Bioremediation of Petrochemicals and Dye Industrial Effluents through Microbial Fuel Cells

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1 Introduction

A fuel cell that uses microorganisms as catalysts for the oxidation of substrates, leading to electricity generation, is termed a microbial fuel cell (MFC). They are deployed for sustainable energy production, treatment of wastes, and minimizing CO_2 emissions. There is a compelling need for energy- and cost-efficient alternates to the conventional wastewater treatment systems due to their high energy requirements. Domestic and industrial effluents contain a multitude of organic compounds that can fuel MFCs, and the microbes in MFCs can accomplish both pollutant degradation and power generation in parallel. Moreover, it minimizes solid disposal by 50–90% [23] and hence is generally accepted as a promising sustainable biotechnological solution to future energy requirements. However, it is yet to be commercially exploited since there are many hurdles to be overcome. This chapter mainly focuses on the application of MFCs toward the removal of environmental pollutants, such as synthetic dyes and polycyclic aromatic compounds. Recent progress in the understanding of system architecture, anode/cathode materials, and biocatalysts employed in MFCs for pollutant degradation to achieve high energy density has been reviewed.

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2 Bioremediation of Petrochemicals Through Microbial Fuel Cells

The ever-growing global economy and population have ensured an increasing demand for petroleum products, thus aiding the growth and multiplication of relevant industries worldwide [101]. The environmental impact of petroleum products is a correspondingly vast and extensive field. Petroleum hydrocarbons are harmful to many soil organisms and humans. This fossilized fuel has become a necessary source of energy since the late nineteenth century due to elevated consumption globally. The petrochemical industry involves the production of solvents, resins, lubricants, and plastics, generating wastewater containing toxic compounds [79]. In the downstream sector, wastewater from petroleum industries and refineries contains phenolic compounds, aromatic hydrocarbons, salts, alkanes, naphthalene, nitrobenzene, oil, and grease. These pollutants are toxic, not biodegradable, and carcinogenic [17]. In the upstream sector, the exploration and production process of conventional crude oil generates the largest volume of waste stream termed Produced water (PW). Typically, PW contains emulsified oils besides a high amount of sodium chloride, carbonates of magnesium, sulfates, and other inorganic dissolved and suspended solids. Benzene, xylenes, ethylbenzene, and toluene are hazardous chemicals, also known as BTEX materials. They are a threat to human health and are common PW water contaminants [121]. Petroleum product contamination degrades the ecological structure and function of the soil, affecting moisture, pH, carbon/nitrogen ratio, and porosity. As the pollutant concentration rises, the hydrophobicity increases, thereby inhibiting the seed germination [39]. The aromatic hydrocarbon groups of compound derivatives have molecular rings, including benzene rings. Usually, instead of a hydrogen atom in the benzene ring, methyl or ethyl groups are found (toluene, methyl/dimethyl benzene and ethylbenzene) [92]. These hydrocarbons, referred to as polycyclic aromatic hydrocarbons (PAHs), can cause musculoskeletal malformation and bone marrow suppression [93]. PAHs toxicology and physicochemical characteristics are varied according to their molecular weight [104]. Recent studies demonstrate that benzenes can easily bind to soil particles, and BTEX chemical compounds need sufficient oxygen to break down slowly. These compounds are volatile and hence can affect the human respiratory system [58]. Hence, researchers have taken great efforts to upgrade the different technologies used to treat wastewater from the petroleum industry, such as adsorption, coagulation, membrane separation process, dissolved air flotation, and chemical destabilization [103]. However, these unit operations are ineffective for the treatment of petroleum wastewater because of the complexity of PAHs. Although incineration and chemical oxidation can remove up to 99.0 and 92.3% of the total petroleum hydrocarbons from wastewater, respectively, both these restoration procedures have disadvantages. Toxic substances, such as dioxins, furans, polychlorinated biphenyls, and volatile heavy metals, are released into the atmosphere as a result of the incomplete burning of crude oils [19]. Therefore, an effective remediation measures should be tailored considering the human health and ecosystem management.

The biological treatment of petroleum wastewater can be accomplished by aerobic and anaerobic processes. However, the anaerobic process is preferred over aerobic due to lower energy consumption and less sludge production, though it is time-consuming [1, 102]. Bioelectrochemical systems (BES) are versatile [89] because, in contrast to typical bioreactors, they can concurrently generate electricity and treat wastewater. Similarly, in recent years, an emerging technology based on biostimulation, namely microbial fuel cells (MFCs), has been demonstrated to remove PAHs from soils and sediments.

2.1 MFCs for Petroleum Product Degradation

2.1.1 In-Situ Bioremediation of PAH-Contaminated Soil in Microbial Fuel Cells

In situ bioremediation is a biological treatment method to clean up PAHs present in the environment. The potential advantages of in-situ bioremediation are (i) minimal site disruption, (ii) continuous treatment of contaminated soil and groundwater, and (iii) economical. The anaerobic degradation of petroleum compounds has been identified in a wide spectrum of microbial pathways, including denitrification [4, 51], metal reduction [52], and sulfate reduction [84]. MFC configurations with different electrode materials for in-situ bioremediation to remove hydrocarbons from soils and sediments as shown in Table 1. Yuan et al. [113] developed a tubular air-cathode MFC (TAC-MFC) with a cloth cathode assembly and utilized organic sediment as the substrate, demonstrating the highest power density of 107.1 ± 8.6 mW/m^2, coulombic efficiency of 17.9%, and the lowest internal resistance (20 Ω). Bioremediation of PAHs in sediments through MFC technology is shown in Fig. 1; the anodic reactions include the oxidation of organic matter, sulfides, and other potential pollutants. The color of the sediment apparently turned from black to brown after MFC operation in the contaminated sites. Huang et al. [25] reported enhanced phenol biodegradation and simultaneous electricity production from organic pollutants in a soil MFC (Fig. 2). An in-situ MFC was inserted into waterlogged anoxic soil for the remediation of organic pollutants, and the power reached 29.45 mW/m^2 with 90.1% removal of phenol. The phenol degradation rate in this BES was approximately 23 times higher than biological treatment. Similarly, another bioelectrochemical treatment (BET) system showed 41.08% petroleum hydrocarbon removal compared to 20.72% obtained using conventional anaerobic treatment with petroleum sludge as the substrate [59]. Wang et al. [105] demonstrated a U-tube MFC for the bioremediation of PAHs-contaminated soil. Due to the presence of exoelectrogenic biofilms on the surface of the anode, it could effectively remove *n*-alkanes and PAHs from the soil. However, the power density was low ($0.85 + 0.05$ mW/m^2), suggesting that the U-tube air-cathode MFC was not as suitable for soil remediation systems.

To increase the degradation efficiency of total petroleum hydrocarbons (TPH) [117], varied the arrangement of anodes from vertical to a horizontal position in a

Table 1 MFC for treatment of petroleum refinery water effluent

S.No	Petroleum products	Co-substrate	Anode	Cathode	Reactor configuration	Power density	Treatment efficiency	References
1.	Groundwater hydrocarbons	Glucose	Stainless steel	Carbon cloth	Single-cell MFC	120 (mW/m^2)	–	Morris and Jin [66]
2.	Quinoline	Glucose	Carbon	Air	Dual-chambered MFC	16.4 (mW/m^2)	Removal: 200 mg/L	Zhang et al. [114, 116]
3.	Hydrocarbon-contaminated groundwater	–	Carbon	Pt catalyst	Dual-chambered MFC	1.25 (mW/m^2)	COD: 65.6%	Adelaja et al. [2]
4.	Petroleum Hydrocarbon-contaminated waste water	–	Carbon cloth	Platinised-carbon cloth	Dual-chambered MFC	1.06 (mW/m^2)	COD Removal: 79.1%; degradation rate: 91.6%	Adelaja et al. [3]
5.	Complex petrochemical wastewater	–	Stainless steel mesh	Graphite plate	Single chambered BET system	Anoxic: 17.1 (mW/m^2) and Aerobic: 14.3 (mW/m^2)	Substrate degradation: 3.34 kg COD/m^3-day	Yeruva et al. [112]
6.	Ethylene glycol	Glucose	Graphite sheet	Pt sheet	Single chamber MFC	5.72 (mW/m^2)	COD: 92–98%	Hosseinpour et al. [24]
7.	Petrochemical wastewater	–	Carbon cloth	Pt coated carbon cloth	Single chamber MFC	225 ± 1.4 (mW/m^2)	COD: 84.4% and 95% oil removal	Srikanth et al. [94]
8.	Petrochemical wastewater	Acetate	Graphite cylinder	Graphite cylinder	Single chamber graphite BES	222 (mW/m^2)	TPH: 90% COD: 34.9%	Mohanakrishna et al. [60–62]

(continued)

Table 1 (continued)

S.No	Petroleum products	Co-substrate	Anode	Cathode	Reactor configuration	Power density	Treatment efficiency	References
9.	Petroleum refinery wastewater	–	Graphite fiber	Pt coated carbon cloth	Single chamber air-cathode MFC	45 (mW/m^2)	Removal of TPH: 89% COD: 41.2%	Mohanakrishna et al. [60–62]
10.	Petroleum refinery wastewater	Acetate	Bioanode	Biocathode	Single chamber hybrid based BES	Batch mode: 725 (mW/m^2) *continuous mode*: 561 (mW/m^2)	Batch mode: COD: 69.2% *continuous mode*: COD: 37%	Mohanakrishna et al. [60–62]
11.	2,4-dichlorophenol	Glucose	Carbon cloth	Platinised titanium (Pt/Ti) catalyst plate	Dual chamber MFC	37 (mW/m^2)	COD: 41%	Hassan et al. [22]
12.	Petrochemical wastewater	–	Polyacrylonitrile coated carbon felt	Polyacrylonitrile coated carbon felt	Dual chamber MFC	850 (mW/m^2)	COD: 40%	Sarmin et al. [87]
13.	Petroleum produced water	–	Graphite brush	Platinum coated carbon cloth	Dual chamber MFC	1089 (mW/m^2)	sulfate: 93.9%, TPH: 53.1%, COD: 60.2%	Mohanakrishna et al. [64, 65]
14.	Petroleum refinery wastewater	–	Carbon fiber brush	Carbon cloth	Osmotic microbial fuel cell and up-flow microbial desalination cell	84 (mW/m^2)	COD: 93%	Sevda and Abu-Reesh [88]

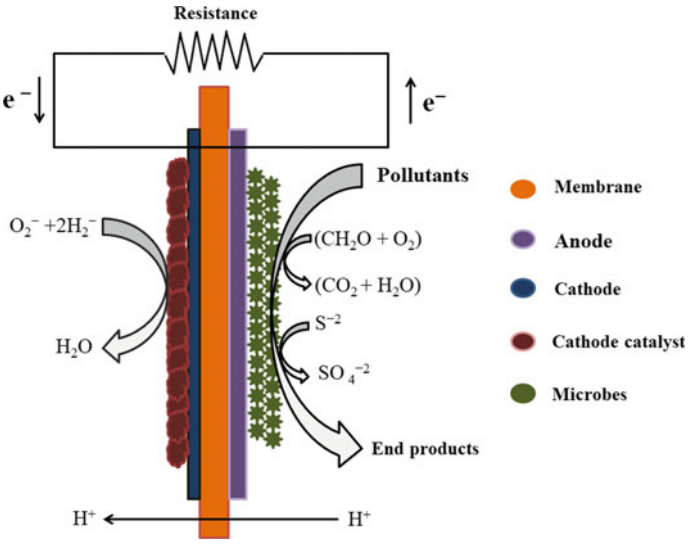

Fig. 1 Illustrative bioremediation mechanics of sediments in MFC. Adapted from Yuan et al. [113]

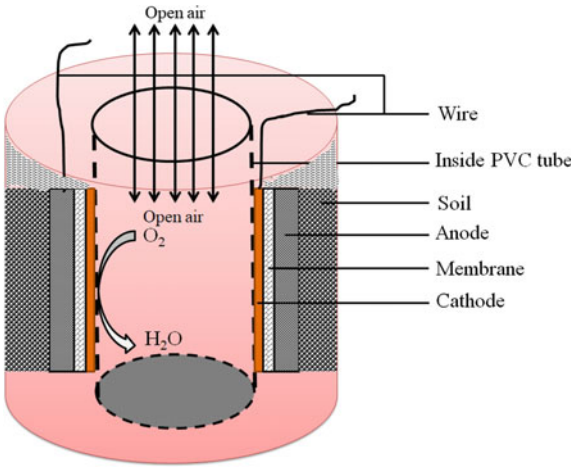

Fig. 2 Schematic representation of Tubular air-cathode MFC (TAC–MFC) Adapted from Huang et al. [25]

soil MFC (Fig. 3 and found that the horizontally arranged anodes (50.6% showed superior performance than the vertical counterparts (8.3%. Charge output was also higher in the horizontal anode configuration (833 C compared with the vertical mode (762 C. The PAH degradation rate increased from 2 to 24% in 66 d; however, low power output was observed in the in-situ MFCs with vertical anodes (an average of 37 mWm^{-3}). In most cases, the internal resistance of the in-situ MFC was high was

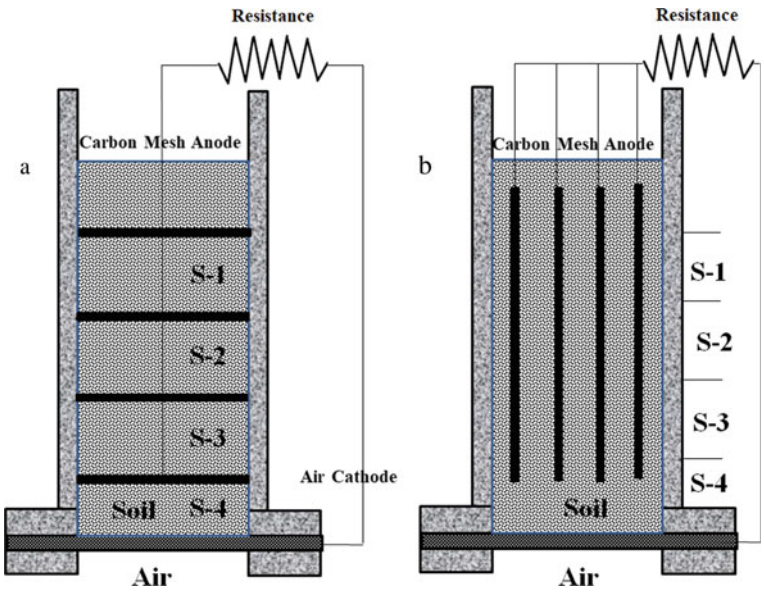

Fig. 3 Figurative representation of soil MFC (A) vertically arranged anodes and (B) horizontally arranged anodes. Adapted from Zhang et al. [117]

mainly due to the high ohmic resistance i.e., low soil conductivity and mass transfer limitations [67]. To overcome these issues, the amendment of PAH-contaminated soil with different additives, such as sand, carbon fibers, and biochar, was adapted to enhance the bioelectrochemical remediation of PAHs. Amendment with sand increased the soil porosity from 44.5 to 51.3%, decreased Ohmic resistance by 46%, and increased the charge output from 2.5 to 3.5 C g^{-1} [42]. The porosity of the soil plays a significant role in increasing both dissolved oxygen content and proton transport, thereby decreasing charge transfer resistance. Moreover, the internal resistance of the soil MFC decreased by 58% after mixing 1% carbon fiber with the soil samples, leading to a 15-fold increment in electricity production and 329% higher removal of petroleum hydrocarbons [44, 45]. Similarly, the addition of biochar with high N content accelerated the selective enhancement of the bacterial community and exhibited the best efficiency in the removal of recalcitrant contaminants, such as aromatics [47]. A plant microbial fuel cell (PMFC) is a promising modified MFC that fosters plant-microbe relationship at the rhizosphere region of a plant, which consequently increases the biofilm adhesion on the anode, thereby improving electron transfer from the biofilm to the anodes in the PMFC. The synergistic action of plants and surfactants significantly enhances the efficiency of the MFC system in the removal of petroleum from the soil and encourages the applications of P-MFCs for the in-situ remediation of petroleum-contaminated soils. Zhao et al. [118] used plant MFC to degrade phenanthrene and pyrene in contaminated soils. The addition of surfactant β-cyclodextrin enabled the desorption of PAHs from the soil to the plant

at a higher rate than Tween 80 and decreased the charge transfer resistance by 63.4%. This suggests that the amendment of contaminated soil with some additives, which possess the characteristics of conductivity and micronutrient, enhances the removal of petroleum from the soil in MFC systems.

2.1.2 Bioremediation of Petroleum Wastewater in MFCs

Petroleum hydrocarbons are very common groundwater contaminants that usually deplete oxygen when they undergo biodegradation. The anaerobic degradation of petroleum compounds has been identified to involve a wide spectrum of microbial pathways, including denitrification [4, 51], metal reduction [52], and sulfate reduction [84]. The bioremediation of petroleum waste, including wastewater from petroleum refineries, petroleum sludge, and oil-contaminated soil, has been studied in MFCs (Table 2). The downstream petroleum refinement processes generate large volumes of wastewater containing hydrocarbons, such as phenols, benzene, and xylenes, as well as inorganic compounds, such as ammonia, nitrite, sulfides, and heavy metals. Wastewater from petroleum refineries can range from 0.4 to 1.6 times the volume produced during crude oil processing [9] The low BOD/COD ratio (<0.2) and high total dissolved solids (TDS) render the petroleum refinery wastewater (PRW) unsuitable for biological treatment.

In 2008, Morris and Jin reported, for the first time, MFC technology for the degradation of hydrocarbon contaminants in groundwater under anaerobic conditions. Using refinery wastewater as the sole substrate, they achieved power generation (as high as 120 mW/m^2 at the cathode) in approximately ~6 d with a single cell MFC. It is known that the larger the electrode spacing, the potential losses in the cell are higher. The authors reported that the cell potential decreased by ~55% over a 9 m-long proton bridge, with a 6.9% decrease in potential per meter of the bridge [66]. To enhance the treatment of PRW in a single-chamber air-cathode microbial fuel cell (MFC), external voltage from 100 to 500 mV was applied to the electrochemically active anodic biofilm [60–62]. The MFC operated with 500 mV supplemental voltage exhibited a maximum power density of 132 mW/m^2, which was three times higher than control MFC (45 mW/m^2). Similarly, the highest substrate removal efficiency (48%) was obtained with the MFC supplemented by 500 mV, followed by those supplied with 300 mV (37%), 100 mV (32%), and the 0 V control (27%). Therefore, the applied potential enhanced the diesel-range organics (DROs)/straight chain-alkane degradation efficiency. [88] proposed the coupling of a hydraulically connected osmotic microbial fuel cell (OsMFC) and an up-flow microbial desalination cell (UMDC) for enhanced PAH removal, along with high power output in the MFC and simultaneous seawater desalination in the UMDC (Fig. 4). Both OsMFC and UMDC were connected to 100 Ω external resistance, and the system achieved 93% chemical oxygen demand (COD) removal from the petroleum refinery wastewater besides 48% salt removal from the seawater. Sarmin et al. [87] treated the petroleum chemical wastewater from an acrylic acid plant using an MFC, which generated 850 mW/m^2 maximum power density at 1500 mA/m^2 current density and

Table 2 In situ MFC for bioremediation of PAHs-contaminated soil

S.No	Petroleum products	Anode	Cathode	Reactor configuration	Power density	Treatment efficiency	References
1.	Organic rich sediment	Carbon felt	Canvas cloth	Tubular air-cathode MFC (TAC-MFC)	107.1 ± 8.6 (mW/m^2)	LOI: 33.1% ROOM: 36% AVS: 94.9%	Yuan et al. [113]
2.	Organic pollutants	Carbon felt	Conductive paint + catalyst	Soil MFC	29.45 (mW/m^2)	SCOD (mg/L): 430 TCOD (mg/L): 35,500	Huang et al. [26]
3.	Petrochemical sludge	Graphite plates	Open-air	Single chamber MFC	53.11 (mW/m^2)	41.08% of TPH	Mohan and Chandrasekhar [59]
4.	Petroleum sludge	Non-catalyzed graphite plates	Air	Single chambered BET system	20.62 (mW/m^2); 198.31 (mA/m^2)	58 (aromatics at 30 g TPH/L) 70 (aromatics at 3 g TPH/L)	Chandrasekhar and Venkata Mohan [7]
5.	Petroleum contaminated sediments	Carbon cloth	Carbon paper/platinum (Pt)	Sediment MFC	2162 (mW/m^3)	16,000 mg/kg sediment	Morris and Jin [67]
6.	Phenanthrene and pyrene in freshwater sediments	Stainless steel	Stainless steel	Sediment MFC	–	*Removal*: Phenanthrene: 99.47 ± 0.15% Pyrene: 94.79 ± 0.63%	Yan et al. [109]
7.	Petroleum Hydrocarbon contaminated saline soil	Carbon mesh	Air	U-Tube MFC	0.85 ± 0.05 (mW/m^2)	TPH: 0.3–2.5%	Wang et al. [105]

(continued)

Table 2 (continued)

S.No	Petroleum products	Anode	Cathode	Reactor configuration	Power density	Treatment efficiency	References
8.	Petroleum hydrocarbon soil	Carbon mesh	Air	Soil MFC	37 (mW/m^2)	TPH: 120%	Li et al. [41]
9.	Hydrocarbon-contaminated soil	Carbon cloth	Air	Soil Tubular BES	39.1 (mW/m^2)	TPH removal: 78.7%	Lu et al. [53]
10.	Polycyclic aromatic hydrocarbons (PAHs)	Plain graphite plate	Graphite felt	Sediment MFC	0.4–0.6 V	22.1% (TOC removal in closed circuit)	Yang et al. [111]
11.	Benzo(*a*)pyrene, pyrene in sediments	Graphite felt	Graphite felt	Sediment MFC	64.8 mV	Degradation rate: 70%	Yan et al. [110]
12.	Petroleum hydrocarbon-contaminated soil	Carbon mesh	Air	Air-cathode soil MFC	–	TPH degradation: 15.3 ± 0.2%	Zhang et al. [117]
13.	Polycyclic aromatic hydrocarbons (PAHs) in sediments	Graphite plate	Graphite plate	Sediment MFC	Anaerobic: 2 (mW/m^2) and Aerobic: 5.8 (mW/m^2)	(Anaerobic environment) removal: 76.9% of Naphthalene, 52.5% of acenaphtene and 36.8% of phenanthrene TOC: 67%	Sherafatmand and Ng [91]
14.	Petroleum hydrocarbon-contaminated soil	Carbon mesh	Air	Soil MFC	–	Degradation of petroleum hydrocarbons(PH) upto 268%	Li et al. [42]

(continued)

Table 2 (continued)

S.No	Petroleum products	Anode	Cathode	Reactor configuration	Power density	Treatment efficiency	References
15.	Petroleum Hydrocarbons in soil	Carbon mesh	Activated carbon air-cathode	Soil MFC	43 (mW/m^2)	–	Li et al. [44, 45]
16.	Petroleum hydrocarbon-contaminated soil	Carbon mesh	Air	Soil MFC	304 (mA/m^2)	Degradation rate: 484%	Li et al. [44, 45]
17.	Petroleum hydrocarbon-contaminated soil	Graphite rod	Carbon air-cathode	Soil MFC	0.8 (mW/m^2)	TPH: 100%	Li et al. [44, 45]
18.	Petroleum hydrocarbon-contaminated soil	Carbon cloth	Carbon cloth	Dual chamber MFC	220 (mW/m^2)	–	Zhou et al. [120]
19.	Petroleum hydrocarbon-contaminated aged soil	Carbon mesh	Air	Soil MFC	153.8 ± 0.1 mA m^{-2}	bioelectricity generation rate: 0.321 C d^{-1} g^{-1}	Li et al. [46]
20.	Petroleum hydrocarbons in soil	Carbon mesh	Air	Soil MFC	–	Total charge generation: 79–86%	Li et al. [47]
21.	Petroleum hydrocarbon-contaminated soil	Graphite brush	Platinum coated carbon cloth	Soil-based MFC	Electrochemically active bacteria	Total dissolved solids (12.08% removal) and sulfates (62.64% removal)	Mohanakrishna et al. [64, 65]
22.	Polyaromatic hydrocarbon (PAH)	Graphite felt	Graphite felt	Single chamber plant-microbial fuel cell	117.3 mV	Removal: 48.04%	Zhao et al. [118]

(continued)

Table 2 (continued)

S.No	Petroleum products	Anode	Cathode	Reactor configuration	Power density	Treatment efficiency	References
23.	Petroleum hydrocarbon-contaminated soil	Plain graphite based plates	Plain graphite based plates	Soil based MFC	Acetate: 1126 (mW/m^2) Sewage: 1145 (mW/m^2)	*TPH degradation*: Acetate: 525 mg/L, 67.4% Sewage: 560 mg/L, 71.8% COD reduction Acetate: 72.1% Sewage: 74.6% *Sulfates removal* Acetate: 62.9% Sewage: 72.6%	Mohanakrishna et al. [63]
24.	Hydrocarbon-contaminated soil	Phenanthrene-coated bioanode	Pt coated carbon cloth	Dual chamber MFC	0.019 (mW/m^2)	Degradation of phenanthrene: 2 mg/cm^2 Power production: 37 (mW/m^2)	Sharma et al. [90]

TPH: Total petroleum hydrocarbon; COD: Chemical oxygen demand; SCOD: Soluble Chemical oxygen demand; TCOD: Total Chemical oxygen demand; LOI: Loss on ignition; ROOM: Readily oxidizable organic matter; AVS: Acid volatile sulfide

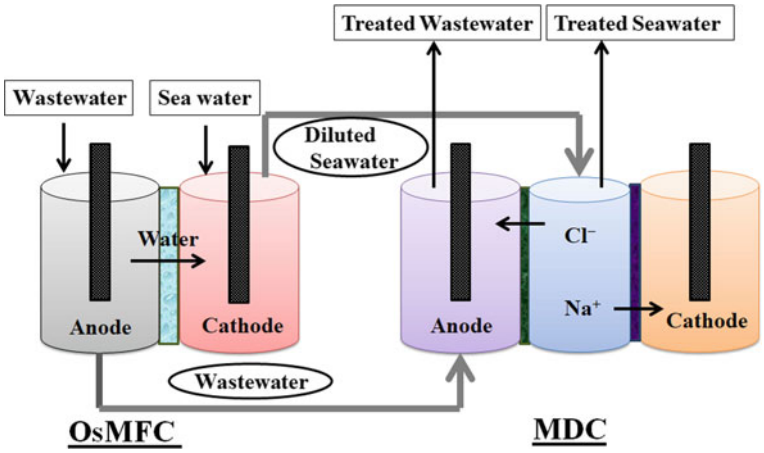

Fig. 4 Schematic representation of the dual system of hydraulically connected Osmotic MFC and MDC. Adapted from [88]

45,000 mg L^{-1} COD. Produced water (PW) refers to the large quantity of wastewater generated during oil and gas extraction. PW is usually characterized by high amounts of dissolved solids (TDS) and residual petroleum hydrocarbons, which are considerably hazardous to the environment. The level of environmental damage is dependent on the geographical location, age of the well, nature of geological formations at the specific location, the type of hydrocarbon products found the well and the stage of oil extraction. The bioelectrochemical treatment of PW was studied by Mohanakrishna et al. [64, 65] in two configurations viz., single- and dual-chamber microbial fuel cells (MFCs), of which the latter configuration displayed superior function with respect to power generation (1089 mW/m^2) and substrate degradation (COD removal efficiency = 60.2%) compared with the single-chamber configuration (PD 789 mW/m^2; COD removal efficiency = 54.7%); However, since the nature of PW varies based on the location and oil extraction stage, intensive studies are required to develop suitable processes with reference to the operating conditions; further, understanding the microbiology in connection with the composition of PW is also essential.

2.1.3 Microorganisms for PAH Treatment in Fuel Cells

A summary of the different microbial species employed in MFC operation for specific PAH substrates and their possible performance parameters are given in Table 3.

The hydrocarbon-degrading bacterial strain *Alcanivorax* has been shown to simultaneously improve bioelectricity generation and bioremediation performance in soil MFCs [42]. Li et al. [44, 45] suggested that employing the *Alcanivorax* strain is an

Table 3 Microbes involved in bioremediation of PAHs in MFC

S.No	Petroleum products	Reactor configuration	Biocatalyst	Power density	Treatment efficiency	References
1.	Refinery waste water	Dual Chambered MFC with aerated cathode	*Paenibacillus sp and Deinococcus sp*	102.93 mW/m^3	Oil removal rate: 85.56 ± 1.10%	Guo et al. [21]
2.	Oil refinery waste water	Single chamber with air-cathode	*Pseudomonas Putida*	0.005 mW/m^3	COD 30%	Majumder et al. [55]
3.	Hydrocarbon-contaminated soil	Soil tubular BES	*Comamonas testosteroni*, *Pseudomonas putida*, and *ochrobactrum anthropic*	39.1 (mW/m^2)	TPH removal: 78.7%	Lu et al. [53]
4.	Hydrocarbon-contaminated groundwater	Dual-chambered MFC	*Shewanella oneidensis* MR1 14,063, *Pseudomonas aeruginosa* NCTC 10,662 and mixed cultures	1.25 (mW/m^2)	COD: 65.6%	Adelaja et al. [2]
5.	Petroleum hydrocarbon soil	Soil MFC	Exoelectrogenes (*Geobacteraceae* sp., *Escherichia* sp.)	37 (mW/m^2)	TPH: 120%	Li et al. [41]
6.	Petroleum hydrocarbon-contaminated soil	Soil MFC	*Alcanivorax*	–	Degradation of petroleum hydrocarbons (PH) upto 268%	Li et al. [42]
7.	Petroleum Hydrocarbons	Soil MFC	*Alcanivorax*	43 (mW/m^2)	–	Li et al. [44, 45]
8.	Petroleum hydrocarbon-contaminated soil	Dual chamber MFC	*Geobacter* sp. and *Ochrobactrum* sp	220 (mW/m^2)	–	Zhou et al. [120]

(continued)

Table 3 (continued)

S.No	Petroleum products	Reactor configuration	Biocatalyst	Power density	Treatment efficiency	References
9.	Petroleum hydrocarbon-contaminated soil	Soil MFC	δ-Proteobacteria (Proteobacteria), Flavobacteria (*Bacteroidetes*) or *Clostridia* (*Firmicutes*)	304 (mA/m^2)	Degradation rate: 484%	Li et al. [44, 45]
10.	2,4-dichlorophenol	Dual chamber MFC	Mixed bacterial consortia (*Arcobacter*, *Cloacibacterium* and *Bacillus* sp.)	37 (mW/m^2)	COD: 41%	Hassan et al. [22]
11.	Petroleum hydrocarbon-contaminated aged soil	Soil MFC	*Proteobacteria*, *Firmicutes*, *Bacteroidetes*, *Actinobacteria*, *Chloroflexi*, *Planctomycetes* and *Acidobacteria*	153.8 ± 0.1 mA m^{-2}	bioelectricity generation rate: 0.321 C d^{-1} g^{-1}	Li et al. [46]
12.	Petrochemical wastewater	Dual chamber MFC	*Pseudomonas* spp. and *Bacillus* spp. Along with methanogenic archea *Methanobacterium* spp	850 (mW/m^2)	COD: 40%	Sarmin et al. [87]
13.	Hydrocarbon-contaminated soil	Dual chamber MFC	*Pseudomonas*, *Rhodococcus*, *Thauera* and *Ralstonia*	0.019 (mW/m^2)	Degradation of phenanthrene: 2 mg/cm^2 Power production: 37 (mW/m^2)	Sarmin et al. [90]

effective approach to eliminate soil contamination in barren areas/extreme environments. Hassan et al. [22] found that various exoelectrogens, including *Arcobacter*, *Aeromonas*, *Pseudomonas*, *Acinetobacter*, *Cloacibacterium*, and *Shewanella* sp., were involved in the degradation of 2, 4-DCP (dichlorophenol). This study discovered that *Cloacibacterium* sp. can degrade phenol in MFCs. Sarmin et al. [87] utilized pre-acclimatized mixed culture inoculum composed of electrogenic genera, such as *Bacillus* sp., *Pseudomonas* sp., and *Methanobacterium* sp. (methanogenic archaea), as biocatalysts in treating petrochemical wastewater (PCW) from an acrylic acid plant. This suggests that MFCs could be potential alternatives to conventional aerobic and anerobic processes, which are high energy-intensive.

2.1.4 Influence of Co-substrates and pH in MFCs for PAHs Degradation

The cultivation medium used for microbial consortia in MFC systems generally contains co-substrates to drive microbial growth during operation. Most studies have reported the use of co-substrates along with petroleum refinery wastewater in the anode chamber [22, 24, 44, 45, 66, 114, 116, 118]. In addition [60–62], used acetate as a co-substrate and it can be inferred that the substrate can stabilize microbial activity in an anode and continue electricity output at a steady current density level. pH is a crucial factor for anaerobic processes and serves as an intrinsic index of the changes in biological systems. Srikanth et al. explained that in batch mode operation, a change in pH toward the acidic range (pH < 4.0) [94] can be attributed to the competing action of mixed consortia in the redox reactions. Mohanakrishna et al. [60–62] reported that a single-chamber air-cathode MFC operated with PRW as the anolyte showed negligible change in the pH of the effluent. However [113], explained that the pH of all sediments slightly decreased, probably due to the fermentation of organics. Mohan and Chandrasekhar [59] Mohan & Chandrasekhar et al., demonstrated that the system pH was strongly controlled by the microbial metabolic rate, bioavailability, and mass transfer. In soil MFCs, the depletion of existing organic acids and the formation of bicarbonate salts leads to an increase in soil pH [26]. Li et al. [42] showed that pH values of the soil effectively correlated with the addition of sand as vast pH shifts were observed with increasing sand quantities. Li et al. [44, 45] elucidated that the addition of glucose substrate can neutralize soil pH and improve conductivity, and then the glucose is partly metabolized to organic acids (such as acetate). Soil pH and conductivity were the prominent factors that controlled the polyphenol oxidase and dehydrogenase activities, thus increasing TPH degradation and power output.

3 MFC for Dyes Degradation

Dyes containing one or more azo bonds are widely used in textile, leather, plastics, cosmetics, and food industries [74]. The color of these dyes is due to the azo bond and associated chromophores; so, the disposal of these dyes into the surface water not only affects the aesthetics, but their breakdown products have often been found to be carcinogenic, mutagenic, or toxic for humans [18]. These dyes can also obstruct the light and oxygen penetration into water, thus affecting the aquatic life. Several methods for the treatment of dye-containing wastewater which broadly fall into three categories: physical (adsorption, coagulation/flocculation, membrane filtration etc.), chemical (chemical oxidation, photo-catalytic oxidation, electrolysis, Fenton reagent etc.), and biological (biosorption, enzymatic degradation etc.) [34]. These physicochemical methods can be quite effective for color removal, but they have inherent disadvantages, including high operational costs and the generation of huge quantities of sludge for disposal. Recently, attempts have been made to address this issue through the biological route, using specific microorganisms for the cleaving of the azo bond. The biological treatment for the decolorization and degradation of azo dyes may be either aerobic, anaerobic, or a combination of both, depending on the type of microorganisms being employed [5, 35]. Most of the azo dyes are decolorized under anaerobic conditions however the anaerobic reduction is generally very slow, and produce methane rich gas. In most cases, the aromatic amines derived from azo dyes are not degraded under anaerobic conditions due to its high redox potential. These product amines released from azo dye decomposition are known, or suspected, to be more carcinogenic for humans [81] than the parent compound azo dyes. Thus, MFC has been employed for performing the dual duty of degrading the textile dyes and generating power. Moreover, such a system can reduce solids generation by 50–90% and reduce the load on their disposal. Based on their structure, dyes are classified into: acid/basic dyes, direct dyes, azo dyes, sulfur dyes, fiber reactive mordant dyes, etc., [38]. Dye molecule consists of two parts: (1) chromophores, which are functional groups of dyes, and (2) auxochromes, which enhance the color. Azo dyes account for more than 70% in the textile effluent. Most of the studies have focused the dye decolorization in the anode of MFC and few reports on dye decolorization in the cathode [27].

3.1 Azo Dye Decolorization and Degradation in the Anode

The co metabolism reaction is probably the main mechanism of dye degradation at the anode in which the reducing equivalents (electrons) are formed during the anaerobic oxidation of co-substrate. Once the substrate was oxidized and transferring the some portion of electrons for the exoelectrogens accumulated on the anode that pass through an external circuit producing current. Simultaneously, the other part of the electrons transferred to split the −N=N− bond in the azo dyes to form

aromatic amines in the anode chamber. Hence, there will be a competition for electrons between dye substances and an anode in an MFC. Different dyes were tested as anolyte with different substrates in MFC as shown in Table 4.

Electricity generation from glucose as co-substrate accompanied by decolorization of azo dye active brilliant red X-3B (ABRX3)was investigated by Sun et al. [97] using a microfiltration membrane air-cathode single-chamber microbial fuel cell (MFC). Around 90% of the dye was removed within 12 h at an dye concentration of 100 mg/l. The dye degradation mechanism via MFC technology as shown in Fig. 5. Anaerobic-aerobic sequential MFC reactor couple system was developed for Congo red degradation [48].

It was observed that the amines are formed in the anode chamber, and they are resistant to further degradation in the MFC under an anaerobic environment. To overcome this, a novel aerobic biocathode was designed by Sun et al. [95], since these intermediates are transferred to the cathodic chamber and further biodegradation takes place under aerobic conditions. Degraded intermediates transferred to the biocathode resulted in an almost 150% increase in open cycle potential (OCP) of the cathode accompanied by a 73% increase in stable voltage output from 0.33 to 0.57 V and a 300% increase in maximum power density from 50.74 to 213.93 mW/m^2. Azo dyes removal in CW has been investigated by Noonpui and Thiravetyan [73] and they demonstrated that a maximum 97% of dye removal in the constructed wetland (MFCCW); however, the removal efficiency was found to be a function of structure and size of the dye molecule. Moreover, they emphasized that the plant, soil, and microorganisms might all influence the efficiency of dye removal in a CW.

Double chambered MFCs are the most common design in MFCs studies which separated by a PEM. The protons transfer through PEM can be a limiting factor due to the suspended solids and soluble contaminants [28]. Moreover, double chambered MFC is difficult to scale-up. Single chambered up-flow membrane-less microbial fuel cell (UFML MFC) was developed by Thung et al. [100] for decolorization of Acid Orange 7 (AO7) and electricity generation simultaneously. Azo bond and naphthalene moieties were completely degraded but aromatic amines were remained in the effluent and however the COD and color removal by the bioreactor was up to 90%. The azo dyes (methyl orange (MO), Congo red (CR), reactive yellow (RY), reactive red (RR), along with nutrient medium were tested as anolytes in the *Pseudomonas-catalyzed* MFC at an external resistance of 220 Ω [29]. In the anode chamber maintained under anaerobic conditions more than 90% of MO and RB could be decolorized by *P. aeruginosa* in 5 d, but RR was decolorized only about 74%. Generally, the microorganisms produce azoreductases which reductively cleave the highly electrophilic azo bond at the expense of a reducing agent, typically NAD(P)H [54]. It is expected that reduction under anaerobic environments will be nonspecific, although the rate of decolorization depends on the organic source and the dye structure (Engineering 2012). It suggested that a simple structure and low-molecular weight azo dye MO degraded faster in MFC than CR, RR, and RY which are highly substituted and of high molecular weight. On the other hand, it was also observed that bacterial decolorization of di azo RB was more superficial when compared with either of the mono azo dyes (RR, RY). The presence of two electron withdrawing group (SO_3^{-1}) at

Table 4 Different dyes for anolyte with different substrates in MFC

S.No	Dye	Anode	Cathode	Co-substrate	Bio-catalyst	Power density	Decolourization efficiency	References
1.	Active brilliant red X-3B	Porous carbon papers	Porous carbon papers with Pt and PTFE	Glucose	Anaerobic sludge	234 mW/m^2	90%	Sun et al. [97]
2.	Methylene blue	Bottom of microcam	Rhizopheric zone-wetland	Sucrose	Wastewater	15.7mW/m^2	80%	Yadav et al. [108]
3.	Reactive Brilliant red X-3B	Granular activate carbon	SS mesh	Glucose	Anaerobic sludge	302mW/m^3	91.24%	Fang et al. [13]
4.	Reactive Brilliant red X-3B	Activated carbon/SS mesh	Activated carbon/SS Mesh	Glucose	Wastewater	852mW/m^3	94.9%	Fang et al. [14]
5.	Methyl orange	Granular activate carbon	Granular activated carbon	Glucose	Synthetic wastewater	688 mW/m^3	75–85%	Fang et al. [15]
6.	Congo red	Porous carbon papers	Porous carbon papers	Glucose	*Pseudomonas sp.* and *Aquamicrobium sp*	29 mW/m^2	20.3% increase	Sun et al. [96]
7.	Congo red	Graphite felt	Graphite felt	Glucose	Anaerobic sludge	0.193 ± 0.0023	70%	Huang et al. [26]
8.	Mordant orange	Toray carbon paper(TCP)	Toray carbon paper(TCP)	Acetate	Bacterial culture	4.9 ± 0.5 Wm^{-3}	89%	Sarma et al. [86]
9.	Acid red(18)	Carbon felt	Carbon felt	Acetate	Mixed consortium	8.67mW/m^2	91%	Oon et al. [75]

(continued)

Table 4 (continued)

S.No	Dye	Anode	Cathode	Co-substrate	Bio-catalyst	Power density	Decolourization efficiency	References
10.	Brilliant green	SS mesh-GO	SS mesh-GO	Glucose	*Geobactor sulfureducens*	0.04 mW/cm^2	80%	Khalid et al. [36]
11.	Methyl orange	Carbon felt	Carbon felt	Acetate	*Proteobacteria, Actinobacteria, Bacteroidetes, Planctomycetes, Firmicutes*	628 mW/m^2	92.5%	Zhong et al. [119]
12.	Acid red (18) Acid orange(7) Congo red	Carbon felt	Carbon felt	Acetate	Mixed consortium	1.58 mW/m^2 1.13 mW/m^2 1.02 mW/m^2	96% 67% 60%	Oon et al. [77]
13.	Methyl red	Carbon cloth	Platinum rod	Glucose	Bacterial consortium	–	71.7%	Krithika et al. [37]
14.	Sunset yellow	Graphite rod	Graphite rod	Acetate	Mixed consortium	46.42 ± 0.01	93%	Tacas et al. [98]

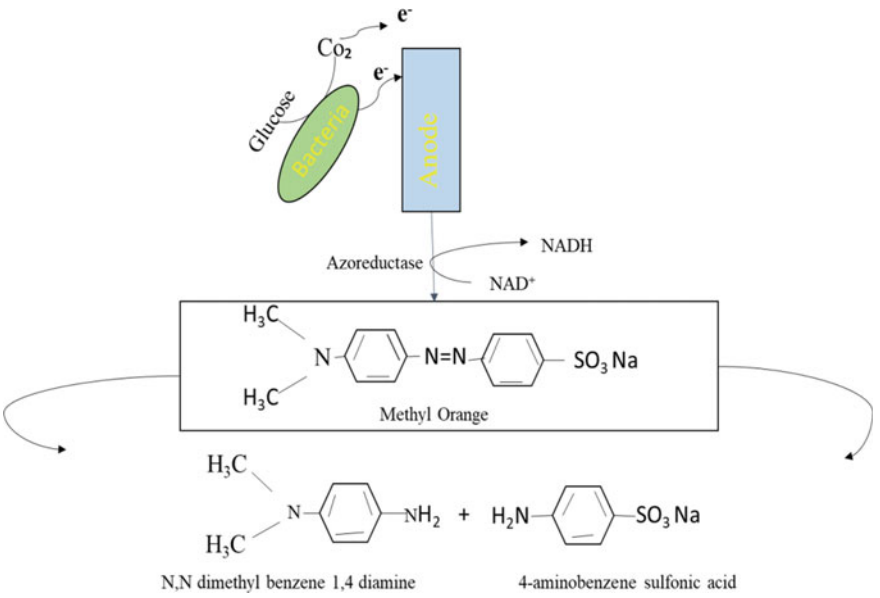

Fig. 5 Degradation mechanism of dye

ortho position to azo bonds and OH group of the naphthol ring in RB were also the main aspect of the azo bond cleavage. The sulfo groups could withdraw electrons from azo bond by resonance, and the environment in the proximity of azo bond could become more electrophilic for decolorization [115].

Reduction of azo dye in the anode chamber might be performed either by direct enzymatic reaction or mediated reaction of enzyme cofactors, which are biologically regenerated by sulfate-reduction bacteria (SRB) to break down the azo bond [85]. Santos et al. [11] used single-chamber air-cathode microbial fuel cells to investigate the interaction mechanisms among Congo red decolorization, sulfide oxidation, and bioelectricity generation. The results showed that effective removal of sulfide (98%) and azo dyes (88%) was achieved at an initial sulfide/dye ratio of 0.9 under neutral conditions, accompanied by a maximum power output of approximately 23.50 mWm^{-2}.

3.2 Azo Dye Decolorization and Degradation in the Cathode

The color of dyes is due to azo bond and associated chromophores, so disposal of dyes into surface water not only affects the aesthetic but cause also biotoxicity. The azo dye was used in the cathode with the dual objective of solving as facilitator for electron acceptor and the possibility of decolorization. The cathode chamber was continuously sparged with N_2 gas to avoid O_2 competition with the azo dyes for

electrons. The reduction reactions in the cathode chamber are described by Goyal and Minocha [20], Menek, Zeyrekl, and Karaman n.d.; Parthasarathy and Narayanan [78] in which the –N=N– double bond was reduced to hydrazo (1) or amine (2), via the consumption of two or four electrons.

$$-\mathrm{N{=}N}- + 2\mathrm{e} + 2\mathrm{H}^{+} \rightarrow -\mathrm{NH} - \mathrm{NH}- \quad (1)$$

$$-\mathrm{N{=}N}- + 4\mathrm{e} + 4\mathrm{H}^{+} \rightarrow -\mathrm{NH_2} - \mathrm{NH_2}- \quad (2)$$

Mu et al. [68] achieved an effective decolorization rate of dye was 2.64 ± 0.03 mol m^{-3} d^{-1} achieved at the cathode of a bioelectrochemical system while acetate was used in the anode [68]. Reduction of azo dyes in the cathode of MFC using electrons produced from metabolic oxidation of *Klebsiella pneumoniae* strain L17 in the anode [50]. Different azo dyes congo red, methyl orange and reactive blue, were tested as catholyte in *Pseudomonas-catalyzed* MFC using a graphite block [29]. Compared to potassium ferricyanide (72.8 ± 17.8 μW/m^2) and potassium permanganate (86.28 ± 16.8), addition of azo dyes enhanced the power output by several fold (congo red-91.73 ± 21.2; Methyl orange 314.07 ± 22.6; Reactive blue 172 294.43 ± 11.9 μW/m^2). Lot of research findings (Table 5), the degradation efficiency was high; however it was found that low power density resulting from the small specific capacitance of the carbon-based materials.

3.3 *Key Parameters Influencing the MFC for Dyes Degradation*

Developing an MFC for dye waste water treatment with a simple configuration at affordable costs for the ease of scale-up still remains the challenge. Many workers have reported that the several factors that affect the MFC performance including the microbes, the proton selective membrane, the material of the electrodes, the cathode/anode catalyst, the electrode spacing, and the system design [31]. It is known that electron transfer between the electron carriers in the bacteria transport chain and the anode/cathode is low, leading to a high internal resistance that reduces the power generated in MFCs.

3.3.1 Effect of Co-substrate

One of the most crucial aspects of MFC is the substrates used due to their effect on the dye degradation and power output. A report by [97], when confectionary and glucose wastewater were utilized as co-substrates in MFC along with a model azo dye, there

Table 5 Different dyes for catholyte with a different substrate in MFC

S.No	Dye	Anode	Cathode	Substrate	Bio-catalyst	Power density	Decolourization efficiency	References
1.	Methyl orange	Carbon felt	Carbon felt	Glucose	*K. pneumoniae strain L17*	34.77 mW/m^{-2}	90%	Liu et al. (2009)
2.	Congo red	Rough plain carbon felt	Carbon felt	Glucose	Anaerobic sludge	364.5 mW/m^{-2}	80%	Li et al. [48]
3.	Reactive blue 221	Graphite bars	Graphite bars	Urea	Anaerobic sludge	27 mW/m^{-2}	83%	Bakhshian et al. (2011)
4.	Active brilliant red X-3B	Porous carbon papers	Porous carbon papers	Glucose	Domestic wastewater	50.74 mW/m^2 to 213.93 mW/m^2	90%	Sun et al. [95]
5.	Acid Orange 7	Carbon plate	Carbon felt	Sodium acetate	Anaerobic sludge	7.07 mW/m^{-2}	96%	Thung et al. [100]
6.	Reactive Brilliant red X-3B	Granular activated Carbon	Granular activated Carbon	Glucose	Wastewater	117mW/m^3	92%	Fang et al. [16]
7.	Acid orange 7	Carbon felt	Carbon cloth	Solidified gel	Ganoderma lucidum BCRC 36,123	13.38 mW/m^2	90%	Lai et al. [40
8.	Orange II	Carbon cloth	Carbon paper	Sodium acetate	Wastewater	72.6 mW m^{-2}	96.3%	Xu et al. [106]
9.	New coccine, Acid orange 7, Reactive red 120 and Reactive green 19	Carbon felt	Carbon felt	Sodium acetate	Wastewater	20.64 mW/m^2	95.1 ± 1.1%	Oon et al. [76]

(continued)

Table 5 (continued)

S.No	Dye	Anode	Cathode	Substrate	Bio-catalyst	Power density	Decolourization efficiency	References
10.	Methyle Orange	Graphite	Graphite	Glucose	*P.aeruginosa*	95.83 ± 4.10 $\mu W/m^2$	87 ± 2.8	Narayanasamy and Jayaprakash [69]
11.	Methyl orange	Carbon felt	Carbon felt	Glucose	Anaerobic sludge	502.5 ± 17.1 mW/m^2	97.1 ± 1.8%	Liang et al. [49]
12.	Orange II	Carbon cloth	Carbon paper	Acetate	Anaerobic sludge	206.2 ± 3.1 mW m^2	80%	Xu et al. [107]
13.	Congo red	Graphite fiber brush	Carbon cloth	Acetate	Anaerobic sludge	23.50 mW/m^2	98%	Dai et al. [11]

is a competition between the anode and azo dye reduction for electrons from co-substrate oxidation. Bioelectricity generation with a simultaneous Congo red degradation can be achieved using glucose, acetate sodium, and ethanol as co-substrate in PEM air-cathode single-chamber MFC. More than 98% of the Congo red (300 mg/L) was decolorized within 36 h for all tested co-substrates during the electricity generation. Glucose produced the highest power densities when compared to Ethanol [6, 57] studied that simultaneous electricity generation and tetra-azo dye (Direct Red 80) decolorization was examined in a dual-chamber MFC. Glucose was identified as a better co-substrate than acetic, propionic, and lactic acid for dye removal and current production (477.8 mW/m^2). It can be understood that the low-molecular co-substrates can be easily degraded, excessive co-substrate addition can lead to high COD, in addition to resource (dye) waste. Anaerobic azo dyes degradation in MFC are strongly influenced by co-substrate type and its concentration suggested that the combination of dye containing waste water and high organic content waste water from food/biorefinery industries that is easily biodegraded simultaneously that can both improve cost and energy.

3.3.2 Effect of PH

Anodic pH microenvironment is one of the important factors which can influence substrate metabolic activity and as well as proton transfer mechanism [80]. pH has a significant factor on dye degradation efficiency because of enzymatic activity depends on the pH. The color of the solution and solubility of the dyes is affected by the pH. The release of metabolic products by a biocatalyst into the anolyte will lower the system pH [31] acidic can bring about alterations in several MFC system parameters such as concentration of ions, membrane potential, proton motive force and adversely affect bacterial growth and adhesion during MFC operation. Raghavulu et al. [80] found that acidophilic pH in anodic chamber was effective performance with respect to power output compared to the corresponding neutral and alkaline operations. However, substrate degradation was observed to be higher in neutral condition followed by alkaline and acidophilic operations. On the other side, though cation exchange membranes (CEM) and anion exchange membranes (AEM) are less expensive separators [72], pH in the cathode chamber increases because of the transport of the cationic species other than protons (particularly of alkali and alkaline earth metals) through the CEM. This increase in pH negatively impacts the performance of the MFC. For every 3 unit increase in pH, a loss of potential of 0.18 V occurs in the cathode, according to [83]. Ideally, the only membranes that can prevent the pH effect on MFC performance are 100% proton selective membrane. Buffers have been commonly used in MFCs in order to (i) maintain a suitable pH for the anodic bacteria biofilm, (ii) increase solution conductivity, (iii) increase the transfer of protons generated at the anode to the cathode, and (iv) reduce the operational pH, which would be beneficial for oxygen reduction. Hence, the primary challenge in scaling-up an MFC application is to ensure a more stable electrolyte environment in the MFC by adding buffers in continuous mode.

3.3.3 Effect of Hydraulic Retention Time (HRT)

HRT is the average amount of time that reactants stay in a reactor and it is calculated by dividing the volume of a reactor by the influent flow rate. Oon et al. [75] studied the effect of HRT on MFC performance and reported that longer HRT allows longer contact time between the substrates and biofilm, which enabled more organic matter to be oxidized by microbes, thus, led to a higher reduction of COD. It was found that the COD removal efficiency at 1 day HRT was 70%, and rose about 7% when MFC operation was extended to 2 days HRT and it also enhanced the azo dye decolorisation and bioelectricity generation.

Oon et al. [75] studied the influence of HRT on the decolorisation and bioelectricity generation can be improved by longer HRT (2 days) and by regulating the external resistance closer to the internal resistance of the MFC system. The low concentration of New Coccine (NC) (25 mg/L) improved 17% of the power density to 20.13 ± 0.37 W/m^3. The internal resistance decreased from 50 to 32 Ω. As dye concentration increases, the decolorisation efficiency maintained over 90% (200 mg/L NC), whereas, power density dropped to 10.83 ± 1.21 W/m^3. Results showed that both decolorisation and power performances were improved with 2 days HRT. However, the HRT was too high, then the depletion of substrate occurs. Due to mass transfer losses, it increased the overpotential of the system thereby reducing the power output. Li et al. [48] studied the influence of HRT on the power density. The maximum power density reached 552.2 mW/m^2, when the HRT was 14.8 h. Increasing HRT from 14.8 to 44.4 h decreased the substrate concentration in the anodic chamber, which increased the open circuit voltage of anode from −431 to −283.8 mV. Longer HRTs reduce the organic loading rate, thereby reducing the cell performance due to the rapid depletion of the substrate, whereas shorter HRTs favor the development of non-exoelectrogenous bacteria, which reduces the CE and electrochemical performance with less effective COD removal, hence optimum HRT is preferred.

3.3.4 Microorganisms in Fuel Cells for Dye Wastewater Treatment

Many dissimilatory metal reducing bacteria possess the ability to transfer electrons directly attracting much attention for the application of MFC in dye degradation. Pure strain such as Proteus hauseri ZMd44 [8] and *Pseudomonas aeruginosa* [31] has been shown to generate electricity in mediator-less MFC systems and degrade the dyes simultaneously. Different azo dyes such as methyl orange (MO), Congo red, reactive blue 172 (RB), reactive yellow 145, and reactive red 2 were investigated in the *Pseudomonas*-catalyzed MFC [29], which is capable of producing pyocyanin and several other electron-shuttling compounds. Several isolates such as *Geobacter sulfurreducens* and *Beta Proteobacteria* have also been shown to produce energy output without the addition of mediators in MFC and dye removal [13]. In some cases, interestingly it has been found that the decolorized intermediates of azo dyes are redox-active chemical species which itself act as a mediator [8].

An overview of different species of microorganisms employed in the MFC operation with specified dye-bearing substrate. Different species involved in the MFC operation and their possible decolorization efficiency with power output is given in Table 4. Bacteria were identified as the members of the genera *Azospirillum*, *Methylobacterium*, *Rhodobacter*, *Desulfovibrio*, *Trichococcus*, and *Bacteroides* were more abundant in the dye degradation process.

3.3.5 Effect of Electrode

Among all the MFC components, the electrode materials play a crucial role in power generation since the cost of the electrode is a key barrier to the translation of MFCs from the laboratory scale to the industrial process [82]. The anodic material must be conductive, highly porous with enhanced active surface area, noncorrosive, and exhibit redox behavior compatible with microbial metabolism [30]. Different types of materials ranging from non-corrosive stainless steel to versatile carbons have been examined as anodes in different configurations [78]. Similarly, cathode materials also have an equally important effect on MFC operation and productivity. Ideally, cathodes are expected to have a high redox potential to accept electrons [71]. However, it is difficult to obtain high cathodic potentials unless metal-oxide-loaded catalysts are used. [33] employed granular activated carbon as the bioanode and biocathode for the treatment of industrial dye wastewater in an MFC to avoid the platinum catalyst and Nafion membrane, which are two tailbacks in MFC in terms of affordable costs. A stable voltage of 0.214 V was attained with 73% and 77% decolorization at the anode and cathode, respectively. Cui et al. [10] filled a cuboid titanium basket (L 13 cm × W 10 cm × H 7 cm) tightly with granular graphite and welded a titanium rod of 30 cm length on the basket for the external circuit connection. The large specific surface area provided by the graphite granules in this configuration was appreciably better for the power generation. Graphite-epoxy resin composite electrodes (MS-GECE) doped with various metal salts (MS) were tested by Jayapriya and Ramamurthy [31, 32] in dye degradation using *Pseudomonas aeruginosa* as the biocatalyst. The best-performing MFC was the one with Mn^{2+} GECE electrodes, producing the highest power density for methyl orange (4676 mW/m^2) and reactive black (2593 mW/m^2).[96] used a biocathode MFC for Congo red decolorization in textile dyeing sludge. The biocathode MFC exhibited excellent performance, achieving Congo red decolorization rapidly in 48 h, with a power density of 9 mW/m^2. Bulk-modified graphite-polyester composite electrodes (GPECE) doped with metal salts (MS-GPECE) prepared by casting [69] displayed redox behavior suitable for bacterial metabolism, and enhanced biofilm formation was observed in a Pseudomonas-catalyzed microbial fuel cell (MFC). The highest power density (1575 ± 223.26 $\mu W/m^2$) achieved with Ni-GPECE as the cathode material was approximately 15-fold higher than that obtained with graphite block, and its methyl-orange decolorization efficiency was 97 ± 1.4%. Nickel cobaltite ($NiCo_2O_4$)- and conductive polyaniline (PANI)-modified carbon cloth (CC) electrodes were synthesized by simple, low-cost hydrothermal and electropolymerization techniques, respectively,

by Narayanasamy and Jayaprakash [70]. Among the different electrode combinations tested, the surface-modified electrode Ni/PANI/CC (NCCP) produced the maximum power density when applied as a cathode (12.194 ± 0.59 mW/m^2), which was 15.9-fold higher than that obtained with CC (0.769 ± 0.0023 mW/m^2), besides a high decolorization efficiency of $80.5 \pm 2.1\%$. The preferred electrodes for dye decolorization seem to be mostly carbon and graphite electrodes. Though the maximum color removal has been achieved with carbon-based electrodes in combination with a Pt-catalyst-modified cathode, the incorporation of carbon nanotubes (CNTs) in conductive polymers has become a promising cost-effective electrode fabrication strategy in the MFC arena [72, 99]. All these efforts aspire to unearth the best electrode combination for effective decolorization of azo dyes while generating power as a by-product in MFCs.

4 Conclusion

If power output in these systems can be increased, MFC technology may provide a novel method to offset the high operating cost of traditional wastewater treatment plants, thereby making water recycling and waste disposal more inexpensive for developing and developed nations. MFCs operated with different synthetic dyes and polycyclic aromatic compounds at the pilot-scale demonstrate feasibility but require technical improvements toward high degradation efficiency and lower cost to be economically competitive at the industrial level. Evident inadequacies in commercialization aspects, including cost and durability, have been detected with respect to some MFC components, such as electrode materials, oxygen-reduction reaction catalysts, and membranes. Nowadays, attention toward the development of alternate materials is mounting, and MFC technology might lay a sophisticated path for the direct management of industrial dye effluents and PAH-rich petroleum waste via bioremediation.

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