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# Comprehensive Review on the Use of Food Industry Wastewater as Substrates in Microbial Fuel Cells



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#### Abstract

Today, the world is facing climate change challenges with environmental protection being a top priority. Optimizing energy consumption due to its high cost and environment protection is a basic human demand. For industries, reduction in production costs is determinative to success. In this regard, Microbial fuel cell (MFC) is a unique promising technology with wastewater treatment and bioelectricity generation. The MFCs will help reduce energy consumption, curb the wastewater pollution, and standardize it for releasing into the environment. The food industry by producing high volumes of biomass with high organic pollution load are highly prone to use in MFCs as a substrate. Various food industry effluents have been tested, in real or synthetic form in the MFCs. Due to the improvements in the process and progress in novel configurations, better results have been increasingly obtained. Now, the MFC can be used in the industries individually or by integration with other technologies. In this review, the latest results from the use of food industry wastewater in MFCs along with effective process conditions are evaluated.

Keywords: Wastewater treatment; Food industry; Substrate; Power density; COD removal; Bio-energy

#### **Highlights**

- a) Using real food industries wastewater as a substrate in MFC are evaluated.
- b) Main wastewater components like pure synthetic wastewater are presented.
- c) Municipal wastewater performance in MFCs is described.
- d) Effect of wastewater sub-components in MFCs are investigated.

# Introduction

Biotechnology is continuously innovating for creating and developing good ecosystems, transformation of pollutions, development of biodegradable material, development and modification of production processes and safe environmental disposal. In this regard, the waste management is one of the influential subjects of sustainable development. Biomass is the fourth biggest energy source worldwide and provides 14% of the world's energy. Extraction of bioenergy from biomass is novel area which has remained on a research scale. Turning biomass into bioenergy can be done through different biological methods. Bioelectrochemical (BE) systems are a promising technology through the Microbial Fuel Cells (MFC) whose major role is electricity generation through directly organic compounds oxidation.

MFC is a novel approach for the wastewater treatment and energy production simultaneously. Hence, two fundamental challenges of today's world can be addressed: available water and

energy. The organic compounds in the wastewater are converted electricity through the electrogenic microorganisms. The main idea of using microbes to generate electricity was first introduced by Potter in 1911 [1]. Since then, more concepts and practical improvements have been achieved through the foundation of Cohen's 35-unit in 1931 [2]. Karube et al. [3] catalyst-related research began in the 1960s [3], followed by further work by Bennetto et al. [4] on kinetic mediators in the 80s and 90s, leading to the development of the term "analytical MFC" which is still in use. At the end of 1990s, a series of discoveries showed that some special microorganisms can be used directly in fuel cells for the purpose of energy production without any involvement in hydrogen mediated procedures [5]. Approximately, through usual methods, more than 10 times energy in the form of carbon (C) compounds is needed to the organic substrate treatment in wastewater. On average, 1kg of carbohydrates shows COD of 1.06kg which can be converted into 4.41kw h<sup>-1</sup> or 13\*10<sup>6</sup> coulombs [6]. In the past two decades, there have been extensive developments

in MFC technology including cost investment reduction, configuration improvement through novel, cheap and stable materials, comprehension of electron transferring mechanisms and their facilities, high-efficiency bio-electrocatalytic interfaces, substrates, microbial communities, biofilms, integrating with other technologies, bottlenecks and challenges identification, large-scale efficiency. With MFC efficiency improvement, the current density (Cd) of MFCs' is close to suitable level for practical operations.

Substrate plays an important role in the biological processes in providing the C source requirements [7,8]. he density of substrates and their chemical structure especially in real wastewater can affect the microbial population along with overall performance of MFC [8]. So far, many studies have been performed focusing on the variety of substrates and wastewaters. At the first step, researchers have focused on using a simple and pure substrate (C source) along with pure microbial culture followed by synthetic and real wastewater with different settings of MFC to examine the various aspects of MFCs. The possibility of electricity generation from complex substrates is a great achievement. The cheap cost of food industrial wastewaters along with biological benefits can make them a suitable choice for biological treatment. The food industry wastewaters mainly contain monomers of high molecular mass complex compounds of carbohydrates (the highest proportion), fatty acids, amino acids, nitrogen (N) compounds, sulfur (S) compounds, and phosphorus (P) compounds. The heavy metals can be effectively removed or recovered using MFC [9,10]. Also many studies have indicated the MFC capability for decreasing some azo dyes as a kind of contamination in dyed wastewater [11]. Based on the interaction between microbes and electrode, different systems have been developed that not only remove organic compounds, but also produce valuable materials from wastewater such as bio-flocculants, bio-plastics, biosurfactants, hydrogen, methane and many other compounds [12]. Nevertheless, a complex wastewater, which might cause problems for bacterial activity with a low conductivity, will be a major challenge in this path. Sometimes for efficiency improvement or removing obstacles in substrate availability especially for food industry wastewater, pre-treatment is required. For example, energy extraction from lignocellulosic based biomass has received much attention due to its accessibility and usability, as well as abundance [13].

MFCs are high potential technology with a great advantage of simultaneous wastewater treatment and bioenergy generation. This leads to changes in waste management, development of resource recovery technologies and development of diversified and efficiently energy. The MFC can be a special and attractive occasion for food industries. Now, given the high cost of energy for the food industries wastewater treatment around the world and the many challenges of standardizing wastewater for releasing, which is itself high cost and energy-intensive, MFCs can attract the attention of industries to find their place. As such, the MFCs

can reduce the costs of production in industries. The energy generated can be used as part of the energy requirement for the production process, treatment process or other parts of the factory installations. This study has been conducted to help greatly the major steps of researchers towards the treatment efficiency improvement and the perfect industries for implementing by identification of wastewater and similar backgrounds. In mentioning the literature, the effort is not to merely report the figures and findings briefly, but to elaborate the important topics and special differences to provide better conclusion and comparison. Using these results, the food industries should be encouraged to invest widely in this field and help expand renewable energy, protect the environment, and reduce their own costs. It is hoped that this technology would be able to rapidly find its place in wastewater treatment plants or food industries.

# **Main Components of Wastewater**

In general, most of the food industry wastewaters are based on C, N, S and P. Specifically, the C often constitutes the majority of pollutants, based on the nature of industrial activity, followed by other compounds. The behavior of these compounds in the MFCs as major components should be thoroughly investigated.

#### Carbon resource

C is one of the most significant organic compounds in food industrial wastewater. It provides favorable conditions for biological treatment for electricity generation. C exists in the wastewater in various forms. One of the main components of food industry is carbohydrates. It is clear that all monosaccharides of acidic hydrolysis of lignocellulosic material in the hydrolysis process can be used to generate electricity [14]. Some performances of MFCs with C resource as substrate are presented in Table 1. In this regard, the power was successfully generated from six hexoses, pentoses and sugar derivatives in an aircathode SC-MFC by a mixed culture and more than 80% reduction was achieved in COD [15]. Even so, with glucose loading rate of 1000mg l<sup>-1</sup> d<sup>-1</sup>, power density maximum (Pdm) of 4310mW m<sup>-2</sup> and CE of 81% were obtained in a DC-MFC [16]. Short chain volatile fatty acids can produce more electricity than long chain volatile fatty acids, due to their faster degradation than long chains. In this regard, acetate and propionate were the preferred options [17]. Regarding ethylene glycol, it was found that ethylene glycol did not have a negative effect even at high concentrations (2000ppm as the only C source). Of course, we could improve the MFC performance when ethylene glycol is used at the same concentration with glucose [18]. A continuous two-stage process with synthetic kitchen food waste and adding domestic wastewater as a balanced diluent was designed which was with hydrogen and volatile fatty acid generation in the first stage and electricity generation by MFC in the second stage. The MFC was operated using a volatile fatty acids-rich effluent from first stage that specifically contained acetic acid. The combined system could reduce the COD load by 90%. The concentration of volatile fatty

acids showed a descending trend over time. The CE of 46% and the Cd of  $65.33 \text{mA} \text{ m}^{-2}$  were obtained with a voltage of 148 mV [19]. The appropriate effect of MFC on volatile fatty acids reduction was indicated. The methanol is not considered as a substrate and it is not applicable in this technology, but ethanol can be used [20]. The environmental and toxic effects of alcoholic sugars are still vague on other living organisms. Poly-alcohols cannot be completely metabolized and used by humans but can be used by MFC. As a byproduct in the ethanol fermentation process, they were used with 92% Pd obtained within the range 1490-2650 mW

m<sup>-2</sup>, with the following order galectitule> ribitol> xylitol> arbitol> sorbitol> mannitol respectively, and COD elimination was 71-92% [21]. The treatment of recalcitrant C source contaminant is very important. Hence, there are some ways for treatment performance improvement. In this regard, by engineering microbial consortia and using fermenter-exoelectrogen like Klebsiella pneumoniae can create a good situation in converting glycerol to lactate. Afterward, lactate can used by S. oneidensis as a C source to generate power [22].

Table 1: MFCs Performance with Carbon Resources.

NO	Substrate	MFC Type	Inoculum	Anode	Cathode	Voltage (mV)	Pd (mW m²)	COD Re- moval%	CE%	Ref
1	Glucose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	390	2160	93	28	[15]
2	Glucose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	350	2090	93	23	[15]
3	Fructose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	310	1810	88	23	[15]
4	Fucose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	350	1760	84	34	[15]
5	Rhamnose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	270	1320	90	30	[15]
6	Mannose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	290	1240	88	25	[15]
7	Sucrose	SC-MFC	AS	C fiber	C cloth	NA	1.79 (W m <sup>-3</sup> )	94	4	[33]
8	Xylose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	380	2330	95	31	[15]
9	Arabinose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	260	2030	93	27	[15]
10	Ribose	DC-MFC	Mixed bacterial culture	C cloth	C cloth	270	1520	86	30	[15]
11	Galacturonic acid	DC-MFC	Mixed bacterial culture	C cloth	C cloth	330	1480	80	22	[15]
12	Glucuronic acid	DC-MFC	Mixed bacterial culture	C cloth	C cloth	440	2770	89	24	[15]
13	Gluconic acid	DC-MFC	Mixed bacterial culture	C cloth	C cloth	280	2050	93	30	[15]
14	Galactitol	SC-MFC Media- tor-less	Mixed bacterial culture	C cloth	C cloth	340	2650	90	13	[21]
15	Mannitol	SC-MFC Media- tor-less	Mixed bacterial culture	C cloth	C cloth	240	1490	91	19	[21]
16	Sorbitol	SC-MFC Media- tor-less	Mixed bacterial culture	C cloth	C cloth	260	1690	71	10	[21]
17	Arabitol	SC-MFC Media- tor-less	Mixed bacterial culture	C cloth	C cloth	260	2030	91	25	[21]
18	Ribitol	SC-MFC Media- tor-less	Mixed bacterial culture	C cloth	C cloth	320	2350	92	28	[21]
19	Xylitol	SC-MFC Media- tor-less	Mixed bacterial culture	C cloth	C cloth	290	2110	91	21	[21]

20	Glucose	Stack- MFCs	Saccharomyces cerevisiae	Graphite plate	Graphite plate	3220	2003	NA	22	[14]
21	Glucose	SC-MFC Media- tor-less	Mixed microbial solution	Graphite fiber felt	Graphite fiber felt	351	218	98.8	26.2	[27]
22	Glucose	SC-MFC Media- tor-less	AS	Graphite fiber felt	Graphite fiber felt	508	456.8	94.3	55.4	[27]
23	Sucrose	SC-MFC Media- tor-less	Mixed microbial solution	Graphite fiber felt	Graphite fiber felt	305	164.6	93.7	20.3	[27]
24	Sucrose	SC-MFC Media- tor-less	AS	Graphite fiber felt	Graphite fiber felt	411	298.9	81.8	59.5	[27]
25	Ethylene glycol	DC-MFC	Mixed anaerobic consortia	Unpolished graphite	Pt sheet	NA	5.72	97	NA	[18]
26	Glucose	DC-MFC	Mixed anaerobic consortia	Unpolished graphite	Pt sheet	NA	2.47	92	NA	[18]
27	Glucose +ethylene glycol	DC-MFC	Mixed anaerobic consortia	Unpolished graphite	Pt sheet	NA	5.26	94	NA	[18]
28	Acetate	SC-MFC Media- tor-less	Domestic waste- water	Toray C paper	C paper +Pt	798	661	99	13.2	[25]
29	Butyrate	SC-MFC Media- tor-less	Domestic waste- water	Toray C paper	C paper +Pt	795	349	98	7.8	[25]
30	Ethanol	DC-MFC	AS	Porous C paper	Porous C paper	213	40	NA	42-61	[20]
31	Ethanol	SC-MFC	Sludge+ bacteria from the DC- MFC	Porous C paper	Porous C paper +Pt	750	488	NA	11-May	[20]
32	Naphthalene+ benzidine+2% NaCl	SC-MFC	AS	C cloth	C cloth	306	156.06	85	NA	[29]
33	Glycerol	SC-MFC	Bacillus subtilis	C cloth	C cloth+4 PTFE diffusion layers	560	0.06 (mW cm <sup>-2</sup> )	NA	23.08	[30]
34	Glycerol (pure, 3.2mg l <sup>-1</sup> )	DC-MFC	AS	C paper	C cloth +Pt	280	65.4	99	34.1	[31]

Depending on the inoculum, the C source of either fermentable (glucose) or non-fermentable C (acetate) can be very important. The output power is a function of wastewater rigidity based on a Monod-type relationship with a half-saturation constant of Ks [23]. The energy conversion efficiency and potential efficiency in MFCs was investigated. The methane was only detected in glucose-fed MFC, which shows that acetoclastic methanogens are pushed out of the competition by the analyte respiring bacteria. Unlike a high biomass density, the anode respiring bacteria density and electron donor concentration were very low causing a large reduction of potential efficiency with low Cd. This condition leads to the slow kinetics for electrons transferring into the anode and the large loss because of substrate concentration gradient in the anode biofilm. The energy conversion efficiency was obtained 42% and 3% with acetate and glucose respectively. Potential efficiency of 6% with the Pdm of 9.8mW m<sup>-2</sup> with low

current for glucose were obtained. As such, the main reason for the reduction of potential efficiency and Pd in glucose is the presence of non-electrogenic bacteria with high-concentration in anode biofilms and low electron donor concentration. In contrast, potential efficiency of 59% for acetate with high current and Pdm of 360mW m<sup>-2</sup> [24]biomass, residual organic compounds, H2, and CH4 gas. The comparison of the two donors allowed us to objectively evaluate the diversion of electron flow to nonelectricity sinks for fermentable donors, leading to different behaviors in energy-conversion efficiency (ECE. Note that unlike acetoclastic methanogens, the anolyte respiring bacteria could not out-compete H<sub>2</sub>-oxidizing methanogens. Therefore, with use of organic complexes as fuel, the control of methane production is so important and the CE can significantly improve. Totally, many of complex sugars cannot be used by common exoelectrogenes such as Shewanella, Geobacter. Specially, S. oneidensis was unsuccessful in power generating from glycerol as a microbial consortium due to lack of compounds as the transporters. Thus, the selection of microbial species based on the type of substrate is very important.

The output power is a function of the substrate density which can be well described with saturation kinetics, although the Pdm was different from the circuit load [25]. The colloidal particles and organic complexes in fermented wastewater not only reduce the Pd, but also has important roles as rate-limiting parameters in the steady output power. Pd reduction is mainly due to the high IR of the complex substrate. In this regard, a Pd 1,884mW m<sup>-3</sup> from real fermented wastewater (obtained from coffee bean processing wastewater), while Pd of 3664mW m<sup>-3</sup> was obtained from acetate with the same OLR. The Pd reached 2981mW m<sup>-3</sup> by doubling the OLR. At result, the effect of complexity and OLR is clear [26]. On the other hand, based on the CE, the MFC performance can be raised with increasing substrate complexity which is due to the fact that some species can use products derived from other species during the process of biodegradation of complex wastewater [27]. The Pd with butyrate as a substrate was obtained 66% lower than acetate with the same open circuit voltage (OCV). The CE and energy recovery values were (31-10%, 3-7%) and (15-8%, 2-5%) for acetate and butyrate respectively suggesting a significant loss of electrons and energy in processes except electricity generations [25].

Operational parameters are the determining factors in system performance. Adding mediators in MFC with can have a significant effect on performance. For example, When toluene as a xenobiotic contaminated was used with pyocyanin (as a mediator), there was 3.64-time increment in the Pdm from 4.69 to 21.7mW m<sup>-2</sup> and 13-time increment in CE from 0.83% to 11.62% than when toluene was used alone. Adding pyocyanin improved electricity production significantly by system impedance reduction, electron density increase, and reducing the IR (from 500 to 100) [28]. The MFCs performance in naphthalene and benzidine treatment increased by 344.77% in relation to power generation when the redox riboflavin mediator (30µM) was added externally [29]. The salinity is an effective method in treatment of a naphthalene and benzidine mixture. The optimal salinity was 2.0% and with salt concentration of 5%, where the performance was significantly improved. The pH can play an important role in treatment efficiency. The investigation during glycerol degradation indicates that the electricity generation during long time was stable under neutral toward alkaline conditions. The electrochemical activity increased via adding pH from 7, 8 and 9, where pH 7 was the most suitable pH for stable current generation [30]. The identification of optimum concentration is very important. With the increase in glycerol concentrate higher than optimum, the Pd can remain, but CE and COD removal decreased [31]. Due to effect of type of substrate and its concentration on microbial community changes, a special attention was paid to the evolutionary stages of the anaerobic inoculum via replacing glycerol with glucose as a

substrate for reaching a steady MFC efficiency with suitable Pd. Even at high concentrations of glycerol, the adaptability of Anaerobic Sludge (AS) with the use of glucose is an appropriate method for maintaining the MFC's stable performance [31]. Identification of optimum temperature is very important. The performance of the system was desirable at 40°C (compared to 30°C and 50°C) where a Pdm of 292.60mW m<sup>-2</sup>, COD elimination of 90%, and naphthalene and benzidine biodegradation of 100% were obtained. The biological degradation of naphthalene, benzidine, COD, Pdm, and CE at 40°C were 178.78%, 196.29%, 215.78%, 185.07mW m<sup>-2</sup>, and 120.53% better respectively compared to the initial temperature (30 °C). However, these cases dropped sharply when the operation temperature reached 50 °C [29]. Generally, the best temperature is dependent on species of microbial community.

Since the integration is one of the suitable methods for increasing the MFC application, a two-phase process of dark fermentation linked with a DC-MFC and the raw glycerol was used. After fermentation, 20% of organic compounds was removed by of hydrogen gas production. The fermentation effluent with COD of 7610mg  $\rm l^{-1}$  used as feedstock for MFC and Pd of 92mW  $\rm m^{-2}$ , COD elimination of 50%, and CE of 14% were obtained. When the fermentation effluents were diluted by 50%, CE was only 27%. Higher concentrations of the substrate led to further electron loss due to other chemical, physical, and microbial reactions [32].

#### Nitrogen resource

One important part of organic compounds in domestic wastewater, food processing, and industrial wastewater is protein as a source of nitrogen (N). Ammonia, ammonium, nitrite, and nitrate are the most important N forms with nitrate being one of the most problematic compounds in wastewater in particular. To remove the N from wastewater, nitrification and denitrification are well-known methods. Some performances of MFCs with N resource as substrate are presented in Table 2. In this regard, the R-group in amino acids plays an effective role in power generation from protein. The R-group of some amino acids include alanine (non-polar amino acids), serine (polar amino acids), asparagine with a side neutral-charged side chain and histidine, lysine and arginine with positively-charged side chain, as well as aspartic acid and glutamic acid with a negative charge. The Pd generation is as follows (L-Serine > L-Arginine > L-Histidine > L-Glutamic acid > L-Aspartic acid > L-Asparagine > L-lysine > DL-Alanine) [34]. In MFCs, the hydrophilic and acidic parts can easily be degraded in comparison to the neutral sections. In addition, the aromatic compounds in the hydrophilic section can preferably be removed more than non-aromatic compounds. The compounds such as tryptophan-protein in all parts and aromatic proteins in neutral part of hydrophilic and hydrophobic can easily be removed. Further, the MFC can easily hydrolyze and biodegrade the hydrophobic amide-1 proteins and aliphatic components. The carboxylic and alcoholic groups in hydrophobic acid and hydrophobic neutral and transphilic acid increase due to the

hydrolysis and fermentation of high molecular organic compounds [35]. Bovine serum albumin is a model of compounds which is characterized by a high weight and high water solubility molecule structure; peptone is a complex mix of various proteins and meatpackaging wastewater generally contains blood, meat, fat tissue, meat extracts and animal abdominal contents with COD. The highest Pd of 354mW m $^{-2}$  from bovine (1100mg l $^{-1}$ ), 269mW m $^{-2}$  from peptone (300mg l $^{-1}$ ), and 80mW m $^{-2}$  from meat-packaging wastewater (1420mg COD l $^{-1}$ ) were obtained. With salinity and adding (300mg l $^{-1}$  NaCl) to increase the conductivity in meat Table 2: MFCs Performance with Nitrogen Resources.

packaging wastewater, the Pd reach increased by 33%. The protein elimination was obtained more than 94% for all substrates. Also, a COD elimination of 87% was achieved for meat packaging wastewater [36]. The concentration of substrate is effective on internal resistance (IR) and totally on power generation. The Pdm reached 19mW m $^{-2}$  with cysteine concentration of 385mg  $l^{-1}$  where IR was about 700-1000. The Pdm increased by 39mW m $^{-2}$  when cysteine concentration reached 770mg  $l^{-1}$  where IR was about 493 [37].

NO	Substrate	MFC Type	Anode	Cathode	Pd (mW m²)	COD Re- moval%	N Remov- al%	CE%	Ref
1	Synthetic wastewater	Coupled MFC	Graphite felts	Graphite felts – oxic -biocathode	14 (W m <sup>-3</sup> )	98.8	97.4	NA	[39]
1			Graphite felts	Graphite felts - anox- ic-biocathode	7.2 (W m <sup>-3</sup> )			NA	
3	University of Connecticut wastewater (3.5 mg l-1 of DO)	Short-cut nitrifica- tion and autotro- phic denitrifica- tion MFC	C brushes	C cloth	294.9	NA	99.9	2.41	[41]
4	Wastewater Treatment Plant	SC-MFC	C felt	C felt	88	97%	96	43	[46]

Using the nitrate as a cathodic electron acceptor is a good way for N removal. The removal of N mainly depends on the sequential nitrification and denitrification in wastewater treatment. The nitrification and denitrification were obtained in the half-cell by completing the cathodic nitrification process through special aeration. The denitrification efficiency increased by reduction of dissolved oxygen in a cathodic chamber of membrane-aerated MFC [38]. The nitrate can be denitrified electrochemically by oxidizing the ammonium in to the nitrate in a coupled oxic-biocathode MFC and anoxic-biocathode MFC. Most of COD is eliminated at the two MFCs anodes. The oxic-biocathode is responsible for nitrate oxidization and anoxic-biocathode is responsible for nitrate denitrification [39]. Another coupled MFC was configured, composed of a nitrifying sulfide elimination MFC and a denitrifying sulfide elimination MFC. The favorable sulfur (S)/N ratio was three. Total N elimination of 58.7% and S production of 27.4-33.3% were obtained. The entrance of nitrifiers and electroactive oxic microorganisms from the oxic-cathode in the anoxic-cathode was successful in N elimination [40]. Next, a short-cut nitrification and autotrophic denitrification MFC was developed, which unlike biological nutrient removal processes, had a simplified operation, reduced operational time, reduced aeration cost, saved C sources, and reached energy-positive situation. In the cathode of MFC with Short-cut nitrification, ammonium was mainly reduced to nitrite instead of nitrate (nitrite to nitrate ratio: 3). The nitrite reduced to N gas through autotrophic denitrification via electrons generated from the anode chamber. It generates 0.007kWh m<sup>-3</sup> of net electricity which is the self-sustained energy-positive and with an excellent potential for removing nutrients [41]. For developing a

high-rate denitrifying, the cathode COD /  $\rm NO_3$ -N ratio was effective on OCV and nitrate elimination significantly. The heterotrophic conditions in the cathode did not change the energy production. The optimal pH for denitrification was 7. In this regard, in MFC cathode, a high-rate denitrification was obtained by cow manure and soil. The best  $\rm NO_3$ -N removal rate of 7.1kg  $\rm NO_3$ -N  $\rm m^{-3}$  net cathodic chamber d<sup>-1</sup> was obtained in the 7.31 COD /  $\rm NO_3$ -N ratio with a Cd of 190mA  $\rm m^{-2}$ , a Pd of 31.92mW  $\rm m^{-2}$  and CE of 9.7. Reduction in OCV and  $\rm NO_3$ -N elimination rates were obtained at COD /  $\rm NO_3$ -N ratio >12 and <7. The overall CE was low in this case which could have been related to the anode unfavorable metabolic products and the high organic level content of cathode [42].

An anaerobic sequencing batch reactor as a pre-treatment was integrated with MFC for increasing ability to oxidize insoluble, polymeric, and complex organics. This system can increase hydrolytic-acidogenic conversion, power recovery, and C and N removal. In this regard, through the sewage and groundwater, soluble organics and volatile fatty acids contents increased by 52% and 120% with sewage pretreatment, respectively. The optimum Pd of 7.1W  $\ensuremath{\text{m}^{-3}}$  and Cd of 45.88A  $\ensuremath{\text{m}^{-3}}$  were obtained indicating 8% and 10% improvements compared to without pretreated sewage. As compared to without pre-treatment system, 217% higher for C elimination and 136% higher for N elimination were obtained with integrated system [43]. In addition, N elimination in cathode can be catalyzed by anaerobic ammoniumoxidizing bacteria (anammox) along with denitrifiers and nitrifiers. Anammox produces dinitrogen gas as a final product by employing nitrite as the electron acceptor [44]. In this regard, a loop MFC was configured which included a denitrifying organics/

sulfide elimination MFC and a nitrification chamber. High total organic carbon/ sulfide ratio in effluent was successful to elemental S recovery and electron generation. Entrance of oxygen into the cathode could enhance N elimination and output power but prevented elemental S recovery. The total organic carbon/ sulfide ratio of 4.69 and cathodic DO of 4.2mg l<sup>-1</sup> were favorable. In this situation, 100% of total organic carbon, 100% of sulfide, and 82.6% of total N were eliminated with S recovery of 35.1% and CE of 53%. Further, prevailing denitrifiers, nitrifiers, and anammox also took part in N elimination in the cathode [45].

#### Sulfur resource

The S compounds such as sulfide and sulfate are harmful and toxic to human health, as well as corrosive with unpleasant odor. These compounds in found some food industrial wastewater such as sugar and alcohol. Sulfate cannot be removed directly by the MFC as it is an electron acceptor. Thus, the sulfate-reducing bacteria (heterotrophic bacteria that uses organic compounds as C and energy sources) could convert the sulfate to sulfide in an anaerobic medium [47]. Then, MFC uses the sulfide oxidizing

acceptor such as nitrate, for oxidation of sulfide) and it oxidizes the sulfide to S<sup>0</sup> [48]. There is no possibility for simultaneous removal of sulfide and organic compounds if there is lack of nitrate or other electron acceptors in wastewaters. While adding the electron acceptors is not an appropriate and eco-friendly option [49]. As such, for the reaction of sulfide oxidation, the anodic level can act as an acceptor for electrons [50]. ulfide and sulfate are removed with electricity generation simultaneously using the MFC with the anodic biofilm SRB (sulfate reducing bacteria) + SOB (sulfide oxidizing bacteria) and anode as an electron acceptor [51]. Thus, this biological reactor can remove sulfide, nitrate, and organic compounds simultaneously [52-55]. It was also found that bacteria associated with S and anaerobic fermentation bacteria interact with each other [56]. The initial concentration of sulfide has a positive relationship with the bioelectricity production. Given that, the anode potential diminishes due to increase in sulfide as it has a low recovery potential [56]. Some performances of MFCs with S resource as substrate are presented in Table 3.

bacteria (they are autotrophic, so they need minerals electron

Table 3: MFCs Performance with Sulfur Resources.

NO	Substrate	MFC Type	Inoculum	Anode	Cathode	Pd(mW m <sup>-2</sup> )	Voltage (mV)	COD Removal%	S Re- moval %	CE %	Ref
1	Artificial wastewater (containing lactate and sulfate)	Cylindrical DC-MFC	SRB+SOB	C felt	C cloth	62	730	NA	NA	Na	[50]
4	Artificial wastewater	SC-MFC	Desulfovib- rio desulfu- ricans	High surface area activated C cloth	C cloth +Nafion ionomer	0.51	NA	NA	99% (sulfate)	NA	[62]
5	Artificial wastewater (60 mg l <sup>-1</sup> of sulfide, 800 mg l <sup>-1</sup> of COD)	Anaerobic baffled stacking MFC	AS	C fiber felt and graph- ite granules	C paper	200-270	2582	73.8	81.2	NA	[56]
6	Artificial wastewater	H-type cylindrical DC-MFCs	Anaerobic granular sludge	C-fiber-felt	C-fiber-felt	396.1	NA	31.7	88.5	11.4	[60]
7	Artificial wastewater	H-type cylindrical DC-MFCs	Anaerobic granular sludge	TiO <sub>2</sub> +C-fi- ber-felt	C-fiber-felt	607.8	NA	56.9	94.4	13.2	[60]
9	Artificial wastewater	H-type cylindrical DC-MFCs	Anaerobic granular sludge	Fe <sub>2</sub> O <sub>3</sub> +C-fi- ber-felt	C-fiber-felt	537.6	NA	55.2	91.4	12.2	[60]
10	Sulfate-rich wastewater	DC-MFC	Pretreated AS	Biotic anode- Non-cat- alyzed graphite	Abiotic cathode- Non-cat- alyzed graphite	240	498	58	56	NA	[63]
11	Artificial wastewater+ lactate,	DC-MFC	SRB	C rod	Pressed activated C+ graph- ite rod	349	≈0.7	77.5	93.2	NA	[59]

MFCs can remove the sulfate significantly. This ability can be further increased using several methods. Particularly, using graphite as an electrode, the sulfate removal was obtained 99.84% [57]. In comparison to glucose, sulfide was alternatively used as an electron donor and the Pdm was obtained 401mW m<sup>-2</sup> at level of 100mg l<sup>-1</sup> with removal of 64% which was more than twice that of the case with glucose in a mediator-less DC-MFC [58]. Elsewhere, the sulfide and COD removals increased when HRT increased from 24 to 60h, but electricity generation was not increased as the electron donor was inadequate at the end of the reactor. The sulfide was mainly oxidized to S and sulfate [56]. Adding electron donor can have a major effect on microbial communities causing enhanced sulfate reduction and electricity generation. The share of each phylum in the microbial community is affected by the electron donor. In this regard, lactate has proved

to be more effective than glucose. The analyses indicated that electron donors especially lactate have a great effect on sulfate-reducing, S-reducing and sulfur-oxidizing bacteria growth [59]. On the other hand, modified anodes can increase sulfide removal. They can provide more active surface for bacterial adhesion by enhancing density of biomass. C-fiber-felt anodes were modified by the Nano-layers of  ${\rm TiO_2}$  and  ${\rm Fe_2O_3}$  in the vertical direction. Compared to MFC with C-fiber-felt anode, Pdm was improved by 1.53 and 1.36 times for MFC- ${\rm TiO_2}$  and MFC- ${\rm Fe_2O_3}$ , respectively and sulfide removal efficiency and total organic C was increased after 48 hours [60]. The pH of 8 as is more effective compared to acidic and neutral condition. Alkalinity and salinity can increase the power generation by sulfate reducing bacteria. The Pdm of 1188mW m<sup>-3</sup> was obtained at 6g l<sup>-1</sup> of NaCl concentration [61].

# Phosphorus resource

Table 4: MFCs Performance with Phosphorus Resources.

NO	Substrate	МГС Туре	Inoculum	Anode	Cathode	Pd (mW m²)	Voltage mV	COD Remov- al%	P Re- moval%	C E%	Ref
1	Synthetic wastewater (3.5mg l <sup>-1</sup> of DO)	Tubular DC- MFC	AS and aerobic sludge	C paper	C cloth +Pt	530	521	81	84-97	52.48	[64]
2	Synthetic wastewater (DO 2mg l <sup>-1</sup> of DO)	Tubular DC- MFC	AS and aerobic sludge	C paper	C cloth +Pt	303	178	77.3	84-97	23.9	[64]
3	Swine waste- water	SC-MFC	-	C felt	C paper +Pt/C	2.3 (W m <sup>-2</sup> )	300	91	82%	47	[66]

P is known as one of the main pollutants of food wastewater. P has also a valuable aspect in wastewater, because as a fertilizer it is a nutrient for plant growth. P as a natural resource is subject to shortage, so recovery of P from wastewater has become an important issue. Accordingly, the MFCs are offered in this regard and it can be a brilliant way of removing and recovering P. In MFCs, cathodic dissolved oxygen is effective on performance due to the loss of the electron acceptor. The drop of power generation is due to the decline in the cathodic dissolved oxygen. With dissolved oxygen reduction, Pd and CE decline sharply. In addition, following chemical precipitation, a large amount (about 80%) of total P was eliminated, by microbial absorption (17-4%) and by the sediments contained phosphate, carbonate and hydroxylate. The COD was first eliminated in the anode (> 70%) followed by in the cathode (<5%). In anaerobic anode, phosphorus-accumulating bacteria absorbed a large part of COD in polyhydroxybutyrate form. Along with entrance of this effluent to cathode with aerobic or oxygenpoor condition, polyhydroxybutyrate is used by phosphorusaccumulating bacteria as the C source. However, most of N can be removed (more than 85%) when cathodic dissolved oxygen was reduced to low levels [64]. Some MFCs performance with P resource as substrate are presented in Table 4. The constructed wetlands MFC (CW-MFC) showed an increase of capacity in

immobilized P with the removal rate of total P and reactivity P of 85-86% and 89-90% respectively from swine slurry. An alum sludge was used while providing a concurrent up flow-down flow system and the Pdm increased by 70% and reached 0.268 W m $^{-3}$  (compared to 0.168W m $^{-3}$  for up-flow system). However, the COD removal efficiency dropped to 64%, compared to 80% which was achieved by a constant up-flow system [65].

Regarding P recovery, MFCs are able to recover phosphate from the digested wastewater sludge by precipitating in the cathode in the form of struvite (MgNH $_4$ PO $_4$ ·6H $_2$ O). The result of production in suspended solid form (instead of the solution form) indicates that P in the suspended solid form is first dissolved, and then it is precipitated on the cathode [66]. The orthophosphate can be obtained from iron phosphate (FePO $_4$ ) available in digested wastewater sludge by reaction with Mg $^{+2}$  and NH $_4$  $^+$ . The process efficiency was achieved to be up to 82% or 600mg l $^{-1}$ . The crystalline fertilizer was analyzed and it was found that the compound is about 90% accurate and there are no toxic metals such as arsenic, cadmium, lead, or chromium [67]. In another study, 70-82% of phosphorous was eliminated and some struvite formed where the amount of P in these was about 4.6-27% of the influent [66].

Integrating the MFC considerably increases the organics, N and P removal in CWs. The optimal operating conditions for COD, NH<sub>3</sub>-N, N and P removal are very different from each other and are affected by several factors. HRT is the most important factor via about 50%. Generally, dissolved oxygen level in the cathode and effluent reflux ratio are significant for pollutant removal and external resistance is the most effective factor for power generation [68]. In addition, ferrous ion, ferric ion, and ferric hydroxide, as metabolic intermediates are useful for phosphate elimination [69]. In another integration, the CW-MFC with pyrite can produce effluents with a stable pH and low sulfate level. It increased power generation 19-28.4% more than CW-MFC without pyrite. Also, nitrate and total P removal efficiencies in the CW-MFC with pyrite were 15% greater than CW-MFC without pyrite and obtained as 70.1% and 91.2%, respectively [70]. A pyritebased CW-MFC can be useful. Pyrite (FeS<sub>2</sub>) is an abundant sulfide mineral including Fe and S. By increasing N removal through autotrophic denitrification, pyrite is a suitable feedstock for CWs [71]. According to the results, the success of this technology in the removal and recovery of valuable phosphorus can be seen, which is still needed to enhance the efficiency with further studies.

#### **Food Industry Wastewater**

The food industry wastewaters are generally based on the main components of the wastewater. Nevertheless, the real wastewaters can be used in this technology. A real wastewater is a collection of components where each of them can have a special effect on the process individually. Some of these pollutants can inhibit microbial process and some of them can facilitate it. As such, evaluation of real wastewater as a complex collection in the MFSs is a proper way to improve and achieve clear results. The results of MFCs performance with some different food industrial wastewaters content as substrate are presented in Table 5. Some important food industry wastewaters, which have been investigated in studies, are described.

Table 5: MFCs Performance with Food Industry Wastewater.

NO	Substrate	MFC Type	Anode	Cathode	Inoculum	Pd (mW m <sup>-2</sup> )	Voltage (mV)	COD Re- moval%	CE%	Ref
1	Composite vegetable waste	Media- tor-less SC-MFC	Graphite plate	Graphite plate	Anaerobic mixed consortia	57.38	250	62.86	NA	[136]
2	Composite canteen-based food waste	Solid phase SC-MFC	Graphite plate	Graphite plate	NA	170.81	463	76	NA	[137]
3	Food wastes	SC-MFC	C cloth	C cloth +Pt	AS	5.6 (W m <sup>-3</sup> )	510	90.3	NA	[35]
4	Food process- ing wastewa- ter	DC-MFC	Graphic sheets	Graphic sheets	Sludge	230	422	86	21	[127]
5	Food waste leachate	DC-MFC	С	С	Anaerobic sludge	15.14 (W m <sup>-3</sup> )	1120	90	NA	[77]
6	Food waste leachate	SC-MFC	C felt	Gas diffusion + Pt	collected from wastewater plant	1.86 (W m <sup>-3</sup> )	250	95.4	11.07	[138]
7	Dairy waste- water	Media- tor-less DC-MFC	Graphite plate	Graphite plate	Activated sludge	621.13	856	90.46	37.16	[139]
8	Dairy waste- water	SC-MFC (pH initially ad- justed to 9)	C cloth	C cloth	Escherichia coli -K-12	1.05 (W m <sup>-2</sup> )	654	95.45	67.53	[140]
9	Dairy waste- water	DC-MFC	Copper	Copper	Anaerobic and facultative mi- crobes in dairy wastewater	NA	644	92.2	NA	[141]
10	Dairy waste- water	Up-flow tu- bular MFC	C cloth	C cloth +Pt	Shewanella oneidensis and Clostridium butyricum	2.4 (W m <sup>-3</sup> )	490	96	4.40%	[80]
11	Dairy waste- water	SC-MFC	C cloth	C cloth +Pt	Shewanella oneidensis and Clostridium butyricum.	0.5 (W m <sup>-3</sup> )	616	95.5	1	[142]

12	Whey (from sterile filtra- tion)	DC-MFC	Plain C paper	C cloth+ Pt	AS	46	NA	94	11.3	[143]
13	Labanah Whey wastewaters	Cylindrical mem- brane-less MFC	Graphite	Graphite	Mixed microbial consortia	23.23 (W m <sup>-3</sup> )	NA	72.76	NA	[144]
14	Yogurt waste- water	SC-MFC	Stainless steel fiber felt	activat- ed C	-	1043	NA	88	23	[84]
15	Orange peel waste	DC-MFC	Graphite felt	Pt + graphite cloth	Anaerobic consortia	358.8	590	80	15.5	[90]
16	Lemon peel waste	DC-MFC	C felt	C cloth	AS	371	580	75.8	32.30%	[91]
17	Citrus waste	SC-MFC	Plain graphite plate	Plain graphite plate	anaerobic bioelectrogenic consortia	71.1	262	63.8	33.2	[145]
18	Brewery wastewater	DC-MFC With chitosan/bio- degradable copolymer	С	С		3882	NA	70	NA	[146]
		membrane	brush	brush	-					
19	Brewery wastewater	DC-MFC	C brush	C brush	-	4245	NA	72	NA	[146]
20	Winery waste- water	DC-MFC	C-felt	C-felt	activated sludge	420	72	650 (mg l <sup>-1</sup> d <sup>-1</sup> )	NA	[147]
21	Winery waste- water	DC-MFC	C felts	C felts	activated sludge	890	178	600 (mg l <sup>-1</sup> d <sup>-1</sup> )	42.2	[148]
22	Vegetable oil wastewater	DC-MFC Operation at 35 °C	Titanium rod	C cloth	AS	6119	5839	90	36.5	[106]
23	Soybean edible oil refinery wastewater	SC-MFC	Graphite brush +tita- nium	Stainless steel mesh + activat- ed C	Activated sludge	746	490	93.60%	22	[105]
24	PMW	SC-MFC (with activated C addition)	C cloth	C cloth	Anaerobic mi- crobial sludge	344.3	267.1	91.9	3.06	[149]
25	Starch processing wastewater	SC-MFC	C paper	C paper +Pt	-	239.4	490.8	98	8	[150]
26	Soluble starch	SC-MFC	graphite fiber felt	graphite fiber felt	AS	277.6	396	79.4	67.7	[27]
27	Sugarcane molasses	DC-MFC (H type)	NA	NA	Brevibacillus borstelensis	188.5	990	81.7	59.8	[151]
28	Molasses wastewater	Water- fall-type MFC + polyvinyl alcohol-hy- drogel as membrane	C felt	C cloth	Activated sludge	16.1	2130	90.1	NA	[152]

#### Acidogeneic food waste leachate

Food leachate contains a large amount of organic compounds, ammonium nitrogen, heavy metals, organic and inorganic chlorine, salts, etc. Leachate is essentially buried through landfill in some countries such as India [72]. The composition and concentration of leachate contamination is mainly under the influence of the landfill's age [73]. Biological treatment may be one of the most ideal methods due to the high bioavailability of leachate, but the presence of ammonia, sulfides and heavy metals will hamper and reduce the efficiency of the biological treatment process significantly [74]. The food leachate alone can generate electricity, but the addition of the inoculum causes a significant reduction (about 7 days) at the start time. A higher efficiency was achieved when AS was used in MFC with food leachate; the analysis of the anode biofilm microbial community indicated which was due to the richness of two fermentative (Clostridium and Bacteroides) and electrogenics (Magnetospirillum and Geobacter) [75]. For strength wastewater like this, the substrate concentration of above 20000mg l-1 can inhibit microbial activity in the anode chamber. In addition, the membrane fouling by microorganisms and cations can significantly affect the ion exchange capacity and MFC performance. Recirculation is a great technique used to increase the output power of anaerobic reactors. Especially recirculation indicated when food leachate was used in MFCs and it was effective to enhance the bioelectricity generation [76]. Acidogenic fermentation can be used as a pre-treatment in this kind of wastewater and at the end of the 58th day of fermentation, the COD and pH of this leachate reached 135.11g l-1 and 3.93 respectively [77].

# Dairy industry wastewater

The dairy industry wastewater with complex compounds consists of degradable organic compounds where 97% of COD contains sugar content [78]. There is a high concentration of fermentable substrates [79]. The compounds in treated MFC effluent may have toxic effects at different ecological levels, which indicates the importance of complete MFC process. As such, toxicity testing in MFC treatment can be useful. In this regard, investigation of acute toxicity indicates the raw dairy wastewater has highly toxic effects, while the MFC effluent is without toxicity effects after complete process [80]. To demonstrate the feasibility of dairy processing wastewater treatment by recovering bioenergy through MFC using dairy wastewater, measuring the amount of extractable energy and highlighting the main sources of energy loss in the system. On average, the MFC especially extracts an energy from 8.95 \* 10<sup>-2</sup> kW h kg<sup>-1</sup> COD with a highest output of 20.53 \* 10<sup>-2</sup>kW h kg<sup>-1</sup> COD [81]. n dairy industry, the clear issue is that the MFC is a valuable alternative to effluent treatment and energy generation, while it needs several experiments on largescale in dairy industry [81]. Using modified anode by nanoparticles can reduce cost investment and increase the performance. In this regard, copper-doped iron oxide nanoparticles are a good choice and characterized for wettability and electrochemical

analysis. The coated anode indicated a good hydrophilic property in wettability. It increased potential, decreased resistance where Pdm of 161.5mW m<sup>-2</sup> and COD removal of 74% were obtained [82]. The yogurt wastewater was found able to start an MFC using its natural microflora [83]. Alkaline conditions (pH 10.5) are useful for treating the yogurt wastewater. However, solids suspension can be removed using a pretreatment precipitate at first. Due to increase of the COD concentration from optimum during treatment in MFC with yogurt wastewater, the IR increase and the viability of bacteria is reduced in the anode biofilm. Thus, the electricity generation diminishes [84].

#### Whey

It is a by-product of the cheese production with a high content of organic compounds, mainly a mixture of lactose and protein. The whey contains 4-5% carbohydrate, up to 1% protein, 0.5-0.4% fat, less than 1% lactic acid, and 1-3% salt [85]. The primary density of microorganisms has a great role in MFC performance where the pre-incubate anode can improve performance in this treatment. Hence, anodes were enriched with microorganisms inherent to whey from 1 to 3 months before MFC activity. The result significantly enhanced and offered CE of 80.9%, COD removal of 92.8%, and Pdm of 1800mW m<sup>-2</sup> [86]. Two biological methods, MFC to traditional technology (fermentation by Lactobacillus bulgaricus) have been compared for treating whey and condensed whey. The results prove that the biological wastewater treatment is a very suitable and efficient alternative for traditional wastewater treatment. In the traditional method, during the continuous process for both whey and condensed whey, the removal of COD was over 70% while with MFC was 95% and 78%. Further, in the MFC method, the Pd values of 188.8mW m<sup>-2</sup> and 288.12mW m<sup>-2</sup> with the CE of 26% and 15% were obtained for whey and condensed whey respectively. Obviously due to the higher content of organic compounds, the power produced of condensed whey is about twice the whey itself [87].

The filter-sterilized (effluent mainly containing lactose) and acidified (effluent mainly containing acids) (after 48h of fermentation at mesophilic temperature) cheese whey are suitable pre-treatments. The initial COD concertation and initial pH are important parameters to reach optimum acidification and energy recovery in the form of hydrogen. In MFC, a Pdm of 1.57W m<sup>-3</sup> was obtained through acidification and 3.26W m<sup>-3</sup> through filtering [88]. Also, dark fermentation can be used as a pre-treatment where the hydrogen and an effluent rich in volatile fatty acids are produced, which are used in MFC. In this regard, a Pd of 0.34mW m<sup>-2</sup>, CE of 14%, and COD elimination of 41% were obtained with raw whey. On the other hand, a Pdm of 439mW m<sup>-2</sup> was achieved by dark fermentation effluent, which is 1000 times larger compared to raw whey, with CE of 24% and COD removal of 100% obtained. The main electrochemical performance difference of both MFCs was analyzed. In this regard, Geobacter, Pseudomonas and Thauera were the dominant electroactive population when fed by dark fermentation effluent, while the clostridium and lactobacillus were the dominant electroactive population when directly fed with raw whey. Also, in MFCs fed with raw whey, the low variety may increase the growth of lactic acid bacteria and cause acidification of the environment by fermentation of lactose. These microorganisms are known to produce bacteriocin; thus, they can succeed in a competition with exoelectrogenics. In addition, the pH in dark fermentation effluent was constant as the volatile fatty acids contained were absorbed completely [89].

#### Juice industry wastewater

The juice industry wastewater is characterized by biodegradability due to sugar as its main contamination (glucose and fructose) [25]. About 50-60% of the weight of the processed fruit is converted into peel waste which includes peel, seeds, and residue of internal coatings [90]. The changes in the concentration of inflow organic compounds may influence the function and composition of the microbial community [83]. The key components of orange peel waste, called pectin and cellulose, were also tested in a pure form. There was a stable current by pectin, while using cellulose alone as substrate did not produce a significant current. Thus, the absence of bacteria, which can degrade cellulose, was clarified and the activity of the enzymes of pectinase and polygalacturonase was detected [90]. Ultra-sonication can also be used as pre-treatment to enhance the power generation due to the increase of soluble COD and the degradation kinetics of the lemon peel substrate [91]. The photosynthetic biocathode could increase system performance with juice industry wastewater. This biocathode provided oxygen in the role of electron acceptor and contained a pure culture of Chlorella Vulgaris. By performing a few experiments, it was determined that the output power was obtained more rapidly and more stably in the continuous flow mode (23.97mW m<sup>-2</sup>) (with microorganisms in biofilm and in suspension) than in batch mode (Pdm of 22.85mW m<sup>-2</sup>). It was also evident that up to 1066mg COD l-1 could be treated successfully in this system. However, the cell voltage dropped from 15 to 10mV when the COD level increased from 343 to 1066mg COD l-1 [92]. An MFC does did start the fermented apple juice using its natural microflora. In this regard, compost leachate as an inoculum was used where Pdm 78mW m<sup>-2</sup> was obtained. Also, the AS was used with the Pdm 43.7mWm<sup>-2</sup> obtained [83]. The OCV of 0.4V and Pdm of 31.58mW m<sup>2</sup> were obtained using cashew apple waste in MFC [93].

#### **Brewery industry wastewater**

The brewery wastewater contains many organic compounds in the water, including sugary, starch and protein compounds produced at various stages, such as saccharification, fermentation, cooling, washing, etc. Generally, the COD of these wastewaters is 3000-5000mg l<sup>-1</sup> [94]. Adding mediators such as phosphate buffer can increase the output power by 185% [95]. In a tubular air-cathode MFC, the treatment efficiency was 93% which is a very good performance, but compared to other reports the output power was 96mW m<sup>-2</sup> which was poor [96]. However,

researchers have designed an MFC pilot-scale combining 40 tubular MFCs with a 10-liter capacity per day, which also showed a treatment efficiency of 86.4%, but the output power did not increase at all and it remained the same [97]. A 20-L continuous flow MFC (containing two 10L MFCs) with low investment cost was designed which worked for over one year with significant and stable performance, where COD removal of 94.6 with Pdm of 18.70mW m<sup>-2</sup> (in 228 day) were obtained [98]. Membrane fouling is considered as a challenge in MFCs especially for this kind of wastewater. The anaerobic baffled reactor including sediment MFC was integrated with anaerobic/aerobic membrane bioreactor (MBR). Every 24h, the back flush with water and aeration with air in anaerobic MBR were able to inhibit the fouling growth. Meanwhile, regular aeration was more effective than back flush, though it could be more effective if both ways were performed at the same time. The aerobic MBR indicated a higher stable effluent quality and higher performance in treatment. In this regard, only  $0.52\% \sim 0.99\%$  of output power was used for fouling mitigation in anaerobic MBR, but 31.80% of power was required in aerobic MBR, showing a very significant difference [99].

#### Winery industry wastewater

There is acidic pH, high organic content with low N and P concentrations, significant polyphenols, macro and micronutrients and heavy metals in all kinds of winery wastewater. The winery wastewater is produced during crushing, pressing, and washing the fermentation tanks, barrels, machines, and production rooms. The characteristics of wastewater winery are described as undesirable proportion of COD / N and COD / P [100]. Even when P and N were added, the COD removal was not effective. Daily removal fluctuated around 1000mg l-1 d-1 and during a complete test period COD removal was 17%. However, the electricity generation was affected positively by increasing the concentration of P and N. Thus, CE increased from about 2% to 15% and Pdm rose from 105mW m<sup>-2</sup> to 465mW m<sup>-2</sup> by adding P and N [100]. When comparing white and red wine, degradation of existing organic compounds is critical, hence BOD / COD of white wine settling was 0.93 while for red wine settling it was 0.33. Also, polyphenols at high concentrations can inhibit the process in red wine settling. Particularly in the anode, different types of substrates lead to formation of the different bacterial consortium. It was found that the COD removal, output power, and CE in white wine are greater than those of red wine [101]. It is clear that in order to achieve the desired performance, modification in the composition of the effluent is required by adding some items. In particular, adjusting the COD / N and COD / P ratios as well as BOD / COD is in demand.

#### Oil industry wastewater

The vegetable oil industry wastewater contains high amounts of sulfate and other organic materials. Some changes in the process will lead to better results; in particular, the membrane plays a key role in various processes. In this regard, wastewater from an oil refinery was treated using an MBR including

ultrafiltration and microfiltration [102-104]. There was a positive correlation between the amount of COD and voltage generation in MFCs. In comparison to some industrial wastewaters and chemical wastewaters with higher COD, a suitable efficiency in voltage generation (890mV) and more COD removal (87%) were obtained using vegetable oil wastewater (Ghee Industries). In addition, phosphate removal of 47% and sulfate removal of 42% were obtained [57]. In addition, the concentration of volatile fatty acids increased in the effluent by elevating the organic loading of COD, which were mainly acetate and propionate. The propionate showed the highest concentrations at the highest COD concentrations in MFC [105]. Regarding the operation temperature, an increase in MFC efficiency was achieved with an increase in the operation temperature and time. The performance of mesophilic microorganisms rose to a certain level by increasing the temperature from 25°C to 35°C [106]. For soybean edible oil refinery wastewater, some pre-treatment can be used for removing the harmful and problematic solvent compounds such as gravitational separation, slag scraping, flotation, emulsion breaking, and flocculation [107]. The olive and olive oil wastewater with toxic properties contain free fatty acids, carbohydrates, and phenolic compounds. Phenolic compounds have a determinant role in biological treatment due to their antimicrobial properties. In order to improve the efficiency, a physicochemical pre-treatment before anaerobic digestion has been proposed for reducing the olive oil toxicity. With olive mill wastewater in a SC-MFC, a maximum voltage of 381mV was obtained. COD elimination of 65% and phenols elimination of 49% were achieved using the process. A great decline was also obtained in the 3,4-dihydroxybenzoic acid, tyrosol, gallic acid, and p-coumaric acid. When treated pure phenols were tested instead of this wastewater, it was proved that 3,4-dihydroxybenzoic acid and caffeic acid do not affect the power generation, while propyl gallate suppresses the voltages production [108].

# Palm oil mill wastewater (PMW)

The complex compounds of PMW with COD of 50,000mg l-1 consists of amino acids, mineral materials, short fibers, organelles, N compounds, organic acids and carbohydrates, such as hemicelluloses as well as simple carbohydrates [109]. PMW is a high strength effluent. In this regard, an effluent with a 1:50 dilution (964mg COD l-1) was compared to effluent without dilution. The Pd decreased to about 22mW m<sup>-2</sup>, but the removal efficiency of COD and ammonium nitrogen and CE rose to about 70%, 75%, and 24% respectively [110]. On the one hand, it is evident that the higher initial COD sample led to greater electricity generation and less COD removal. On the other hand, the lower initial COD sample indicated more COD removal, but electricity generation was dropped. It reveals an inverse relationship between electricity generation and COD removal. In comparison to anaerobic microflora, controlled inoculum can increase the output power but with a weaker result in COD removal. This difference is due to the absence of a wider range of the microorganisms such

as fermentative microorganisms in controlled inoculum which are able to use more organic compounds. In this regard in a DC-MFC, the controlled inoculum (isolated from AS) was compared with AS inoculum. A Pdm of 107.35mW m<sup>-2</sup> was obtained with controlled inoculum which was about three times greater than that of the usual AS inoculum (33.62mW m<sup>-2</sup>). Also, CE of 74% was obtained with controlled inoculum which was 50% higher than that of the usual AS (24%). The COD removal efficiency for controlled inoculum was less than 32%, which was lower than the usual AS [111]. Further, the PMW natural anaerobic microflora and pure isolated culture of PMW, where Pseudomonas aeruginosa ZH1 was identified, as an inoculum were used in a DC-MFC. Using Pseudomonas aeruginosa ZH1, a Pdm of 451.26mW m<sup>-2</sup> was reported, 5 times greater than MFC with AS PMW (85.11mW m<sup>-2</sup>). The maximum COD elimination of 13% was obtained from AS PMW and 3% was obtained with Pseudomonas aeruginosa ZH1 [112]. Some integration methods can be used for PMW treatment. The MFC integration into anaerobic MBR can be effective on filtration efficiency. The MFC has a pre-treatment role before the anaerobic MBR. The anaerobic MBR can further treat the effluent from MFC. In the integrated system with direct feeding, the filtration efficiency indicated a significant development. In addition, decrease in polysaccharide concentration was observed which in turn suggested a better filtration performance [113]. Immobilized biological aerated filters have a great effect on the removal of COD and NH<sub>2</sub>-N. A two-phase cylinder MFC and immobilized biological aerated filters were used. The COD removal of 96.5% and NH<sub>3</sub>-N removal of 93.6% were obtained and the Pdm was obtained 44.6mWm<sup>-2</sup> [114]. Ultrasound can reduce the delay time in bioelectricity generation and improve the performance. It increased by 25% and reached 18.3 W m<sup>-3</sup>. In addition, COD removal increased from 30% to 54% [115].

#### Starch industrial wastewater

The starch industry produces extensive high-strength wastewater within a COD range of 16870 to 22800 mg l-1 with wide levels of carbohydrates, cellulose, protein, and other nutrients [116]. Biological conversion of starch processing wastewater is beneficial for recovery of resources such as the microbial biomass protein and biopesticide [12]. Electrogenesis species such as Shewanella oneidensis alone are unable to initiate the process and generate electricity using starch. The electricity can be produced by two-stage fermentation of starch. Streptococcus bovis 148 can produce lactic acid by metabolizing starch where Shewanella oneidensis MR-1 can generate power using lactic acid. In separate fermentation (first by Streptococcus bovis 148 followed by Shewanella oneidensis MR-1), a Pdm of 49.9mW m<sup>-2</sup>, and CE of 10.2% were obtained. In parallel fermentation (using both bacteria at the same time), a Pdm of 12.1mW m<sup>-2</sup> and CE of 4.7% were obtained [117]. The results indicated that two-stage fermentation has a higher performance in comparison with parallel fermentation in one stage.

#### Molasses wastewater (beet and sugarcane)

The molasses from sugarcane and sugar beet are inexpensive and available. Hence, it is used in the fermentation industry such as molasses-based distillery industry as a raw material. The molasses process and sugar beet process can produce a wastewater with high COD (65000-130000mg l-1) [118] and it seems to have suitable properties for use in MFC as substrate. Low substrate conversion rates are a major constraint on MFC performance using molasses wastewaters [119]. As a result, integration with other systems is a great way to overcome this problem. An anaerobic baffled stacking MFC with a combination of four units can improve the output power. The Pd average of 115.5mW m<sup>-2</sup> was obtained for four units, where a total COD removal efficiency of 50-70% was obtained [118]. An integrating system including three parts of an up-flow AS blanket reactor-MFC-biological aerated filter could enhance the treatment efficiency of beet sugar molasses. In this system, COD and sulfate majorly were reduced by up-flow AS blanket reactor; the MFC was responsible for the oxidation of produced sulfide and power production, where the dye and phenol derivatives were removed by biological aerated filter, where a Pdm of 1410.2mW m<sup>-2</sup> was obtained. The initial COD was 127500mg l-1 and removal efficiencies of COD, sulfate and color were 53.2%, 52.7% and 41.1% respectively [120]. Totally, it can be stated that PEM can reduce the recovery of electrons in the form of electricity more than expected, where electrons may be lost due to release of oxygen (totally 28%) [37] Integration of the dark fermented sugar cane molasses with MFC can improve the energy recovery. After fermentation, the remaining energy in the substrate can be extracted by MFC. The total carbohydrates and COD reduction in MFC were about 88% and 85% respectively, along with a Pd of 3.02W m<sup>-3</sup>. A slightly alkaline anodic pH was more effective in the MFC and could improve its performance. Thus, using an alkaline pre-treatment, the process was carried out at pH 7.5 [119]. In this regard, the system performance was enhanced to maximum when pH increased from 6 to 8, which may be related to the efficient extracellular electron transfer at pH 8. The distillery wastewater from sugarcane molasses was used where the Pdm and COD removal were obtained 63.8mW m<sup>-2</sup> and 63.5 % at pH 8, respectively [121]. Another study examined the effect of fed pH (5.4-10) and buffer situations on power generation and treatment performance in DC-MFC with distillery wastewater from sugar beet molasses. A Pdm of 168mW m<sup>-2</sup>, COD elimination of 68.2%, color elimination of 26.4%, and TDS elimination of 15.4% were obtained at pH 8. The Pdm of 194.7mW m-2 was obtained when borate buffer was used in the anode. The results revealed that the activity of exolectrogenic microorganisms was significantly influenced by pH and also led to increased conductivity; as such, the alkaline conditions of pH 8 and borate buffer produced the best efficiencies of treatment and power generation [122]. It is clear that slightly alkaline conditions could improve the system performance.

To compare SC-MFC and DC-MFC in molasses wastewater treatment, their performance in treatment and output power

should be considered. Two types of SC-MFCs with/without the PEMs and a DC-MFC were investigated. In both SC-MFCs, the values of COD removal and output power were similar. This indicates that the PEM does not increase the performance. The COD removal was achieved 90% and 50% for SC-MFC and DC-MFC respectively. However, a Pd of 17mW m<sup>-2</sup> was obtained in DC-MFC which is 2.2 times higher than in SC-MFCs. The Proteobacteria value in DC-MFC was 2 to 3 times higher than SC-MFC which is a determinant factor in power generation [123].

#### Chocolate industry wastewater

The chocolate industry wastewater is known as a non-toxic effluent with high total solid content and high COD. In DC-MFC, membrane or salt bridge can be used for connecting two chambers, which are effective on performance, where chocolate industry wastewater was used with activated sludge. Via membranes, a Pdm of 1.5W m<sup>-2</sup> was obtained, while vie salt bridge Pdm of 0.94W m<sup>-2</sup> was obtained with  $100\Omega$  external resistance, and finally MFC with glucose generated Pdm of 1.6W m<sup>-2</sup>. Totally, a significant reduction of 65% was observed in COD. The total solids content decreased by 68% and soluble solids by 50%. In addition, chocolate industry wastewater was used in cathode, as catholyte and it had a good result with a Pd of 1.02W m<sup>-2</sup> as compared to an air-cathode with Pd of 0.58W m<sup>-2</sup>. This increase reflects the interference of some of the additional factors of wastewater (chemical and microbial). Meanwhile, the results were improved using the ferricyanide, but the ferricyanide is a known catholyte suffering limitations such as high costs and the continuous replacement need in MFC. With adequate electricity generation through wastewater in the cathode, the cathode can be considered as an appropriate biotic cathode in which bacteria make the process more stable and cheaper where there is no need for an additional catalyst [124]. Some operational parameters can change to increase the system performance. In this regard, an annular SC-MFC with the chocolate wastewater treatment was used. Optimizing the distance between the electrodes and 46.15% distance reduction from 1.3 cm to 0.7 cm which resulted in a decrease in IR from 100 to  $50\Omega$ . Thus, the Pd increased from 7.98 to 22.898W m<sup>-3</sup> and the COD removal of 90% was obtained [125]. The same group in the following, continued to optimize the system. The system's performance was increased about 88% by elevating the temperature from 25°C to 35°C. The Pd of 16.75W m<sup>-3</sup> was the highest in the pH 7 with CE of 45.1%, which is the preferred pH for maximum microbial activity. In the effluent, a significant reduction in COD of 91% and turbidity of 78% were obtained [126].

#### Cereal industry wastewater

The wastewater of the protein-based food industry can be regarded as non-toxic due to low dangerous compounds with high BOD as well as high organic compounds content. It may also consist of simple sugars and starches [127]. The cereal industries wastewater can be considered by hydrogen production and MFC technologies. With fermentation of sugars, hydrogen is produced, but many organic compounds remain in the effluent and they

can be used by MFC. It is evident that integrating an MFC to the hydrogen production system (with high-sugar effluent where the major bio-hydrogen by-product is propionic acid) can increase the MFC performance. A Pdm of 371mW m<sup>-2</sup> was obtained by SC-MFC and cereal industry wastewater [128].

#### Tomato industry wastewater

The peel and grain in the tomato pomace contain a high nutritional value and have great potential for energy extraction by MFC. The seeds have a high thermal value and are composed of protein, lipids and carotenoids. The peel is rich in amino acids and carotenoids. Thus, they are rich in C and N. One gram of cull tomato contains half a microgram of riboflavin and thiamine, both of which can be used as an electron transfer mediator to improve exoelectrogen extracellular respiration in MFCs. As a result, redox mediators in MFC are endemic pomace. The tomato pomace has been used recently in MFC with Pdm of 132mW m<sup>-2</sup> obtained. The low efficiency of tomato pomace MFC compared to glucose (Pd of 169mW m<sup>-2</sup>) can be due to the slow kinetics of hydrolyzing, such as limited availability of COD in the electrolyte and its particle characteristics [129].

#### Seafood processing wastewater

The wastewater of seafood processing industry consists the high organic content which includes blood, fish head, intestinal and meat remains [130]. A tubular up-flow MFC can be used for treatment of seafood processing wastewater. A Pdm of 105mW m<sup>-2</sup> and CE of 28.03% were obtained, where the total and soluble COD removal were obtained 83% and 95%, respectively. Also, the Pdm was obtained 222mW m<sup>-2</sup> when the phosphate buffer was used as a catholyte [131]. Salty seafood wastewater can be treated by MFC, higher than 3 to 5% of salt distinguishing it from other industrial wastewaters. A Pdm of 16.2W m<sup>-3</sup> with CE of 15% were obtained at 4.2-hour HRT, while up to 80% removal of the COD content and biological nitrification were obtained. It is evident that HRT is effective on the Pd and a high HRT can improve the ability of removing the soluble COD as well as biological nitrification. High salinity and the other factor in wastewater can activate a low and constant IR about 100 in all experiments.

In addition, the wastewater pH was close to the normal level in both chambers [132]. To solve the blockage problem of seafood wastewater, an MFC with up-flow bio-filter circuit was developed as non-chemical without additives. A Pdm of 9.47mW  $\rm m^{\text{-}3}$  and maximum COD removal efficiency 94.37 % were obtained with pH 5.6-6.5 and aeration of 2.0  $\rm lmin^{\text{-}1}$  [133]. This configuration was able to overcome the blockage problem effectively.

#### Mustard tuber wastewater

Fuling mustard tuber is a popular pickle around the world. It produces high strength wastewater and salinity in large volumes during the production process. This effluent was used in a DC-MFC with the entire experiment divided into 4 sections characterized by concentration elevation of primary clarifier effluent. A Pdm of 246mW m<sup>-2</sup>, CE of 67%, and COD removal of 57% were obtained for the first sections. COD removal of 85% was achieved from the fourth sections as the highest COD removal; however, the power recovery efficiency was very low [134]. It can be concluded clearly that there is a significant negative correlation between the concentration of the primary clarifier effluent and the maximum cell voltage, and there is a significant positive correlation between the concentration of primary clarifier effluent and the IR. Accordingly, the colloidal complexes and particles in the primary clarifier effluent increased the IR gradually and subsequently reduced the power output. Further, this effluent can be used as a catholyte when a combination of different biocathodes is used. There was simultaneous removal of chemicals and N in cathodes, where most of the contamination was reduced during the acclimating period. Given the removal of N, aerobic and anaerobic microenvironments can form inside the cathodic biofilm and perform both heterotrophic denitrification and BE denitrification. A Pdm of 1.32W m<sup>-3</sup> was obtained where the COD and ammonium both reduced by 99% [135]. The MFC had similar performance in the same steps, which may suggest that this wastewater is an adequately self-buffered catholyte. The oxygen in a cathode leads to low bacterial activity by the over-potential and it can create essential constraints for energy recovery which should be reduced.

**Sub-Components of Wastewaters** 

Table 6: MFCs Performance with Sub- Components of Wastewater.

NO	Substrate	MFC Type	Anode	Cath- ode	Inoculum	Pd (mW m-2)	Voltage (mV)	Substrate Removal %	CE%	Special Note	Ref
1	2,4-DCP	DC- MFC	C cloth	C cloth	Bacillus Subtilis	9.5	450	60	23	Potassium Persulfate as catholyte	[159]
2	Nitro- benzene wastewa- ter	CW- MFC	Graph- ite felt	Graph- ite felt	AS	19.5	590	93.9	NA	Scirpus validus was used	[160]

In addition to the main components, the food industrial wastewaters may also contain sub-components such as phenol compounds and nitrobenzene. The presence of these compounds in wastewater may have various effects such as inhibition on

microbial activity which should be investigated. The results of MFCs performance with some wastewaters with sub-components as substrate are presented in Table 6.

#### Phenolic compounds

There are toxic contaminants such as phenolic compounds in industrial effluents. In many industrial wastewaters, phenolic compounds are major toxic contaminants such as chemical, pharmaceutical, textile, food, and oil refining. Compared to the conventional anaerobic degradation, MFCs efficiency are comparable to the high efficiency of 2,4-Dichlorophenol (2,4-DCP) degradation. Phenol as a substrate was used in air-cathode SC-MFCs. The results showed that using the initial concentration of 600mg phenol l-1 with glucose inoculation, Pdm was obtained 31.3mW m<sup>-2</sup> which was 1.9 times higher than using phenol alone, as a substrate. Also using closed circuit voltage and OCV, the phenol degradation values were 89% and 77% respectively with CE obtained 3.68%. Sodium acetate and sodium propionate were other compounds tested for inoculation which were weaker than glucose in all cases. Examining the IR of these cases, the MFC with glucose produced the lowest IR, even compared to MFC with phenol alone; as a result, it is one of the reasons for producing higher Pd. Glucose is a fermentable substrate and during the process in SC-MFCs, it can be broken down into some non-fermentable substrates (such as acetate and ethanol). Glucose may lead to a stimulated growth of the microbial population of the entire anode, which increases the activity of electrochemical biofilms of anode causing different performances [153]. The effluent coconut husk retting contains recalcitrant compounds such as phenol. Thus, it must be treated before being released in the environment. It was used in DC-MFC with batch mode and a Pdm of 362mW m<sup>-2</sup> was obtained. The COD removal of 91% was obtained with CE of 25%. In addition, a high phenol removal of 93% was achieved using a primary concentration of 320g phenol m<sup>-3</sup> [154]. For using phenol in the cathode, investigations were carried out using the ammonium and phenol fed to an aerobic cathode in a SC-MFC. The results showed that the phenol had no certain inhibitory effect on the nitrifying even up to a level of 600mg l-1. Compared to the conventional aerobic bioreactor with the same MFC under open circuit conditions, nitrification and higher rate of N elimination was less suppressed by phenol in MFC. Bacterial analysis showed that the electrochemical active bacteria and denitrifiers in the anaerobic chamber have an important role in the output power and anaerobic denitrification, while in aerobic cathode the degrading bacteria of phenol are responsible for phenol oxidation, with aerobic nitrification performed by nitrifiers and aerobic denitrification by denitrifiers [155].

#### Nitrobenzene

Nitrobenzene is widely used to produce pesticides, paints, explosives, rubber, and other chemical compounds. In the groundwater, the nitrobenzene compounds are highly toxic, bio-accumulative, and are stable in groundwater. In CW, the high concentration of nitrobenzene can inhibit the activity of microorganisms and reduce the nitrobenzene removal efficiency

[156]. SC-MFCs designed with pre-enriched anodes can be useful for nitrobenzene elimination and power generation. A Pdm of over 25W m<sup>-3</sup> was obtained when the nitrobenzene concentration was 1.2-6.2mol m<sup>-3</sup> d<sup>-1</sup>, and steady Pd of over 13.7W m<sup>-3</sup> was generated within nitrobenzene concentration of 1.2-14.7mol m<sup>-3</sup> d-1. A nitrobenzene elimination over 97% was obtained even when the nitrobenzene-loading rate was reached 17.2mol m<sup>-3</sup> d<sup>-1</sup>. The potential product of nitrobenzene reduction such as aniline can be eliminated efficiently [157]. Xie et al [158] combined a membraneless air-cathode SC-MFC with CW for finding a possible treatment with good nitrobenzene removal efficiency. The wastewater was tested on a typical MFC and CW-MFC. Pdm values of 0.59mW m<sup>-2</sup> and 1.53mW m<sup>-2</sup> were achieved for MFC and CW-MFC respectively. The CE for MFC and CW-MFC was 13.9% and 16.4%, respectively. The highest nitrobenzene elimination was obtained 92.89% at CW-MFC while the nitrobenzene concentration to COD was 1:16 with HRT in 24 hours. The COD removal obtained ranged from 67.92% to 78.30% where the CW-MFC was more effective for COD reduction. Also, glucose was applied as a C source in this wastewater, where co-metabolism due to microorganisms and glucose can simulate the nitrobenzene reduction.

# **Municipal Wastewater**

The municipal wastewater is a complete wastewater in terms of structure and constituents; it is a collection of fundamental factors and component. This wastewater emanates from any domestic, industrial, commercial or agricultural activities, surface runoff, storm water, and any sewer inflow or wastewater infiltration. Food waste forms a large part of municipal wastewater which is about 27% [161]. The loss or wasting of food products is a global issue claiming one-third of total wastes [162]. The chemical energy of municipal wastewater is at least about 13kJ g-1 COD and it is approximately 9 times more than the prevalence required to treat this wastewater [163]. To treat the municipal wastewater, approximately 35.2kWh per population equivalent and year is required in Germany [164]. The effluent of the primary sedimentation reservoir is one of the best choices to use as substrate in MFC or integrate MFC into treatment plants; This effluent is relatively low solid content and it minimizes the risk of clogging or sloughing in MFC [164].

Some large-scale MFCs are investigated for municipal wastewater treatment. The stackable horizontal MFC was designed which consisted of several 250L units. During the sustained operating period, A Cdm of 0.435 A was seen in each module where Pdm of 116mW was obtained. The COD removal of 79% and total N removal of 71% were obtained and CE was 5% [165]. In addition, the energy extraction via an MFC with 200L system (effective volume of 100L) was investigated. There were three rows of MFCs in the connection with the highest output power of 114mW. The organic compounds removal of 75% and solids removal of 80% were obtained [166]. Further, in the largest research ever done in terms of scale, a 1000 L modularized MFC

was developed which worked for more than one year (a stack containing 50MFC, with 20L volume for each one). The removal rate of 70-90% was obtained. A Pdm of 125W m<sup>-3</sup> was generated with synthetic wastewater, while it ranged from 7 to 60W m<sup>-3</sup> municipal wastewater. The CE was obtained within the range of 41-75%. Using HRT of 2hr, energy recovery was obtained 0.033kWh m<sup>-3</sup> from municipal wastewater [167]. A 45L pilot MFC includes four membrane-less SC-MFCs combined into a full-scale municipal wastewater treatment plant for 9 months, perhaps it is one of the best ways for integration. The best results were obtained by removing 24%, 40%, and 28% for COD, TSS, and N respectively in HRT of 22 hr. The average Pd was more than one cycle for quaternary cells in the same HRT where Pd of 82mW m<sup>-2</sup> was obtained. The mean normalized energy recovery was obtained 0.36kWh kg<sup>-1</sup>COD degraded and CE was 24.8% [164]. The results indicated that an MFC with balanced function among the individual units would be very important for energy extraction. As such, the poor performance of an MFC had a negative effect on the entire efficiency of the connected MFCs [166].

#### **Domestic wastewater**

Domestic wastewater is kind of wastewater derived from plumbing fixtures and appliances such as toilets (urine, stool and toilet paper), bathrooms, laundry, dishwashers, garbage disposal and cleaning which forms part of municipal wastewater. Given the nature of this wastewater, it seems to be a suitable option for use in MFC. So far, clear and suitable results have been obtained due to the use of this wastewater in MFC. It has even been shown in some cases where the electricity generated by MFC with this wastewater can be used in a practical way in the homes. Now researchers are seeking better results with novelty in configurations, designs, operational parameters and large scales. In this regard, a stack of MFC including 40 individual air-cathode units was examined with a continuous flow. It is evident that the removal of pollutants, electricity production, and microbial communities changed significantly when the MFC stack was changed from unrelated to series or parallel connections. When the units were unconnected, Bacillus and Lysinibacillus were the main bacterial species in the anode. Then, after changing from unconnected to series and parallel connections, Pseudomonas aeruginosa followed by different Bacilli classes formed the dominant species in the stacked MFC. The voltages drop occurred in the MFC stack, which were mainly limited by the cathodes. This loss of voltage indicates that the high IR the MFC stack generated a parasitic cross current. The voltage of 0.08-1.1V in an OCV, COD removal of 74%, NH<sub>4</sub>-N removal of 62% were obtained from individual MFC units. In series connection mode, a Pd of 2500mW m<sup>-2</sup>, voltage of 4.9V, COD removal of 84% and NH<sub>4</sub>-N removal of 80% were obtained. In parallel connection, the Pd of 5.8mW m<sup>-2</sup>, COD elimination of 78% and NH<sub>4</sub>-N removal of 73% were obtained [168]. Further, the power saving, low solids formation and high treatment efficiency are the major benefits of applying temperature-phased

in MFC series configurations [169]. With temperature decline from 27.5°C to 8°C with mesophilic inoculum from a digester, MFC indicated strong proteolysis at low temperature compared to anaerobic digestion. The anaerobic digestion indicated a better performance at warmer situations. A lipases activity test indicated that hydrolyze lipids of anaerobic digestion and MFC were similar at 27.5°C and MFC indicated better lipolysis potential at low temperatures than anaerobic digestion. The switch from anaerobic digestion to MFC altered the microbial community by 15% where the MFC showed the maximum diversification. On the other hand, temperature drop can change the community by 40% [170].

Integration of MFC with other technologies for this kind of wastewater can be useful. In this regard, the effluent of an up-flow membrane-less MFC was used for feeding of photobioreactors. MFC alone generated Pdm of 481mWm<sup>-3</sup>. The COD removal of 77.9%, total P removal of 23.5%, and NH<sub>4</sub>\*-N removal of 97.6% were also obtained. On the other hand, when MFC combined with photobioreactor, 99.3% of total P and 99% of total NH,+-N were removed [171]. In another integrating, an algae biofilm with an MFC was integrated and the removal of total N, total P and COD were obtained 96%, 91.5% and 80.2% respectively which was far superior to MFC or algae biofilm alone. The Pdm was obtained 62.93mW m<sup>-2</sup>, which was 18% greater than when MFC was used alone (52.33mW m<sup>-2</sup>). The CE was 17.01% and it is 36.7% higher than when MFC used alone (12.44%). An energy of 0.094kWh m<sup>-3</sup> was generated overall [172]. The new configurations with novelty for achieving better results or reducing investment cost are always encouraged. In this regard, different spacing of electrodes in some configurations with more than one anode and flow rates affect the current generation and COD removal. The MFC generated 0.22 kWh kg-COD<sup>-1</sup> from low strength domestic wastewater by a flat-panel air-cathode MFC. This kind of MFC is well known for overcoming effluents with the low conductivity and biodegradability. Current generation was same at various spacing, although COD elimination was influenced by flow rates. According to the volume of effluent treated, the normalized energy recovery indicated a good correlation with the flow rates in entire anode different spacing. According to the COD elimination performance and apart from the anode different spacing, the normalized energy recovery indicated a negative correlation with the COD elimination rates [173] Given the importance of investment costs, an up-flow MFCs with polyvinylidene fluoridebased activated C air-cathode which is low cost, easy to configure and with higher stability for low strength wastewater. The average total COD removal rates of 5.11kg COD m<sup>-3</sup> d<sup>-1</sup> and Pd of 3.96W m<sup>-3</sup> were obtained which were even greater than MFCs with Pt cathode. Also, high and steady suspended solid elimination > 90% was obtained without any clogging during the whole operation. In addition, a low voltage booster was applied to improve low voltage generation. The voltage rose from < 0.4V to 4.35-5.2V

while there was no voltage reversal. It was adequate to drive three LED bulbs for>12 days [174].

#### Urine

The urine is a highly abundant waste product that forms part of municipal wastewater. The urine with a high concentration of organic compounds (10g l-1 of COD) mainly includes organic acids, N compounds, and carbohydrates [175]. Proper results have been achieved using raw or diluted urine in MFC. In this regard, an anaerobic microbial community, which was enriched with electroactive degrading urine microorganisms, was utilized. The Cd, Pd, COD removal, and CE during operation on undiluted urine were obtained as 495mA m<sup>-2</sup>, 306.5mW m<sup>-2</sup>, 75.5%, and 26.5% respectively. In comparison to control group (without microbial enrichment), it was found that the addition of microbial acclimation resulted in elevated Cd production by 80%. After 10 days of operation, most organic compounds were reduced except acetate [176]. During 45 days of operation in a SC-MFC with human raw urine, COD was degraded effectively with the COD elimination efficiency being 25-40%, 35-60%, and 60-75% in 1-day cycles, 2-day cycles, and 4-day cycles respectively [177]. Elsewhere, a large scale MFC with 64 units in two stacks was utilized. MFCs with a terracotta ceramic membrane were used, which formed with two stacks where each one had 32 individual units. By collecting the output current of 32 units in parallel connection and by adding the potential of 2 stacks in series connection for more than 120 days operation, an average Pd of 23mW m<sup>-2</sup> was generated with an efficient Cd of 65mA m<sup>-2</sup>. In addition, COD removal of 70% was obtained in parallel connection of stacks, less than 50% in series connection of stacks and 80% when the stacks were separate [178].

The Struvite (magnesium ammonium phosphate-MgNH,PO, 6H,O) is normally formed in the urine because of chemical reactions. Its recovery has two great benefits. The first is containing N and P, which can simultaneously remove them and improve the quality of the effluents and the scond is reduction in operating costs. Further, it can cause reduced blockage of tubes in the wastewater treatment plants [179,180]. In addition, the struvite can be applied in the role of a slow-release fertilizer [181,182]. At such, an integrated system combining the struvite process with an MFC can recover P from the urine successfully and treat the urine [183]. The struvite precipitation mainly depends on the wastewater condition such as pH and ammonium level [184]. As an acceptor, the nitrate can be used in a cathode, which allows the concurrent elimination of C and N in the anode and cathode respectively [185]. In general, in the domestic wastewater, N is in NH, +-N form; therefore aeration in MFCs is necessary for ammonium oxidation, nitrification, and denitrification [185]. The precipitation of struvite crystals increases in the urine via addition of magnesium. Several sources of magnesium, including MgCl<sub>2</sub>, synthetic sea water and a mixture of commercial sea salts are used for sea water preparation to be combined with real and fresh urine. Commercial sea water showed the best results in

terms of struvite precipitation with the amount of struvite in the collected solids growing from 21% to 94%. In addition, the sea water increased the maximum power efficiency of MFC by more than 10% and elevated pH, conductivity, concentration of chloride ions, and it changed the properties of the collected catholyte [186]. A three-phase MFC system and the struvite extraction process were developed with two MFC groups used (each group containing 4 MFC units). In the first phase, the untreated urine was entered into the first MFCs group. MFCs improved the urea hydrolysis and it was useful for the struvite precipitation process in the second phase. In the second phase, the MgCl, was added to the effluent of these MFCs and the struvite precipitation process was performed. In the third phase, the effluent was introduced into the second group of MFCs, after separating the struvite sediments. Using a three-phase system, 82% of PO<sub>4</sub><sup>3</sup>-P and 20% of COD were eliminated from undiluted human urine. Also during the operation, Pd of 14.32W m<sup>-3</sup> at phase 1 and 11.76W m<sup>-3</sup> were obtained at phase 3 [187]. The results were appropriate when urine was treated by the MFC, as well as with integration of struvite process with MFC.

#### Stool

The human stool wastewater contains plentiful organic compounds. An automated MFC was designed operating with astronaut's stool on a day. It was used in MFC. When the DC-MFC operation was performed with real human stool wastewater for 190 hours, Pdm of 70.8mW m $^{-2}$ , COD removal of 71%, soluble COD removal of 88%, and NH $_4^+$  removal of 44% were obtained [188]. The pre-treatment is method useful for increasing the performance. For adjusting the pH, the HRT was 5h. The Pdm of the MFC fed with pre-treated human stool was obtained 22mW m $^{-2}$ . This was 47% higher than the non-pre-treatment control with a Pd of 15mW m $^{-2}$  [188]. Considering the configuration tips, which were mentioned, it was found that the capacity of electricity generation increased by enhancing electrode surface area and shortening the distance between the electrodes. The total power of 787.1mW and Pdm of 240mW m $^{-2}$  were also obtained [188].

#### Conclusion

The community has completely recognized that the wastewater can be a valuable resource for energy generation and other compounds recovery, rather than a source of useless or harmful pollution. The MFCs have brought new opportunities as an emerging innovative technology. The MFCs apply organic compounds directly to produce energy. At the same time, they convert the substrate into electricity and treat the wastewater via their oxidation. Food industry wastewaters as a substrate meet most of the requirements of this technology in wastewater treatment. Note that due to the biological nature of the process, some parameters are determinant. In this regard, adjusting the temperature, pH, IR, HRT, OLR, conductivity and dissolved oxygen are very important. Each of them can affect the process with inhibitory effect. Sometimes pre-treatment is required for performance improvement or starting time reduction. Changes

in designing are an appropriate method for performance improvement and investment cost reduction. Integration with the other technologies can also expand the implementation of MFCs on large scales of application. All these issues have been addressed in the text. With great progress, the output power of MFCs has increased significantly over the past decade which is due to scientific and technological advancements. However, this technology still has weaknesses to be recognized as an independent treatment and energy generation system.

Finally, more investigations should be performed to better understand the potential, stable MFCs' capability, answering to the challenges and identifying the future route. The MFC technology has not yet found its position on operational scales due to major challenges including high investment costs, important bottlenecks, increasing system scales, long-term operation, output power, energy recovery, microbial process and integrating. So far, good progresses have been made in all of these cases, but as MFC plays a significant role in the pattern of future changes of wastewater treatment and bioenergy production, more innovative research for improvement and better performance are required.

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