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Review

Recent advances in the use of different substrates in microbial fuel cells toward wastewater treatment and simultaneous energy recovery

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HIGHLIGHTS

- Comprehensive and state-of-the-art information on various wastewater substrates in MFCs is provided.
- Most recent reports on complex substrates in MFC studies are consolidated.
- MFCs performance is compared with different substrates in terms of power density, coulombic efficiency and COD removal rate.
- Major operational parameters affecting the MFC performance has been evaluated.
- Existing bottlenecks in enhancing the process efficiency are discussed.

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ABSTRACT

The interest in the use of electroactive microorganisms for different applications by means of bioelectrochemical systems (BES) has been constantly increasing since the last decade. The main application of BES among others, which has received a widespread attention and researched extensively, is the microbial fuel cell (MFC) technology that relies on the electrogenic nature of certain bacteria to simultaneously treat different wastewaters and produce electric power. Various types of wastewaters have been examined as substrates for feeding bacteria in MFCs. The number and complexity of wastewaters have increased rapidly over the last few years thus necessitating the need for documenting this data further. This review provides a comprehensive and the state-of-the-art information on various wastewater substrates that have been used in MFCs. The performance of different types (designs) of MFCs in terms of electric current and power outputs together with the wastewater treatment efficiency in terms of chemical oxygen demand (COD) removal and columbic efficiency (CE) is presented. Some of the challenges and future perspectives with regard to the energy recovery from wastewaters using MFCs are briefly discussed.

Contents

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1. Introduction

Biotechnology is constantly explored for well-being of ecosystems, pollutant transformation, generation of biodegradable materials, developing environmentally safe manufacturing and disposal processes thereby providing a new dimension to the sustainable development research [1]. Addressing waste management and global climate change issues effectively is important for the sustainable development. In this context, achieving energy neutral treatment of municipal and industrial wastewaters is necessary [2–4]. With the energy needs increasing exponentially and fossil fuels resources being finite, the search for new alternative sustainable energy solutions have increased tremendously. One of the revived bio-electrochemical concept and promising technology that is proposed to address these aspects is microbial fuel cell (MFC), which principally produce electricity from the anaerobic oxidation of biodegradable organic substrates [5,6]. Microorganisms are capable of converting a wide variety of biodegradable organic compounds into CO_2, water and energy [7]. MFCs harvest this microbially produced energy and also provide habitat to maintain their growth and metabolic activities [8]. A general layout of a two chambered MFC is such that in the anodic compartment, microorganisms can bring about oxidative conversions and simultaneous chemical and/or microbial reductive processes can occur in cathodic compartment (Fig. 1). Electrodes of both compartments are usually separated by a proton or cation exchange membrane and are interconnected through an external circuit having an external resistor or load [9].

Power output by MFCs has increased considerably over the last decade due to several scientific and technical advances. Applications for the microbe–electrode interactions have also been expanded to waste/wastewater treatment, bio-remediation, toxic pollutants/xenobiotics removal, recovery of commercially viable products, i.e. resource recovery, sequestration of CO_2, harvesting the energy stored in marine sediments, and desalination [10–15]. MFCs have several unique advantages over the conventional bioenergy production technologies such as convenience in transmission and utilization of electricity, high substrate energy to electricity conversion efficiency and opportunities to develop as new electrical infrastructures for remote areas [15]. Recently, interlinking possibilities of MFCs with other technologies for the generation of two or more energies have attracted considerable attention [16–18].

In the last few years, several developments in MFC technology have been achieved concerning various designs and configurations [19–28], electrodes and electrode surface modifications [29–36], microbial communities [21,37–41], operational conditions for performance and biofilm formation [42–44], challenges and possibilities [45–49], fundamental electron transfer mechanisms [6,50], and applications [21,51–56]. Besides, considerable efforts have been made toward homogenizing the field and creating uniformly accepted parameters [57,58]. The growing interest in MFC

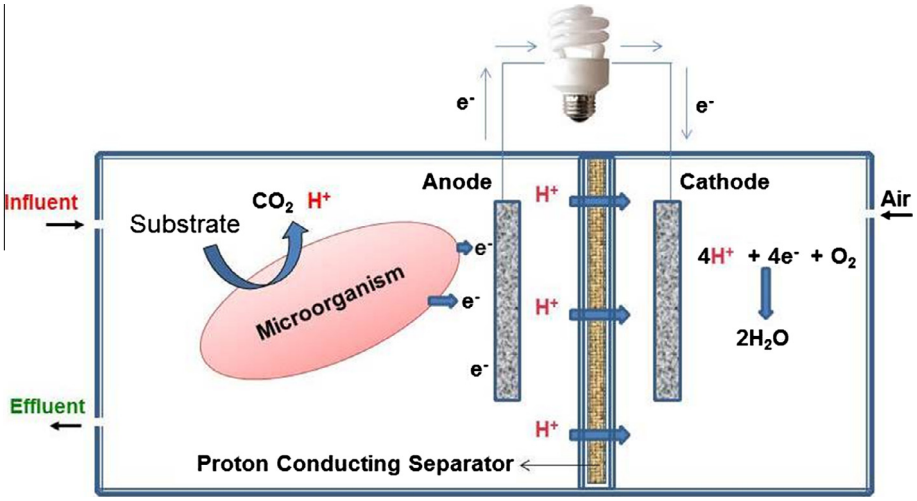

Fig. 1. A general layout of a two chambered microbial fuel cell.

technology among the scientific and engineering fraternity during the last few years is not only because of its potential application in energy harvesting but also because of its positioning as a more eco-friendly alternative to conventional anaerobic digestion and activated sludge waste treatment processes.

Based on microbe–electrode interactions, various systems are being developed with time to not only remove organics, but also for the production and recovery of value-added products like bioflocculants, bioplastics, biosurfactants, chemicals, electricity, hydrogen, methane and many other compounds from wastewater. In this paper, the focus is confined to the application of MFCs in wastewater treatment with simultaneous electricity harvesting. An overview of both low molecular weight and high molecular weight substrates emphasizing mainly various wastewaters as potential feed sources for MFCs with respect to recent advances, existing challenges, and future perspectives is presented. Recent literature pertaining to various wastewaters as potential source of electricity generation and their treatment using MFCs has been extensively reviewed.

2. Recent advances in the use of wastewaters as substrates in MFCs

Researchers across the globe have investigated several types of wastewaters with different MFC designs and operational conditions, and reported on various parameters such as columbic efficiency (CE), chemical oxygen demand (COD) removal rate and the effects of substrate concentration on the maximum voltage output and power density (P_d) [14]. Hence, comprehensive tables for both defined and undefined substrates have been included for providing a comparative information on various aspects of MFC performance, even though it is often difficult to tabulate all parameters because of use of different MFC designs, operational conditions, various measurement units and representation styles to name a few. The data presented for some cases is recalculated from their original source papers.

2.1. Simple or defined substrates

Various organics can be utilized in MFCs for electricity generation. Major metabolic fuels – carbohydrates, fatty acids and amino acids are the monomers of all complex and high molecular weight wastewaters. By far the most abundant group of these organics is carbohydrates. Fuel and energy generation from lignocellulosic biomass such as agricultural residues and woody biomass has drawn considerable attention because of their readily availability and abundance [59]. The electricity generation with various simple molecules as electron donors has been tested over the years (Table 1). The use of pure substrates has been the choice of researchers focusing on different fundamental aspects of MFCs operating mainly with pure cultures. The growth medium with pure substrate (carbon source) and other micronutrients is usually referred to as synthetic wastewater. Comparative table of simple substrates also provide a rough estimate for predicting power output of complex wastewater substrates.

2.1.1. Carbon source wastewater or substrates

The effect of carbon sources on the performance of MFCs was first tested by Kim et al. [76]; using *Proteus vulgaris* as biocatalyst. Electricity generation by direct glucose oxidation in MFCs was investigated then by some researchers [77,78]. An average loading rate of 1000 mg l^{-1} d^{-1} of glucose in dual-chambered MFC resulted in 4310 mW m^{-2} P_{dmax} (maximum P_d) and corresponded to 81% CE [78]. Afterward, the electricity generation from carbohydrates has attracted sufficient attention [79]. For instance, electricity from six hexoses (glucose, galactose, fructose, fucose, rhamnose, and mannose), three pentoses (xylose, arabinose, and ribose), and three sugar derivatives (galacturonic acid, glucuronic acid, gluconic acid) in single-chambered air cathode MFCs was tested successfully using a mixed bacterial culture [60]. The highest power density (P_{dmax}) of 2770 mW m^{-2} was obtained with glucuronic acid, which was followed by xylose and glucose. Mannose produced the lowest P_d about 1240 mW m^{-2}. The CE ranged from 22% to 34% with over 80% COD removal. In a novel stack of MFCs (consisting of four units and operated in a continuous mode), 30 g l^{-1} of pure glucose, 200 μmol l^{-1} natural red and *Saccharomyces cerevisiae* as an active biocatalyst in anode and 400 μmol l^{-1} of potassium permanganate as oxidizing agent in the cathode resulted in 6447 mA m^{-2} as maximum current and P_d of 2003 mW m^{-2} [80]. CE of 22% was achieved at parallel connection. These results indicated that all monosaccharides in a hydrolysate from acid hydrolysis of lignocellulosic materials could be used for electricity generation.

Commercial foodstuffs sweetened with sugar alcohols such as isomalt, lactitol, maltitol, sorbitol, and xylitol are widely available today as sweeteners for diabetics. Digestion and metabolism of sugar alcohols in humans is partial and intake exceeding 10–20 g d^{-1} may cause harmful effects on humans [81]. The environmental and toxic impacts of such sugar alcohols on other living beings are not yet clear. Water-soluble materials of corn stover (14–27% of the total biomass on a dry weight basis) contain certain amount of polyalcohols that account for about 3–7% of the dry weight of extractives [82]. The ethanol fermentation process also produces polyalcohols as by-products. Such alcohols have been tried as substrates in MFCs. First time demonstration of electricity generation from polyalcohols (xylitol, arabitol, ribitol, galactitol, mannitol, and sorbitol) was carried out using single-chamber, air–cathode and mediator-less MFCs and using a mixed culture enriched from domestic wastewater [62]. Power densities were in the range of 1490–2650 mW m^{-2}. The P_d series in descending order was Galactitol > Ribitol > Xylitol > Arabitol > Sorbitol > Mannitol. COD removal efficiencies for all polyalcohols ranged in between 71% and 92%. These results indicated that microbes could utilize polyalcohols for electricity generation, which, on the other hand could not be metabolized completely by humans.

2.1.2. Nitrogen source wastewater or substrates

Protein is an important constituent in organic wastewaters such as domestic, food processing and industrial wastewaters. The different nitrogen containing organic wastewaters vary in types and concentrations of amino acids. Yang et al. [63] examined the possibility of direct electricity generation using single chamber air–cathode MFCs of eight representative amino acids based on different side chains (–R group). These included non-polar amino acid alanine and polar amino acids such as serine and asparagine with –R groups with no charges; histidine, lysine, and arginine with positive charges, and aspartic acid and glutamic acid with negative charges. Results of this study clearly showed the dependence of P_d on –R groups in amino acids. Serine resulted in the highest P_{dmax} of 768 mW m^{-2} and alanine produced the lowest P_d of 556 mW m^{-2}. The P_d showed following trend in descending order: L-Serine > L-Arginine > L-Histidine > L-Glutamic acid > L-Aspartic acid > L-Asparagine > L-lysine > DL-Alanine. COD removal was over 91% and CEs ranged from 13% to 30%. These results indicated that –R group plays an influential role in power generation from amino acids in MFCs.

Heilmann and Logan [83] examined electricity generation from proteins and a protein-rich wastewater. P_{dmax} achieved in a single chamber MFC using bovine serum albumin (BSA, 1100 mg l^{-1}) and peptone (300 mg l^{-1}) was ≈354 mW m^{-2} and ≈ 269 mW m^{-2}, respectively. The recovery of organic matter as electricity was

Table 1
An overview of performance of MFCs with simple or defined wastewater substrates.

S. No.	Substrate type	Conc.	Inoculum	Type of MFC	Working volume (l)	Anode	Cathode	OCV Max. (V)	CE%	COD Rem. %	P_{dmax} (mW m^{-2})	Ref. No.
Carbohydrates: Monosaccharides												
HEXOSES												
1	D-Glucose	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.39	28	≈93	≈2160	[60]
2	D-Galactose	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.35	23	≈93	≈2090	[60]
3	D-Fructose	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.31	23	≈88	≈1810	[60]
4	L-Fucose	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.35	34	≈84	≈1760	[60]
5	L-Rhamnose	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.27	30	≈90	≈1320	[60]
6	D-Mannose	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.29	25	≈88	≈1240	[60]
7	Sucrose	0.1 g l^{-1}	Anaerobic sludge	Single chambered	NA*	Carbon fiber veil	Carbon cloth	NA*	4	94	1.79 W m^{-3}	[61]
PENTOSES												
8	D-Xylose	8.0 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.38	31	≈95	≈2330	[60]
9	D-(-)Arabinose	8.0 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.26	27	≈93	≈2030	[60]
10	D-(-)Ribose	8.0 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.27	30	≈86	≈1520	[60]
Sugar derivatives												
SUGAR ACIDS												
11	D-Galacturonic acid	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.33	22	≈80	≈1480	[60]
12	D-Glucuronic acid	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.44	24	≈89	≈2770	[60]
13	D-Gluconic acid	6.7 mM	Mixed bacterial culture	Two chambered	0.012	Carbon cloth	Carbon cloth	0.28	30	≈93	≈2050	[60]
Polyalcohols												
HEXITOLS												
14	Galactitol	6.7 mM	Mixed bacterial culture	Single chambered mediatorless	0.012	Carbon cloth	Carbon cloth	0.34	13	≈90	≈2650	[62]
15	Mannitol	6.7 mM	Mixed bacterial culture	Single chambered mediatorless	0.012	Carbon cloth	Carbon cloth	0.24	19	≈91	≈1490	[62]
16	Sorbitol	6.7 mM	Mixed bacterial culture	Single chambered mediatorless	0.012	Carbon cloth	Carbon cloth	0.26	10	≈71	≈1690	[62]
PENTITOLS												
17	Arabitol	8.0 mM	Mixed bacterial culture	Single chambered mediatorless	0.012	Carbon cloth	Carbon cloth	0.26	25	≈91	≈2030	[62]
18	Ribitol	8.0 mM	Mixed bacterial culture	Single chambered mediatorless	0.012	Carbon cloth	Carbon cloth	0.32	28	≈92	≈2350	[62]
19	Xylitol	8.0 mM	Mixed bacterial culture	Single chambered mediatorless	0.012	Carbon cloth	Carbon cloth	0.29	21	≈91	≈2110	[62]
Amino acids												
20	L-Serine	2.0 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.37	20	≈93	768	[63]
21	L-Asparagine	1.5 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.39	15	≈93	595	[63]
22	L-Asparitic Acid	1.5 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.25	25	≈94	601	[63]
23	L-Glutamic Acid	1.2 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.33	≈ 28	≈95	686	[63]
24	DL-Alanine	2.0 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.28	30	≈96	556	[63]
25	L-Lysine	1.0 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.32	25	≈93	592	[63]

(*continued on next page*)

Table 1 (*continued*)

S. No.	Substrate type	Conc.	Inoculum	Type of MFC	Working volume (l)	Anode	Cathode	OCV Max. (V)	CE%	COD Rem. %	P_{dmax} (mW m^{-2})	Ref. No.
26	L-Histidine	1.0 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.42	25	≈93	718	[63]
27	L-Arginine	1.0 mM	Domestic Waste-water	Single chambered air cathode	0.028	Carbon cloth	Carbon cloth	0.36	13	≈91	727	[63]
Organic acids												
28	Acetic acid	1 g l^{-1}	Domestic Waste-water	Single chambered	0.028	Graphite fiber brushes	Wet-proofed carbon cloth	0.475	19.9	NA*	835	[64]
29	Formic acid	1 g l^{-1}	Domestic Waste-water	Single chambered	0.028	Graphite fiber brushes	Wet-proofed carbon cloth	0.180	3.9	NA*	62	[64]
30	Lactic acid	1 g l^{-1}	Domestic Waste-water	Single chambered	0.028	Graphite fiber brushes	Wet-proofed carbon cloth	0.400	13.4	NA*	739	[64]
31	Succinic acid	1 g l^{-1}	Domestic Waste-water	Single chambered	0.028	Graphite fiber brushes	Wet-proofed carbon cloth	0.400	16.2	NA*	444	[64]
32	Acetate	800 mg COD l^{-1}	Domestic Waste-water	Single chambered	0.028	Toray carbon paper	Carbon paper	0.79	7	>99	506	[65]
33	Butyrate	1000 mg COD l^{-1}	Domestic Waste-water	Single chambered	0.028	Toray carbon paper	Carbon paper	0.79	5	>98	305	[65]
34	Formate	20 mM	Anaerobic digested fluid	C-type mediatorless MFC	0.005	Graphite felt	Graphite felt	NA*	6.5	83	NA*	[66]
35	Propionate	5 mM	Anaerobic sludge	Two chambered	NA*	Graphite felt	Graphite felt	0.800	31.5	NA*	115.6	[67]
36	Lactate	20 mM	Anaerobic enrichment cultures with soil	Two chambered	0.025	Graphite felt	Graphite felt	0.750	12.5	NA*	320	[68]
Alcohols												
37	Ethanol	1.5 mM	Anaerobic sludge	Single chambered	0.028	Toray carbon paper	Carbon paper	0.750	10	NA*	488	[69]
38	Glycerol	0.5 ml l^{-1}	*Bacillus subtilis*	Single chambered	0.500	Carbon cloth	Carbon cloth	0.560	23.08	NA*	600	[70]
Nitrogenous heterocyclic compounds												
39	Pyridine	120 mg COD l^{-1}	Anaerobic sludge	Two chambered	0.250	Carbon paper	Pt coated carbon paper	0.524	<8.0	86	142.1	[71]
40	Quinoline	120 mg COD l^{-1}	Anaerobic sludge	Two chambered	0.250	Carbon paper	Pt coated carbon paper	0.494	<8.0	93	203.4	[71]
41	Indole	120 mg COD l^{-1}	Anaerobic sludge	Two chambered	0.250	Carbon paper	Pt coated carbon paper	0.413	<8.0	95	228.8	[71]
Miscellaneous												
42	Phenol	600 ml l^{-1}	Anaerobic sludge	Single chambered	0.330	Carbon felt	Pt coated carbon cloth	0.631	3.68	88.9	31.3	[72]
43	Sulfide	NA*	*Pseudomonas sp. C27*	Two chambered	NA*	Carbon felt	Pt-coated carbon cloth	0.669	25.60	NA*	29.3	[73]
44	Tetrathionate	2 g l^{-1}	Original aerobic process water sample	Two chambered	0.100	Graphite plate	Carbon cloth + Graphite plate	0.175	4.9	NA*	13.9	[74]
45	Furfural	6.68 mM	Anaerobic and aerobic sludge	Single chambered	NA*	Carbon cloth	Carbon cloth	0.420	30.3	68	361	[75]

OCV max = open circuit voltage maximum, COD = chemical oxygen demand, CE = coulombic efficiency, P_{dmax} = power density maximum, mW = miliwatt, V = volt.

* NA: data not available.

Table 2
An overview of performance of MFCs with undefined wastewater substrates.

S. No.	Waste water	Substrate Conc.	Inoculum	Type of MFC	Working volume (l)	Anode	Cathode	OCV Max.	CE%	COD Rem. %	P_{dmax}	Ref. No.
1	Acidogenic food waste leachate	1000 mg COD l^{-1}	Anaerobic sludge	Two chambered	0.400	Carbon felt	Carbon felt	0. 400	20	>87	432 mW m^{-3}	[90]
2	Alcohol distillery wastewater	28,400 mg COD l^{-1}	Alcohol distillery wastewater	Two chambered	7.27	Carbon fiber paper	Carbon fiber paper	0.611	NA*	88.38	124.03 mW m^{-2}	[91]
3	Animal carcass wastewater	11.18 g COD l^{-1}	Anaerobic granular sludge supernatant	Up-flow tubular air cathode	1.2	Graphite felt	Carbon fiber cloth	0.55	0.25	50.66	2.19 W m^{-3}	[92]
4	Biodiesel wastewater	1400 mg COD l^{-1}	Domestic waste water	Single chambered open air cathode	NA*	Carbon brush	Carbon cloth	0.47	18	90	2110 mW m^{-2}	[93]
5	Brewery wastewater	1501 mg COD l^{-1}	Brewery Anaerobic mixed consortia	Single chambered open air cathode	0.100	Carbon fibers	Activated carbon powder and PTFE containing Pt catalyst	0.51	2.58	20.7	669 mW m^{-2}	[94]
6	Bad Wine	7.8 g COD l^{-1}	*Gluconobacter roseus* and *Acetobacter aceti*	Two chambered	0.125	Carbon felt	Graphite	0.823	45	41	3.82 W m^{-3}	[95]
7	Cattle wastes (manure wash-water)	NA*	*Escherichia coli* strain K-12 and manure leachate	Three compartment air cathode	1.85	Graphite fiber brush	Pt coated carbon cloth	0.57	5.2	NA*	216 mW m^{-2}	[96]
8	Cassava mill wastewater	16,000 mg COD l^{-1}	Mixed culture sludge	Two chambered	30	Graphite plates	Graphite plates	0.18	20	72	1771 mW m^{-2}	[97]
9	Cellulose	2 g COD l^{-1}	Domestic wastewater	Single chambered Air cathode	0.026	Carbon paper	Pt coated Carbon cloth	≈0.6	50	70	1070 mW m^{-2}	[98]
10	Cheese whey	6.7 g COD l^{-1}	Anaerobic sludge	Two chambered	0.310	Carbon paper	Carbon cloth	NA*	11	94	46.00 mW m^{-2}	[99]
11	Chocolate industry	1459 mg COD l^{-1}	Activated sludge	Two chambered	0.400	Graphite rods	Graphite rods	0.498	NA*	75	1.5 W m^{-2}	[40]
12	Coal Tar wastewater	2013 mg COD l^{-1}	Activated sludge	Single chambered	0.600	Graphite felt	Graphite felt	0.543	NA*	88	4.5 mW m^{-2}	[100]
13	Coking wastewater	3150–3200 mg COD l^{-1}	Coking wastewater	Single chambered	0.300	Graphite-fiber brush	Carbon cloth	≈ 0.6	17	50	538 mW m^{-2}	[101]
14	Composite vegetable	52 g COD l^{-1}	Anaerobic mixed consortia	Single chambered	0.43	Graphite plates	Graphite plates	0.25	NA*	62.86	57.38 mW m^{-2}	[102]
15	Corn stover hydrolysate	1000 mg COD l^{-1}	Domestic wastewater	Single chambered	0.028	Carbon paper	Carbon cloth	0.5	26.90	70	861 mW m^{-2}	[103]
16	Crude glycerol	7610 mgl^{-1}	Anaerobic sludge	Two chambered	0.025	Graphite brush	Carbon cloth	0.210	≈13	50	92 mW m^{-2}	[104]
17	Dairy industrial wastewater	53.22 $kgCODm^{-3}$ d	Activated sludge	Two chambered	2.00	Graphite plate	Graphite plate	0.856	37.16	90.46	621.13 mW m^{-2}	[105]
18	Distillery wastewater	3200 mg COD l^{-1}	Isolated broth culture	Two chambered	0.250	Graphite plate	Graphite plate	0.680	NA*	71.8	202 mW m^{-2}	[106]
19	Domestic wastewater	238.7 mg COD l^{-1}	Activated sludge and effluent of primary clarifier	Upflow membrane-less MFC	0.850	Carbon fiber brush	Carbon fiber brush	0.200	14	77.9	481 mW m^{-3}	[107]
20	*Enteromorpha prolifera* hydrolysate	1000 mg COD l^{-1}	Pre-domesticated bacteria from other dual chambered MFC	Single chambered	0.027	Carbon cloth	Vulcan XC-72, PFTE coated carbon cloth	0.72	69.1	76.1	1027 mW m^{-2}	[108]
21	Ethanol stillage	37,890 mg COD l^{-1}	Anaerobic sludge	Two chambered	4.0	Graphite rods	Carbon cloth	NA*	NA*	81.50	93.0 W m^{-2}	[109]
22	Fermented apple juice	3501 mg COD l^{-1}	Compost leachate	Two chambered	0.500	Graphite felt	Platinum mesh	0.6	4.70	NA*	78 mW m^{-2}	[110]
23	Fermented corn stover hydrolysate	5300 mg l^{-1}	Anaerobic mixed consortia	Single chambered	0.016	Carbon-felt	Pt-deposited carbon	NA*	≈10	NA*	1180 mW m^{-2}	[111]
24	Food processing	1900 mg COD l^{-1}	Activated sludge	Catalysts and mediatorless	1.5	Graphic sheets	Graphic sheets	0.475	21	86	230 mW m^{-2}	[112]
25	Human urine	17 g COD l^{-1}	Human urine	Single chambered	0.130	Carbon brush	Carbon cloth	0.498	NA*	75	55 μW	[113]
26	Human feces wastewater	≈ 650 mg COD l^{-1}	Anaerobic sludge	Two chambered	1.0	Carbon paper	Pt coated carbon paper	0.548	NA*	71	70.8 mW m^{-2}	[114]
27	Molasses wastewater	127,500 mg COD l^{-1}	Anaerobic granular sludge	Up-flow anaerobic sludge blanket reactor	1.08	Granular graphite	Carbon paper	0.5	1	53.20	1410.2 mW m^{-2}	[115]
28	Mustard tuber wastewater	287 mg COD l^{-1}	MTWW	Two chambered	0.150	Non-wet proof carbon cloth	Non-wet proof carbon cloth	0.696	67.7	85	246 mW m^{-2}	[116]

(*continued on next page*)

Table 2 (*continued*)

S. No.	Waste water	Substrate Conc.	Inoculum	Type of MFC	Working volume (l)	Anode	Cathode	OCV Max.	CE%	COD Rem. %	P_{dmax}	Ref. No.
29	Palm oil mill effluent sludge	2680 mg COD l^{-1}	*Pseudomonas aeruginosa* strain ZH1	Two chambered	0.100	Carbon graphite	Carbon graphite	NA*	NA*	3	451.26 mW m^{-2}	[117]
30	Petroleum refinery wastewater	250 mg COD l^{-1}	Activated sludge	Dual-chambered GC-packing-type	0.400	Carbon rod	Graphite flake	0.305	NA*	64	330.4 mW cm^{-3}	[118]
31	Piggery wastewater	1500–3250 mg COD l^{-1}	Anaerobic sludge	Two chambered loop configuration MFC	1.0	Hexahedral stain-less steel mesh filled with graphite granules	Pt coated graphite felt	NA*	1.7	NA*	1415.6 mW m^{-3}	[119]
32	Purified Terephthalic acid wastewater	8000 mg COD l^{-1}	Anaerobic sludge	Single chambered	0.250	Graphite–fiber brush	Waterproof carbon cloth (Pt catalyst)	0.0309	2.05	74	31.8 mW m^{-2}	[120]
33	Real dye wastewater	2200 mg COD l^{-1}	Real dye wastewater	Granular activated carbon based single chamber MFC	2.5	GAC bed	GAC bed	0.39	NA*	71	8.0 W m^{-3}	[121]
34	Real field Dairy waste water	4.44 kg COD m^{-3}	Anaerobic mixed consortia	Single chambered open air cathode	0.48	Graphite plates	Graphite plates	0.3	4.30	95.49	1.10 W m^{-3}	[122]
35	Recalcitrant Pharmaceutical wastewater	7.98 kg COD m^{-3}	Anaerobic mixed consortia	Single chambered open air cathode	0.43	Graphite plates	Graphite plates	0.346	NA*	85	205.61 mW m^{-2}	[123]
36	Rice Mill Wastewater	2250 mg COD l^{-1}	Anaerobic sludge	Earthen pot MFC	0.40	Stainless steel	Graphite plate	0.304	21	96.50	2.3 W m^{-3}	[124]
37	Rice straw	1000 mg l^{-1}	Mixed bacterial culture	Two chambered	0.160	Carbon paper	Carbon paper	0.705	37	NA*	190 mW m^{-2}	[125]
38	Red wine lees wastewater	10,100 mg COD l^{-1}	Denitrification tank wastewater	Single chambered	0.028	Graphite brushes	Pt and PTFE coated carbon cloth	0.34	9	27	111 mW m^{-2}	[126]
39	Retting wastewater (Coconut husk)	2.69 g COD l^{-1} d^{-1}	Retting wastewater	Continuous up flow dual-chambered	0.600	Graphite sheet	Graphite sheet	0.880	0.8–8	32	254 mW m^{-2}	[127]
40	Saline seafood wastewater	NA*	Marine sediments	Anoxic/oxic – MFC	0.226	Packed granular graphite	Packed granular graphite	0.780	15	NA*	16.2 W m^{-3}	[128]
41	Sewage wastewater	400 mg COD l^{-1}	Anaerobic sludge	Membrane –less	4.6	Graphite rod	Graphite rod	0.188	NA*	82.70	6.73 mW m^{-2}	[129]
42	Slaughterhouse wastewater	4850 mg COD l^{-1}	Anaerobic mixed-sludge	Dual-chambered	0.125	Carbon cloth	Platinised titanium mesh	NA*	64	93	578 mW m^{-2}	[130]
43	Starch processing	4852 mg COD l^{-1}	Starch processing waste water	Air cathode single chambered	NA*	Carbon paper	Carbon paper	0.49	8	98	239.4 mW m^{-2}	[131]
44	Steroidal Drug Industrial effluent	1340 mg COD l^{-1}	Anaerobic sludge	Air cathode single chambered	0.028	Carbon brush	Carbon cloth	0.52	30	82	22.3 W m^{-3}	[132]
45	Swine wastewater	8270 mg COD l^{-1}	Swine wastewater	Cube-shaped air cathode	NA*	Carbon paper	Carbon paper	0.4	NA*	84	228 mW m^{-2}	[133]
46	Synthetic acid-mine drainage	NA*	NA*	Two chambered	0.0014	Carbon cloth	Carbon cloth	0.63	98	NA*	290 mW m^{-2}	[134]
47	Synthetic penicillin	1 g l^{-1} glucose + 50 mg l^{-1} penicillin	Mixed consortia	Air cathode single chambered	0.100	Carbon felt	NA*	0.517	NA*	90	101.2 W m^{-3}	[135]
48	Synthetic (potato) starch wastewater	1933 mg COD l^{-1}	*Escherichia coli*	Two chambered	NA*	Platinised titanium strip or mesh	Platinised titanium strip or mesh	0.900	18.5	61	502 mW m^{-2}	[136]
49	Tetrachloroaurate wastewater	2000 ppm	Anaerobic sludge	Two chambered	0.060	Carbon brush	Carbon cloth	0.851	57	NA*	6.58 mW m^{-2}	[11]
50	Toxic refractory organic pesticide, hexachlorobenzene	40 mg kg^{-1}	Anaerobic sludge and Soil	Single chambered	0.140	Granular activated carbon	Carbon cloth	0.326	NA*	71.15	77.5 mW m^{-2}	[137]
51	Yogurt wastewater	8169 mg COD l^{-1}	Anaerobic sludge	Two chambered	0.500	Graphite felt	Platinum mesh	0.7	9.6	NA*	53.8 mW m^{-2}	[110]
52	Wheat straw hydrolysate	NA*	Domestic wastewater	Two chambered	NA*	Carbon paper	Carbon paper	0.703	17	NA*	148 mW m^{-2}	[138]
53	White wine lees wastewater	6400 mg COD l^{-1}	Denitrification tank wastewater	Single chambered	0.028	Graphite brushes	Pt and PTFE coated carbon cloth	0.42	15	90	262 mW m^{-2}	[126]
54	Winery wastewater	2200 mg COD l^{-1}	Anaerobic sludge	Single chambered	0.028	Graphite carbon brushes	Carbon cloth	0.441	18	65	31.7 W m^{-3}	[139]

OCV max = open circuit voltage maximum, COD = chemical oxygen demand, CE = coulombic efficiency, P_{dmax} = power density maximum, mW = miliwatt, V = volt, PTFE = polytetrafluoroethylene, Pt = platinum, GAC = granular activated carbon.

* NA: data not available.

comparable to that obtained with other substrates with CE 20% for BSA and 6% for peptone.

2.1.3. Organic acids and miscellaneous substrates

Acetic and butyric acids are the primary fermentation end products in the process of biohydrogen production. Such organic acids have been tested as substrates in MFCs. For example, Liu et al. in the initial study reported 506 mW m^{-2} and 305 mW m^{-2} with acetate and butyrate in MFCs, respectively [65]. Thereafter, acetate has been widely used as a substrate for MFCs operated with *Geobacter sulfurreducens*. Mixed volatile fatty acids (VFAs) have also been used in MFCs, which generated P_{dmax} of 240 mW m^{-2} [84]. Short-chain VFAs supported higher power generation than longer chain VFAs because of their rapid degradability. A combined system comprised of dark fermentation reactor producing biohydrogen and MFC producing electricity using the mixture of VFAs has been reported to reduce the COD load by 90% [5].

Kim et al. [69] investigated two different MFC configurations for electricity production from alcohols (ethanol and methanol). The P_{dmax} using ethanol as substrate was 488 mW m^{-2} and CE was 10%. Kiely et al. [64] also reported higher P_d of 820 mW m^{-2} with ethanol. Examination of methanol as possible substrate did not result in appreciable electricity generation [69]. Several other pure substrates that have been investigated as substrates in MFCs include succinate, dextran, glycerin etc. However, because of insufficient data some of them are not included in Table 1.

Energy-conversion efficiencies with the most commonly used fermentable (glucose) and non-fermentable (acetate) substrates in MFCs have also been evaluated in several studies. For instance, using continuous mode operation and ferricyanide as catholyte, Min and Logan [85] reported the P_d of 212 and 286 mW m^{-2} by glucose and acetate, respectively. Rabaey et al. [86] reported 66 and 90 W m^{-3} with glucose and acetate, respectively in another study. In a study of Lee et al. [87], the P_d in the acetate-fed and glucose-fed MFC in batch fed mode were 360 and only 9.8 mW m^{-2}, respectively. The presence of high density of non-exoelectrogenic bacteria in anode biofilms has been reported to be the main reason in achieving low current production with fermentable substrates.

Al-Shehri recently evaluated the recalcitrant mixture of naphthalene and benzidine that resulted in P_{dmax} of 292.60 mW m^{-2} and 100% mixture removal at optimal conditions [88]. Toluene supplemented with pyocyanin achieved a 3.64-fold increase in P_{dmax} from 4.69 to 21.7 mW m^{-2} and a 13-fold increase in CE from 0.83% to 11.62% in comparison to only toluene feed in a study by Wu and coworkers [89].

2.2. Complex or undefined wastewater substrates

Attributed to the ability of MFCs for wastewater treatment with simultaneous energy recovery, several waste streams have been tried as substrates so far. An overall account of the performance of MFCs with different wastewaters is presented in Table 2. Some wastewaters are not presented in table but discussed in subsections because of insufficient available data to tabulate. A few wastewater substrates are discussed under specific categories in subsequent sub-sections.

2.2.1. Food and food-processing industry wastewater

Food losses occurring at the end of the food chain (retail and final consumption) are termed as food wastes [140,141]. Throughout the world, huge quantities of food wastes that are rich in carbohydrate content are produced. About 27% of total municipal solid waste (MSW) is composed of food waste [142]. Globally, wastage or loss of roughly one third of edible part of food produced for human consumption (estimated 1.3 billion ton per year) is a concern [141]. This has prompted several researchers to investigate food wastes as potential substrates in MFCs.

2.2.1.1. Composite vegetables and food wastewater. The feasibility of composite vegetables waste as substrate in single chambered mediator-less MFC was evaluated by Venkata Mohan and coworkers [102]. The MFC operated with this waste resulted in P_{dmax} of 57.38 mW m^{-2} with effective removal of 62.86% COD. In a subsequent year the same group [143] evaluated the electrical potential of solid phase MFC (graphite electrodes; open-air cathode) by stabilizing composite canteen based food waste. Maximum OCV of 463 mV and P_d of 170.81 mW m^{-2} were obtained. At organic loading rate (OLR) of 1.74 kg COD m^{-3} d^{-1} this substrate resulted in maximum potential, P_d, and COD removal of 295 mV, 107.89 mW m^{-2}, and 64.83%, respectively [144].

2.2.1.2. Food processing wastewater. The efficiency of catholytes on bioelectricity production and substrate removal were evaluated by employing food-processing wastewater as anolyte by Sangeetha and Muthukumar [145]. Experiments with dual chambered, salt bridge MFC in this study resulted in P_{dmax} of 123.8 mW m^{-2}, current density of 54.3 mA m^{-2}, power yield of 110 mW kg^{-1} and COD removal of 98.9%. Mansoorian et al. [112] reported P_{dmax} of 230 mW m^{-2} with COD removal of 86% with food processing wastewater. Miran and coworkers [146] recently reported P_{dmax} of 371 mW m^{-2} with lemon peel (synthetic) wastewater used at an initial concentration of 1.0 g l^{-1}.

Starch products manufacturing industry utilizes a large amount of water; consequently, resulting in a large amount of starch processing wastewater (SPW) characterized by high COD ranging from 16,870 to 22,800 mg COD l^{-1}. SPW has relatively high contents of carbohydrates, cellulose, protein, and nutrients, and it can impose heavy loads on the environment [147]. Bioconversion of SPW is advantageous in terms of resource recovery such as microbial biomass protein [148,149] and biopesticide [150]. SPW of COD 4852 mg l^{-1} in MFCs resulted in P_{dmax} of 239.4 mW m^{-2}, maximum CE of 8% and COD removal efficiency of 98.0% [131]. In another study, potato extracts (starch) synthetic wastewater was tested by Herrero-Hernandez et al. [136] by using *Escherichia coli* in mediator-less MFC. The P_{dmax} 502 mW m^{-2} was achieved with titanium mesh electrodes with significant reduction of initial oxygen demand of wastewater by 61%.

2.2.1.3. Protein food industry wastewater. Protein food industry wastewater can be characterized as non-toxic because it contain very few hazardous compounds, high BOD, and high organics comprising of simple sugars and starch [151]. Such type of waste for example, cereal wastewater having 595 mg COD l^{-1} has been evaluated as substrate in two chambered MFC. It resulted in 81 mW m^{-2} P_d and 95% COD removal [152].

2.2.1.4. Tomato industry pomace. The tomato industry generates large amount of pomace waste on an annual basis globally. Fogg et al. [153] recently demonstrated use of pomace as the feedstock for electricity production in MFC and P_{dmax} reported in this case was 132 mW m^{-2}.

2.2.1.5. Acidogenic food waste leachate. Acidogenic food waste leachate has also been evaluated as potential resource [154]. Leachate characterized by complex structure and high pollution load may cause ground and surface water contamination. Rikame et al. [155] obtained using 5000 mg COD l^{-1} as substrate and anaerobic sludge as an inoculum, the P_{dmax} of about 15.14 W m^{-3} and 90% COD removal. In another study, 1000 mg COD l^{-1} food waste leachate resulted in 432 mW m^{-3} P_d and more than 87% COD removal [90].

2.2.2. Beverages industry wastewater

Brewery wastewater has high COD, is nontoxic, and is characterized by presence of high organics in water consisting of sugar, starch, and protein components. Aerobic sequencing batch reactor [156], cross-flow ultrafiltration membrane anaerobic reactors [157], and up-flow anaerobic sludge blanket reactors [158] are routinely used biological methods for brewery wastewater treatment. Biological treatment is effective but requires a high-energy input due to the need for aeration [159]. In this context, MFCs have been tried for treatment of such wastewaters. Winery wastewater also has high-COD and has been evaluated as a feed in MFCs by researchers.

2.2.2.1. Beer brewery wastewater. Beer brewery wastewater with COD of 2240 mg l^{-1} in single chambered MFC resulted in P_{dmax}, COD removal, and CE of 205 mW m^{-2}, 87%, and 10%, respectively. Further P_d was increased by 158% to 528 mW m^{-2} by adding 200 mM phosphate buffer [160]. In another study, experimental results showed that the full strength wastewater of 2239 mg COD l^{-1} and 50 mM phosphate buffer solution (PBS) resulted in P_{dmax} of 483 mW m^{-2} (12 W m^{-3}) [161]. Further, the air–cathode MFC displayed a maximum power of 669 mW m^{-2} (24.1 W m^{-3}) running on raw continuous brewery wastewater (COD = 1501 mg l^{-1}) [94].

2.2.2.2. Winery wastewater. Using this wastewater (2200 mg COD l^{-1}), a maximal electrical energy of 31.7 W h m^{-3}, 65% COD removal and 18% CE has been reported [139]. Rengasamy and Berchmans [95] reported 3.82 W m^{-3} P_d, 41% COD removal and 45% CE in two chambered MFC with bad wine (7.8 g l^{-1} of COD) as substrate with *Acetobacter aceti* and *Gluconobacter roseus* as biocatalysts. Penteado and coworkers [162] reported 105 mW m^{-2} P_d, COD Removal about 17% and 2% CE, and demonstrated that the unbalanced nutrients/COD ratio is a major challenge in the winery wastewater treatment and efficient electricity production. Electricity generation using white and red wine lees (262 mW m^{-2} and 111 mW m^{-2} respectively) in air cathode MFCs was recently reported by Sciarria et al. [126]. COD removal and CE performance was greater in white wine lees compared to red wine lees.

2.2.2.3. Fermented apple juice. Fermented apple juice, another brewery product has been tested in MFC as substrate. Cercado-Quezada et al. [110] reported P_{dmax} of 78 mW m^{-2} using compost leachate as inoculum with fermented apple juice.

2.2.3. Confectionary industry wastewater

The chocolate manufacturing industry wastewater has high total solids (TS), biochemical oxygen demand (BOD) and COD. Using activated sludge as a source of microorganisms, chocolate manufacturing industry wastewater was investigated in two-chambered MFC that resulted in P_d of 1.5 W m^{-2} and 75% COD removal [40].

2.2.4. Dairy industry wastewater

Dairy wastewater is complex in nature and is composed of biodegradable organics of which 97% of total COD is sugar [163]. It contains high concentration of fermentable substrates [3]. Real field dairy wastewater was evaluated in single chamber MFC that resulted in 1.1 W m^{-3} P_d and 95.49% COD removal [122]. Increased P_{dmax} 20.2 W m^{-3} was obtained using spiral stainless steel mesh anode with graphite coating in another study [164]. In a study by Mansoorian et al. [105], dairy industry wastewater at organic loading rate of 53.22 kg COD m^{-3} d^{-1} in catalyst-less and mediator-less membrane MFC resulted in $P_{dmax,}$ CE and COD removal of 621.13 mW m^{-2}, 37.16%, 90.46%, respectively.

2.2.4.1. Cheese whey. Treatment of cheese whey in MFC has also been examined by several researchers [165–168]. The liquid fraction after precipitation and removal of milk casein is cheese whey and it is characterized by 4–5% carbohydrates, proteins up to 1%, fats at about 0.4–0.5%, lactic acid less than 1% and salts 1–3% [169]. Using cheese whey, one-month-old pre-incubated anodes in MFC showed 0.13% CE, 88.3% COD removal efficiency and P_{dmax} was 29.1 mW m^{-2}, whereas the three-month-old pre-incubated anode showed 80.9% CE, 92.8% COD removal, and 1800 mW m^{-2} P_{dmax} [170]. In another report, two-chambered MFC using 6.7 g COD l^{-1} filter sterilized cheese whey yielded a P_{dmax} of 46 mW m^{-2} approximately with over 94% COD removal and about 11.3% CE [99].

2.2.4.2. Yoghurt wastewater. Yoghurt wastewater was also examined as suitable fuel using anaerobic sludge and compost leachate as inoculum sources. A P_{dmax} of 54 mW m^{-2} resulted from sludge MFCs while 37 mW m^{-2} from compost leachate MFCs [110].

2.2.5. Agro processing industry wastewater

2.2.5.1. Rice milling industry wastewater. With rice milling industry wastewater maximum sustainable volumetric P_d and COD removal of 2.3 W m^{-3} and 96.5% respectively were reported using earthen pot MFC [124]. Hassan et al. [171] used rice straw without pretreatment and inoculated with a mixed culture of cellulose-degrading bacteria in MFC. At an initial concentration of 1 g l^{-1}, the P_d reached 145 mW m^{-2} and corresponding CE of 45.3%. Increased P_{dmax} of 190 mW m^{-2} was reported recently using same concentration of rice straw feed by other group [125].

2.2.5.2. Cassava mill wastewater. About 0.2 ton of starch, 0.4–0.9 ton of residue, and 5–7 l of wastewater is produced in the starch production process from a ton of fresh cassava root [172,173]. Cassava mill wastewater is a carbohydrate-rich starch waste having high COD, BOD, total solids, and low ammonium–nitrogen concentrations [174]. It contains large amounts of natural cyanoglycosides which can be enzymatically hydrolyzed to cyanide. Cyanide concentration has been reported as high as 200 mg l^{-1} depending on cassava varieties [175]. Presence of varying amounts of cyanogenic glucosides and their breakdown products found to be higher than the safe levels for agriculture and other purposes and is cause of concern for consequential potential deleterious effects [176]. Where large scale cassava processing activities occur, it should be subjected to treatment before being discharged to surroundings. The biological treatment processes can be disrupted and inhibited due to toxic nature of cyanide [177]. The results of MFC experiments with full strength cyanide laden wastewater (16,000 mg COD l^{-1} as wastewater strength, 86 mg l^{-1} in terms of cyanide content) and mixed culture sludge inoculum showed that the MFCs could generate a P_{dmax} of 1771 mW m^{-2} [97]. With raw Sago-processing wastewater (11,328 mg COD l^{-1}), the P_d of 300 mW m^{-2} (4500 mW m^{-3}) has been reported in MFCs [178].

2.2.5.3. Palm oil mill effluent. Palm oil mill effluent (POME) is characterized by complex substrates comprising of amino acids, inorganic nutrients such as sodium, potassium, calcium, magnesium, short fibers, organelles, nitrogenous constituents, free organic acids, and a mixture of carbohydrates ranging from hemicelluloses to simple sugars [179]. More than 2.5 tons of POME can be produced in 1 ton crude palm oil production process [180]. Typically, the COD and BOD are up to 50,000 mg $l^{-1}$and 25,000 mg l^{-1}, respectively. It also has a high acidic content which causes environmental concern [181]. Economically viable technological solutions such as simple skimming devices [182], land disposal [183], chemical coagulation and flotation [184], aerobic [185] and anaerobic biological processes [186] are used for POME treatment.

However, these treatment methods are highly energy consuming [187]. Furthermore, due to low reaction rates and high solubility of produced methane, anaerobic digestion generally fails [188,189]. A double-chamber MFC has been tried to treat raw POME inoculated with anaerobic sludge. It showed the P_{dmax} and volumetric P_d of about 45 mW m^{-2} and 304 mW m^{-3} respectively, but low CE and COD removal efficiency of about 0.8% and 45% respectively were obtained [190]. Earlier, another group examined POME treatment using a two-stage MFC system integrated with immobilized biological aerated filters. It resulted in P_{dmax} of about 44.6 mW m^{-2} and more than 90% of total COD removal rate [191]. Baranitharan et al. reported the P_{dmax} of 107.35 mW m^{-2} with POME feed [192]. The corresponding CE and COD removal efficiency of 74% and about 32% respectively were obtained with polyacrylonitrile carbon felt anode and controlled inoculum in this study. Using *P. aeruginosa* strain ZH1 as an inoculum resulted in P_{dmax} of 451.26 mW m^{-2} which was five times higher compared to that of MFC using anaerobic POME sludge inoculum [117].

2.2.5.4. Mustard tuber wastewater. One of the three most famous pickles worldwide is Fuling mustard tuber. During its production process, high-strength and high-salinity containing mustard tuber wastewater (MTWW) is discharged in large volumes [193]. Anaerobic process followed by aerobic and/or physicochemical technology are the costly methods used currently to treat this effluent [193]. At a current density of 50 mA cm^{-2}, a dilution ratio of 1:2, without pH adjustment and 240 min of processing, electrochemical oxidation of MTWW resulted in 80.4% COD removal and the corresponding specific energy consumption was 45.8 kW h m^{-3}, despite its usual resistance to biological treatment [194]. Guo et al. reported using dual-chamber MFCs a P_{dmax} of 246 mW m^{-2} (6.6 W m^{-3}), 67% CE and 85% COD removal with MTWW [116]. It also generated P_{dmax} of 1.32 W m^{-3} and adequately self-buffered when used as catholyte with mixed-species biocathodes in MFCs [195].

2.2.6. Livestock industry wastewater

2.2.6.1. Meat industry wastewater. Slaughterhouse wastewater is characterized by high-strength wastewater, mainly constituted of biodegradable organic materials. The possibility of using slaughterhouse wastewater in MFCs has been reported [83,130]. For instance, slaughterhouse wastewater in dual chambered MFC resulted in P_{dmax} of 578 mW m^{-2} [130].

2.2.6.2. Animal carcass wastewater. Alkaline hydrolysis is a treatment and disposal method in which animal carbohydrates, lipids, proteins, nucleic acids, as well as any pathogenic microorganisms, including viruses are converted into a sterile solution composed of amino acids, small peptides, sugars, and soap, along with the minerals [196]. This sterile solution i.e. animal carcass wastewater is coffee-colored alkaline solution characterized by high amounts of BOD (70 g l^{-1}), COD (105 g l^{-1}), ammonia (1 g l^{-1}), organic nitrogen (8 g l^{-1}) and total phosphorus (0.4 g l^{-1}) [197]. Bioelectricity generation with animal carcass wastewater was evaluated using upflow tubular air cathode MFCs. It resulted in P_{dmax} of 2.19 W m^{-3} with 50.66% COD removal [92].

2.2.6.3. Swine wastewater. In livestock industry, swine wastewater treatment is important for sustainable animal production [198]. In methane gas generation during anaerobic treatment, ammonia, and odor producing chemicals removal is a cause of concern [199]. Volatile organic acids are the main chemicals in generation of nuisance odors and are controlled by stimulating dissimilatory iron reduction [200]. Integrated real-time control strategy for nitrogen removal in swine wastewater treatment using sequencing batch reactors was demonstrated by Kim et al. [201]. Preliminary tests using swine wastewater in aqueous cathode dual chambered MFC showed P_d of 45 mW m^{-2} [133]. In addition, more extensive tests with single-chambered air cathode MFC produced a P_{dmax} of 261 mW m^{-2} [202], 228 mW m^{-2} [133] and 382 mW m^{-2} [203]. The use of alkaline-thermal pre-treated sludge as a bioflocculant and its application in swine wastewater pre-treatment is promising because the bioflocculant could reduce COD, ammonium and turbidity by several orders of magnitude [204–206]. Use of this pretreated swine wastewater seems to be suitable in enhancing MFC efficacy and thus can open a door for novel interlinking possibility for enhanced resource recovery, energy recovery and swine wastewater treatment.

2.2.7. Refinery and distillery industry wastewater

2.2.7.1. Biorefinery wastewater. In usual manufacturing process, biodiesel is manufactured through transesterification of lipids with alcohol [207]. Acyl groups of triglycerides produce 1 mol of glycerin for every three moles of ester [208]. During biorefinery process typically 4–10 times more water is utilized than the amount of biofuel generated. Biorefinery wastewater is characterized by residual sugars, 5-furfural, phenolics, and other pretreatment and fermentation byproducts. Post-fermentation biorefinery stream containing phenolic compounds and furan aldehyde derivatives conversion has been tried as substrate in MFCs. Using biocathode MFCs, electricity generation from glycerin-containing biodiesel side stream achieved maximum P_d of 23 W m^{-3} [209]. In another study, P_d of 2110 mW m^{-2} (cathode surface area) with biodiesel waste blended with 200 mM PBS with the heat-treated carbon brush anode has been reported [93].

2.2.7.2. Corn stover neutral and acid steam-exploded liquid hydrolysate fraction. Neutral and acid steam-exploded (hydrolysis processes that convert the hemicellulose to soluble sugars) corn stover waste biomass contains 70% cellulose and hemicellulose, and 15–20% lignin [210]. Xylose is among the major constituents of corn stover acid hydrolysates [60]. Hydrogen can be produced from sugar-enriched liquid hydrolysate fraction but most of the COD remains as fermentation end products consisting primarily of acetic and butyric acids. Energy intensive neutral and acid steam-explosion converts <47% of carbohydrates into ethanol [211]. By using MFCs, direct generation of electricity up to 371 mW m^{-2} was demonstrated in fed-batch test at 1000 mg COD l^{-1} and further up to 971 mW m^{-2} with acid hydrolysate by increasing solution conductivity and using a cathode containing diffusion layer [103]. The single chamber, air–cathode MFCs fed with such wastewater and inoculated with mixed culture inoculum produced maximum power of 331 mW m^{-2}. The use of residual solids from the steam exploded corn stover produced 8% more P_d (406 mW m^{-2}) than the raw corn stover [212]. Fermented corn stover hydrolysate produced P_{dmax} of 1180 mW m^{-2} at 5300 mg l^{-1} (8% dilution) of waste [111].

2.2.7.3. Ethanol stillage wastewater. Ethanol stillage wastewater contains lignin, which is good component to suppress methanogens so it is also a good source for bioelectricity production. Sakdaronnarong et al. [109] achieved using ethanol stillage wastewater, the P_d of 93 W m^{-2} with 81.5% COD removal.

2.2.7.4. Molasses wastewater. Zhang et al. [115] tested molasses wastewater, which is by-product from sugar beet process and a raw material for ethanol production, and reported P_{dmax} 1410.2 mW m^{-2}.

2.2.8. Mining and allied industry wastewater

Acid-mine drainage (AMD) is caused by biological oxidation of metal sulfides to metal sulfates. AMD is characterized by low pH and solubilized metals such as lead, copper, cadmium, and arsenic causing serious threat to aquatic environment. AMD treatment using MFC technology was investigated using an AMD fuel cell which generated a P_{dmax} of 290 mW m^{-2} with CE of more than 97%. These results also suggested that not only electricity production, but also removal of metals, production of useful products, and recovery of metals from AMD are possible using MFC [134]. In investigation of gold recovery from tetrachloroaurate wastewater, Choi and Hu [11] achieved 6.58 mW m^{-2}. Other wastes such as coal tar [100] and coking [101] wastewaters generated P_{dmax} of 4.5 and 538 mW m^{-2}, respectively in MFCs .

2.2.9. Pharmaceutical industry wastewater

2.2.9.1. Synthetic penicillin wastewater. Synthetic penicillin wastewater in an air–cathode single chamber MFC was evaluated by Wen et al. [135]. 1 g l^{-1} glucose + 50 mg l^{-1} penicillin resulted in P_{dmax} 101.2 W m^{-3} which was 6-fold higher than the sum of that for 1 g l^{-1} glucose (14.7 W m^{-3}) and 50 mg l^{-1} penicillin (2.1 W m^{-3}) as the sole fuel. These results indicated that toxic and bio-refractory organic matter containing wastes such as antibiotic wastewater might also be a good resource for MFC technology.

2.2.9.2. Recalcitrant pharmaceutical industrial effluent. Recalcitrant pharmaceutical industrial effluent is characterized by complex composition and high toxicity. In MFCs, it resulted in P_d of 205.61 mW m^{-2} at organic load of 7.98 kg COD l^{-1} [123]. Recently, paracetamol containing toxic effluent has been reported to produce P_{dmax} of 217.27 mW m^{-2} [213].

2.2.9.3. Steroidal drug production wastewater. Hydrocortisone production is an important intermediate of steroidal drug production. At the acid hydrolysis step, a large amount of highly toxic refining wastewater is produced [214,215]. Such steroidal drug production wastewater has been investigated in MFCs and resulted in P_{dmax}, CE, maximum COD removal efficiency of 22.3 W m^{-3}, 30% and 82% respectively [132].

2.2.10. Paper recycling industry wastewater

Wastewater from paper industries contains soluble organics and particulate matter such as cellulose, which can be ineffectively treated with traditional wastewater technologies. Sustainable agriculture and bio-based industries have led to use of efficient method for treating cellulose-containing wastewater. In MFCs, treatment efficiency of such wastewater was limited by its conductivity [216]. Evaluation of full-strength paper mill effluent for electricity generation in mediator-less MFC resulted in P_{dmax} of 24 mW m^{-2} [217]. To overcome this problem, different buffers were tried. 50% PBS reached P_d of 501 mW m^{-2}, CE of 16%, and total COD removal of 76%. Cellulose removal was 96% [216]. Higher power densities, for instance P_{dmax} of 1070 mW m^{-2} (cathode surface area) in single-chamber and 880 mW m^{-2} in two-chamber air–cathode MFCs with CE up to 50% and COD removal up to 70% [98] have been reported with cellulose which is a by-product of the paper manufacturing industry.

2.2.11. Textile industry wastewater

Textile industry produces one of the most complex industrial wastewater. Dye wastewater amounting to more than 7×10^5 tones per year [218] contains recalcitrant organic molecules [219]. Along with recalcitrant organics, toxic, mutagenic or carcinogenic chemicals [220] usually characterize the dye effluent. The 60% of the total dyes manufactured are azo dyes [221]. Usually, for treatment of this recalcitrant wastewater physical or chemical and electrochemical methods are used [222]. Development of toxic intermediates, lower removal efficiency, and higher specificity for a group of dyes are some of the operational limitations of these methods [223]. Concerning these issues, MFCs have been tried to treat such wastewater. For instance, granular activated carbon based single-chamber MFC produced a P_d of 8 W m^{-3} [121]. In earlier study, granular activated carbon based MFC produced a P_d of 1.7 W m^{-3}. In this case, the COD removal was 71% at the anode and 76% at the cathode with almost nontoxic cathode effluent and threefold less toxic anode effluent within 48 h operation [224]. Fang et al. [225] demonstrated electricity production from azo dye wastewater using a MFC coupled with constructed wetland (CW-MFC), an adapted device to treat the wastewater and produce energy which has more wastewater treatment volume and is easier to maintain than other MFCs. The highest P_d reached in this case was 0.852 W m^{-3}.

2.2.12. Petrochemical industry wastewater

Various investigations have been carried out for the possible application of MFC technology in the effective treatment of petroleum hydrocarbons contaminated sites and refinery effluents. For instance, real-field petroleum sludge has been reported as an electron donor leading to power generation of 53.11 mW m^{-2} [226]. Around 31 mW m^{-2} (cathode surface area) P_d was generated during diesel degradation in the anode compartment of dual chambered MFC [227]. In a sediment MFC containing total petroleum hydrocarbons concentration at approximately 16 g kg^{-1}, up to 24% degradation and 2162 mW m^{-3} power has been reported [228].

Purified terephthalic acid (PTA) is a raw material for petrochemical products manufacturing with a high strength in organic materials. Generation of 1 ton of PTA produces 3–10 m^3 wastewater [229]. Foad Marashi et al. [120] for the first time examined the raw wastewater of PTA from a petrochemical plant in a membraneless single chamber MFC. They achieved 31.8 mW m^{-2} (normalized per cathode area) and CE of 2.05% for a COD removal of 74% during 21 days at acidic pH (4.45) while higher P_d (65.6 mW m^{-2}) was achieved under alkaline condition (pH 8.5) [230].

2.2.13. Domestic and municipal wastewater

Energy recovery from domestic wastewater is one of the major subjects of interest among researchers. A decade ago, domestic wastewater was evaluated as substrate using single-chambered MFCs [85,231]. For example, using domestic wastewater (initial COD of 200–300 mg l^{-1}), P_{dmax} up to 26 mW m^{-2} was achieved while up to 80% COD was removed [232]. Air–cathode MFCs fed with 345 mg COD l^{-1} domestic wastewater produced 22.5 W h m^{-3} corresponding CE of 18% and 83% COD removal [139]. Jiang et al. [107] evaluated effectiveness of an upflow membrane-less MFC and a photobioreactor coupled system for continuous wastewater treatment and electricity production. Alone, the upflow MFC produced a P_{dmax} of 481 mW m^{-3}, and achieved 77.9% COD, 23.5% total phosphorus, and 97.6% $NH_4 \pm N$ removal. When combined with the photobioreactor, the system achieved 99.3% total phosphorous and 99.0% total $NH_4 \pm N$ removal. At substrate concentration of 54 g COD l^{-1} d^{-1}, the highest P_d of 422 mW m^{-2} (12.8 W m^{-3}) was produced while achieving 25.8% COD removal [233]. Lee and Nirmalakhandan [234] recorded P_{dmax} of 36.6 mW m^{-2} and 67 mW m^{-2} when digested anaerobic sludge was used as the seed in a single compartment combined membrane-electrodes (SCME) design MFC, and hydrogen-generating bacteria were used as the seed in the twin compartment brush-type anode electrodes design MFC, respectively by using livestock organic solid waste as fuel. P_{dmax} of 6.73 mW m^{-2} (13.65 mW m^{-3}) was obtained in a membrane-less MFC during

actual sewage treatment inoculated with pre-heated mixed anaerobic sludge with successful removal of organics [129]. Sciarria et al. [235] with the mixture of domestic and olive mill wastewater (in ratio 14:1, w/w) obtained comparatively improved P_{dmax} (124.6 mW m^{-2}) than domestic wastewater alone. Fly ash leachate (the residue from incineration of municipal solid waste (MSW)) has been also investigated as substrate in dual chambered MFC and resulted in P_{dmax} of 80 mW m^{-2} (cathode surface area) [236].

Ammonium recovery and simultaneous electricity generation from urine was proven possible by Kuntke et al. [237] in MFCs with gas diffusion cathode. An ammonium recovery rate of 3.29 g N d^{-1} m^{-2} (vs. membrane surface area) was achieved at a current density of 0.50 A m^{-2} (vs. membrane surface area). Single compartment MFCs treating raw human urine were extensively investigated over 45-days operation as a cost-effective process for power production, contaminant removal, and nutrient recovery [113]. Such MFCs effectively degraded COD in urine, with the COD removal efficiency of 25–40% in 1-day cycles, 35–60% in the 2-day cycles, and 60–75% in 4-day cycles.

The feasibility of human feces wastewater for electricity generation in MFC was investigated by Fangzhou and coworkers [114]. With this waste P_d of 70.8 mW m^{-2} and 71% COD removal was achieved.

3. Existing challenges concerning the use of wastewaters as substrates in MFCs

Rapid evolution of MFC technology on many fronts, including reactor designs, selection of materials, renewable substrate utilization, and selection and understanding of biocatalyst has brought it much closer to realize its full potential and application for bioenergy production and simultaneous wastewater treatment. Until a few years ago, the technology was considered far from commercialization but recently some commercial prototype MFCs have appeared in the market. EcoVolt from Cambrian Innovation is one such wastewater treatment system based on the MFC technology (http://cambrianinnovation.com/solutions/ecovolt/). Other companies such as Emefcy [238] and Pilus Energy (http://www.pilusenergy.com/) have also planned for launching a commercial MFC based wastewater treatment system. Although power densities from MFCs have risen by several orders of magnitude over the past decade, many challenges as discussed further below still remain. Not only limitations and problems of electric current outputs, but also the selection of suitable wastewater in terms of molecular complexity and resistance (due to low conductivity) is still a major challenge. Adding external buffering systems is not the sustainable way to increase the conductivity of wastewaters. Wastewater concentration and its composition are among the most important factors that affect the COD removal and P_d in MFCs. Unwanted biomass growth, incomplete biodegradation of the substrates, hydrogen production, methanogenesis, aerobic degradation processes, and neutral metabolites diffusing to the cathode chamber are some of the factors that affect COD removal efficacy in MFCs. Microbial activity is slower in pH less or more than the optimum pH, therefore, consideration of pH and its changes during operation is among important factor in selection of substrate. However, the low current intensity around the optimum pH may also be due to poor transfer of protons thus limiting cathode kinetics. Thorough chemical and microbial analysis of wastewater can help in understanding the chemical and microbial reactions that happen in the anode and the cathode compartments. The use of mixed cultures that utilize complex substrates is favorable for MFCs. Understanding their ecology and influencing factors affecting their acclimation can help to achieve desirable augmentation in COD removal and P_d.

CE gives an indication of the proportion of electrons from substrate (organic matter) that is recovered as electricity. Low CE has been a general issue in the MFC systems fed with real or complex wastewaters. Several factors can be associated to achieving low CE in MFCs. First factor is the diversion of electrons into non-exoelectrogenic biomass growth. Substrate consumption via other competing metabolic processes, like fermentation, methanogenesis or the metabolic inhibition of the exoelectrogens due to a build-up of acid or other toxic by-products from microbial activity is a second reason behind low CE. Third, large number of electrons are not available since the electrons are locked in substrates by other electron acceptors. Fourth reason is low electron transfer efficacy due to such limitations associated with electrodes as the difficulty of accessibility of electrons to anode or cathode (for instance, due to deposition of interfering or insulating substances on electrodes). Separators can also reduce CE, for instance, lower thickness favored diffusion of electrolyte can result in reduced CE. Finally, biocatalyst grazing can also affect the CE in MFCs. All these aspects need to be addressed in order to increase the CE of wastewater treating MFC systems.

Inadequate amount of bioelectricity in terms of voltage and P_d generated is a major concern in scaling-up of MFC technology. Scaling up challenges, electrodes, membranes, maintaining a good biofilm etc. are well-discussed existing challenges which have somewhat improved in recent years. Thus far, most of the research has been done with the lab scale reactors. One of the practical difficulties faced with scale-up is that the surface area increases with increase in power output only when a high internal resistance does not limit the system [239]. For membrane-less MFCs, it is still not clear whether the larger-scale systems can produce power densities that are comparable to the laboratory scale systems [240]. The performance of electrode materials limits due to fouling of their active surfaces, loss of activity, corrosion and other degradation mechanisms. Influence of electrode surface chemistry or biointerfaces, electrode spacing and packing per unit volume of the reactor are some important factors that need thorough consideration in scaling up of MFCs [241,242,240]. Furthermore, lack of in-depth knowledge on diverse and undefined community of microbes and their electrogenic pathways is also a major biotechnological concern in MFC research. To produce higher power outputs that can be used to drive an electronic device using MFC technology, substantial advancements in these directions are also needed [243].

The high cost of the MFC components to support suitable energy production and scalability has been another major concern during the last decade and still remains a matter of a great concern among researchers. New materials for MFCs are being constantly explored and developed to improve their economic feasibility and performance. For instance, a wide variety of materials such as carbonaceous, metal and metal oxides, and composite materials as well as configurations such as 2D and 3D [29,36,244] have been studied as the anodes. The cathode is a major performance determining component of the MFCs. The cathodic reaction capacity can be enhanced by using platinum-group metals (PGMs) but the cost is a major concern in using Pt based cathodes. Among novel cathodic materials, carbon nanofibers are regarded as one of the most promising alternatives [245]. Nitrogen-doped graphene, a newly emerging nitrogen-doped oxygen reducing catalyst synthesized by chemical route, has also been reported to be a low cost alternative to PGMs. Recently, the stable power generation with nitrogen-doped graphene cathode based MFCs was demonstrated [246]. Ma et al. [247] demonstrated that graphitic carbon and silver/iron oxide composite material based catalyst can be a low cost alternative to platinum/carbon. These studies demonstrate the possibility of using graphene based cathodes for potential large-scale application of MFCs. For more details on issues such as new

materials used in MFC (cathode, anode, and membrane/separator), types of configurations and MFC modeling, readers are directed to a recent review by Hernández-Fernández et al. [248]. In addition, other issues such as membrane fouling, electrode blockage, life of electrode and robustness with changing environment and wastewater composition need to be improved for longevity and stability of the MFCs treating wastewaters. Longer operational times required for the treatment and longer start up times with low power output with high COD containing wastewaters in MFCs are other concerns in the commercialization of the technology. The requirement of number of MFC units to power a small DC pump efficiently is a limitation [249]. Several hundreds of MFC are needed to power pump so efficiency of MFCs in terms of reasonability of energy management is still questionable [250]. Recent study demonstrated that MFC scale-up may be better achieved by stacking of miniaturized MFCs rather than increasing the size of an individual unit [251]. Although the low voltage provided directly from MFCs is not enough to power an electrical device, Lobo and coworker's study shows that MFC stacks may be able to do so without significant energy loss [252]. Reduction of internal resistance of systems at least by an order of magnitude is necessary to power larger AC pumps by MFCs. In addition, transformers can be coupled at the output of the DC–AC converter to boost the voltage to a usable level. However, many associated scaling-up challenges, such as potential voltage reversal in MFC stacks and more efficient energy harvesting from MFC reactors still remain unresolved. It is demonstrated by Ledezma et al. [253] that substrate imbalances and starvation do not necessarily result in voltage reversal. MFC-cascade stacks can maximize COD reduction and avoid voltage reversal under adverse conditions. The issues associated with the stacking of MFCs need to be resolved further.

As far as improving biofilm activities at anodes are concerned several strategies have been proposed by researchers. An interesting method was proposed recently by Liao et al. [254] to improve the power output of MFCs by rotating the carbon-brush anode. They reported based on Tafel plots analysis that the rotating anode can improve the electrochemical activity of the biofilm. The enhancement of endogenously produced biosurfactant by genetic engineering is one of several alternative strategies proposed to improve MFC performance by improving the extracellular electron transfer rate and energy extraction efficacy [255]. Holmes et al. [256] reported recently that eukaryotic grazing can be a major issue related to low power output in sediment MFCs. Potential of these recent research findings with regard to the scale-up challenges need to be further explored. Although studies over the past few decades have investigated the role of microbial communities at anodes, the involvement of microbial communities at cathodes has come to the fore only since the past few years [257]. This provides an opportunity to use both bioanode and biocathode for wastewater degradation and electrosynthesis processes [258–260] in BESs leading to simultaneous bioremediation, resource recovery, and energy harvesting applications.

4. Conclusions and future perspectives

Based on the extensive review of existing literature, this article presented an overview of recent advances in the use of a wide range of wastewaters as potential substrates in MFCs. Substrates rich in mixture of carbon sources – glucuronic acid, galactitol, ribitol, xylose, glucose, galactose, gluconic acid, xylitol, arabitol etc.; nitrogen sources – serine, histidine, arginine, etc., and organic acids – acetic acid, lactic acid, etc. have been proven to show a better performance in terms of power output. Ethanol stillage, cassava mill, chocolate industry, molasses, biodiesel, cellulose wastewaters have shown efficient capacity for bioelectricity production. In particular, agricultural, brewery, domestic, and food wastewaters are generated throughout the world in huge amounts. So, they offer potential feedstocks for fueling MFCs. Among these, distillery industry along with agro processing industry wastewaters have better efficiency because of presence of methanogenic inhibitors and natural presence of electron transferring mediators like lignin in them. Thus substrates derived from lignocellulosic biomass and hydrolysates seem to be good choice as feed for MFCs. Food and dairy industry wastewaters are also good but are limited by the presence of other electron acceptors and of non-exoelectrogenic microorganisms such as fermenters. Animal processing industry associated wastewaters particularly from slaughterhouse and swine yards have good efficacy because of the presence of blood, which is a good substrate for microorganisms, as a major constituent in these wastes. However, further research is needed in order to enhance the power outputs on several accounts such as pretreatment and bioaugmentation with such nutrient rich wastewaters as these have a huge potential as substrates in MFCs.

The consideration of factors such as the selection of wastewater and its ionic strength are important for achieving maximum power outputs in MFCs. Operational conditions such as flow mode, temperature, pH, organic loading rate, *hydraulic retention time*, microbial selection and colonization efficacy, internal resistance etc. plays an important role in MFCs treating wastewaters. Finding cost effective and sustainable alternatives to proton or cation exchange membranes should also be focus of the further research efforts. Even after extensive research efforts it is very difficult to identify the most promising MFC designs, electrodes, substrates, and biocatalysts because of inconsistency in research outputs and data representation. Therefore consistency in uniform data reporting and suitable conversion formulas for comparisons is also necessary to further the research and development of MFC at larger scales.

Technological improvements for reducing consumption of energy for wastewater treatment and suitable mathematical model will also be required for MFC optimization. Optimization of the process of microbial energy harvesting and substrate utilization efficiency need to be considered. Optimum utilization of substrates by microbes will lead to maximization of the fuel cell performance. There is very limited comparative literature for simple substrates. Further extensive research is needed on simple or defined substrates and its influence on electrogenic microbes; to understand and optimize the substrates, which are complex in nature. Understanding internal resistance and losses and overcoming them might help to enhance the MFC performance in near future. Another most important area where the research needs to focus on is the understanding of the exoelectrogens in particular in mixed cultures. One of the major limitations associated with mixed culture is overgrowth of non-electrogens in biofilms on electrodes. Eukaryotic grazing of exoelectrogens is another issue of consideration. Understanding these microbial aspects will help to plan some decisive strategies for bioengineering or biotechnological improvements in MFCs. Computer simulation prototype MFC startups are needed to be developed to achieve complete real time monitoring including wastewater composition; operational conditions; population dynamics, cell–cell communication, molecule–surface interactions and gene-expression profiles of electrode-associated biofilms. Interlinking possibilities of MFCs with other technologies should also be considered to enhance the chances of their integration into existing wastewater treatment plants and thus to advance the prospects of MFCs for practical applications.

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