]
rGO/PDA/CC	Solvent immersion	Mixed	Dual	2047 mW m^{-2}	[35]
Mo_2C/Gr/CC	Soft template-assisted assembly/calcination	*Shewanella putrefaciens*	Dual	1697 mW m^{-2}	[36]
Fe_2O_3/Gr/CF	Hydrothermal reduction	Mixed	Dual	334 ± 4 mW m^{-2}	[37]
GO/ZnO/graphite rod	Solvothermal	Mixed	Dual	2.55 mW m^{-2}	[38]
GO/ZnO/graphite rod	Solvothermal	Mixed	Dual	1350 × 10^{-3} mW m^{-2}	[39]
GO/TiO_2/graphite rod	Solvothermal	Mixed	Dual	608 × 10^{-3} mW m^{-2}	[40]
GO/$NiWO_4$/CC	Hydrothermal	Mixed	Single	1128 ± 42 mW m^{-2}	[41]
N-GO/CeO_2(1:2)/CA (carbon aerogel)	Hydrothermal	Mixed	Dual	1468 mW m^{-2}	[42]
3D-Gr/NiO	Hydrothermal	*Shewanella putrefaciens*	Dual	3632 mW m^{-2}	[43]
rGO/MnO_2/CF	Dip Coating/Electrodeposition	Mixed	Dual	2065 mW m^{-2}	[44]
Gr-Foam/CC	Freeze Drying/Pyrolysis	*Shewanella putrefaciens*	Dual	786 mW m^{-2}	[45]
Gr-aerogel/CC	Hydrothermal	*Shewanella putrefaciens*	Dual	679.7 mW m^{-2}	[46]
Gr/CNT-biofilm/CC	Reduction	*Shewanella oneidensis*	Dual	0.095 mW m^{-2}	[47]
TiO_2/rGO/CC	Sol-gel/Solvothermal reduction	*Shewanella putrefaciens*	Dual	3169 mW m^{-2}	[48]
Hierarchical porous-Gr/Ni	Freeze drying	*Shewanella putrefaciens*	Dual	3903 mW m^{-2}	[49]
rGO/PANI/Pt/CC	In-situ polymerization	*E. coli*	Dual	2059 mW m^{-2}	[50]
GO–SnO_2/CC	Hydrothermal/microwave assistant	*E. coli*	Dual	1624 mW m^{-2}	[51]
Gr/TiO_2/Nickel foam	Microwave-assisted solvothermal	*Shewanella oneidensis*	Dual	1060 mW m^{-2}	[52]

enhances the catalytic characteristics of such systems [153].

By simply extracting the A-element from the Mn+1AXn (n = 1,2,3) or MAX phase (where M is an early transition metal, A is a group IIIA or IVA element, and X is C/N), MXene can be manufactured easily in a single step and on a large scale [53]. Titanium carbide ($Ti_3C_2T_x$), which can be produced by submerging pre-phase Ti_3C_2 in Hydrofluoric acid (HF) or (HCl + LiF) [54], and has been employed in numerous applications, is currently the most researched MXene (high capacity electrodes, supercapacitors, optoelectronic and flexible wearables) [154]. MXenes are an excellent conducting transporter to enhance electron transmission, making MXene a perfect fit to increase anodic conductance [155]. An ideal 3D arrangement may enhance the number of active sites in nanocomposite materials that promote EET and bacterial adhesion. Merging MXene with other nanomaterials improves the MXene's overall structure while simultaneously attaining outstanding electric conductance and chemical stability. Being negatively charged, the MXene surface guarantees robust interface interaction upon merging with other constituents, successfully inhibiting aggregation of nanomaterials and improving stability [156–158]. The anode in an MFC requires mass transport enhancement, reduced electron charge transfer resistance, selective microbial enrichment, improved bacterial adhesion, and good chemical stability [159,160]. Based on the above requirements, MXenes are regarded as a perfect anodic nanomaterial for the fabrication of MFC due to their distinct spatial arrangement and electrophilicity.

An MFC anode was recently modified using multi-layered Ti_3C_2 MXene coated onto carbon cloth (Ti_3C_2/CC) [53]. The addition of Ti_3C_2 increased the anode's total surface area, wettability, and surface roughness. In comparison to the carbon cloth (CC) alone (2050 mW m^{-2}), MXene was utilised as the anode and effectively started up the MFC after a single day of inoculation with a microbial population to provide a maximal power density of 3740 mW m^{-2} [53]. Ti_3C_2 MXene 3D arrangement, increased surface area, and superior conductance, which can enable bacterial growth, rapid mass transfer, and additional active sites for bio-catalytic redox processes, are primarily responsible for enhancing MFC efficiency. Additionally, the outer membrane cytochromes, a densely packed web of bacterial nanowires, and a vast proportion of extracellular polymeric compounds (the lowered gap between microbial species and MXene surface) enabled direct EET between the biofilm and the Ti_3C_2/CC anode, efficiently enhancing the MFC output. Compared to CC (18.6%), the developed Ti_3C_2/CC anode produced 23% more coulombic efficiency.

Metallic ions and organic ligands comprise the porous materials known as metal-organic frameworks (MOFs) [54]. Due to the ability of various synthesizing percentages or solvents to precisely control their sound structure, orientation, particle size, and pore diameter, MOFs exhibit distinct benefits such as significant specific surface area, crystalline nature, and stable tuneable crystal arrangement [54]. MOFs are routinely used as precursors of nanostructures because of their unique characteristics [161]. Due to its unique structural arrangement, the zeolite imidazolate framework-67 (ZIF-67), dependent on imidazole ligands and metallic ions, has lately been explored as a precursor for effective catalysis [162]. The ZIF-67 contains many microspores and N elements, which enrich the microbes on the surface of the electrode, increasing the anodic biocompatibility. However, its weak conductance and ease of aggregation restrict ZIF-67's wide range of applications as an

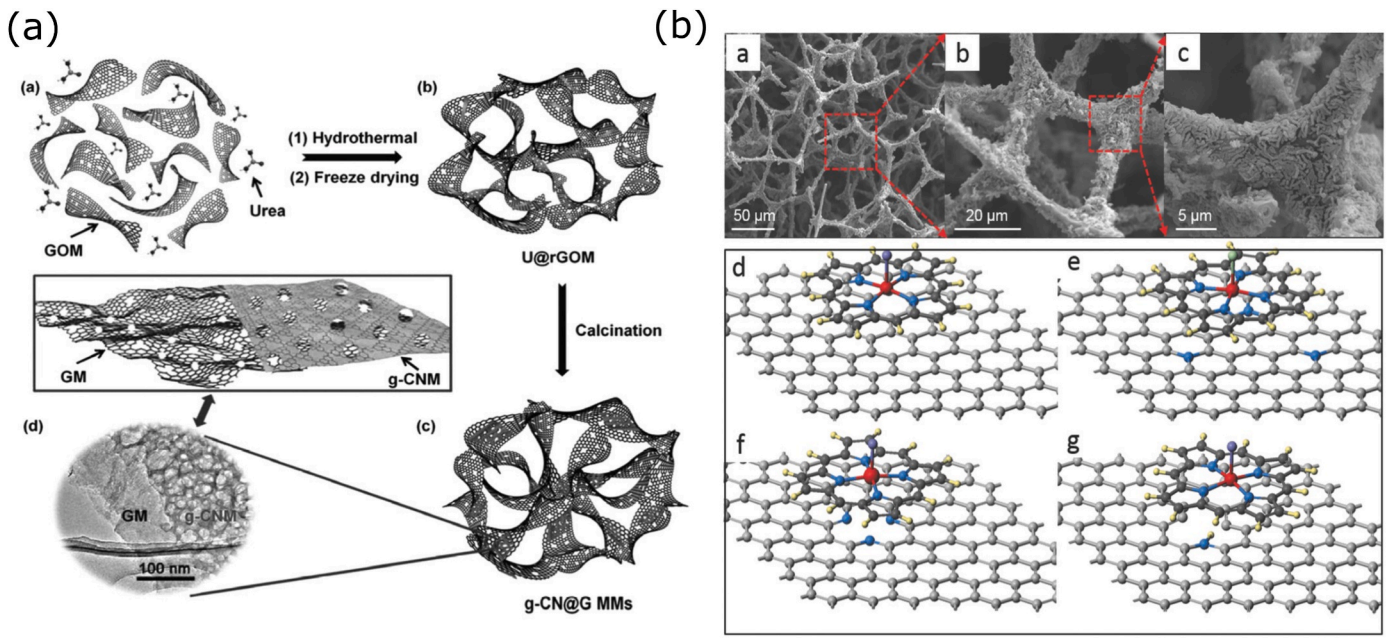

Fig. 7. (a) Schematic representation of graphene layer fabrication on g-CN nanosheets in Mesh-on-Mesh (g-CN MMs) with, a). Showing urea deposited in a graphene oxide mesh (GOM), b). 3D Urea@GOM formed after hydrothermal process followed by freeze drying, c). g-CN@G MMs and, d). Enlarged image of a section of g-CN@G MMs. Adapted with permission from Ref. [146] Copyright 2017, Wiley-VCH GmbH, (b) SEM imaging of biofilm upon the on NGC scaffold a–c), DFT calculation of energy adsorbed between porphyrin iron and d) graphite, e) graphitic nitrogen, f) pyridinic nitrogen, and g) pyrrolic nitrogen. (Ball colour key: Yellow-H atoms, Blue- N atoms, Red- Fe atoms, and Gray- C atoms). Adapted with permission from Ref. [149] Copyright 2016, Wiley-VCH GmbH.

MFC anodic material.

In order to resolve this problem, ZIF-67 is mixed with other nanostructures to create a highly efficient anode in MFC to overcome these deficiencies. On the carbon felt, a solid electrocatalytic anode composed of MOF and MXene was created by self-assembling of imidazole-based ZIF-67 [54]. The negatively charged MOF was used to impregnate the positively charged MXene, increasing its specific surface area and amount of active sites. This increase ultimately facilitates the colonisation of microbes and the catalytic reaction. In addition, Ti_3C_2-ZIF-67's "sandwich-like" layer structure may facilitate the extracellular electron transfer and boost the transportation of ions, electrolyte permeability, and microbial adherence to create a strong biofilm [54]. A higher volumetric power density of 5700 0.12 mW m^{-3}, which was found to be 25.59%, 56.14%, and 63.16% greater than bare Ti_3C_2, ZIF-67, and CF anodes, respectively, was produced by the MFC using the Ti_3C_2-ZIF-67 anode [54]. An innovative and effective path for the continued development of anodic materials for MFC power production would be represented by using Ti_3C_2-ZIF-67 anodes. Similarly, a straightforward dip-and-dry/hydrothermal technique was employed in a different study to alter ordinary carbon felt with $NiFe_2O_4$-MXene ($NiFe_2O_4$-MXene@CF) insted of MOF [55]. To compare the difference in power density, $NiFe_2O_4$@CF along with MXene@CF were prepared. The electrochemical efficiency of the synthesized $NiFe_2O_4$-MXene@CF was distinctly superior, with a maximum power density of 1385 mW m^{-2}, which was 5.6, 2.8, and 1.4 fold greater than the power output of CF, $NiFe_2O_4$@CF, and MXene@CF anodes respectively [55]. Additionally, compared to CF (898 Ω), the $NiFe_2O_4$-MXene@CF anode provided a reduced charge transfer resistance (198 Ω) and a significant increase in current density over time. Moreover, many exoelectrogens were supported by $NiFe_2O_4$-MXene@CF, which enabled biofilm enrichment and led to an increased current output.

The anode's properties are crucial parameters for MFC efficiency because of their effect on the formation of biofilms and EET. Conducting polymers can find a niche as an alternate material to prepare MXene nanocomposites in order to alter the CC electrode surface. With this idea, Without using a binder, Jiang et al. [56] synthesized PANI-MXene to alter the CC (PANI-MXene/CC) anode. Via electrostatic adsorption between the positively charged PANI and negatively charged MXene groups, PANI-MXene was synthesized using this technique. The PANI-MXene adorned CC surface, facilitated bacterial adhesion and colonisation, creating dense and complete biofilm layers as a bio-anode. The PANI-MXene/CC bio-anode successfully performed an EET procedure with a maximal power density of 737.3 mW m^{-2} [56]. Similar results were obtained using poly (diallyl dimethylammonium chloride) (PDDA) to construct MXene@PDDA/carbon cloth (CC) as the MFC anode in place of PANI [57]. High electric conductance, biocompatibility, super hydrophobicity, and increased bacterial load at the anode were all characteristics of the CC surface treated with MXene@PDDA. In comparison to the bare CC anode (580 mW m^{-2}) and MXene/CC anode (731 mW m^{-2}), the MFC coupled with MXene@PDDA/CC anode achieved an output voltage of 587 mV and a higher power density of 811 mW m^{-2}. The W_2N-MXene composite has recently been fabricated on a CC anode, which performs best compared to other MXene-based MFC-based anode using wastewater (Fig. 8a) [163]. The developed nanocomposite provides a power density (548 W m^{-2}) with an 83% reduction in COD. The practical electron transfer efficiencies also increased to 161 % compared to bare CC. This enhancement in power density is acquainted with the adhesion of microorganisms through nanowires which contribute to electron transfer.

The hydrophobic nature of carbon anode sometimes hinders microbial adhesion and fail to form biofilm on the surfaces. To overcome these issues carbon cloth modified with suitable material can enhance electron conductivity and at the same time promote biofilm. Recently, MnO_2@MXene coated CC (MnO_2@MXene/CC) was fabricated as an anode material for MFC (Fig. 8b) [58]. The MnO_2 is loaded onto the MXene surfaces allowing microbes to have contact with the MnO_2@MXene and the electrons are rapidly transported to the external circuit through flagella (Fig. 8c). The developed structure promotes bacterial colonies formation on the surface of the electrode which increases the overall EET. The multi-layered nanosheet of MXene and flower-like MnO_2 were combined resulting in a honeycomb structure with a rough and undulating surface which provide a large area for the

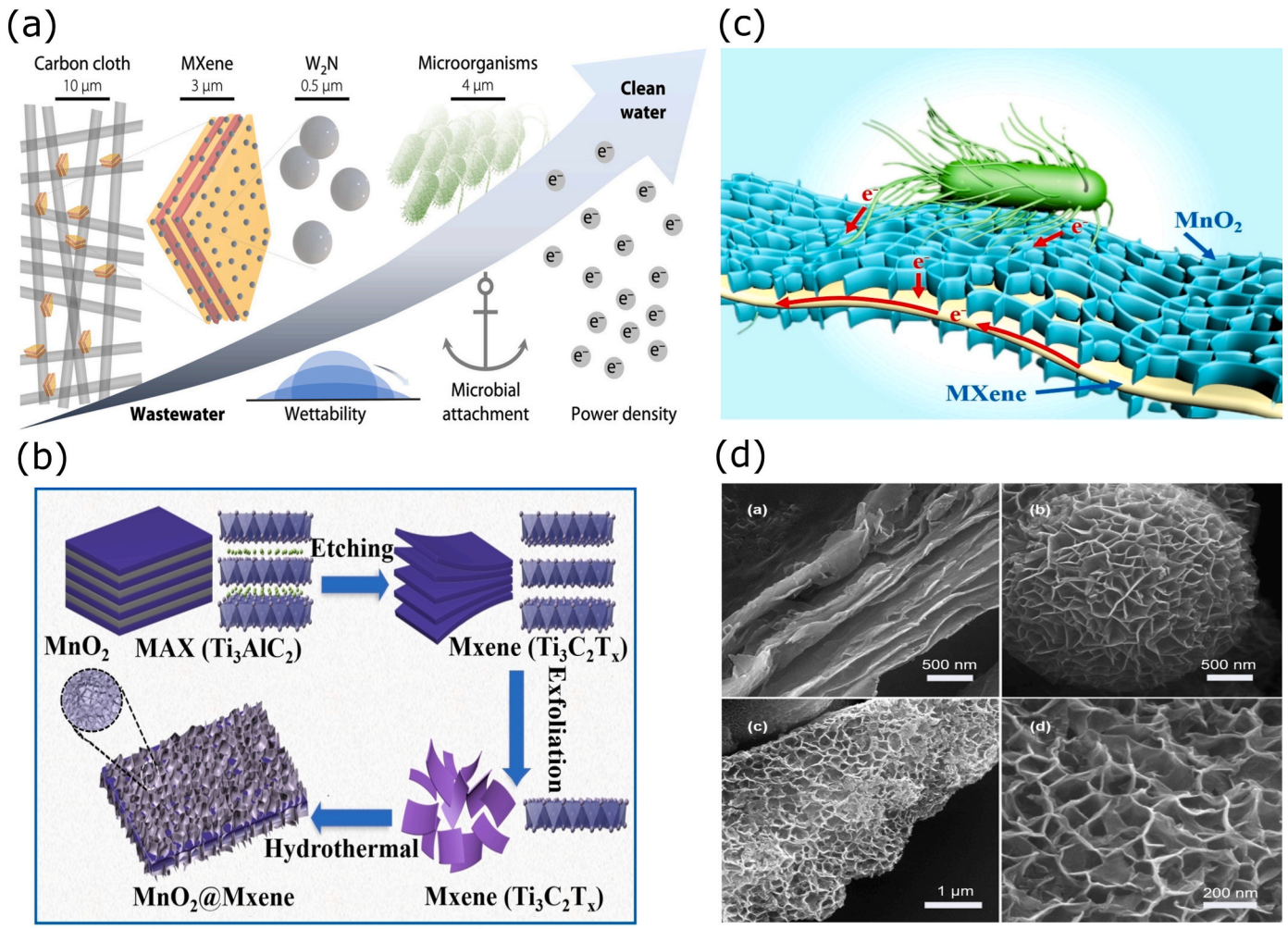

Fig. 8. Schematic illustration of MXene synthesis and electron transport process. (a) PANI/MXene (Adopted with permission from Ref. [163] Copyright 2023, with permission from Elsevier), (b) different stages of MnO_2/MXene synthesis, (c) electrogenic bacteria on MnO_2/MXene with possible electron transport mechanism, (d) SEM imaging (a-MXene, b- MnO_2, *c*-d MnO_2/MXene (Adopted with permission from Ref. [58] Copyright 2022, Wiley-VCH GmbH).

enrichment of bacteria (Fig. 8d). This type of structure provides abundant hydrophilic sites for bacterial attachment and biofilm formation. Topological porosity accelerates electrolyte diffusion and provides food for bacterial reproduction. The MFC device with MnO_2@MXene as anode material attained a high maximal power density of 746.3 mW m^{-2}.

The performance of an anode depends on the bacterial attachment, which accelerates the transport of protons to the cathode from the anodic surface. Poor anodic performance is one of the criteria for developing an efficient MFC. Multi-layered Ti_3C_2 MXene (*m*-MXene) is another alternate material that may be used as an anode in MFC because of the layered structure, which provides a wide surface area for the adhesion of microbes and biofilm formation [59]. In order to overcome these issues, Yang et al. created an efficient anodic material (*n*-MXene) by restacking and aggregating MXene using a solution phase flocculation method (ammonium ion technique). Compared to the *m*-MXene anode, the *n*-MXene utilised as an anode enhancer had a larger surface area, hydrophobicity, and electr-positivity. The maximum power density with *n*-MXene was 686 mW m^{-2}, which was greater than the control, *m*-MXene, and *s*-MXene by 22.9 %, 27.1 %, and 34.8 % (509 mW m^{-2} < 626 mW m^{-2} < 647 mW m^{-2} respectively) (Table 3).

To maintain the power and voltage stability in MFC for consistent and reliable longer-term function for longer time. Some of the strategies are highlighted to achieve stable power and voltage in MFC.

1. Anode design: The anode design is critical for power and voltage stability [164]. Increase surface area via novel nanomaterials, modification through chemical treatment, and promote biofilm

Table 3
Details of the recently developed MXene-based anode nanocomposites.

Anode material	Method	Inoculum Type	Reactor	Areal/Volumetric Power density	Ref.
Ti_3C_2/CC	Coating	Mixed	Dual	3740 mW m^{-2}	[53]
Ti_3C_2/ZIF67/CF	Precipitation	Mixed	Dual	5700 mW m^{-3}	[54]
MXene/$NiFe_2O_4$//CF	dip-and-dry/hydrothermal	Mixed	Dual	1385 mWm^{-2}	[55]
MXene/PANI/CC	Etching/Soaking	Mixed	Dual	737.3 mWm^{-2}	[56]
MXene@PDDA/CC	Etching/Soaking	Mixed	Dual	811 mWm^{-2}	[57]
MXene/MnO_2/CC	Hydrothermal/Spin-coating	Mixed	Dual	746.3 mWm^{-2}	[58]
Ti_3C_2-MXene/NH_4^+/CF	Dipping	Mixed	Single	686 mW m^{-2}	[59]

formation and microbial attachment leading to enhanced EET and higher output current. Optimizing and configuring electrode spacing can help in minimise internal resistance, improving voltage in MFC.
2. The concentration of substrate in a controlled manner: Substrate concentration (organic compounds or wastewater) needs to be precisely controlled. More inflow or less can cause enhance/low microbial activity with substrate depletion/low concentration, which results in unstable power output [165]. Finding a balance substrate supporting a balanced microbial population and steady power supply is most crucial in MFC.
3. Maintaining pH: Maintaining pH is essential for microbial activity and proliferation. Extreme pH shifts can case inhibit or slow down the metabolic activities leading to a lower power supply. Monitoring appropriate pH using different solutions and buffers can help provide a stable microbial environment and power generation [166].
4. Temperature: Maintaining a stable temperature for microbial activity is also vital for the performance of MFC towards power and voltage stability. Thermal insulator systems can regulate the temperature avoiding fluctuations, which might affect the growth of the microbial community and stable power output [167].
5. Cleaning and Maintaining: Regular cleaning and maintaining of electrodes is essential to prevent biofouling and any surface contamination. Biofouling reduces the power and voltage stability by improper EET. Brushing, and chemical cleaning can promote electrode selflife and reduce degradation with time [168].
6. Material selection: Conductive materials and their choice can impact power and voltage stability. As mentioned above, 2D materials have relatively better electrochemical properties, and are commonly used as anode materials in MFC. These materials can enhance EET and contribute towards stable power and voltage. Selecting the proper electrode for MFC, which shows better durability, good conductivity and compatibility, can help maintain stable power and voltage.

9. Challenges and future perspectives

MFCs are a revolutionary technique since they can use wastewater and biocatalysts to generate stable power density and voltage from chemical energy [71,169–172]. Cleanliness, efficiency, and renewability are a few of the advantages of MFC energy production, which does not release toxic by-products. Additionally, the microorganisms utilised in MFCs are numerous and widely accessible in the biosphere. Despite earlier enhancements and innovations in MFC anode material fabrication, there are still certain limitations in raising the net output of these systems.

- The development of advanced MFC techniques is constrained by the inadequacies related to the characteristics of the electrode fabrication materials, such as catalytic behaviour, surface area, charge transport resistance, and cost. Making efficient MFC electrodes would most likely need innovative materials with superior charge transfer capabilities, durability, and large surface area. In order to facilitate the development of biofilms on their surfaces, there is a pressing need to investigate novel hybrid 2D nanomaterials with high conductance, permeability, and biocompatibility. Using commercially available nanostructures like CNTs and graphene oxide as anode materials showed excellent results but are expensive and unstable for large-scale and commercial applications. As mentioned in this review, adoption of greener materials (g-C_3N_4 and MXene) is a better candidate for building MFC with superior qualities.
- Output current stability and lower power generation are prime challenges of MFC when it comes to scaling up. Research addressing this issue regarding cost and material is still in its infancy. The major market acceptance of MFC is associated with its overall cost of the material and unit, which triggered losses during power generation [173].
- Available electrode systems used in MFC highly depends on various autotrophs and exoelectrogenic strains, reflecting the low power densities. The selection of appropriate bacterial strains/mixed colonies capable of forming a biofilm on the nanomaterial surface is required to enhance the electron transport phenomenon which increases the electrical efficiencies of MFC.
- Screening appropriate microorganisms is a crucial criterion for enhancing of MFC's overall performance. Therefore, research should focus on identifying novel microorganisms (bacteria or yeasts), capable of producing high EET with improved adaptability towards harsh environmental conditions. New metagenomic work can help researchers to uncover emerging stains of microorganisms and their associated metabolic activities for stable power density.
- Metabolic engineering in microorganisms is crucial in altering the pathways to increase their electroactive properties. Genetic manipulation methods can be introduced to EET, which improves substrate utilization and regulate the electron transport chain. Strains with improved current production, tolerance to environmental stress, and effective electron transfer will be the critical focus of research in the future.
- Genetic engineering techniques (gene knockout, overexpression, or horizontal gene transfer) will create recombinant strains with stable power production capabilities.
- The optimization process for the proper functioning of MFC is essential for practical applications. Several parameters (electrode material, MFC design, working conditions and appropriate substrate choice) are responsible for enhancing power output, improving stability and reducing the overall cost of the MFC.
- A new approach towards synthetic biology must be designed for MFC development by designing and fabricating microorganism systems with specific functions. More recently, researchers have been working on engineering the synthetic microbial consortia involving mixing various microbial strains that work synergistically to improve the performance of MFC.
- In recent years, innovation and improved design in anode materials have helped improve maximum power density and performance. Due to their increased porosity, large surface area, and electric conductance, 2D nanocomposites are extensively employed for anode fabrication. Various 2D nanomaterials with improved characteristics, such as increased mechanical strength, high electric conductance, and excellent thermal stability, have been hypothesized and synthesized in recent times to enhance the overall efficiency of electrodes.
- The design and size of electrodes are critical parameters in developing of anodes. In MFC, bacterial adherence/colony formation and electron transmission to the cathode from the anode are caused by appropriate electrode distancing and optimum surface area [174].
- Surface enhancements of an anode using combinational 2D nanocomposites have improved MFC efficiency concerning maximum power density and wastewater remediation. However, the in-depth principles and proper guidelines remain unclear, which is an open question for researchers to explore advanced mechanisms to develop efficient modification techniques.
- The stability of the anode is another challenge for long-term use in distinct environmental conditions. Most recent reports focus on power output and wastewater treatment, with no published work on electrode stability over long-term [175]. The stability of the anode is the major limitation of MFC industrial-scale applications, and more emphasis should be given to novel electrode fabrication techniques.
- Future research should also concentrate on the effect of these nanomaterials on the environment and human health which can be used as a salient component in an MFC. The effect of the 2D and 3D structure of the nanomaterials on the electron transfer rate also needs to exploring in connection with the effects of nanocomposites prepared from the above materials.

- Commercialization to scale-up the process is vital for the MFC industry. While MFC works fine at laboratory-scale experiments, scaling up and commercialization remains challanging. In future, efforts should be centred on MFC design, engineering challenges and the development of greener cost-effective materials enabling large scale production and implementation
- Working closely and collaborating between scientists, engineers, and industry is vital for narrowing the gap between laboratory research and real-world applications

10. Conclusion

Emerging 2D nanomaterials with large surface area, electroactive regions, chemical and thermal stability, and increased porosity are promising choices in MFC applications. Furthermore, these qualities can further be improved by combining them with other nanomaterials to create nanocomposites.

The present review is centred on the application of 2D materials as efficient anode modifications for the adhesion of microorganisms and biofilm formation, which further enhances the maximum power density of the MFC. As presented above in different sections, 2D materials demonstrates that anode modification (CNTs, graphene, graphitic carbon nitride, and MXene) is the superior choice to construct highly efficient MFC. These reported anode nanostructure materials possess unique electrical, optical, physical, and/or chemical properties making them ideal in MFC applications.

Together with the recent literature presented in the review, our experience working with different nanomaterials provides a layout for formulating the need for MFC using 2D materials. We have been actively working on different nanomaterials, including graphitic carbon nitride [145,176], graphene [177–179], CNTs [180], MXene [181], and other nanomaterials [182–186]in different applications (remediations, biosensors, diagnostics, nanotoxicity, electrode fabrications, transducers, batteries and solar panels). This combined cumulative field of nanomaterials was put forth to develop this comprehensive review, including 2D nanomaterial as a potential candidate for constructing MFC anode.

There will undoubtedly be more nano carbon-based anodes and catalysts accessible due to ongoing research advancements in nanocomposites of CNTs, graphene, g-C_3N_4, and MXene. The price could significantly decline as the magnitude of supply rises and increasing demands within the fuel cell material market is satisfied. Also, doping 2D materials with other nanostructures may be improve their electrical characteristics for efficient catalytic applications. Combining a 2D material with polymers, nanoparticles, hydrogels, oxides, and the formation of aerogels in single and multicomponent nanocomposites pave the wave to building more efficient MFCs [22,49,187]. For instance, as reported, CF/N-CNT/PANI/MnO_2 nanocomposites demonstrated an excellent maximal power density of 13,800 mW m^{-2} [20]. Additionally, a hierarchical porous Gr/Ni nanocomposite anode with a porous structure has a superior power density of 3903 mW m^{-2} in MFC [49]. A Greener approach for the producing these anode materials, eg. Graphitic carbon nitride, which is non-toxic, earth-abundant, and environmentally friendly, provides an alternative material compared to conventional metal-based materials. Moreover, because of the low conductivity of this material, appropriate doping with other metal catalysts or superior 2D nanomaterial will overcome these issues. The result will be lower cost and better performance of the MFC in sewage treatment and power generation at the industrial level. MXene, an upcoming material not much explored in MFC as discussed above, can find its nice because of its excellent 3D structure, semiconductor nature, and large surface area which can accommodate bacterial colonisation producing more power.

Enhancing the interplay of microbes from sewage treatment with the anodic material and understanding their compatibility are the latest research priorities for MFC. To generate more power from sludge, waste organic products, and industrial effluents, additional research is still required. One of the main concerns of scientists is the need for less expensive, safer, and eco-friendly materials since MFCs should be environmentally benign and sustainable. It is predicted that producing effective, economical, and efficient MFCs utilising novel nanocomposites with economical manufacturing, will be crucial in further developing the MFC market in the future.

Author contributions

Raghuraj Singh Chouhan: Conceptualization, Writing-Original Draft, Writing, Reviewing and Editing, Visualization, Software, and Funding Acquisition. Sonu Gandhi: Writing, Reviewing and Editing, Visualization, Investigation. Suresh K. Verma: Writing, Reviewing and Editing. Ivan Jerman: Reviewing and Editing. Syed Baker: Reviewing, Writing, Software and Editing. Marko Štrok: Writing- Reviewing and Editing, 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.

Data availability

No data was used for the research described in the article.

Acknowledgment

The Slovenian Research and Innovation Agency is acknowledged through funding of projects J7-2597, BI-US/22-24-162, J1-3033, and programs P1-0143 and P2-0075. The copyright permissions have taken for all the figures used in the article.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2023.113813.

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