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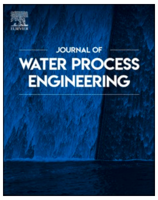

An overview of agro-industrial wastewater treatment using microbial fuel cells: Recent advancements

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ABSTRACT

Discharge of agro-industrial wastewater causes severe damage to the environment due to the various hazardous components it contains. Progressive circular agriculture and industry involve closed cycles and zero-waste principles; therefore, waste treatment for subsequent reuse is of great interest. The microbial fuel cell (MFC) is a promising sustainable technology capable of treating wastewater at low cost without greenhouse gas (GHG) emissions while simultaneously producing electricity. The principle behind MFCs is based on the conversion of chemical energy into electricity using electrochemically active bacteria (EAB) as a biocatalyst. This article presents an overview of the progress and recent advancements in MFCs as a pivotal technology for agro-industrial wastewater treatment with production of sustainable electricity, compared to conventional treatment processes. Furthermore, the fundamental aspects of the design, configuration, operation, and application of MFCs in wastewater treatment (i.e., removal of inorganic nutrients, nitrate, phosphate, sulfate, sulfide, ammonium, organics, dyes, carbohydrates, fatty acids, and petroleum) are comprehensively discussed. Despite the viability and potential of MFCs in treating agro-industrial wastewater and producing electricity, they face various limitations and significant challenges, which are highlighted in this review. Future opportunities and perspectives on MFC applications are also discussed. Further research is required to address MFC limitations, which could make them feasible and practicable for real-world applications. However, MFC technology is clearly an excellent option for simultaneous wastewater treatment and electricity production without the need for external energy sources.

1. Introduction

In the past decades, wastewater has been the subject of extensive research and concern due to its negative impact on human, animal, and environmental health [1]. The conventional processes used in wastewater treatment require high energy consumption and are expensive

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Abbreviations: $Al_2(OH)_5Cl$, aluminum chlorhydroxide; BES, bioelectrochemical system; BOD, biochemical oxygen demand; CaO, calcium oxide; $Ca(OH)_2$, calcium hydroxide; CEM, cation exchange membrane; CFU/g, colony-forming unit per gram; COD, chemical oxygen demand; DC-BES, dual-chamber bioelectrochemical system; DC-MFC, dual-chamber microbial fuel cell; DWW, domestic wastewater; EAB, electrochemically active bacteria; $FeCl_3$, iron chloride; $FeSO_4$, ferrous sulfate; GHG, greenhouse gas; IWW, industrial wastewater; K, potassium; MFC, microbial fuel cell; N, nitrogen; $(NH_4)_2SO_4$, ammonium sulfate; NO_2^-, nitrite; NO_3^-, nitrate; P, phosphorus; PBD-MFC, pretreated buffalo dung microbial fuel cell; PO_4^{3-}, phosphate; S^0, elemental sulfur; SC-MFC, single-chamber microbial fuel cell; stacked MFC, stacked microbial fuel cell; Pt, platinum; SC-BES, single-chamber bioelectrochemical system; SMFC, sediment microbial fuel cell; TC-MFC, triple-chamber microbial fuel cell; TPH, total petroleum hydrocarbon.

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[2]. They also release significant greenhouse gas (GHG) emissions, including carbon dioxide (CO_2) and methane (CH_4) [3], which contribute to global warming and climate change.

On the other hand, agro-industrial wastewater contains many hazardous materials, e.g., ammonium, nitrates, nitrites, and phenol, and high loads of chemical oxygen demand (COD). Still, it also has a high level of chemical energy and nutrients, which could be exploited as a resource through eco-friendly and low-cost processes compared to traditional methods, such as tank aeration [4,5]. Wastewater treatment processes emit a large amount of GHGs, such as CO_2, nitrous oxide (N_2O), and CH_4 [6].

Bioelectrochemical systems (BESs) have emerged as environmentally friendly and integrated technologies that could potentially eliminate organic and inorganic contaminants in wastewater [7]. The microbial fuel cell (MFC) is a derivative of BESs that has gained researchers' attention due to its feasibility in eliminating contaminants in wastewater without harming the environment [8]. Since its development, the MFC has been investigated for wastewater treatment and simultaneous energy recovery [9–16]. MFCs have great potential for converting chemicals, such as sugars, into electricity using microbes as biocatalysts [17].

Several reviews have considered using MFCs and constructed wetlands for pollutant removal [2,8,18]. But they have some omissions, which are revealed in this article. For example, in a review by Santoro et al. [8], the design configuration aspects of the MFCs were not considered. Gupta et al. [18] did not discuss in detail the effects of MFC design configuration on the reactors' performance and mechanisms for waste treatment and electricity production. In addition, the reviews by Bolognesi et al. [19] and Pandit et al. [20] did not address aspects of the removal of inorganic compounds, particularly nitrogen (N), sulfur (S), and phosphorus (P). In summary, the findings of these reviews showed that MFCs are a sustainable form of wastewater treatment due to their ability to (1) eliminate pollutants from various types of wastewater and (2) produce electricity. However, comprehensive reviews of the recent advances in the design, performance, and mechanisms of MFCs for agro-industrial wastewater treatment are still lacking.

To address this gap, this review presents a discussion of the performance and mechanisms of various MFC design configurations for electricity generation and contaminant removal, covering several types of agro-industrial wastewater.

2. Overview of recent advancements in MFCs

2.1. Designs, types, and configurations of MFCs

MFC technology has sparked great debate among researchers worldwide regarding the designs, types (e.g., single chamber, dual chamber, triple chamber), and configurations [21,22]. An understanding of these three parameters is essential to increasing the efficiency of MFCs when operating in the long term, whether under controlled or uncontrolled conditions [23]. An MFC comprises two compartments or chambers, i.e., an anode and cathode separated by a membrane, such as a proton exchange membrane [24]. The anode and cathode are connected to an external circuit using an electrical wire to recover electrical energy [18]. In an MFC, the anode electrode plays a crucial role [25] because it is responsible for bacterial activity. Therefore, it is an essential component in the operation of MFCs. In addition, biofilm formation occurs on the MFC anode's surface and is the necessary motor in electron transfer and MFC performance by EAB activities [26,27]. The electricity-generating capacity of MFC systems also depends mainly on the kind of substrate [28,29] and bacterial community (type of culture), as recently reviewed by Wang et al. [5]. The critical review by Apollon et al. [4] examined the different factors improving BES performance in zero-waste recovery and simultaneous electricity production. They reported that the potential of MFCs to produce significant power density is directly dependent on the kinetics of the reactions of the electrodes within the fuel cell system.

Even though MFCs are promising sustainable systems that have been successfully used to eliminate pollutants in wastewater at the water-energy nexus [30], they face significant challenges [4,31], including (1) low performance, (2) biofouling, (3) scaling up, (4) high-cost materials, and (5) difficulty operating over the long term. These challenges can be overcome by using various methods [32]. MFCs' low performance can be solved by using suitable (new) microorganisms [33] and electrode materials [27,34,35]. In addition, anode surface modification, MFC configuration, and suitable electron acceptors and substrates are vital factors in improving power generation efficiencies in MFCs. For anode modification, pure and mixed cultures have been developed in the laboratory to enhance MFC power output, and a detailed discussion was presented in a recent review by Wang et al. [5]. To date, researchers have undertaken extensive efforts to implement MFCs on a large scale. However, implementing MFCs so that they can operate in more complex conditions, i.e., in a long-term operation without registering low performance, remains a significant challenge for researchers in the bioenergy field. Further research is required to overcome these limitations and challenges that MFCs face, which is essential in broadening this technology toward practical application and commercialization [36]. Here, we summarize the most recent advances in terms of design and configuration.

2.1.1. Single-chamber MFCs

A single-chamber MFC (SC-MFC) usually comprises an anode and an air cathode (Fig. 1). Recent studies used SC-MFCs to carry out complex wastewater treatment processes [37,38]. These studies show the efforts that researchers all over the world are making to bring MFCs to the field of large-scale application. Wastewater contains large amounts of nutrients and organic matter that can be reused in sustainable agriculture. In addition, the wastewater's organic matter helps improve MFC efficiency in terms of power output.

In the study by Syed et al. [39], an SC-MFC was proposed, designed, and constructed using 100 mL glass bottles to evaluate electricity production and cattle manure pretreatment simultaneously. The SC-MFCs had an anode chamber made of carbon paper (2.5 cm in length and 2.5 cm in breadth) and an air cathode made of platinum (Pt)-coated carbon paper with the same characteristics as the anode. Subsequently, the SC-MFCs were operated with pretreated samples of cow and buffalo dung in a batch mode for five days. The system showed great potential to remove organic pollutants; specifically, it removed 80 % of COD, 87 % of

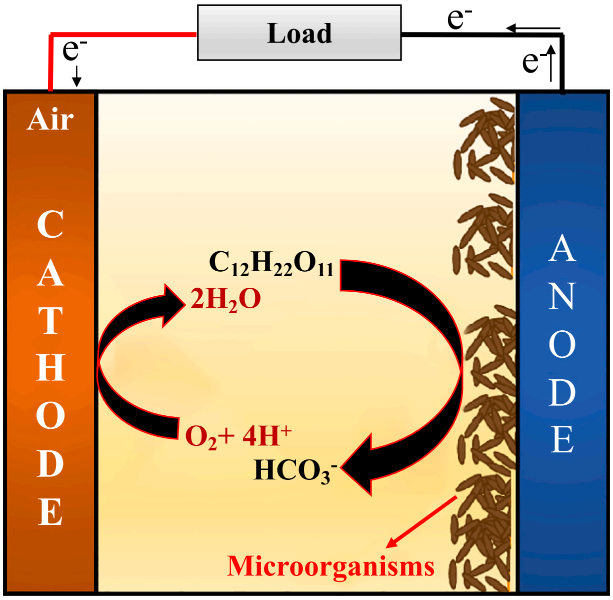

Fig. 1. A typical diagram of an SC-MFC with an air cathode.

biochemical oxygen demand (BOD), and 198 mg/L of total organic carbon in pretreated buffalo dung (PBD). In addition, it recovered nutrients, including 1.9 ± 0.02 mg/L of P, 98.3 ± 1.5 mg/L of N, and 141.2 ± 2.5 mg/L of potassium (K), from PBD. A maximum power density of 12.75 mW/m^2 was reached with MFCs. These outcomes indicate that the SC-MFC is a good option for removing organic pollutants from cattle manure before using it as a biofertilizer. These promising results achieved by Syed et al. [39] showed the feasibility of using MFCs when configured and operated with a single chamber.

However, in one study by Logroño et al. [40], a prototype SC-MFC showed an 18 % higher performance for pollutant (dye) degradation in wastewater than the performance achieved in the previously mentioned study [39]. An SC-MFC with an air biocathode was built to eliminate natural textile dyes from wastewater and generate power simultaneously [40]. The anode and cathode electrodes were constructed using carbon fibers. Subsequently, the anode and cathode were treated before starting the experiment. The SC-MFCs were inoculated with 125 mL of dye wastewater. Removal efficiencies of 54–80 % chromium (Cr), 92–98 % COD, and 98 % zinc (Zn) and a maximum volumetric power density of 132 ± 27.5 mW/m^3 were recorded in this study. According to the study by Fadzli et al. [41], releasing large amounts of dye has adverse environmental effects, such as blocking light and O_2 transfer into the water.

Recently, a novel single air-cathode MFC was built for both wastewater treatment and electricity production [15,42]. The air cathode (surface of 6.7 cm^2) was made of carbon cloth with 0.3 mg/cm^2 of Pt. Then, bamboo charcoal (surface area of 75 cm^2) was used as an anode electrode. The working volume of the SC-MFC was 500–530 mL [42]. The system was suitable for removing contaminants in wastewater, showing removal efficiencies of 80 % COD, 32 % nitrate (NO_3^-), and 11 % phosphate (PO_4^{3-}) [15].

A novel air-cathode clay-cup-membrane (3 mm thickness, 9 cm diameter, with a surface area of 0.0693 m^2) SC-MFC was recently built for the production of electricity and nutrient recovery from livestock urine waste (cow, goat, and sheep) [43]. Graphite felt (648 cm^2) was used for the anode electrode and stainless steel mesh (270 cm^2) for the air cathode. The system's anode had a total volume of 1000 mL, with a working volume of 643 mL. Before operating the SC-MFCs, urine samples were collected and preserved at 4 °C for 72 h. Subsequently, the SC-MFCs were inoculated with cow, goat, and sheep urine and operated for 43 days. The SC-MFC proved a feasible mechanism, recovering 94 % ammonium (NH_4^+), 98 % PO_4^{3-}, and 33 % K in cow urine. After adding the different livestock urines, the system recorded a maximum power output of 46.97 ± 0.67 mW/m^2, which led to a 98 % increase in bioelectricity production. The system had a more effective recovery rate of nutrients than that reported by Paucar and Sato [15]. This difference is due to several factors, including MFC configuration type, membrane, electrode, operating time, and catholyte or anolyte. Another critical factor to consider is the working conditions of both systems; one operated under controlled conditions (lab) [15] and the other operated under uncontrolled conditions (e.g., open field) [43].

In summary, the above study showed that livestock urine is a potential inoculum in the SC-MFC. In addition, the potential to use low-cost materials, such as bamboo and ceramics (clay cups), as electrodes was demonstrated. These results were interesting due to the SC-MFCs' performance regarding nutrient recovery rate and energy production. At the same time, the use of low-cost electrodes points to potential applications in low-income settings. Therefore, further research on MFCs with different anolytes/catholytes is required to improve their performance.

2.1.2. *Dual-chamber MFCs*

A dual-chamber microbial fuel (DC-MFC) is a BES made of two compartments, i.e., an anode and a cathode (Fig. 2). Anaerobic reactions occur in the anode compartment/chamber. In contrast, aerobic reactions occur in the cathode compartment/chamber. More recently, Cui et al. [25] constructed a single-chamber bioelectricochemical system (SC-BES) and a dual-chamber BES (DC-BES) with 650 mL and 325 mL working volumes, respectively. These systems were designed to degrade emerging contaminants such as 2,4,4′-trichloro-2′-hydroxydiphenyl ether (Triclosan). The SC-BESs were made of cylindrical glass and had a bioanode and a biocathode placed at the top and the bottom, respectively. The DC-BESs were configured with plexiglass; pretreated graphite fiber brushes were used for both anode and cathode materials, separated by a cation exchange membrane (CEM). The reactors were operated with domestic wastewater. The reactors were operated in a long-term experiment using a 10 Ω external resistance for over three months. During the investigation, Cui and colleagues also evaluated biofilm formation at the anode and cathode chambers. Biofilms were successfully formed on the electrode surfaces, improving the performance of the reactors. This kind of observation serves as a benchmark for measuring the efficiency of BES systems. It gives researchers an idea of when there is more significant bacterial activity on the surface of the electrodes (i.e., anode or cathode). The SC-BES and DC-BES proved to be viable options for removing the emerging pollutant [25].

In another study, a DC-MFC-based biosensor was used in municipal wastewater treatment [44]. The DC-MFC-based biosensor operated with a total working volume of 300 mL (200 mL artificial wastewater and 100 mL anaerobic sludge) under 300 mg/L COD for wastewater. The system recovered up to 40 mg/L of NH_4^+–N. These findings indicate that,

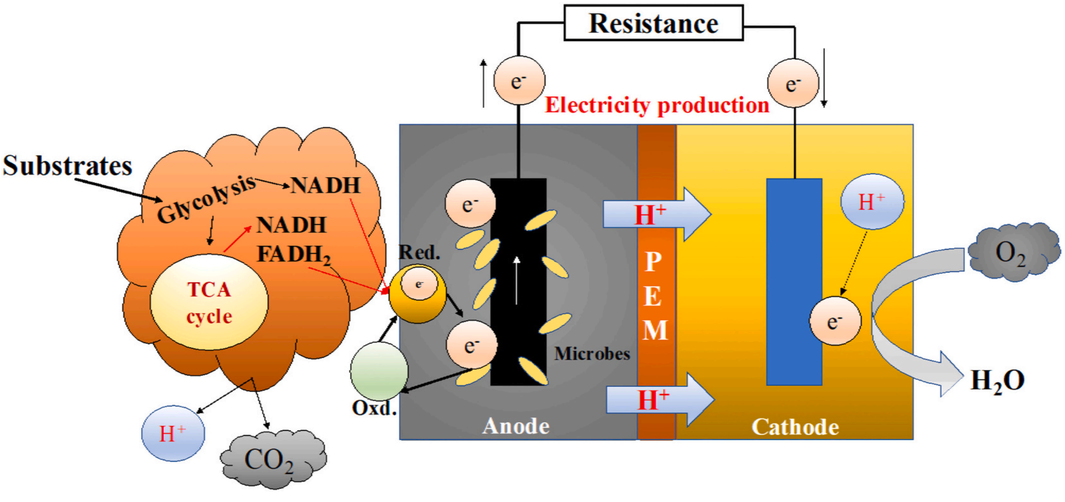

Fig. 2. Typical DC-MFC technology and anode electrode reactions.

like other systems, both the SC-MFC and DC-MFC are feasible to remove contaminants from wastewater. Thus, the present authors argue that any BES performance depends mainly on the type of configuration.

2.1.3. Triple-chamber MFCs

A triple-chamber MFC (TC-MFC) is a device built with two cathode compartments and one anode compartment (Fig. 3) [45]. In a study by Zheng et al. [45], a TC-MFC made of plexiglass with a working volume of 28 mL was proposed, designed, and configured to remove pollutants (toxic metals) from synthetic municipal wastewater. The TC-MFC system had a different design configuration than an SC-MFC or DC-MFC; two membranes were used as separators during the system assembly. The system had two cathode chambers; the first was separated from the anode by a CEM, while the second had an anion exchange membrane (Fig. 3). The pretreated electrodes were built with carbon felt. Subsequently, the TC-MFCs were inoculated with a pure culture (*Acidithiobacillus*) previously isolated from sewage sludge from a municipal wastewater plant. The pure strains must be ready before starting the MFC system. One of the advantages of using previously identified strains is that it helps with monitoring phenomena during investigations. That means the bacterial population is known, and predictions can be made when analyzing the bioinformatics data. The TC-MFC registered high removal efficiencies of bioleaching of 90.2 % S° and 93.1 % $Fe(OH)_3$, respectively [45]. Recently, Sarkodie et al. [46] reported that a bioleaching process is a good option for eliminating toxic metals due to its low cost and eco-friendliness. However, like other methods, bioleaching is limited by factors such as CO_2 supply, pH, O_2 level, nutrients, and temperature. Despite that, the bioleaching process remains one of the most promising sustainable methods using BESs. However, it involves further work to recover and recycle the bioleaching elements.

2.1.4. Stacked MFCs

Stacking MFCs first arose in the 20th century (1931) when a Cambridge researcher (Cohen) connected several MFCs in series. The system produced an output of up to 30 V, with a current of 2 mA [47]. However, the prototype could maintain a different voltage output over long-term operation. Table 1 presents an overview of several studies (in chronological order) that have evaluated the performance of stacked MFCs in removing pollutants from industrial, domestic, and municipal wastewater. As can be seen, all the systems successfully removed contaminants from wastewater through COD.

The first full-scale experiment (stacked MFCs, i.e., 24 units) was carried out in 2011 by Cusick and colleagues (Table 1), who built a pilot-scale prototype with a working volume of 1000 L operating over 100 days [48]. As time passed, the system voltage increased. A maximum volumetric current density of 7.4 A/m^3 (7400 mA/m^3) was reached. The system also indicated a 62 % removal of COD in winery wastewater. Later, Feng et al. [49] used another prototype made of six stacked MFCs, with an operating volume of 250 L, for municipal wastewater treatment. This system was the most effective in terms of COD elimination (79 %) compared to the previous study [48]. However, when stacking 10 units, with an operating volume of 90 L, and 150 units, with a working volume of 1000 L, maximum COD removal efficiencies of up to 87 % [50] and 80 % [51], respectively, were reached when treating wastewater. These results show that factors such as system configuration and volume did not influence the performance of the reactors. These systems had different operating times (Table 1).

In addition, for the treatment of domestic wastewater (DWW), 18 MFC units were stacked in a continuous mode of operation [54]. The stacked MFCs, with a working volume of 700 L, achieved a maximum COD removal of 87 % and power of 136.55 mW. In contrast, Das et al. [55] showed the same COD removal value for DWW when stacking six MFC units with a working volume of 720 L. The above studies differed in electrode materials and configuration; however, they had the same objective, i.e., improving the performance of MFCs for DWW treatment. Zhang and Liu [56] recently reported the highest COD elimination efficiency at 99.28 % using stacked MFCs for wastewater treatment. This performance was better than all other studies to date (Table 1).

In one study by Gajda et al. [52], 560 MFC units were stacked in the module to improve the power generation efficiency for possible practical application. Terracotta (small and large) was used to configure and operate the MFCs. The anode electrodes consisted of carbon veil fiber (20 g/m^2), and the cathode electrodes were fabricated by mixing activated carbon. The study indicated that stacking MFCs with a larger anode surface area is feasible for increasing MFC performance in power output. There are two ways to connect MFCs in stacking mode: (1) MFCs stacked in parallel (Fig. 4a) and (2) MFCs stacked in series (Fig. 4b). MFCs stacked in series have higher bioelectricity production [42,60] than MFCs stacked in parallel. Prototype stacked MFCs were constructed for simultaneous urine pretreatment and energy recovery [53]. Thirty-two individual MFCs were connected in two batch modes (parallel and series) for >120 days in a public restroom on the California Institute of Technology (Caltech) campus. Stacked MFCs used in the previous study could simultaneously remove total organic carbon of 0.75 ± 0.25 g C L^{-1} and COD of up to 5 g O_2/L [53]. According to the study, stacked MFCs involve scaling up wastewater treatment and electricity

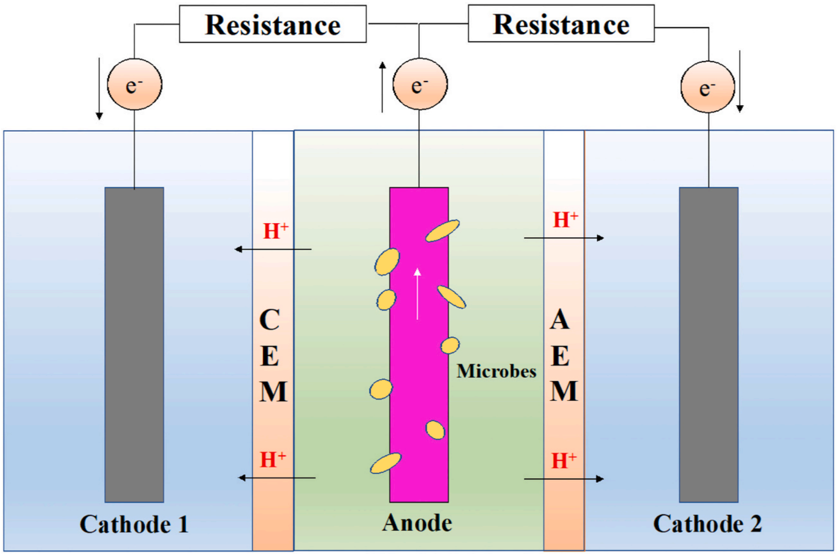

Fig. 3. Typical design of a triple-chamber MFC. The first cathode is separated by a CEM, and the second one by an anion exchange membrane (AEM).

Table 1
Stacked MFCs used in previous works in wastewater treatment.

MFC configuration	Volume (L)	Operation time (days)	Type of wastewater	Chemical oxygen demand removal (%)	Performance	Reference
Stacked MFCs (24 units)	1000	100	WWW	62 ± 20	7.4 A/m^3	Cusick et al. [48]
Stacked MFCs (6 units)	250	10	MWW	79	116 mW	Feng et al. [49]
Stacked MFCs (10 units)	90	7	BWW	78–87	171 mW/m^2	Dong et al. [50]
Stacked MFCs (150 units)	1000	365	MWW	70–80	7–60 W/m^3	Liang et al. [51]
Stacked MFCs (560 units)	0.2	80	Activated sewage sludge	N/A	245 mW	Gajda et al. [52]
Stacked MFCs (32 units)	3.5	120	Urine waste	N/A	23 mW/m^2	Cid et al. [53]
Stacked MFCs (18 units)	700	60	DWW	87 ± 4.5	136.55 mW	Valladares Linares et al. [54]
Stacked MFCs (6 units)	720	~15	DWW	87.29 ± 7.28	61 mW	Das et al. [55]
Constructed Wetlands-MFC (vertical subsurface)	16.7	180	RMWW	80 ± 7.98	25.71 mW/m^3	Mittal et al. [17]
Stacked MFCs (unit stacking)	NA	NA	Nylon Wastewater	99.28	3201 mW/m^3	Zhang and Liu [56]
Stacked MFCs	36	92	DWW	93.52	47.7 mA	Suransh et al. [57]
Stacked MFCs (plug flow channels; 6 anodes)	NA	NA	IWW	82	57.59 mW/cm^2	Opoku et al. [58]
Stacked MFCs (multi-electrode modules)	1000	180	MWW	34	100 mW/m^2	(Heinrichmeier et al. [59]

BWW: brewery wastewater; DWW: domestic wastewater; IWW: industrial wastewater; MFC: microbial fuel cell; MWW: municipal wastewater; RMWW: real municipal wastewater; WWW: winery wastewater.

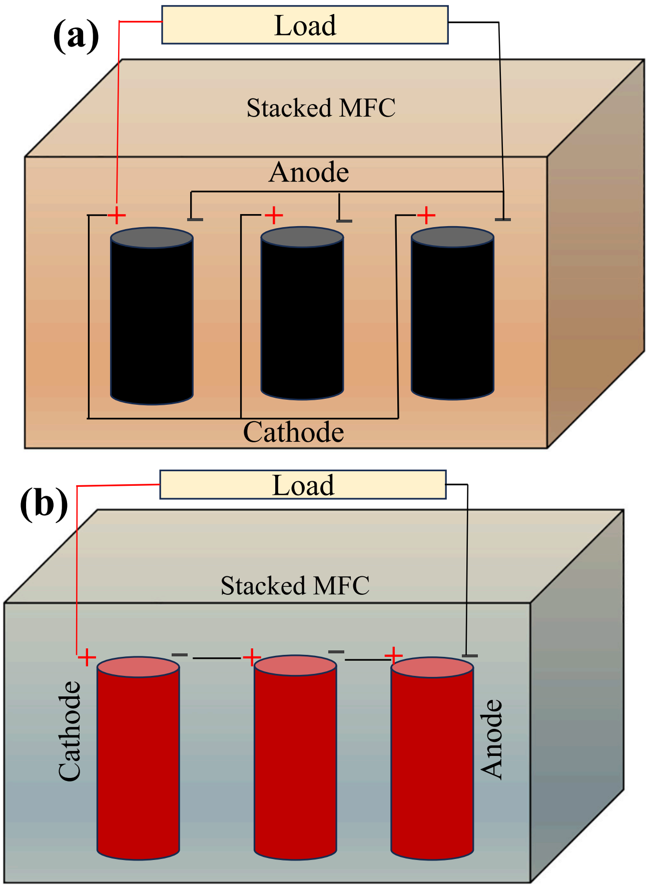

Fig. 4. Schematic diagram of stacked MFCs connected in: (**a**) parallel and (**b**) series.

production for practical applications toward commercialization.

In summary, the MFC designs, types, configurations, and operations discussed above have shown great strides over the past decade. All these advances have aimed to increase MFC efficiency in terms of power output and wastewater treatment efficiency. In addition, these studies also agreed on the anode surface modification as well as the appropriate materials to carry out said modifications. From these facts, the present authors can argue that BES performance depends not only on the type of configuration, but also on other factors, such as pH, temperature, anolyte or catholyte concentration, type of matter (organic or synthetic), bacterial community (including their capacity to carry out electrons transport), and environmental conditions during the system's operation. The authors of the present review suggest more in-depth studies on MFC hybrids to carry out the application of MFCs on a large scale, correcting the limitations of these systems at low power. A comparison of the performance among different configurations of MFCs is still problematic due to differences in electrode materials, wastewater, and operating conditions. This calls for systematic comparative studies of MFC configurations with all other factors or aspects fixed.

3. Agro-industrial wastewater treatment using MFCs

3.1. Nutrient removal/recovery

Many methods are available for treating wastewater containing inorganic contaminants, all with their advantages and disadvantages. The choice of treatment method depends on the type of wastewater, limiting factors, and further application. Reverse osmosis, electrodialysis, and ion exchanger methods do not depend on temperature and do not use additional reagents, but their vast disadvantage is the need for expensive membranes. Moreover, these methods can change water quality (for example, corrosivity and aggressiveness). Physicochemical methods, on the other hand, are easy to use, but the critical disadvantage is the use of chemicals and the need for additional purification. A promising alternative method for treating agricultural wastewater is the use of BES, which makes it possible to combine various production processes and generate electricity. This is an environmentally friendly resource-saving technology allows the reuse of resources.

3.1.1. Nitrate, nitrite, and ammonium

Nitrates (NO_3^-), nitrites (NO_2^-), and ammonium salts (NH_4^+) are nitrogen-containing substances. Nitrates and ammonium can enter water bodies through the discharge of raw or partially treated water. Farmers are actively adding nitrate-based fertilizers to the soil as the demand for food increases and crop rotation needs to be more sustainable. Nitrogen-containing substances are formed in water due to the breakdown of organic protein impurities, mainly urea and proteins from fecal contamination [61]. NO_2^- is a clear indicator of fresh fecal water contamination, especially when both ammonia and nitrite are elevated [62].

Nitrogen does not threaten humans and intensifies agricultural production when appropriately managed. However, if the amount applied to the soil exceeds what the plants need, the N can leach into groundwater, polluting it. According to the World Health Organization, the maximum allowable limit for nitrate in drinking water is 10 mg/L. The presence of NO_3^- in drinking water can have negative consequences on human health [63]. When interacting with bacteria, NO_3^- can turn into NO_2^-, which causes methemoglobinemia [64]. They react with blood hemoglobin, forming methemoglobin. Unlike hemoglobin, this substance does not carry oxygen, which leads to oxygen starvation in tissues. As a result, the state of health worsens, accompanied by lethargy. NO_3^- can form compounds with carcinogenic potential, such as nitrosamines and nitrosamides [65]. NO_3^- has a detrimental effect on the nervous system, cardiovascular system, gastrointestinal tract, and other organs. Therefore, it is essential to purify water and soil from excess nitrate.

The microbiological reduction of nitrates to nitrites and further to gaseous oxides and molecular nitrogen is called denitrification. The successful removal of N in wastewater involves two processes: (1) nitrification and (2) denitrification. The denitrification process proceeds in stages:

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$$

Biological and bioelectrochemical wastewater treatment for nitrogen compounds can be integrated into the MFC [66]. In 2007, NO_3^- was used as a substantial electron acceptor in an MFC cathode chamber for the first time by Clauwaert et al. [67]. The general view of chemical reactions in an MFC is shown in Fig. 5. A biofilm is formed on the anode, which oxidizes carbon compounds, while denitrification occurs on the cathode. Electron transfer occurs either directly via the outer membrane of c-type cytochrome, the flavin bound to c-type cytochrome, or aided by mediators [68]. *Geobacter* and *Shewanella* species can also form nanowires that transfer electrons [69]. Bacteria capable of external electron transfer also include *Comamonas*, *Desulfovibrio*, *Dysgonomonas*, *Enterobacter*, *Klebsiella*, *Pseudomonas*, and *Rhodopseudomonas* [70].

Ammonium degradation is also possible in an MFC. The recommended ammonium ion level for drinking water, according to the World Health Organization, is 50 mg/L. The pH increases due to the synthesis of protons in the anode and their transport in the cathode. High pH drives NH_4^+ ions to form ammonia ($NH_4^+ \rightarrow NH_3$), which can penetrate outside through the air-cathode gas diffusion [71,72]. Consequently, the concentration of total ammoniacal N in wastewater can be reduced. This process works well with an air cathode [73,74] but worsens with an aqueous cathode, where air purge is essential for proper NH_4^+ removal [75].

Another way to remove NH_4^+ is nitrification, followed by denitrification. Nitrifier microorganisms include *Aquamicrobium*, *Nitrobacter*, *Nitrococcus*, *Nitrocystis, Nitrosococcus*, *Nitrosolobus*, *Nitrosomonas*, *Nitrosopira*, *Nitrosovibrio*, *Nitrospina*, *Nitrospira*, *Phycisphaera*, and *Truepera* [70]. Denitrifier microorganisms include *Acidovorax*, *Azoarcus*, *Bellilinea*, *Comamonas*, *Devosia*, *Diaphorobacter*, *Ignavibacterium*, *Limnobacter*, *Nitratireductor*, *Nitrosomonas*, *Oligotropha carboxidovorans*, *Paracoccus*, *Petrimonas*, *Pseudomonas*, *Rhodopseudomonas*, *Thauera*, and *Thermomonas* [70].

Chemoautotrophic nitrifying bacteria carry out nitrification under aerobic conditions in two separate reactions. First, ammonia-oxidizing bacteria oxidize ammonia to nitrite ($NH_3 \rightarrow NO_2^-$) using ammonia monooxygenase (enzyme). Second, nitrite-oxidizing bacteria oxidize nitrite to nitrate ($NO_2^- \rightarrow NO_3^-$) using nitrite oxidoreductase. It has been noted that nitrifying bacteria naturally grow and function on an air cathode [76] or on electrodes in a medium with or without organic substrates [77,78]. However, in most cases, the growth of nitrifying bacteria is slowed due to energy release, and an additional step of nitrifying bacteria enrichment in a mineral salt medium is needed. Further, denitrifying bacteria can use NO_3^- as electron acceptors in the absence of O_2, leading to the formation of gaseous nitrogen-containing chemicals, such as N_2, NO, and N_2O.

An MFC is effective in providing electrons to the denitrifying biofilm and can decrease NO_3^- at a rate of 146 g NO_3^-N/m^3/day [67]. Kondaveeti and Min revealed the removal of nitrate at a rate of 2.04 g NO_3^-N/m^2/day, as nitrate is converted to NH_4^+ or NO_2^- as the predominant product [79]. Some MFC parameters with the possibility of removing nitrogen from wastewater are shown in Fig. 6. Virdis et al. [80] used a DC-MFC with granular graphite electrodes and successfully performed simultaneous nitrification and denitrification in the cathode chamber. Xie et al. [81] achieved high nitrogen removal efficiency of up to 97.3 % and a high volumetric power density of 14 W/m^3 in coupled MFCs, but glucose oxidation had low coulombic efficiency. Zhang and He [82] showed that energy generation is higher than energy consumption, but performance is poor at high N-loading rates and NO_3^- diffusion through separators. A high nitrogen removal at a COD-to-N ratio of 5:1 was demonstrated, but there was poor coulombic efficiency [83]. Wu et al. [84] performed high N removal at a low COD-to-N ratio of 2.8 but also observed poor coulombic efficiency. Li et al. [85] achieved the highest nitrogen removal at up to 99 %. Zhang et al. [86] showed no need for pH control in the anode and cathode electrodes.

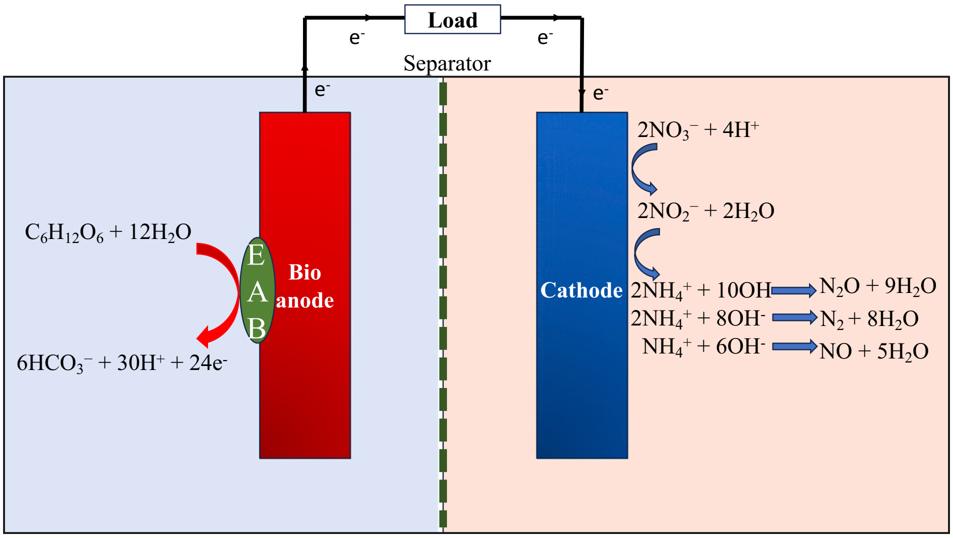

Fig. 5. Schematic diagram of the N-removal mechanism in an MFC.

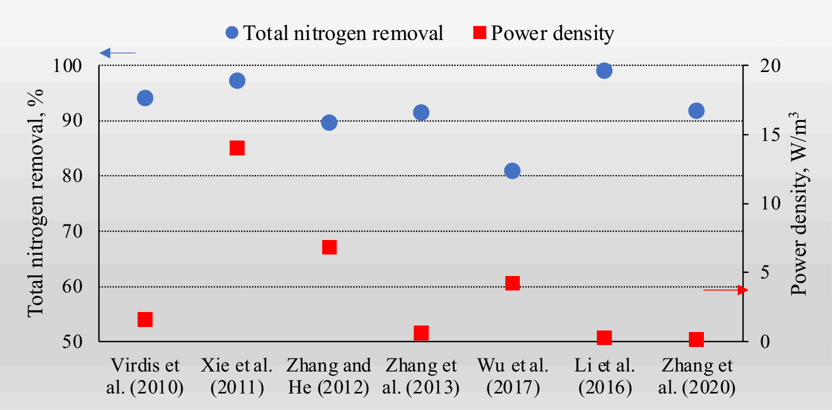

Fig. 6. Ability of some experimental MFCs to remove nitrate and generate power.

Thus, the best results for complex NH_4^+-N wastewater treatment and power generation are obtained using mixed nitrifying sludge – ammonium was oxidized to nitrate and 14.0 W/m^3 of electricity was generated using an oxic-biocathode, and nitrate was electrochemically denitrified and 7.2 W/m^3 of electricity was generated using an anoxic-biocathode in an MFC [81].

3.1.2. Phosphate

Phosphorus is an essential element in various areas of human life and a necessary element for organisms. It is a crucial component of cell structure, bones, and genetic materials [87,88]. In addition, it regulates biological activity by participating in biochemical reactions, such as adenosine triphosphate formation [89]. One of the most critical applications of this element is in the production of fertilizers for the agro-industry, since phosphorus, along with potassium and nitrogen, stimulates the growth and fruiting of many crops. In addition, phosphorus compounds are used in the food industry as acidifiers (for example, in carbonated drinks), thickeners (in the bakery business), and preservatives for oils and frozen vegetables. The domestic use of phosphorus compounds is also pervasive, as they are part of the surfactants for detergents and washing powders. Thus, domestic wastewater contains phosphorus due to human activity and the widespread use of synthetic detergents, which contain polyphosphates.

However, excess phosphorus can be harmful to humans and nature. No direct evidence is available on the human health effects of phosphates contained in washing detergents and other household chemicals, but they do have an indirect damaging effect on the environment. For instance, they triggers the processes of water body eutrophication where wastewater is discharged, O_2 depletion in water reservoirs, and hypoxia of aquatic organisms [90].

The primary source of phosphorus entering water bodies is raw and partially treated domestic and industrial wastewater. The most common forms of phosphorus in wastewater are organic compounds, polyphosphates, and orthophosphates (PO_4^{3-}). The limit for phosphate in drinking water prescribed by the World Health Organization (1999) is 1 mg/L. Methods for removing phosphorus from wastewater are more accessible to implement, compared with nitrogen, but also create many undesirable consequences during operation.

In abiotic electrochemical devices, phosphorus removal is induced by an external current [91]. An external power supply is needed to drive non-spontaneous reactions at the cathode. This usually results in high energy consumption. A BES can be used to reduce energy costs. The biological method using MFCs is based on removing phosphorus and generating power with the help of electroactive microorganisms.

Fig. 7 shows a schematic mechanism for removing phosphorus from wastewater with an MFC. The physicochemical method using a reagent is one effective method of wastewater treatment. $Ca(OH)_2$ is widely used as a coagulant. Calcium is a metal active in displacing heavy metals from

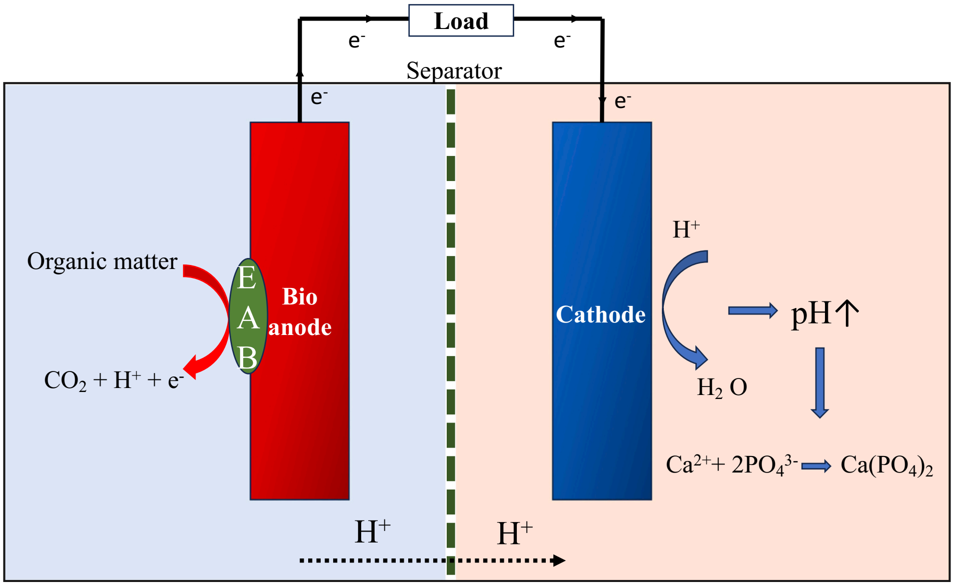

Fig. 7. Schematic of the phosphate removal mechanism in an MFC.

soluble compounds, converting them into insoluble ones, such as calcium monophosphate (CaP). Various salts are precipitated in this situation, including phosphates, sulfates, and chlorides. The pH value determines the degree of precipitation.

With bioelectrochemically mediated phosphorus deposition, energy costs are significantly lower than with abiotic electrochemical deposition [92,93]. A BES can also effectively prevent the formation of chlorinated organic compounds that are highly toxic to living organisms and possibly carcinogenic [94]. Fischer et al. [95] demonstrated that an MFC could provide recovery of phosphorus from digested sewage sludge. But in their work, the MFC technology was only used to dissolve and release phosphate from iron phosphate ($FePO_4$). Phosphorus reduction still required additional chemicals (magnesium and ammonium salts) and pH adjustment. Subsequently, Ichihashi and Hirooka [92] and Hirooka and Ichihashi [96] showed the potential of an air cathode in a single chamber MFC with approximately 70–82 % phosphorus removal.

In the study by Hirooka and Ichihashi [96], P in artificial wastewater was removed in the 19–55 % range as struvite. However, this process depended on the concentration of Mg^{2+} and NH_4^+ ions. Ye et al. [97] tested three types of membranes (CEM, direct osmosis membrane, and non-woven membrane) and discovered that MFCs with CEM provided the best phosphorus removal at 95 % efficiency.

The amount of readily biodegradable organic carbon in domestic wastewater is insufficient for microorganisms to provide sufficient electrical current and a high coulombic efficiency to fully recover phosphorus [93]. Thus, additional organic compounds are usually added to the BES when treating the actual wastewater [96,98]. Phosphorus recovery will rise with increasing levels of organic compounds [94,99]. However, adding organic compounds presents a problem for the application, as this significantly increases the overall cost and may cause unexpected interactions in the aqueous matrix.

When using swine wastewater, natural struvite precipitation was observed on an air cathode. In this BES, 70–82 % of the original total P is removed, and about 4.6–27 % of the total P is recovered by being deposited on the surface of the cathode [92]. Removal of phosphorus through struvite at the cathode is also achieved in single-chamber MECs. The struvite sedimentation rate is 0.3–0.9 $g/m^2/h$, with the removal of up to 40 % of soluble phosphate from the inflow [98]. Cathode deposits need to be cleaned regularly if MFC power generation is a priority [96]. The principle of raising the pH for precipitation was proven early to work in electrolytic cells [100]. However, the electrolysis process requires a significant external energy supply and is less attractive than MFCs. The recovery of P from wastewater in the form of struvite points to the potential of coupling wastewater treatment by MFCs with the production of a biofertilizer.

3.1.3. *Sulfate and sulfide*

In many factories, water used in production becomes contaminated with sulfates, both inorganic and organic, which are harmful to human health. For this reason, almost all industries carry out complex wastewater treatment for sulfates, which allows the acid-base balance of water to be restored and used for further technical or drinking needs.

Sulfates are a natural component of water, and their content depends on the composition of the underlying soil. Sulfates are one of the three main anions of the salt composition of water; that is, the excess of their content co-occurs with an increase in the total hardness and salt content. Based on this, water purification from sulfates is carried out as part of a complex water treatment task.

During the production processes of some industries, wastewater with a high concentration of sulfates is formed. This is primarily where sulfuric acid is used – mining, pulp and paper, oil refining, production of mineral fertilizers, in particular ammonium sulfate $(NH_4)_2SO_4$, and reagent treatment of wastewater, which results in the formation of sulfate ions. Such effluents require mandatory processing and treatment. The World Health Organization has set a limit of 250 mg/L of sulfate ions in drinking water.

Methods for wastewater treatment from sulfates include the physicochemical method (use of reagents at an anion concentration SO_4^{2-} above 2000 mg/L), nanofiltration, reverse osmosis, biological treatment using anaerobic sulfate-reducing bacteria, thermal method, geochemical barriers, and artificial systems (bioplato).

Sulfate serves as an electron acceptor under anaerobic conditions and is thus reduced to sulfide by sulfate-reducing bacteria [101]. In bioreactors, metal ions are precipitated by sulfate-reducing hydrogen sulfide. There are two ways to achieve this: (1) the biological and chemical cleaning process is conducted in parallel – in this case, sulfate-reducing bacteria are not affected by the acidic pH environment; and (2) the process is conducted in two stages – in the beginning, sulfates are converted into sulfides, thereby increasing the pH to an alkaline environment, with precipitation of heavy metals. Then, the sulfides are oxidized to elemental sulfur (S^0), which can later be used in chemical production. To increase the concentration of sulfate-reducing bacteria, easily digestible organic nutrition is used or microorganisms are combined into particular or unique groups.

Desulfurization of sulfate wastewater requires an electron donor to convert sulfate to sulfide, and then an electron acceptor converts the sulfide to S^0. However, in sulfate-carbon wastewater, the end product of sulfate formation is sulfide, which is harmful to living organisms and the reactor. Lee et al. [102] proposed to use the anode surface as an electron acceptor for the sulfide oxidation reaction. Presumably, in an MFC, sulfate ions are effectively reduced by sulfide-reducing bacteria in the biofilm, and the formed sulfide is converted to inert S^0 by sulfide-oxidizing bacteria with the production of electrons by the anode surface (Fig. 8).

Sulfide is corrosive and releases biogas containing hydrogen sulfide, which has an unpleasant smell and must be removed before the biogas can be used in downstream plants [101]. Highly concentrated hydrogen sulfide waters, as a rule, are of anthropogenic origin and are effluents of various industries. Dutta et al. [103] confirmed the oxidation of sulfides with the help of the sulfate-reducing bacteria *Desulfovibrio desulfuricans* on the anode surface to solid sulfur. A laboratory scale experiment is developed based on the abiotic sulfide oxidation. This allows the simultaneous generation of electricity and continuous removal of sulfide from wastewater for two months at a rate of 0.62 kg S/m^3/day [104]. Thus, it is possible to distinguish the advantages of using MFCs for wastewater treatment from various inorganic compounds. This technology is promising, and its potential is now being unlocked and explored for scaling up. Agro-industrial wastewater treatment using MFCs eliminates the use of chemicals, reduces the cost of contaminants removal, and meets the goals of sustainable development. A BES allows the conversion of nitrates to gaseous nitrogen during the removal of nitrogen-containing compounds, provides for the absence of heavy

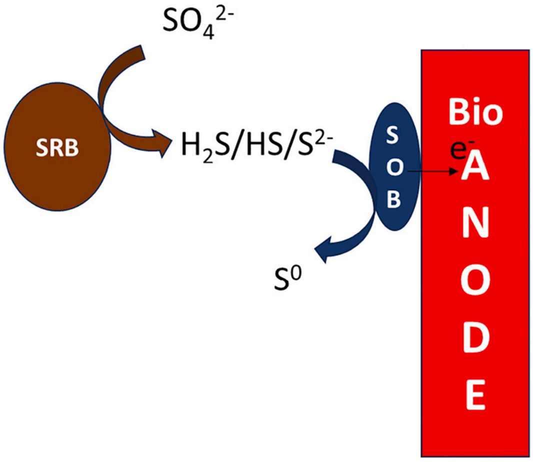

Fig. 8. Schematic of a sulfate-removal mechanism in an MFC.

metals re-contamination during phosphate removal, and allows the activated sludge formation during the sulfate and sulfide removal.

3.2. *Organic compound removal*

3.2.1. *Carbohydrates and organic fatty acids*

The main methods for organic contaminant removal from wastewater include: (1) mechanical methods – used to remove insoluble impurities; (2) biological methods – liquid purification is carried out without the use of chemicals; (3) biochemical methods – microorganisms that feed on pollutants are used along with chemical reagents; (4) chemical methods – used to remove various acids and alkalis from wastewater; (5) physicochemical methods – includes several ways to remove contaminants.

A significant portion of the carbon from organic compounds produced as a result of biological treatment is converted into carbon dioxide and into living bacterial cells, which in themselves are already harmless and often even beneficial to the environment since they can be a source of all the nutrients needed by the soil.

Most agro-industrial wastewater is rich in readily biodegradable carbohydrates and organic fatty acids [105]. These substances represent a promising substrate for MFCs and the field of clean energy. Biodegradable organic matter bacteria include *Desulfovibrio*, *Dysgonomonas*, *Enterobacter*, *Geobacter*, *Klebsiella, Pseudomonas*, *Shewanella*, and *Thauera* [70].

MFCs show a fast rate of organic decomposition at ambient temperature, wherein a small yield of biomass is observed, which might account for the generation of approximately 10 % of sludge in activated processes [106]. To improve performance, MFCs are integrated with advanced membrane processes [107,108]. One exciting example is the development of an osmotic MFC system, which uses a direct osmosis process. An osmotic MFC allows the simultaneous removal of organic pollutants, generation of bioelectricity, and production of high-quality water for possible discharge or reuse [108–110].

The maximum power density of osmotic MFCs reaches 13.6 W/m^3 with breweries, 10.5 W/m^3 with sweetener processing, and 1.3 W/m^3 with swine wastewater. In addition, the soluble COD removal comes to 93.2 % for breweries, 84.1 % for sweetener processing, and 19.3 % for swine wastewater [105]. Some parameters of the MFCs with the possibility of organic removal from brewery, swine, sugar, rice mill, and seafood processing wastewater are shown in Fig. 9.

3.2.2. *Petroleum*

Petroleum remains an important aspect of the economy for many countries, accounting for nearly 32.9 % of worldwide energy consumption [111]. The petroleum-refining industry produces motor fuel, diesel and gasoline, aviation fuel, kerosene, boiler fuel (fuel oil), motor oils and lubricants, etc., from crude oil. The petroleum industry consumes a large amount of water, with the production of abundant wastewater containing recalcitrant hydrocarbons in high concentrations, posing potential environmental threats [112]. A large extent of oil pollution occurs because of faulty exploitation technologies and transportation, which inevitably result in the leakage of petroleum and its derivatives into the environment [113]. Accordingly, the remediation of petroleum hydrocarbon-polluted wastewater, including polluted soil, is relevant due to toxic and harmful components.

Microorganisms are known for their ability to degrade or remove a wide range of organic pollutants, such as antibiotics and pesticides, even such environmentally stable compounds as petroleum hydrocarbons [114–116]. MFCs capable of degrading petroleum are an innovative remediation method that use electroactive bacteria in combination with petroleum-degrading bacteria that both biodegrade hydrocarbons and produce bioelectricity [117,118]. Complex organic oil hydrocarbons, such as polycyclic aromatic hydrocarbons, are initially converted into smaller molecules via hydrocarbon-degrading bacteria, and further, their intermediates serve as electron donors for exoelectrogenic anode bacteria [119]. Thus, using a metabolite of hydrocarbon conversion, the electrogenic microorganisms generate electrons and protons, resulting in bioelectrochemical potential.

Global research on various kinds of MFCs that exploit petroleum has been actively conducted recently. However, at the beginning of 2000, only a couple of works dedicated to this topic were available in the ScienceDirect database. Still, during the last five years, their number has exceeded several hundred every year (Fig. 10). Fig. 10 presents the number of manuscripts found with the terms MFC and petroleum in each year. Numerous works demonstrate that the performance of an MFC in the degradation of hydrocarbons depends on many factors, such as (1) design (e.g., chamber configuration, anode, and cathode materials); (2) chemical (redox mediators and co-substrate); and (3) biological (microbial community).

The idea behind using MFCs in petroleum degradation while producing electricity is fascinating since the researchers noticed improved oil-removal efficiencies. Despite the increased interest in using petroleum as a substrate for MFCs, few studies have investigated MFCs' accelerating effect on petroleum degradation in wastewater. Table 2 presents the latest progress over the last five years, which has not been presented in any other review, and shows both the bioelectrical parameters and the removal efficiency of petroleum hydrocarbons from different types of wastewater.

Table 2 represents the set of leading technical factors – single- and dual-chamber configurations, electrical stimulation, aeration and anaerobic conditions, and illumination – that impact the performance of MFCs in degrading petroleum hydrocarbons from different types of wastes. Previous studies have considered the design and configuration of MFC systems with the primary objective of scaling up this technology to practical application. Consequently, Permana et al. [120] developed two MFCs, i.e., an SC-MFC and DC-MFC, for removing oil in contaminated sediments. The data indicate a higher total petroleum hydrocarbon (TPH) removal rate of 46.15 %, with a maximum power density of 50,570 mW/m^2, in the SC-MFC. In contrast, the DC-MFC showed an elimination rate of 25.64 %, with a maximum power density of 5760 mW/m^2, compared to the control, with the MFC not reaching 10.26 % of

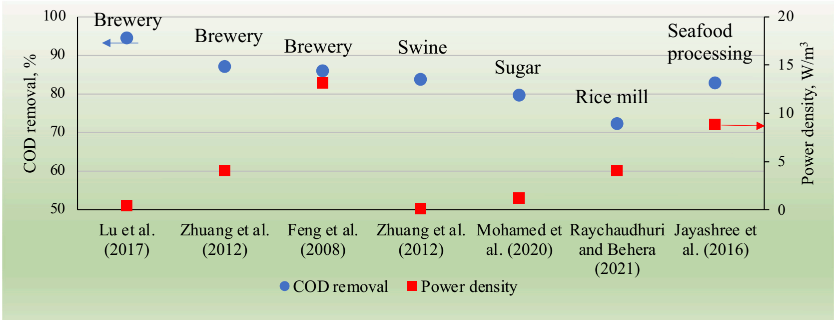

Fig. 9. Ability of some experimental MFCs to remove COD and generate power.

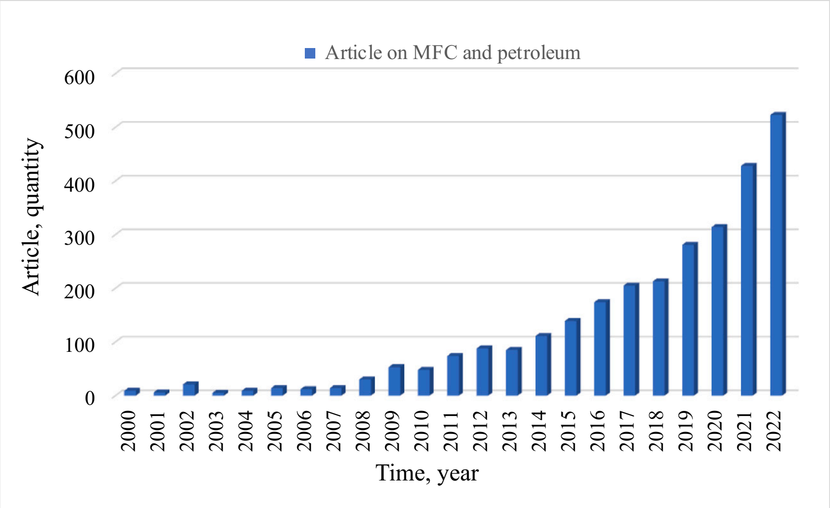

Fig. 10. Number of articles retrieved using search key words “MFC and petroleum” in the ScienceDirect database for the period 2000 to 2022.

Table 2
Performance of recently developed petroleum-degrading MFCs depends on MFC design and/or comparison with the biological removal process.

Configuration of MFC	Electrode material	Type of waste treated	Volume	Petroleum Hydrocarbon	Removal efficiency (%)	Power density (mW/m^2)	Reference
SC-MFC	Carbon as anode and air cathode	Contaminated sediment	N/A	TPH	46.1	50,570	Permana et al. [120]
DC-MFC	Copper (Cu) (0.008 m^2) as anode and cathode	Contaminated sediment	1 L	TPH	25.64	5760	Permana et al. [120]
Control (without MFC)	–	Contaminated sediment	N/A	TPH	10.26	N/A	Permana et al. [120]
SMFC	Graphite felt (10 × 80 mm)	Oily sludge	2 L	TPH	44.73	N/A	Guo et al. [121]
Control (without MFC)	–	Oily sludge	2 L	TPH	4.42	N/A	Guo et al. [121]
BES 0.1 V	Graphite rod (1 × 7.5 cm)	Wastewater	200 mL	Phenanthrene	58.74	N/A	Wang et al. [122]
BES 0.2 V	Graphite rod (1 × 7.5 cm)	Wastewater	200 mL	Phenanthrene	88.27	N/A	Wang et al. [122]
BES 0.3 V	Graphite rod (1 × 7.5 cm)	Wastewater	200 mL	Phenanthrene	53.54	N/A	Wang et al. [122]
Control (without BES)	–	Wastewater	200 mL	Phenanthrene	61.07	N/A	Wang et al. [122]
MFC	Graphite plates (9 × 4 × 0.5 mm)	Mill wastewater	800 mL	Phenol	68	11	Bagheri et al. [123]
DC-MFC	Uncoated carbon felt anode and Pt/C coated cathode	Wastewater	100 mL	Total alkanes	25.5	38	Nandy et al. [124]
DC-MFC	MnO_2 coated carbon felt anode and platinum/carbon-coated cathode	Wastewater	100 mL	Total alkanes	36	47	Nandy et al. [124]
Aerobic SC-MFC with a photocatalytic-BEA	Carbon felt (4 × 5 cm)	Wastewater	120 mL	o-Chlorophenol	67.1	255	Wang et al. [125]
Anaerobic SC-MFC with a photocatalytic-BEA	“	Wastewater	120 mL	o-Chlorophenol	31.4	184	Wang et al. [125]
Aerobic SC-MFC	“	Wastewater	120 mL	o-Chlorophenol	20.5	175	Wang et al. [125]
Anaerobic SC-MFC	Carbon felt (4 × 5 cm)	Wastewater	120 mL	o-Chlorophenol	17.7	134	Wang et al. [125]
Anaerobic bioreactor	Carbon cloth as anode and cathode	Groundwater	N/A	Phenanthrene	32.16	N/A	Liu et al. [126]
Anaerobic ES-bioreactor	Carbon cloth as anode and cathode	Groundwater	N/A	Phenanthrene	90.94	N/A	Liu et al. [126]
DC-MFC	Graphite plate as anode and cathode	Real-field petroleum refinery wastewater	1 L	Furfural and Phenol	99	70.53 (552 mW/m^3)	Jabbar et al. [127]

BEA: Bioelectrochemical Anode; SMFC: Sediment Microbial Fuel Cell; SC-MFC: Single-Chamber MFC; DC-MFC: Dual-Chamber MFC; TPH: Total Petroleum Hydrocarbon.

TPH. According to this study’s findings, the best configuration to remove TPH in contaminated soil is the SC-MFC.

In addition, recently, it was shown that illumination and dissolved oxygen facilitate the cathodic oxidation-reduction reaction, upgrading electricity generation from 134 to 255 mW/m^2 [125]. Moreover, MFC technology and electric stimulation were considered simple ways to accelerate the treatment of oily sludge and wastewater while generating electricity. As seen in Table 2, the rate of oil hydrocarbon degradation while using an MFC could be up to 10.1 times more efficient than without a BES [121].

What are the mechanisms of an MFC's accelerating effect on petroleum degradation? A few ways are known to impact the operation of BESs on degrading microorganisms. The MFC technology increases the efficiency of the biodegradation process via (1) increasing microbial diversity and (2) enhancing extracellular electron transfer. In 2012, the quantity of hydrocarbon-degrading bacteria in the anode area was shown to increase by nearly 50 times (373 ± 56 × 103 CFU/g soil) in the soil of an MFC, compared with the control without MFC and electric operation (8 ± 2 × 103 CFU/g soil) [128]. After the sediment MFC treatment, the amount of hydrocarbon-degrading microorganisms *Bacteroidia* and *Pseudomonadales*, which are capable of secreting redox mediators, as well as the number of electrogenic bacteria *Chloroflexi*, was significantly higher than that in the original sludge [121]. These findings demonstrate strong evidence for a positive relationship and synergism between hydrocarbon-degrading and electroactive bacteria in the degradation of crude oil hydrocarbons. The predominance of bacterial communities participating in hydrocarbon degradation and electrogenesis in an MFC, such as *Rhodococcus*, *Alkaliphilus*, *Delftia*, and *Luteimonas* [124], signifies a prerequisite of successful petroleum removal of up to 36 %, with simultaneous power generation of 47 mW/m^2.

The microbial abundance of bacteria degrading chlorophenol *Azospirillum* sp. and electrogenic bacteria *Comamonadaceae fam.* Was increased from 0.3 % to 17.0 % and 0.48 % to 32.1 % with illumination and aeration, respectively, during electricity generation (of 255 mW/m^2) in an MFC with a photocatalytic-bioelectrochemical anode [125]. MFC technology can also enhance the degradation of petroleum pollutants in anoxic environments [118,129]. Electric stimulation was found to promote anaerobic bioremediation of phenanthrene contaminants in groundwater, hence opening up a new way of bioelectrochemistry application [126]. The interaction between the microbes of biofilm attached to the electrodes or anode and cathode provides fast electron transfer, while the hydrocarbons compound degradation and, as a result, facilitate biodegradation [119,130].

A third mechanism by which the electrical flow of an MFC can accelerate bacterial degradation of petroleum products is through effects at the gene level. Wang et al. [122] showed that micro-electric stimulation upregulated nahAc, pcaH, and xylE gene expression, which are responsible for hydrocarbon degradation. A micro-electric field increased phenanthrene degradation in a modified BES: phenanthrene degradation efficiency was 27.2 % superior to biological control, i.e., without electrical stimulation [122]. This important study showed the physiological and metabolic effect of micro-electrical stimulation of a BES on a phenanthrene-degrading strain and promotion of phenanthrene degradation-related enzyme activity.

Electric stimulation and the cooperation and synergism of microorganisms play a crucial role in enhancing the degradation of hydrocarbons aided by MFCs. MFCs are superior in degrading petroleum wastes compared to conventional biological remediation processes. Therefore, MFC-degrading petroleum hydrocarbons present an excellent sustainable tool to treat various kinds of waste while simultaneously serving as a source of bioelectricity.

In summary, the oil industry and oil fuel exploitation are still powerful worldwide. Oil consumption worldwide increased by 6 % in 2021 compared to the previous year [131]. Hydrocarbons are substances that do not break down easily naturally – a high level of oil pollution has accumulated throughout the years of oil exploitation. And even in the case of a gradual decrease in the use of oil, the remediation of water bodies polluted by oil industrial waste will remain relevant for many years to come.

4. Role of microbes in MFC performance

To discuss BES technology, one must consider the microbial consortia and associated biochemical processes. The performance of a BES system, such as an MFC, is closely associated with microorganism activity [132]. Microorganisms are primarily responsible for carrying out the oxidation reactions on the anode surface as well as the reduction reactions that take place in the cathode electrode. During the start-up of an MFC, microorganisms have the primary function of oxidizing organic matter, and this is mediated by a reduction in bacteria. The bacteria then move the electron to the cathode chamber, producing electricity with the aid of an external circuit [14]. As reported in a literature review, not all microorganisms have the same capacity or characteristics to transfer electrons from the substrate to the anode electrode [133,134]. Therefore, EABs in agro-industrial wastewater must possess the potential to oxidize organic matter in wastewater. An explanation for the difference between each microorganism could be the lack of contact with the anode surface or the absence of active proteins on the electrode surface, as reported elsewhere [135]. The role of microorganisms' cooperators in exoelectrogens, which can convert different compounds, such as antibiotics, heavy metals, dyes, and petroleum hydrocarbons, is considered crucial for agro-industrial wastewater treatment through MFC use.

Microbial removal of pollutants in each type of MFC operating with agro-industrial wastewater directly connects with electricity generation, since pollutants or their metabolites serve as a donor of electrons to exoelectrogens and electrotrophs. The organic compounds are the key component of various kinds of wastewater, mainly represented by COD. A positive correlation has been found between COD removal efficiency and electricity generation [136–138]. Obviously, the higher level of organic matter oxidation results in the enhanced transfer of the electrons to the anode and, consequently, the increased power density [136,137,139]. However, the long retention times lead to a decrease in substrate concentration, demonstrating a negative correlation between COD removal and power production [140,141].

The most effective bacterial species in MFCs are reported to be *Shewanella* spp. and *Geobacter* spp. [142]. These two bacteria have been extensively studied because of their great potential in treating wastewater and a higher electron transport rate, as described in Nawaz et al. [134]. However, the use of a consortium of microorganisms was more effective than pure culture due to the natural formation of optimal microbe combination, which enhances the electrochemical reaction by more efficiently oxidizing the complex organic compounds in wastewater [139]. Even in full-scale wastewater treatment plants with functional stability, bacterial communities have been shown to be variable and dynamic [143]. The stability of a microbial ecosystem is not the result of population diversity as such, but of functional redundancy, which is ensured by the presence of species reservoirs capable of performing the same ecological function [144].

5. Challenges and opportunities in MFC applications

MFCs are systems that address wastewater treatment and sustainable energy production simultaneously, with low energy consumption [20]. Research efforts have focused on optimizing electrode materials and the kinetics of electrode reactions to improve their power output [145]. MFCs are highly effective at removing pollutants, including heavy metals such as hexavalent chromium, from wastewater [146]. An MFC's organic matter removal rate, in comparison with other wastewater treatment methods, is estimated at between 10.32 and 13.73 kg COD/m^3/day [147].

While MFC technology needs further improvement to become economically viable, it could be another method for organic matter removal from the effluent of various industries. MFCs play an essential function in wastewater treatment, providing different strategies as a secondary means of bioenergy generation and a positive way for technological upscaling in wastewater treatment, especially for agro-waste oxidation [148]. Recovery of bioenergy through the wastewater treatment process can help reduce GHG emissions (especially CO_2) and air pollution [149]. However, MFCs still face challenges, such as low pilot-scale power performance, which limit their commercial viability [150]. Proton transfer and electrode material optimization are crucial for commercialization, and wire selection is another important factor affecting MFC performance [151]. Furthermore, MFCs face challenges, such as toxicity in anode biofilm [152], system instability, membrane biofouling (biological contamination of membrane), cost-effectiveness, robustness, longevity, substrate effects, suitability of materials for their design, and large-scale application [4]. Large-scale MFC applications have been presented, indicating that the technology is evolving toward practical applications [48].

MFCs can remove contaminants from various types of wastewater and degrade environmental pollutants [153]. However, scaling up MFCs and addressing material problems are still the most significant challenges in their application for sewage treatment [134]. The production of renewable energy using MFCs is a promising strategy that requires more attention worldwide to carry out the large-scale application of these systems. Overall, MFCs provide the simultaneous advantage of bioelectricity production and wastewater treatment, and optimizing their performance and materials is necessary for their successful commercialization.

6. Future perspectives

The application of MFCs in wastewater treatment is still in its infancy. Thus, several knowledge gaps and challenges exist, critically analyzed in the references. [4,134,154]. Here, several thematic topics were identified for further research. Addressing these knowledge gaps, including low power output, potential toxicity of contaminants, and voltage losses, is necessary for development [4]. Furthermore, most of the work on MFCs has been limited to laboratory-scale studies, while pilots operating for longer time periods are still lacking [150]. Therefore, pilot studies of various designs of hybrid or integrated MFCs are required to provide comprehensive data on such systems' technical and economic feasibility at realistic scales. Such pilot studies will also provide important information on the operational boundaries or limits of the MFC technology: hybrid or integrated MFCs with the capacity for simultaneous generation of bioelectricity. Several potential solutions were suggested in an earlier review [4], but further work is required to develop and evaluate the feasibility and capacity of these solutions to overcome the limitations.

The performance of MFCs is controlled by complex interactions among the various components [155], including microorganisms, solid and liquid matrices, and often heterogeneous biowastes or wastewaters. However, the microbial ecology of MFCs has received limited research attention. For example, the interactions among microbial species and their environment, such as electrode materials and feedstocks, still need to be better understood. Both bioelectricity production and wastewater treatment efficiency using MFC technology were comprehensively reviewed in the references [134]. Thus, there is a need for systematic optimization studies to determine optimum design and operating conditions for typical biowastes or wastewaters used for MFCs. In addition, applying MFCs for bioelectricity generation and wastewater treatment has a potentially lower environmental footprint than conventional technologies [156]. However, data on the environmental impacts and footprint of MFCs concerning water, energy, and GHG emissions are scarce. This calls for comparative life cycle assessment studies of MFCs relative to current and competing technologies [157].

Moreover, increasing interest in minimizing the environmental footprint of technologies calls for the development and application of novel green and renewable materials, such as biochar and its composites, as electrodes and additives in MFCs [112,158,159]. However, the application of biochar and its composites is a recent development in MFCs, and further work is required to develop and test such materials. On the other hand, low-income countries experience high energy poverty coupled with severe environmental pollution due to a lack of advanced water and wastewater treatment technologies. Together, this creates an ideal environment for the development and application of hybrid or integrated MFCs as a potential low-cost technology for simultaneous wastewater treatment and bioelectricity generation. Therefore, research is required in low-income countries to translate current knowledge on MFCs into practical solutions using realistic wastewaters or effluents, such as those from pit latrines and septic tanks, which are commonly used in low-income countries.

Biowastes and wastewater used as feedstock in MFCs often contain a mixture of legacy (e.g., trace metals, pesticides) and emerging (e.g., pharmaceuticals) pollutants [160]. Studies on MFCs have often focused on the fate of legacy contaminants, such as organic (COD, BOD) and nutrient loads [161–163]. However, the behavior and fate of other legacy contaminants and emerging ones commonly detected in biowaste and wastewater need to be better understood. Therefore, the behavior and fate of contaminants in MFCs, including their phase partitioning among biotic and abiotic components, requires further investigation. Such information is critical for developing the final safe disposal, reuse, and recycling of spent MFC substrates/wastes. More information on appropriate enterprise models is also needed to promote the uptake, adoption, and upscaling of MFCs for bioelectricity generation and wastewater treatment. Thus, a determination on whether MFCs could be used as a stand-alone technology or as part of a technology mix among current ones needs to be made. Further work is required to understand the appropriate models to introduce and implement MFCs as part of a broader circular bio-economy strategy based on biorefineries designed to convert biowastes to value-added products.

Finally, in some countries, stakeholders, including consumers, regulators, and policymakers, tend to have negative attitudes or perceptions about waste-based products, especially those derived from human fecal matter [164–166]. These negative perceptions often arise from a lack of knowledge, solid religious and socio-cultural beliefs, and the potential environmental and human health risks associated with waste handling and waste-based products [164,165]. For example, in some religious and cultural settings, products, including energy derived from human excreta, are considered taboo and unacceptable [167]. Therefore, an understanding of stakeholder knowledge, attitudes, and perceptions that could influence the uptake and acceptance of MFCs as a technology for wastewater treatment and bioelectricity generation is needed.

7. Conclusions

This critical review discusses the recent advancements in MFCs for wastewater treatment, with a particular focus on agro-industrial wastewater treatment, and energy recovery, as well as an overview of different studies on microbial fuel cells in degrading contaminants from wastewater. Furthermore, we argue that BESs, such as MFCs, offer a feasible and sustainable alternative for removing organics and nutrients (i.e., phosphorus and nitrogen), inorganics (including sulfate, sulfide, heavy metal removal), and other compounds, such as antibiotics, dyes, and petroleum, from agro-industrial wastewater.

Specifically, this review presents an analysis and summary of recent innovative advances in MFC technology in agro-industrial wastewater treatment, including configurations and functional parameters of single-chamber, dual-chamber, and triple-chamber stacked MFCs. Such analysis will allow the MFC technology to be used in real energy production and agro-industrial wastewater treatment. In addition, it will enable a comparison of standard wastewater treatment methods, with MFCs as an environmentally friendly technology using electrochemically active bacteria. To achieve this, the following aspects have been considered: (1) removal of nitrogen-containing substances in terms of compliance with standards for drinking water, (2) use of MFCs for phosphate, sulfate, and sulfide removal and recovery as nutrients using various strains of bacteria, (3) generation of electricity during the wastewater treatment process, and (4) removal of organic compounds (carbohydrates, organic fatty acids) and degradation of petroleum hydrocarbons.

Mechanisms of an MFC's accelerating effect on petroleum degradation were reviewed and summarized for the first time. In addition, the critical role of microorganisms capable of wastewater treatment and bioelectric power generation was discussed. Finally, we presented the challenges, opportunities, and future perspectives regarding MFC development and application.

CRediT authorship contribution statement

Wilgince Apollon: Conceptualization, Methodology, Writing – original draft. **Iryna Rusyn:** Writing – review & editing, Methodology. **Tatiana Kuleshova:** Methodology, Writing – review & editing. **Alejandro Isabel Luna-Maldonado:** Supervision, Writing – review & editing. **Jacques Fils Pierre:** Writing – review & editing. **Willis Gwenzi:** Writing – review & editing. **Vineet Kumar:** Writing – review & editing.

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.

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