

S. Brunschweiler, T. Hofmann and K. Glas

Industrial wastewater treatment with simultaneous energy recovery using microbial fuel cells – a review

The dual benefit of microbial fuel cells (MFC) for wastewater treatment with simultaneous power generation opens new perspectives and possibilities for industrial wastewater treatment. At present, research focuses on municipal wastewater instead of industrial wastewater treatment, which may be more attractive due to savings in discharge fees, if the required treatment efficiency can be achieved. Accordingly, the objective of this literature review is to clarify whether MFCs can be used for industrial wastewater and whether they are currently competitive with conventional anaerobic-aerobic wastewater treatment plants. Therefore, an overview of MFCs treating industrial wastewater (e.g. breweries, dairy, paper) on a laboratory or pilot scale will enable an assessment of possible applications. Self-sufficient wastewater treatment and further nutrient removal (nitrogen, phosphorus, etc.) will be of additional interest due to possible energy recovery and the tightened discharge limits. The assessment of MFCs with regard to conventional anaerobic or aerobic treatment was pointed out, and combination possibilities were explored. The feasibility of MFCs for the treatment of industrial wastewater and their possible self-sufficient treatment has been demonstrated by several laboratory and pilot scale studies. MFCs can treat both low and high chemical oxygen demand (COD) concentrations, but do not require aeration as in aerobic treatment and, unlike anaerobic treatment, can be operated on a small scale. In addition, the removal of nitrogen, total suspended solids (TSS) and sulphates is possible with MFCs, only the removal of phosphorus is not expected. The modularity and the wide range of operation are the main advantages of MFCs, making MFCs an alternative or complement to conventional industrial wastewater treatment.

Descriptors: microbial fuel cell, industrial wastewater treatment, energy recovery, nutrient removal

1 Introduction and MFC principles

The main application of microbial fuel cells (MFC) is in biological wastewater treatment, where they simultaneously generate electricity from the organic compounds in wastewater. MFCs consist of an anode, where exoelectrogenic microorganisms can settle in the form of a biofilm, and a cathode, on which reduction takes place (Fig. 1). A proton exchange membrane (PEM) can separate the anode and cathode chamber. The cathode can be either an air cathode (single chamber MFC) or be placed in a chamber (double chamber MFC). As a result of the biofilm with exoelectrogenic microorganisms on the anode, oxidation of the organic wastewater components takes place, which produces both electrons (e^-) and protons (H^+). The electrons and protons are transferred to the cathode, where oxygen is reduced to water. [56]

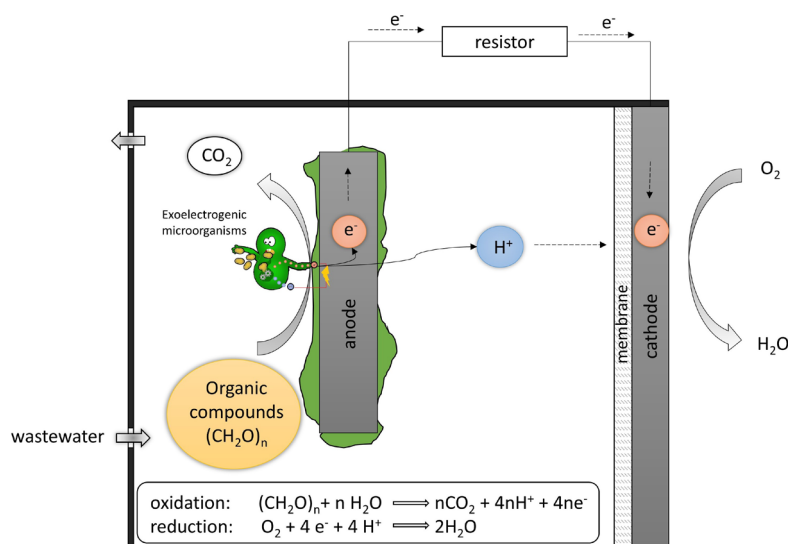


Fig. 1 A schematic representation of a MFC (adapted from [43])

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Authors

Sarah Brunschweiler, Thomas Hofmann, Karl Glas, Chair of Food Chemistry and Molecular Sensory Science, Technical University of Munich, Freising, Germany; corresponding author: sarah.brunschweiler@tum.de

A wide range of exoelectrogenic microorganisms or electroactive bacteria can degrade a variety of wastewater from different sources like domestic, industrial, agricultural etc. [68]. *Geobacter sulfurreducens* and *Shewanella oneidensis* are most commonly studied as electroactive bacteria [22]. Mixed cultures (e.g. anaerobic sludge as inoculum) dominated by different phylogenetic groups

like *Firmicutes*, *Acidobacteria*, *Proteobacteria* etc. can also be used [13]. The type of microbial community dominating a mixed biofilm depends on the source of inoculum, operating conditions, substrate availability and MFC system architecture [13].

In general, competitions in anaerobic mixed cultures can be controlled by parameters such as pH or hydraulic retention time (HRT) [27]. The most important concurrent organisms are methanogens, which can metabolize acetate and hydrogen to form methane [27]. This results in a trophic competition with exoelectrogenic microorganisms and a decrease in coulombic efficiency (CE) [27]. The CE value describes the fraction of the degraded substrate effectively converted into electrons [56].

$$CE = \frac{M \times \int_{t_0}^t I(t)dt}{F \times n \times V_{MFC} \times \Delta COD} \quad (Eq. 1)$$

- M molar mass of oxygen (32 g/mol)
- I produced current (A) integrated over time t (s)
- F Faraday's constant (96,485 C/mol)
- n number of electrons exchanged per molecule oxygen (4)
- V_{MFC} liquid volume of the MFC (L)
- ΔCOD degraded concentration of the COD (g/L)

MFCs usually generate less operating voltage with respect to the electromotive force, because of potential losses or overpotentials [68]. The overpotentials can be divided into the (a) activation overpotentials, (b) ohmic losses and (c) concentration overpotentials [68].

The voltage efficiency (VE) is the ratio between the produced voltage (E_{MFC}) and the theoretical maximum voltage (E_{emf}), which can be calculated using the Nernst equation. E_{emf} can be assumed to be 1.1 V considering MFC being used as acetate as substrate and oxygen as an electron acceptor. [90]. The overpotentials of the MFC determines the difference between E_{emf} and E_{MFC} .

$$VE = \frac{E_{MFC}}{E_{emf}} \quad (Eq. 2)$$

MFC performance and efficiency is generally measured in terms of power density (normalised to anode or cathode surface area or the treated volume) and chemical oxygen demand (COD) treatment efficiency [13, 56].

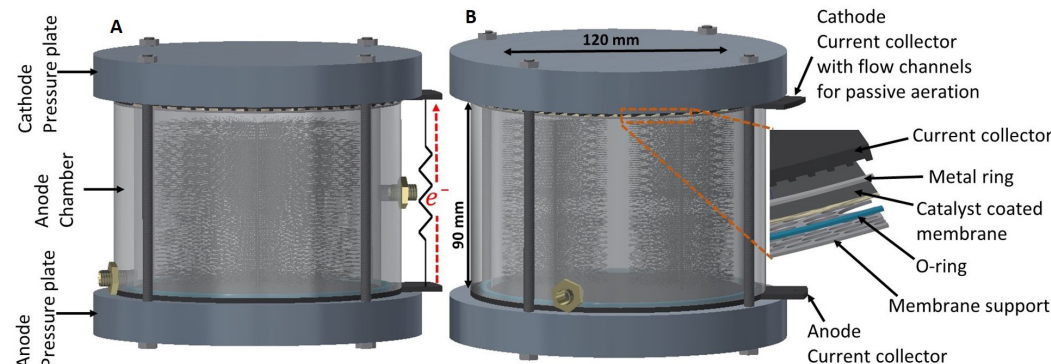


Fig. 2 Sketch of a 1 litre single-chamber MFC with A) one carbon fiber brush B) two small carbon fiber brushes for brewery wastewater treatment [9]

$$PD_{cat./an} = \frac{E_{MFC} \times I}{A_{cat./an.}} \quad (Eq. 3)$$

$$PD_{vol} = \frac{E_{MFC} \times I}{V_{MFC}} \quad (Eq. 4)$$

$$\Delta COD = \frac{COD_0 - COD_t}{COD_0} \times 100 \quad (Eq. 5)$$

$PD_{cat./an}$ power density normalised to the anode or cathode surface area (W/m^2)

$A_{cat./an.}$ surface area of the anode or cathode

PD_{vol} power density normalised to the volume (W/m^3)

COD_0 COD concentration (g/L) in the beginning

COD_t COD concentration (g/L) after time t

Normalised energy recovery (NER) combines electrochemical performance with the efficiency of wastewater treatment. This parameter can describe either the power generated by the MFC system from the degraded COD concentration (NER_{kgCOD}) or from the treated wastewater volume (NER_{vol}) [30].

$$NER_{kgCOD} = \frac{P \times t}{V_{MFC} \times \Delta COD} \quad (Eq. 6)$$

$$NER_{vol} = \frac{P \times t}{V_{MFC}} \quad (Eq. 7)$$

The main challenges of MFC is scaling up the energy output in relation to the amount of wastewater treated [13]. The overall performance of MFCs can be improved either by increasing the volume of MFCs or by connecting several MFCs in electrically stacked systems (serial or parallel) [13], because one MFC can generate a maximum of 1.1 V [89].

Figures 2 and 3 show an example of a sketch of a 1 litre MFC and a picture of a scale-up system of a 3 x 33 L MFC stack for the treatment of brewery wastewater.

In this case, carbon fiber brushes with a high surface area serve as anode and the so-called membrane cathode assembly (MCA), consisting of a membrane coated on one side with 0.5 mg/cm² activated carbon, as cathode [9].

The MFC research is often focused on the treatment of simple substrates (e.g. acetate), synthetic or municipal wastewater. A smaller number of studies utilised industrial wastewater and

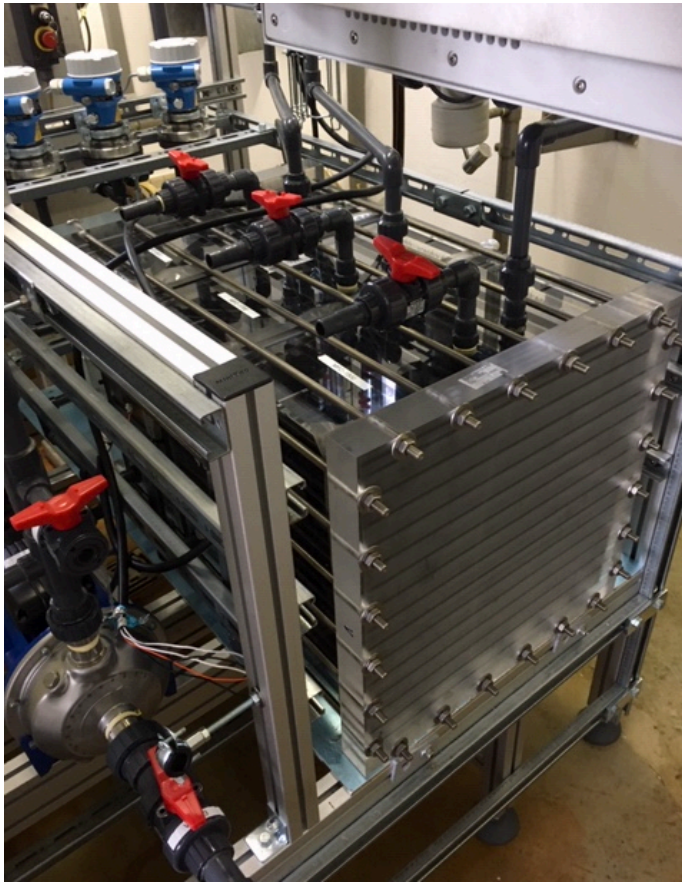


Fig. 3 Picture of a 100 litre MFC (3 x 33 L stack system) for brewery wastewater treatment [85]

only a few dealt with swine or paper wastewater [1]. Industrial wastewater differs from municipal wastewater mainly by its higher COD concentrations (e.g. brewery wastewater) [34]. The application of conventional anaerobic treatment is mainly used for industrial wastewater treatment to reduce high COD loads [80]. In MFCs, the organic contents are oxidised under anaerobic conditions, so that this application should also be suitable for industrial wastewater. Furthermore, the discharge regulations and thus the limit values in industrial wastewater treatment deviate from the requirements of municipal wastewater, so that more opportunities can probably be derived for the use of MFCs.

This paper clarifies the possible applications and requirements for MFCs for the treatment of industrial wastewater (as opposed to municipal wastewater). Accordingly, it aims to answer the hypothesis whether MFCs can be used for industrial wastewater and whether they are competitive with conventional anaerobic-aerobic wastewater treatment plants. A current overview is provided of the state of the art of MFCs for the treatment of industrial wastewater, considering different type of wastewaters (e.g. brewery, dairy and swine wastewater). The performance (power densities & COD degradation) of MFCs on a laboratory and especially on a pilot scale is considered under the aspect of commercialisation of MFCs. A self-sufficient treatment and the further removal of nutrients will therefore be of interest in finding application niches for MFCs. Consequently, an evaluation of MFCs with respect to conventional biological wastewater treatment will be possible and it will be shown how MFCs can be used standing alone or in combination.

2 Delimitation from municipal wastewater treatment

Industry and municipal wastewater can be treated with the same biological treatment systems (anaerobic and/or aerobic). However, due to different compositions and discharge limits, the wastewater treatment systems must be adapted accordingly.

2.1 Requirements for industrial wastewater

Industrial wastewater treatment is regulated to comply with municipal bylaws and to reduce the load on the municipal treatment plant. Discharge fees may be imposed on effluent volume, as well as on suspended and organic loads [84]. The tightened requirements on industrial wastewater quality lead to increased cleaning and wastewater costs, including high pollution surcharges [34]. The type of pre-treatment or the complete treatment depends on the wastewater quality and the opportunities for connection to the public sewage system, so that there is a difference between (A) indirect and (B) direct dischargers [79].

In the case of indirect dischargers, the plant manager of a municipal treatment plant determines the quantity and quality requirements to meet the statutory obligations [79]. In Germany, the DWA M 115 information sheet, which can be consulted for support, defines the requirements for indirect discharges of non-municipal wastewater [19]. Consequently, a company with its own pre-treatment facilities can already reduce the charges for heavy polluters but does not have to meet the tighter requirements of a direct discharger.

The threshold for wastewater for direct dischargers in Germany are set by the Wastewater Ordinance, which also meets all the requirements of the EU's water-related best available techniques (BAT) [79]. The requirements are specified for several branches so that, for example, in breweries with direct discharges, the following thresholds have to be reached: 25 mg/L Biological oxygen demand (BOD_5); 110 mg/L COD; and 10 mg/L NH_4-N , 2 mg/L P_{ges} , 18 mg/L N_{ges} [3].

2.2 Treatment of industrial wastewater in contrast to municipal wastewater

To meet the requirements for direct or indirect discharge, anaerobic or aerobic treatment or a combination of the two is used as a conventional biological method.

Anaerobic treatment of municipal wastewater is less common because of the comparatively low temperature and low COD concentrations [80]. In contrast, more than 258 large-scale anaerobic plants have been built in the field of industrial wastewater treatment (especially in the beverage and food industry sector) in Germany (Status: 03/2012) [80]. With anaerobic reactors, the highly polluted and easily degradable industrial wastewater can be easily decomposed and, at the same time, biogas can be produced [80].

Compared to aerobic wastewater treatment, anaerobic treatment requires fewer chemicals, less sludge production, less space

Table 1 Overview of MFCs treating industrial wastewater with a volume < 10 L

Type of wastewater	MFC volume (anode) [L]	HRT [h]	Total operating time	COD conc. ^b [mg/L]	COD removal [%]	Max. PD _{vol} [W/m ³]	Max. PD _{cat} [mW/m ²]	Max. CE [%]	Ref.
Brewery	5.4	15	5 months	1168	33	5.0	–	4	[28]
	1	48	180 days	3380	83	0.2	21*	–	[9]
	0.5	144	75 days	1410	> 96	0.5	62*	12	[38]
	0.275	24	45 days	2200	30	0.4*	158*	1	[52]
	0.25	12	87 days	2250	82	0.2*	8a	–	[11]
	0.225	168	130 days	510	100	2.9*	552	41	[100]
	0.18	2	–	1501	21	24.1	669	3	[97]
	0.17	–	–	2125	91	29.9	854*	65	[107]
	0.1	15	–	1250	49	0.8	23	–	[96]
	0.05	72	–	3226	64*	5.7*	191 ^a	–	[101]
	0.028	< 96	–	2250	87	5.1	205 ^a	10	[26]
0.028	–	20 days	2250	79	11.0	435 ^a	29	[95]	
Winery	0.125	72	–	7800	59	3.8	–	45	[76]
	0.07	–	–	6850	17	4.7*	465 ^a	15	[72]
	0.028	144	–	2200	65	0.2*	9*	18	[12]
Dairy	2	72	30 days	3620	90	5.8*	621 ^a	37	[61]
	1.5	96*	48 days	1900	86	1.7*	230 ^a	21	[60]
	0.55	–	35 days	3700	96	1.10	75*	14	[92]
	0.48	8	20 days	2360*	55	1.90	92 ^a	10	[24]
	0.45	–	–	2216	56	7.8*	–	14	[8]
	0.435	–	72 days	–	85	27.0	–	56	[66]
	0.43	–	72 days	–	80	27.0	–	46	[10]
	0.35	192	–	1600	91	2.7	161 ^a	17	[21]
0.09	72	450 hours	1000	91	20.2	161*	27	[62]	
Starch processing	0.425*	840	140 days	4852	98	1.4*	239 ^a	8	[59]
Chocolate	0.4	–	–	1459	75	–	1500 ^a	–	[71]
Potato processing	0.4	400	–	–	87	–	–	2	[18]
Cheese whey	0.31	50	–	730	100	0.1*	18 ^a	2	[6]
Slaughterhouse	0.684*	30	120 days	1000	99	–	165 ^a	–	[46]
Swine	3.7	22	62 days	1058	77	–	9 ^a	<1	[104]
	0.295 x 5	60	–	5845	77	2.2*	176	<1	[106]
	1	–	21 days	5400	86	–	88 ^a	–	[20]
	0.1	17	185 days	7000 – 8000	59	30.0*	750	–	[49]
	0.028	–	–	8320	86	–	261 ^a	8	[65]
Paper	0.3	350	–	1460	29	0.2	144	–	[45]
	0.028	6	–	506	26	5.9	210	30	[44]

* Calculated on the basis of data in paper

Some key figures have been rounded to ensure consistency

^a Power density was normalised to the anode surface area

^b Initial COD concentration treated by MFCs

and less energy, since instead of energy-intensive aeration being performed, energy is actually obtained in the form of biogas [34, 80, 84]. However, aerobic treatment is often required for direct discharge after anaerobic treatment, as the COD degradation is often only about 65 – 95 % and the nitrogen cannot be removed [80].

In summary, for industrial wastewater treatment a combination of anaerobic-aerobic systems is often used unlike with municipal

wastewater treatment [84], in which aerobic wastewater treatment is preferred.

2.3 Targets for industrial wastewater treatment

The industry's intention to pre-treat its wastewater, as opposed to municipal wastewater treatment, results from different motives, requirements and costs. Municipal wastewater treatment focuses

Table 2 MFC studies treating different wastewater with the same MFC configuration

Type of wastewater	MFC volume (anode) [L]	HRT [h]	Total operating time	COD conc. ^b [mg/L]	COD removal [%]	Max. PD _{vol} [W/m ³]	Max. PD _{cat} [mW/m ²]	Max. CE [%]	Ref.
Bakery	0.045	60	350 hours	651	86	0.1*	3*	2	[91]
Brewery				661	85	0.1*	3*	2	
Paper				600	78	2.7*	60	26	
Dairy				700	82	0.3*	6*	2	
Brewery	0.6	240	15 days	1778	87	3.4*			[64]
Sugar				1229	90	3.7*			
Dairy				1487	81	2.0*			
Municipal				1235	85	2.5*			
Paper				1581	76	1.8*			
Agriculture	-	-	-	397	68*		12	<<1	[67]
Domestic				671	72*		42	<1	
Paper				1250	79*		82	2	
Food / dairy				1562	88*		150	3	
Brewery	0.25	96–120	-	3574	93	0.3*	188*	2–3	[5]
Brewery + swine (50:50)				5028	53	0.3	213	11	

* Calculated on the basis of data in paper

Some key figures have been rounded to ensure consistency

^a Power density was normalised to the anode surface area

^b Initial COD concentration treated by MFCs

on reducing energy consumption [42], whereas industry focuses on COD reduction to meet the discharge qualities. Therefore, the targets for in-house wastewater treatment include the reduction of municipal wastewater charges and overheads, a cut in the high pollution surcharges, a reduction of wastewater charges through compliance with the minimum requirements for discharge into the aquatic environment and an improved guarantee to comply with the requirements for the discharge of wastewater into the public sewage system (indirect discharge regulation) [34].

pilot plants for municipal and industrial wastewater treatment have been realised, so that commercialisation can be evaluated based on the key figures (Table 3, see page 116).

A direct comparison of MFC studies is not possible due to several influencing parameters. To obtain an overview, tables 1 – 3 are beneficial in determining the range in which power densities and COD removals are possible. In these studies, the type of inocula,

In theory, both discharge options can be investigated with MFC technology, since modularity allows the required limit values to be adjusted, which will be discussed in detail in Section 4. These two discharge options increase the flexibility of using the MFC option as compared to municipal wastewater treatment. Depending on the wastewater composition and the different municipal requirements, niches may arise, in which MFC technology has a high potential.

3 Performance of MFCs in industrial wastewater treatment

Research on MFCs for wastewater treatment often focuses on the application of municipal wastewater. However, there are some studies on the wastewater treatment of various industrial effluents (Table 1 & 2). A major challenge for many innovative technologies is still the scale-up from laboratory to pilot scale. A few

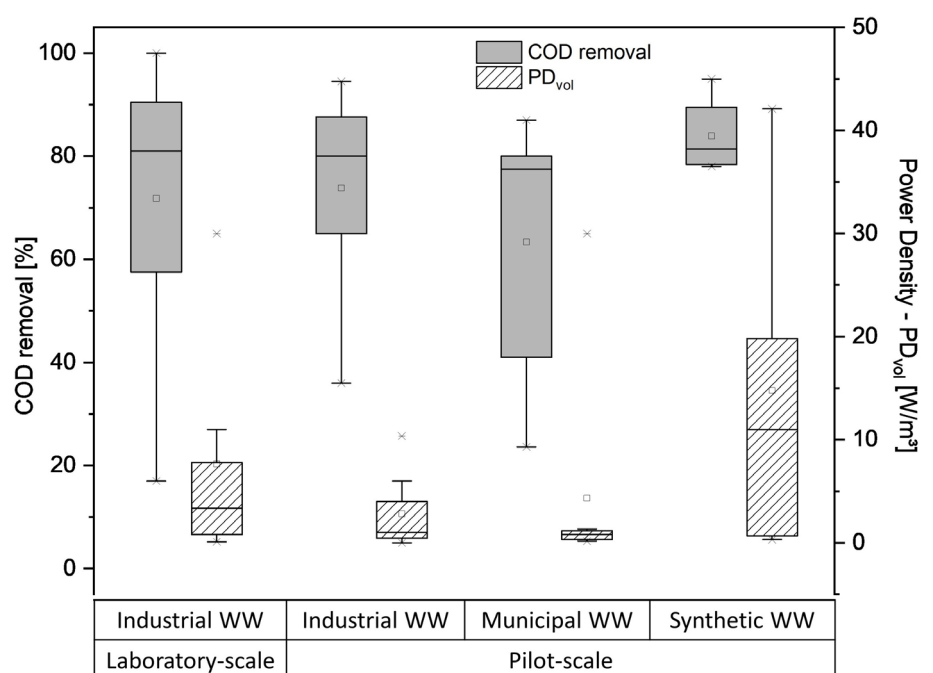


Fig. 4 COD removal efficiencies and power densities (PD_{vol}) of different wastewaters (WW) in laboratory and pilot scale

Table 3 Pilot-scale MFC treating wastewater

Type of wastewater	Total MFC volume [L]	HRT [h]	Total operation time	MFC stack system	COD conc. ^b [mg/L]	COD removal [%]	Max. PD _{vol} [W/m ³]	Max. PD _{cat} [mW/m ²]	Max. CE [%]	NER _{vol} [kWh/m ³]	NER _{COD} [kWh/kg _{COD}]	Ref.
Brewery	90	144	6 months	5 x 18 L SC	3317	88	1.0	159.0	8.0	0.097	0.033	[17]
	20	313	> 1 year	2 x 10 L SC	3197	95	0.4	1.6	13.9	0.120	0.058	[58]
	10	48	180 days	40 x 0.25 L SC	2120	87	6.0	0.1	7.6	0.288*	0.157*	[105]
Distillery	64	151	90 days	1 x 64 L SC	15000	72	2.6	0.01	–	0.387*	0.036*	[51]
	60	96	70 days	1 x 60 L SC	10000	80	10.4	0.02	–	0.995*	0.124*	[51]
Industrial	15	10	–	1 x 15 L SC	1275	90	0.001 ^a	–	–	0.024	0.320	[23]
Swine	100	120	120 days	2 x 50 L SC	7200	52	0.03*	10*	3.0	0.096	0.026	[35]
	90	4	> 2 years	12 x 8 L DC	1000	65	0.9	105	7.0	0.040*	0.110	[7]
	65	9.6	> 6 months	6 x 10.8 L DC	2470	36	4.0	–	17.0	0.034	0.038	[93]
Municipal ^c	1000	2	> 1 year	50 x 20 L DC	250	70 – 80	30	0.73	75.0	0.033	0.048	[53]
	720	36	255 days	6 x 120 L SC	2331*	87	–	7.29**	–	–	–	[14]
	255	43	98 days	1 x 255 L SC	205	41	0.176 ^a	43 ^a	29.5	0.007	0.184	[42]
	250	144	1 year	1 x 250 L SC	333*	79	0.47	58*	3.0	0.054*	0.206*	[25]
	192	12	> 1 year	96 x 2 L SC	155	38	1.0 ^a	6.6 ^a	–	0.013*	0.218*	[29]
	96	12	–	48 x 2 L SC	156	79*	1.35*	17.25*	–	0.016*	0.496*	[31]
	85	264	–	1 x 85 L SC	376	76	0.7*	83.0	27.0	–	–	[81]
	80	288	9 months	1 x 80 L SC	200	50	–	176.0	–	–	–	[87]
	45	22	9 months	4 x 11,2 L SC	118	24	0.88 ^a	82 ^a	24.8	0.012	0.360	[41]
20	20	≈ 105 days	1 x 20 L DC	1000	80	0.17 ^a	0.06 ^a	0.3	0.003*	0.007*	[47]	
Synthetic ^c	72	1.25	6 months	6 x 12 L DC	800	78	42.10	–	–	0.05*	0.337*	[98]
	50	18	–	4 x 25 L DC	700	95	19.8 ^a	–	29.1	0.356*	0.536*	[54]
	45	16	> 1 year	1 x 45 L DC	500	84	0.32*	79.3*	2.03	0.005*	0.012*	[33]
	26	8*	14 months	3 x 8.7 L SC	1000	79	0.69*	–	5.10	0.005*	0.007*	[32]
	20	0.125	–	4 x 5 L DC	–	–	11.0	–	–	0.001*	–	[15]

* Calculated on the basis of data in paper ** Power density measurements have been conducted for each MFC (MFC1-MFC6)

^a Average power density not maximum ^b Initial COD concentration treated by MFCs

^c Municipal includes municipal, domestic and sanitary wastewater; synthetic includes defined medium similar to municipal wastewater composition;

SC: single chamber; DC: double chamber, PD_{vol}: power density (volumetric), PD_{cat}: power density (cathode surface area), CE: Coulomb Efficiency, NER: Normalised Energy Recovery;

Some key figures have been rounded to ensure consistency

the electrode materials, configuration (single or double chamber) and operating conditions are not considered, which sometimes results in large differences in the key figures (COD removal efficiencies, power densities, normalised energy recovery (NER) values). However, some trends can be identified on a laboratory and pilot scale. Furthermore, energy self-sufficient wastewater treatment and the additional removal of other nutrients (nitrogen, phosphorus, TSS, sulphate) in combination with COD degradation can be demonstrated by using MFCs.

3.1 MFCs on laboratory scale

For the treatment of industrial wastewater, which includes wastewater from breweries, wineries, dairies and distilleries, but also swine and paper wastewater, several laboratory-scale MFC studies with a volume of 0.028 – 5.4 litres were summarised in table 1.

The common feature of these wastewater types is the high COD concentration (up to 8320 mg/L), which needs to be removed with MFCs. The process management of MFC can be batch, fed-batch or continuous with currently a median hydraulic retention time (HRT) of 60 h within a wide range of 2 h to 840 h, in which the median COD removal efficiency of 81 % could be achieved. In contrast to COD removal, the power densities are much more dependent on the electrode materials and the MFC configuration (Fig. 4). The median power density amounts to 3.5 W/m³ or 171 mW/m², with the volume or the electrode surface being decisive due to the normalisation of the power output. The Coulomb Efficiency (CE) fluctuates greatly (within a range of 1 – 65%) due to the different inocula and wastewater compositions. This results in a median CE value of 15 %. The wide range of key figures is independent of the type of wastewater, as confirmed by several studies on the treatment of brewery wastewater. Here, the COD

removal efficiencies vary between 21 – 100 % and power densities between 0.2 – 27.0 W/m³.

The suitability of different types of wastewater treated with the same MFC configuration and operating conditions has been investigated in a few studies [5, 63, 64, 67, 91], allowing a direct comparison of different wastewater matrices (Table 2).

According to *Nimje* et al., food/dairy wastewater achieved the highest COD removal rate with 88 % and power density of 150 mW/m² in comparison with paper, domestic and agriculture wastewater [67]. In addition, a higher CE value was measured, so that a larger community diversity of either fermentative and/or exoelectrogenic bacteria at the higher COD concentrations and slightly acidic pH value could settle in food/dairy wastewater [67].

With the simultaneous treatment of brewery, sugar, dairy, municipal and paper wastewater by MFCs, similar current densities and COD degradation rates could be achieved. The influencing factors of temperature, agitation and COD concentration of the wastewater had the same effect on each type of wastewater. [64]

Angosto et al. showed that with a mixture of brewery wastewater and swine manure (50:50) a higher power density and CE value with 11 % could be achieved instead of just 2 – 3 % for pure brewery wastewater [5]. Therefore, at a high COD load, high conductivity and in an environment with a higher nitrogen content than standard brewery wastewater, the COD is likely to be reduced by the more electroactive species. However, the COD removal in the mixed sample (53 %) was much lower than in the pure brewery wastewater (93 %). Some possible reasons for this could be antibiotics in swine faeces that inhibit microbial growth or the higher initial COD concentration. [5]

The reason for varying power output or COD removal is difficult to determine due to the different initial COD concentrations and other influencing parameters (pH, nitrogen, etc.). Therefore, *Velasquez-Orta* et al. adjusted brewery, dairy, bakery and paper wastewater to the same initial COD concentration and also considered influencing parameters such as conductivity [91]. Regardless of the type of wastewater, the COD removal was in the same range (78 – 86 %). An improvement in the current production was observed for all four types of wastewater due to the increase in wastewater conductivity. However, treating paper wastewater resulted in a ten times higher current density than with brewery, bakery and dairy wastewater. Different anodic biofilm communities were detected by microbial and electrochemical analysis, with the highest exoelectrogenic bacterial content found at the anode of the MFC for treating paper effluents. Furthermore, the CE values for MFCs fed with paper wastewater were 26 %, an order of magnitude higher than for MFCs fed with dairy, brewery and bakery wastewater (2 %). [91] In contrast, in the study by *Mathuriya* et al., in which the initial COD concentration was also adjusted, the COD removal of paper wastewater was lower than dairy and potato wastewater [63].

The answer to the hypothesis of MFCs being able to remove high initial COD concentrations as in conventional anaerobic treatment can be confirmed regardless of the type of wastewater. Differences in power output could be caused by differences in the composition

of the biofilm, which can settle in the type of wastewater. Therefore, to increase the power output, the biofilm must be analysed, and the wastewater must be adjusted (e.g. pre-acidification) so that the exoelectrogenic fraction is increased. The MFC configurations and electrode materials are essential for an overall high performance.

3.2 Pilot-scale MFCs

There are currently only a few publications on pilot scale MFCs (10 – 1000 litres), so that in addition to the MFCs for industrial wastewater treatment, municipal wastewater treatment is also shown in table 3.

The median COD removal of MFCs treating industrial wastewater is comparable to laboratory-scale MFCs, at 80 % in an HRT range of 4 – 313 h (Fig. 2). Due to the higher internal losses, the median power densities are considerably lower (1.0 W/m³ or 1.6 mW/m²). In addition, the cathode specific surface area is often reduced in larger reactor sizes, resulting in lower power density. [57]. Therefore, MFC stacking systems are implemented on a pilot scale to minimise the losses (Table 2). The long total operating time of MFCs, usually several months up to several years, indicates overall valid key figures.

Regardless of the type of wastewater a high median COD removal of approx. 79 % was achieved. For industrial and municipal wastewater, the HRT varies between 10 – 313 h and 2 – 288 h, respectively, but the median HRT for municipal wastewater is much shorter at 29 h than industrial wastewater treatment at 120 h. Consequently, similar COD removal efficiencies can be obtained. Calculating the organic loading rate (OLR) and organic reduction rate (ORR) the differences become obvious because the median OLR and ORR of industrial wastewater treatment is much higher with 1.4 and 0.9 kg/(m³/d) in contrast to municipal wastewater with 0.11 and 0.04 kg/(m³/d).

As there are not many pilot-scale studies on wastewater treatment and different MFC configurations, as well as process conditions, it is difficult to compare CE values and power densities. However, the variation in power densities is shown in comparison with synthetic wastewater in figure 2. This shows that higher COD removal and performance densities can be achieved with a defined medium without interfering or unknown substances compared to industrial or municipal wastewater.

No significant conclusions can be drawn on the use of pilot-scale MFCs, as there are only a few studies with different MFC configurations on industrial wastewater treatment. However, according to *Brunschweiler* et al. a further pilot-scale MFC is to be analysed for brewery wastewater treatment with a total volume of 100 L [9].

3.3 Self-sufficient wastewater treatment with MFCs

The power density of MFCs does not indicate energy recovery and does not include factors such as wastewater flow rate or organic removal rate, so making a direct comparison between different studies almost impossible [30]. The information on NER helps to establish an energy balance in order to assess a possible self-sufficient treatment process using MFC technology [30].

In relation to the pilot-scale MFCs treating industrial wastewater, a median energy recovery of 0.097 kWh/m³ (0.058 kWh/kg_{COD}) was calculated on the basis of the references in table 3. In contrast, the median energy recovery of MFCs treating municipal wastewater is approx. 0.013 kWh/m³ (0.206 kWh/kg_{COD}). The same discrepancy between industrial and municipal wastewater was reported by Ge et al. [30]. The average energy recovery of MFCs treating domestic wastewater resulted in 0.04 kWh/m³ (0.17 kWh/kg_{COD}) and the treatment of industrial wastewater resulted in 0.1 kWh/m³ (0.04 kWh/kg_{COD}) [30]. The higher NER_{vol} of industrial wastewater is probably the result of higher conductivities and better electrochemical conditions. On the other hand, the lower NER_{COD} is the consequence of higher COD removal rates or ORR in industrial wastewater.

A few pilot-scale studies [7, 17, 29, 35, 103] not only monitored the energy recovery of MFCs, but also have already compared energy demand and energy recovery with regard to self-sufficient treatment.

Dong et al. and Zhang et al. achieved positive energy balances with 0.034 kWh/m³ (brewery wastewater) and 0.0092 kWh/m³ (municipal wastewater), respectively [17, 103]. In addition, up to 78 % of the theoretical energy needed, including the aeration of the cathode, could be supported by MFCs in the study of Ge et al. [29]. Pumps for feeding and recirculation or other devices in the circuit mainly caused further energy consumption [17, 29, 103].

While treating swine wastewater, Babanova et al. and Goto et al. calculated the energy recovery in comparison with the conventional aerobic and anaerobic treatment [7, 35]. According to Babanova et al. with 0.11 kWh/kg_{COD} more energy could be recovered than with anaerobic digestion, with the energy recovery of methane operating the same HRT of 4 h with only 0.04 kWh/kg_{COD} [7]. Here, the energy recovery is not only competitive with conventional anaerobic treatment but is in fact sufficient to sustain the system's operational energy requirements [7]. However, the NER value recovered by anaerobic digestion cited by Babanova et al. is extremely low. In contrast, the 100 L MFC of Goto et al. produced only 0.026 kWh/kg_{COD}, which was less efficient than using biogas-based electricity generation [35]. To achieve the same COD removal efficiency, an energy consumption of about 4.3 kWh/m³ by aerobic treatment is necessary [35]. 54 % of this energy requirement can also be reduced by combining MFC with a post-aeration [35].

The energy recovery of MFC systems is still low, but it has already been possible to show a self-sufficient treatment of brewery and municipal wastewater by Dong et al. and Zhang et al., respectively. The energy recovery and treatment performance of MFCs compared to conventional anaerobic and aerobic treatment is given in Section 4.

3.4 Further nutrient removal

In addition to COD removal and energy generation in wastewater treatment with MFCs, other removals of nutrients such as nitrogen, phosphorus, TSS and sulphate have been reported in some cases. Sometimes these nutrients must be removed as well to meet the requirements for direct or indirect discharge.

Nitrogen

Nitrogen removal is of great interest in wastewater treatment because of the tougher regulations on nitrogen discharge [103]. Limits are defined for direct and indirect discharge for several nitrogen compounds (ammonia, nitrate, nitrite). Nitrogen is usually removed by biological nitrification and denitrification [7]. Oxidation of ammonium to nitrate in the presence of oxygen and nitrifying bacteria is called the nitrification. The denitrification of produced nitrate to nitrogen gas takes place under anaerobic conditions; only electrons should be provided by the oxidation of organic material, so a reduction of nitrate to nitrogen takes place.

In the most MFC studies regarding nitrogen removal while treating wastewater, removal of NH₄ in the anode chamber was observed as being between 50 – 91 % [20, 29, 59, 61, 65, 87, 104-106]. For example, when treating brewery wastewater with a 10-litre MFC, the NH₄-N was reduced up to 3.6 – 4.6 mg/L [105], so that the limit value for direct discharge was reached (10 mg/L). In addition to NH₄-N removal, NH₃-removal by up to 73 % in the anode chamber has been recorded when treating dairy or swine wastewater [20, 60, 61]. With the decrease in NH₄ in the anode chamber, a simultaneous increase in NO₂ and NO₃ was measured in a few studies [20, 29, 59, 65, 104].

The increase in NO₃ and NO₂ suggests that nitrification of NH₄ was occurring, likely as a result of oxygen diffusion through the cathode [65, 104]. In total, the NO₃ formation is much higher than NO₂ formation, with, e.g. an increase from 0.3 to 40.5 mg/L NO₃ with a parallel increase from 0.08 to 4.12 mg/L NO₂ [104]. However, in most cases NH₄ removal is much higher than the NO₂/NO₃ formation [59, 65]. Therefore, in addition to nitrification, other nitrogen removal processes also take place. There may be several possible pathways, including denitrification, anaerobic ammonia oxidation, nitrite reduction by lithotrophic ammonia oxidisers or by some previously unobserved method of ammonia oxidation coupled to electricity generation [59, 65]. Zhuang et al. [105, 106] suggested that the NH₄ removal is caused by volatilisation due to a localised pH increase at the cathode surface. Furthermore, NH₄ has also been demonstrated to function as a substrate either directly or indirectly for electricity generation [40, 105].

Measuring the TN (total nitrogen) removal, it cannot distinguish between different forms of nitrogen, but if there is a TN removal not only nitrification to NO₃ takes place. The TN removal is sometimes focused using a double chamber MFC, in which the nitrification takes place in the aerobic cathode chamber and the denitrification and COD removal in the anaerobic anode chamber.

In the pilot-scale MFC of Liang et al. the synthetic wastewater was fed through two anode chambers and after that through two cathode chambers [54]. In total, the removal efficiencies of COD, NH₄-N and TN were about 95 %, 97 % and 84 %, respectively. For improved nitrification, Ge et al., Ismail et al. and Vilajeliu-Pons et al. used an additional aerated reactor [29, 46, 93].

Surprisingly, TN removal in membrane-less MFCs reached almost the same percentage as COD removal, although no biocathodes were used in the MFCs of Hiegemann et al. [41]. Treating municipal

wastewater, a TN removal of 18 % and 29 % in 22 h, up to 71 % in 6 days was achieved using membrane-less MFCs [25, 41, 42]. In addition, TN removal of 47 % in the anode chamber of a MFC with membrane treating brewery wastewater was observed [11].

Analysing the biofilm which has built on the cathode, the abundance of ammonia oxidising bacteria and nitrite oxidising bacteria was in the range of 4 % [41], so that the nitrogen was probably degraded via the conventional nitrification-denitrification pathway. The evidence of nitrogen removal in a single chamber MFC was already made, with or without pre-enrichment of nitrifying biofilm on the cathode [70, 99].

In summary, the main effects reported are NH_4 removal and NO_3/NO_2 increase. In a few studies, a TN removal could also be achieved.

Phosphorous

Unlike nitrogen removal, the MFC system did not perform well in removing phosphorous. A little reduction of phosphorous can be reported about 20 % uptake during the 18-hour residential time, resulting in a final concentration of 4.3 ± 1.0 mg/L [29] and about 18 % or 31 % of dissolved phosphorous [60, 61] during dairy wastewater treatment. Biomass uptake could be the major mechanism accounting for the reduced concentration [29, 47].

In contrast, while treatment of swine wastewater by *Egbadon* et al. an increase in phosphates, phosphorus and orthophosphates about more than 50 % was measured [20]. Also a slight increase of 17 % was measured in the MFC treatment of *Min* et al. [65].

It was probably due to low redox potential that causes microorganism stimulation and phosphorus release or the conversion of organic phosphorus in the wastewater to different forms of phosphate [20, 60, 61, 65].

As a conclusion for the performance of MFCs, a low phosphorus content in the anode chamber is necessary, so that according to *Penteado* et al. phosphorus does not become a limiting factor for exoelectrogenic microorganisms [72], but a high phosphorous removal rate is not expected.

TSS

Depending on the type of wastewater, the initial total suspended solids (TSS) concentration can vary a lot. For example, the TSS content is much higher in swine wastewater (400 to 500 mg/L) than in domestic wastewater streams (< 100 mg/L) [7].

Pilot studies by *Hiegemann* et al. on municipal wastewater treatment reported TSS removal efficiencies of 36 % and 40 % at an HRT of 43 h and 22 h to approximately 73 and 49 mg/L [41, 42]. In addition, *Ge* et al. even achieved 83 % TSS removal in the anode chamber and a further 49 % in the cathode and additional aeration tank (73 mg/L → 12 mg/L → 6 mg/L) in only around 18 h HRT [29]. Furthermore, high-strength swine wastewater with TSS concentrations (up to 31 g/L) was degraded with an efficiency of 99 % within an HRT of 504 days [20]. In regard to studies with a

shorter HRT of 4 h or 17 h, a degradation of 50 – 80 % or up to 63 % (5.4 → 2.2 g/L) is still possible [7, 49].

Besides TSS removal, a reduction in volatile suspended solids (VSS) and total dissolved solids (TDS) was also measured during treatment of dairy, brewery or domestic wastewater with an efficiency up to 86 % in 6 days HRT VSS removal in pilot-scale and a TDS removal of 44 % in a small-scale study treating brewery wastewater for 72 h HRT [17, 47, 60, 61, 101].

A possible reason for the TSS removal may be hydrolysis (particulate to soluble substrate), in which colloidal and complex organic matter is degraded due to biologically catalysed processes [17, 41, 60, 61]. Settling of suspended solids inside the MFC [17] is another probable explanation, but, e.g. *Babanova* et al. [7] only needed sludge removal in the reactor after 200 days of operation. The settling and accumulation of solids should be carefully monitored when using wastewater with high TSS concentrations because COD removal efficiency could be significantly impacted by the settling and removal of solids in the reactors [49].

Sulphates

Only a few studies detected the sulphate content during MFC wastewater treatment. Some sulphate removal was reported during dairy wastewater treatment with maximum sulphate removal rates of 39 % and 30 % [60, 61]. When treating swine wastewater, a removal rate of up to 70 % was measured [7]. Sulphates can probably be reduced to sulphides [60, 61] – the identification of sulphate-reducing bacteria, which were correlated with the removal of sulphates was observed around the same time [7].

In summary, it is possible to remove nitrogen, TSS and sulphates. Merely the removal of phosphorus is not expected. In conventional anaerobic treatment, only a slight removal of nitrogen is possible, making further aerobic treatment necessary for wastewater with a high nitrogen content [79]. The TN removal via conventional nitrification-denitrification requires intensive aeration and in particular COD and TSS mainly affect excess sludge production [41]. Therefore, besides COD removal, additional nutrient removal can be a further advantage in the treatment of wastewater using MFCs.

4 Assessment of MFCs with regard to conventional biological treatment

Some comparisons and theoretical studies with conventional biological treatment have already been made for the evaluation of MFCs. There are comparisons with aerobic and anaerobic treatment, as well as possible combinations of aerobic/anaerobic treatment with MFC technology. Therefore, niches could arise for the use of MFCs in treating wastewater.

4.1 Comparison with aerobic and anaerobic treatment systems

MFC vs aerobic treatment

The fact that aerobic treatment, unlike treatment with MFCs, is an

Table 4 Characteristics of aerobic, anaerobic and MFC treatment of wastewater (adapted from [48, 69])

Parameter	Aerobic treatment (activated sludge)	Anaerobic treatment	MFC	Ref.
Energy input for aeration	very high	none	none	[69]
Energy production (kWh/kg)	- 1.2	3.8 heat or 1.5 electrical	0.8 today, 4.4 theoretically	[48]
Loading rate (kg _{COD} /(m ³ *d))	0.5 – 2	8 – 20	3.3	[69]
Removal rate (kg _{COD} /(m ³ *d))	1	20	3	[48]
Operational temperature	ambient	mesophilic, thermophilic	ambient, low temperature	[69]
Gas treatment	low	required	none	[48]
Sludge production	high	low	low	[69]
Nutrient removal	yes	none	possible	[69]
Effluent quality	good	poor	good	[48]
Reactor scale-up	volume	volume	modular	[48]
Capital cost	medium (high volume cost)	medium (high peripheral cost)	high	[48]

energy-intensive biological treatment also opens up prospects for MFCs. Using MFCs wastewater treatment could be turned into a self-sufficient or even net energy-generating process.

According to *Abourached et al.* and *Trapero et al.*, the replacement of the conventional activated sludge process by MFCs will be a promising alternative for fruit or juice processing wastewater treatment [2, 88]. *Abourached et al.* analysed the economic feasibility of MFCs in treating fruit-processing wastewater for irrigation [2]. Consideration of the revenue from water reuse and the electricity generated at the same time, MFCs offer energy, environmental and economic benefits [2]. In addition, the economic assessment of an MFC implementation regarding three different cases (optimistic, most likely, pessimistic) by *Trapero et al.* based on the maximum power density of the MFC treatment of the wastewater from a juice processing plant then, considering all the scenarios, MFC is the most attractive option [88]. Hence, the implementation of MFC as a promising alternative to aerated activated sludge treatment is already under discussion for industrial wastewater treatment with high COD concentrations.

Regarding the more important key value for industrial wastewater treatment, the COD degradation of MFC must be competitive with aerobic treatment. *Fallgren et al.* directly compared COD and TSS degradation in aerobic and MFC wastewater treatment by operating two 15 L reactors in parallel [23, 88]. In order to achieve 90 % COD removal, an HRT of 15 days and eight days was needed for MFC and aerated, respectively [23]. The HRT appears to be very long in both cases, but here the two treatment efficiencies can be compared directly under the same operational conditions. Although the MFC, consequently needs a longer HRT, the TSS concentrations are significantly lower, and an energy gain is possible [23]. A pilot-scale MFC with a 250-litre volume and an long-term operation period of one year has confirmed these findings [25].

Therefore, in addition to sludge reduction, the same cleaning objective can be achieved with an energy gain instead of an energy loss through the MFCs, because there is no need for aeration and sewage sludge disposal [23, 25].

MFC vs. anaerobic treatment

In contrast to aerobic treatment, energy can be produced in the form of methane when wastewater is treated anaerobically. In theory, 1 kg of carbohydrates represents 1.06 kg of COD, containing 4.41 kWh of energy [73, 74]. At present, about 1 kWh can be generated by the anaerobic treatment of 1 kg carbohydrates (1 kg COD → 360 L methane → 1 kWh_e) [74]. MFC produces electricity from organic waste in a direct way, without any gas treatment, so that MFC can theoretically convert 1 kg of COD to 4 kWh of electrical energy [73]. Comparing the net electrical energy yield from various outlets, MFCs have the potential to capture the highest energy value of COD, since combustion can be avoided. [77].

Thus, in theory, an anaerobic digester can produce electricity from organic matter with an efficiency of 30-35 % [39]. The efficiency of the MFC (η_{EE}) depends on the product of (2) Voltage Efficiency (VE) and (1) Coulomb Efficiency (CE) [39, 86].

$$\eta_{EE} = VE \times CE \tag{Eq. 8}$$

In order to be energetically competitive with anaerobic digesters, the total efficiency between 30 – 40 % has to be reached with MFCs [39, 86]. Consequently, the efficiency of VE and CE must have a high percentage.

The CE values of the pilot-scale MFC studies ranged from 3 to 17 % when treating industrial wastewater. The average COD removal efficiencies with $74 \pm 19\%$ are competitive with anaerobic treatment efficiencies. Direct comparison of potato-processing wastewater treatment confirms the comparable COD treatment efficiency as having an 87 % COD removal rate in 400 h by MFCs and only a 75 % removal rate in 1800 h by conventional anaerobic treatment so that, in this case, the degradability by MFCs was much more efficient [18]. The HRT appears to be very long in both cases, but here the two treatment efficiencies can be compared directly under the same operational conditions. But in contrast, the CE of the MFC was much lower, so that the energy recovery of the anaerobic treatment was higher [18].

An overview of the different characteristics of aerobic, anaerobic and MFC treatment of wastewater is given in table 4.

According to *Rozendal et al.*, the MFC system might become a cost-effective alternative to aerobic treatment, but because of higher capital costs, it will be hard to compete with anaerobic treatment [82]. In the cost analysis made by *Gude et al.*, it was shown that MFCs are able to meet 50 % of the total costs, because of the high capital costs for anode and cathode materials [37]. However, regarding some particular benefits over anaerobic digestion, such as operation on a small-scale, as well as the low COD concentrations, low temperatures and integrated nitrogen removal [94], the MFC does have a perspective [82].

4.2 Combination of biological treatment systems with MFCs

In contrast, to consider the conventional treatment system as competitive technologies, a combination of such technologies with MFC systems might be beneficial [73]. Several possibilities have already been suggested either for replacing conventional anaerobic or aerobic treatment steps or as an additional treatment step (Fig. 5).

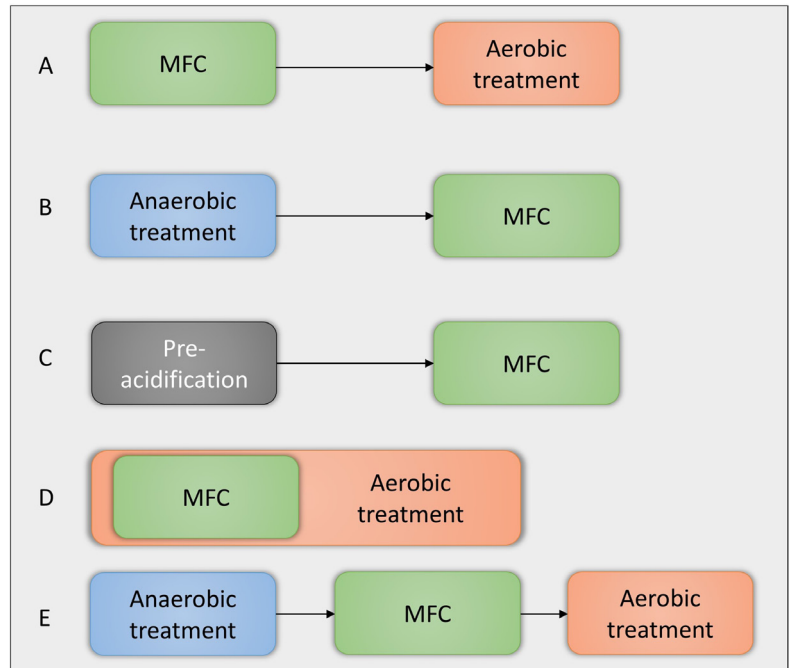


Fig. 5 Combination possibilities between MFC, aerobic and anaerobic treatment

- A) After MFC treatment, aerobic treatment is carried out to reach the required limit values and cut the high energy costs of the aerobic treatment. [55, 78]
- B) Before MFC treatment, the use of an anaerobic digester as pre-treatment for high-strength wastewater, so that only low COD concentrations must be treated with MFCs. [4, 17, 28, 73]
- C) For especially high rates of anaerobic treatment of industrial wastewater such as brewery wastewater, a conventional pre-acidification step is used, which can also be installed before an MFC [78]. Consequently, higher CE values and COD removal rates can be achieved.
- D) The integration of MFCs into activated sludge tanks [37].
- E) A combination of anaerobic digestion, MFC and aerobic treatment for high-strength and complex wastewater [102].

The combination of a MFC with an additional aerobic step (A) has already been investigated; here, the wastewater composition could be reduced for direct discharge and an energy neutral process could be reported in laboratory-scale with domestic wastewater [75]. In addition, the cathode chamber and an external aerobic tank were used for efficient nitrogen removal [29, 46, 93]. The feasibility of treating the wastewater of an anaerobic digester with an MFC (B) has also been examined in several studies, in which a stable flow could be generated by MFCs [11, 16, 18, 36, 50, 83]. For high-strength molasses wastewater in particular, the combination of MFC with conventional anaerobic-aerobic technology was demonstrated [102]. The anaerobic unit was responsible for COD degradation and sulphate removal, the MFC unit was used for oxidation of sulphide to sulphur and electricity generation and the aerobic unit was used to remove colour and phenol derivative [102]. It is an example of a complete wastewater treatment with respect to a number of pollution factors.

5 Conclusion

The feasibility of simultaneous electricity generation and industrial wastewater treatment has been shown in several MFC laboratory cases and even in some pilot-scale studies. The type of waste water has not been the decisive factor. In addition, the self-sufficient treatment of brewery wastewater has already been proven [17].

The flexibility of the modular MFC systems is one main advantage for their application in the direct or indirect discharge of industrial wastewater. Furthermore, the MFC becomes an attractive option for wastewater treatment due to a) operation on a small-scale, b) at low COD concentrations, c) at low temperatures, d) further nutrient removal (nitrogen, TSS and sulphate), e) the fact that no energy is needed for aeration, f) the direct electricity generation, g) low sludge yield and h) centralised or decentralised applications. Efficiently scaling up and reducing the investment costs are the major challenges for any possible commercialisation, but the combination with conventional biological treatment will represent another opportunity.

The overview of MFC treatment of industrial wastewater provides a basis for the practical application of MFCs on a laboratory and pilot scale. A realistic baseline for possible MFC application niches could be provided by a detailed analysis of industrial wastewater treatment plants in terms of energy recovery and treatment efficiencies. In short, MFCs are suitable for the treatment of industrial wastewater, but improvements in efficiency are required.

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