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2   Bioelectrochemical treatment of municipal waste liquor in microbial fuel cells for

3   energy valorization

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Abstract

Microbial fuel cells (MFCs) are recognized as promising applications to produce bioelectricity by utilizing various waste materials. In this study, dual-chamber microbial fuel cells were employed for energy valorization of an untested substrate, the liquid fraction of pressed municipal solid waste (LPW). This by-product is potentially applicable as a substrate in MFCs because of its high organic matter content. In the course of the experiments, the anodic biofilm response and energy production efficiency have been investigated by experimental design approach, taking substrate and fresh inoculum – mesophilic anaerobic sludge (MAS) – addition into account as factors. It was observed that reinoculation could result in a negative effect on the energy production, especially at low substrate (LPW) dosing levels. However, when the LPW to fresh MAS ratio in the anode chamber exceeded a particular value, the biofilm-associated electrical utilization dominated against the degradation in the bulk phase. Furthermore, the results indicated that the highest energy yields (8-9 J g^{-1} ΔCOD d^{-1}) could be attained at the lowest input COD concentrations. The maximal and average COD removal efficiencies were 94% and 87 %, respectively, which indicate the excellent biodegradability of LPW. As for COD removal rate, 1.2-1.9 kg COD m^{-3} d^{-1} could be reached.

Keywords: microbial fuel cell, organic waste, power generation, experimental design, bioelectricity, waste utilization

1. Introduction

Nowadays, bioelectrochemical systems such as microbial electrohydrogenesis cells (Rivera et al., 2015) and microbial fuel cells (MFCs) (Rózsenberszki et al., 2015) are increasingly mentioned in the joint subject of waste treatment and energy generation . MFCs are able to convert waste-bound chemical energy into electricity directly via bioprocesses catalyzed by exoelectrogenic microorganisms (Logan, 2008; Logan et al., 2006; Lovley, 2006). A general, laboratory-scale MFC system consists of three main structural elements, (1) an anode and (2) a cathode chamber and (3) a proton selective membrane separating the two compartments and ensuring the proton transport between them (Huang et al., 2015). As a result of substrate decomposition by anode-living whole cell biocatalysts, electrons are released, captured by the anode and subsequently transferred to the cathode via an external wiring. In the cathode, electrons and protons are combined with oxidative agent e.g. oxygen and in turn, water is formed.

A variety of substrates can be used for bioelectricity generation in MFCs such as saccharides (Chaudhuri and Lovley, 2003; Kim et al., 2000; Vajda et al., 2014), organic acids (Bond and Lovley, 2005; Liu et al., 2005; Min and Logan, 2004), alcohols (Kim et al., 2007; Vajda et al., 2011) as well as inorganic substances e. g. sulphate (Rabaey et al., 2006). In addition, there is a significant research interest towards complex materials i.e. industrial and municipal waste streams (Leaño and Babel, 2012), whichare potential starting materials of power generation in MFCs because of their high organic matter content (Angenent et al., 2004; Pant et al., 2010;

Liu et al., 2004). Therefore, due to the possibility of simultaneous waste management and energy production, MFCs are considered as prosperous applications facilitating sustainability. In the wide range of microbial fuel cells, the ones relying on mixed cultures have been demonstrated as feasible solutions to accomplish efficient "waste to energy" conversion (Jung and Ragen, 2007).

MFCs are powered by electrochemically active bacteria which are the heart and soul of the technology. Among them, organisms such as *Shewanella*, *Geobacter*, etc. species have been identified as strains capable of converting chemically-bound energy into bioelectricity (Wrana et al., 2010). However, when their pure cultures are employed as seeding source to colonize MFC anodes, maintaining sterility is a strict requirement. The applications, which are started-up and operated under sterile circumstances, suffer from considerable economic and technological drawbacks especially if industrial waste streams or bioreactor effluents are utilized. This is attributed to the risk of contamination by indigenous microorganisms carried in the influents which presumably reduce the overall MFC performance. Due to the abovementioned limits of pure cultures, mixed communities are suggested to start-up MFC systems, as performed in this study using MAS.

When non-sterile MFCs are seeded with microbial consortia, e.g. anaerobic digester sludge, waste water sludge, etc., exoelectrogenic strains are enriched on the anode surface and form a bioactive layer. The profile of the anodic microbial population is a crucial issue, which can be changed by competing bacteria in the feed streams or in the seed inoculum itself, possibly leading to altered process performance (Bakonyi et al., 2014).

To assess the impact of process disturbances on MFC efficiency – related to the appearance of non-electricity generating, invader microorganisms – direct methods (electron microscopic investigation, identification of strains based on genetic information, cyclic voltammetry, etc.) can be employed, but they mostly require the deconstruction of the MFC. Nevertheless, indirect methods such as in-situ bioelectric measurements – when the whole MFC plays the role of a "biosensor" – can be chosen for more convenient and quick evaluation. Therefore, in this work, this latter technique was used to analyze the effect of MFC reinoculation (fresh sludge addition to the already developed anodic consortia) on bioenergy production. In the course of the reinoculation tests, an uncommon substrate, the liquid fraction of pressed solid municipal waste (LPW) was applied. Substrate concentration has a decisive role on any bioprocesses, e.g. MFC, since it affects the growth rate and the metabolic activity of microorganisms (in the biofilm) and hence determines the production intensity (in MFC, bioelectric potential). Moreover, the capability of the anode-living, exoelectrogenic bacteria to manage as high organic loadings as possible is a well-known and desired process indicator. LPW, due to its origin (landfill), is a complex mixture and might contain inhibitory components. Thus, its feasibility for MFCs especially in higher concentrations – because of the lack of experiences – is uncertain. Therefore, assessing the behaviour of MFCs to various substrate (LPW) doses was one of our key-interest in order to reveal the robustness and feasibility of this application.

Overall, the aim of this research was twofold. Besides the feasibility study of LPW, the response of the anodic biofilm – formed during preliminary adaptation process – to the reinoculation of MFC anode compartment by mesophilic anaerobic

sludge (MAS) was also investigated. Thereby, the appropriate substrate feeding rate could be estimated for scaled-up waste processing MFC systems. In addition, the reactions taking place at the anode surface and in the bulk phase could be addressed to obtain information about the degradation-capacity of the anodic biofilm. To our knowledge, no research has been conducted with LPW in microbial fuel cells, which brings the novelty of the work.

2. Materials and Methods

2.1. Inoculum and substrate

MAS (the inoculum of fuel cells) was collected from a domestic biogas plant. The biogas fermenter processes kitchen wastes and cattle manure and its effluent is suitable as a seed source providing a good microbial community. The initial total COD and pH values of MAS were 30 g L^{-1} and 7.8, respectively.

LPW is originated from a regional solid waste treatment plant (located in Királyszentistván, Hungary) and was obtained by pressing from the biofraction of municipal solid garbage. The main stages of waste processing at the plant are presented in Fig. 1. The facility handles 120 000 t waste on annual basis from villages and towns located in its neighbourhood. As a first step, the garbage is subjected to a mechanical treatment. After shredding, iron is separated and the rest of the mass stream is further sorted. Subsequently, a certain part of it is recycled, meanwhile others

are sent to additional techniques (Sarkady et al., 2014). The most essential characteristics of LPW material are demonstrated in Table 1.

2.2. MFC design and start-up

The laboratory-scale, dual-chamber MFCs were made of plexiglass. The cells were constructed with a volume of 60 mL for anode and cathode compartments. The electrode chambers were divided by Nafion N 115 proton-selective membrane (Sigma-Aldrich). The surface area and thickness of the membrane were 7 cm^2 and 127 μm, respectively. Carbon cloth fixed on a graphite rod (1.5 mm diameter) was used as electrode material. The apparent surface area (A) was 25 cm^2 for both electrodes (with 5.0 cm $^\times$ 2.5 cm dimensions). The electrode surface specific values were calculated on the basis of apparent surface area, i.e. by leaving the electrode thickness and the area of the graphite rod out of account. Five MFCs were operated in parallel with 100 Ω fixed external resistors (R). The schematic diagram of the experimental MFC and occurring processes can be seen in Fig. 2.

In the start-up phase, the anode chamber was filled with MAS (55 mL), purged by N_2 to ensure anaerobic conditions and fed with Na-acetate (2 mL, 25 g L^{-1} stock solution) after the initial voltage had decreased. Na-acetate was injected in two steps to promote the colonization of the electrode surface by anode-respiring exoelectrogenic microorganisms. After a ~15 days long adaptation period, when the gradual voltage had decreased back to the initial value of inoculation, the experiments with LPW and MAS feeding were carried out. In the course of these measurements, LPW and fresh

MAS were loaded in various ratios (Table 2) and simultaneously, while equal volume of anolyte was removed. Besides LPW and MAS, no other materials (nutrients) were added to the MFCs.

The cathode compartment contained water and was continuously sparged by air. The anode and cathode sides were connected by copper wire and the potential difference was on-line followed using a data logger device (National Instruments USB-6008) and LabView software. The MFCs were incubated at 37 °C. Neither the anodic nor the cathodic chamber was mechanically stirred.

2.3. Experimental design

Quality feature of the adaptation substrate supporting the biofilm formation have significant effect in the start-up phase as well as in further operation. The electrodes coated by sufficiently stable biofilm layers – and thus the complete MFC – might be used for long-term. Na-acetate is a simple, readily accessible substance with fair biodegradability and excellent electron donor capacity (Dulon et al., 2007). Hence, it is recognized as a potential substrate candidate for MFC start-up and was applied in this study to promote the development of biofilm on the anode surface. After MFCs start-up, an experimental design (Table 2) was carried out to study the biofilm response in terms of energy valorization of LPW.

Table 2 presents a complete 2^2 experimental design, having 2 low and 2 high values and one central point, as illustrated in Fig. 3. The two-level designs are particularly useful to obtain reliable information and make firm conclusions about the

effects of factors of primary importance (Bakonyi et al., 2011). In this study, as it can be seen in Table 2 and Fig. 3, the anodic concentrations of LPW substrate and that of fresh MAS were chosen as independent variables to get a feedback about their influence on waste utilization efficiency, which can be correlated with the biofilm response.

2.4. Analysis and calculation

COD (COD_0 initial and COD_t final) values were determined following APHA (APHA, 1995). The COD input (in grammes) of the mixture feeding was calculated based on the known COD values of the components (LPW and MAS) and their injected volume. In the experimental design, the reinoculation ratio was defined as the amount of the MAS injected divided by the working volume of the anode (Table2).

According to Ohm's law, current data (I) and thus electrical power (P) could be calculated based on the voltage measured. Cumulative energy data (E) were calculated from integrating the time-dependent power curve (Eq. 1).

$$E = \int P(t)\mathrm{d}t \qquad \text{(Eq. 1)}$$

Specific values, namely current density (jI), power density (jP) were derived by taking into consideration the electrode surface area (Eq. 2 and Eq. 3).

$$jI = U(t)\,R^{-1}\,A^{-1} = I(t)\,A^{-1} \qquad \text{(Eq. 2)}$$

9

$$jP = U(t)\ I(t)\ A^{-1} = P(t)\ A^{-1} \qquad \text{(Eq. 3)}$$

The energy yield (jE) was a product of the amount of substrate eliminated ($\Delta\text{COD} = \text{COD}_0 - \text{COD}_t$), the time of operation (designated by τ in Fig. 4) and the cumulative energy generated (Eq. 4).

$$jE = E\ \Delta\text{COD}^{-1}\ \tau^{-1} \qquad \text{(Eq. 4)}$$

The Coulombic efficiency (*CE*) of the MFCs can be calculated based on the ratio of the total Coulombs obtained from the substrate and the theoretical maximum of Coulombs when all of the electrons from the substrate generate electricity (Eq. 5).

$$CE = \text{M} \int I(t)\mathrm{d}t\ \text{F}^{-1}\ b^{-1}\ V^{1}\ \Delta\text{COD}^{-1}\ 100\ \% \qquad \text{(Eq. 5)}$$

where M is the molar weight of oxygen, F is the Faraday's constant, b is the number of exchanged electrons per 1 mole of O_2 (equaling to 4) and V is the volume of the liquid in anodic chamber (Logan et al., 2006). The results presented throughout in this work are the mathematical averages of triplicates and the standard deviations were below 5 %.

3. Results and Discussion

3.1. The effect of LPW and MAS ratio on MFC performance

In this work, the effect of MAS addition and LPW (substrate) loading on the performance and behaviour of two-chamber microbial fuel cell was assessed. As for MAS addition, it was preliminary assumed that the fresh microbial consortia fed into the anode chamber might disturb the biofilm and consequently, the electricity generation. Furthermore, it was hypothesized that the nutrients available in the feed can be consumed by microbes (supplemented with the fresh MAS) that are not localized on the electrode surface and hence does not contribute to useful bioelectricity production.

A typical voltage output is demonstrated in Fig. 4, where it can be observed that the feeding strategy in the MFCs is comprised of 4 individual stages according to the recorded voltage pattern, as follows. In the first stage, the MFC unit was inoculated with raw MAS and initial voltage had increased, but shortly it turned into a strong decrease. At that point, in the second stage, sodium acetate was added to help the establishment of the bioactive layer on the anode which resulted in prompt and sharp jump of voltage followed by a gradual reduction in the cell potential. Subsequently, during the third step, the MFC was supplemented again with Na-acetate. As it can be noticed, it induced an observable increment of bioelectric potential, which continuously fell, presumably due to the depletion of adequate substrates. It would also appear that the second adaptation resulted in a more stable voltage signal as compared

to the first one. This was a positive feedback that may correlate with biofilm formation. In the fourth step, after two-times acclimatization with acetate, the MFCs were loaded with the mixture of LPW and fresh MAS in accordance with the experimental design matrix (Table 2).

As it can be concluded from Fig. 4, there was a growing cell potential after the injection of LPW-MAS mixture, which had reached a definite peak range where quite steady voltage (roughly 40 mV) could be measured (approximately between the 650th and 950th hour of MFC operation). Voltage fluctuations may be associated with the complexity of the feed since various fractions of the raw materials could become accessible and biodegradable at different periods of times.

In case of microbial fuel cells (and generally fuel cells) measuring current density (or current) is suitable to determine the gross rate of the electrochemical reaction according to Faraday's law. Thus, specific current- or power density values are useful to compare the efficiency of different MFC systems, as well. Based on the voltage data collected, the main characteristics were calculated which can be seen in Table 3. The outcomes are comparable with other literature studies. For instance, Cercado-Quezada et al. (2010) tested various food industry wastes as well as compost leachate to extract energy in microbial fuel cells. Using the latter substrate (that shows certain relation with landfill-derived LPW), 232 mA m^{-2} current density was reported applying MFC seeded by anaerobic sludge, meanwhile in this research 152-218 mA m^{-2} could be achieved. However, the obtained power densities were notably different, since the 54 mW m^{-2} value by Cercado-Quezada et al. (2010) exceeded our power densities ranging between 5.8 and 11.9 mW m^{-2}. In another paper, Ganesh and

1   Jambeck (2013) operated MFCs fed by leachate and attained 114 mA m^{-2} current

2   density which was slightly lower than the results in our experiments as indicated in

3   Table 3. Tugtas and co-workers (2013) employed MFC to biodegrade pre-digested

4   landfill leachate. It turned out that the steady current densities of 418-548 mA m^{-2}

5   could be obtained, meaning a salient system performance. The compared data were

6   collected in Table 4. The contradiction in MFC efficiencies can be attributed to a

7   number of biological (e.g. the source of inoculum) and architectural (e.g. the external

8   resistance used, the distance of electrodes, the conductivity of anolyte, etc.) reasons.

9   Fig. 5 shows the response of MFC performance to various amounts of LPW and

10   fresh MAS additions. According to the results, it can be pointed out that the MFCs

11   were able to generate bioelectricity and microbes responded positively to the presence

12   of LPW since higher substrate concentrations allowed to gain more (cumulative)

13   energy. However, it would appear that there could be a joint effect of simultaneous

14   LPW and MAS supplementation, as follows. On one hand, the analysis of the

15   response surface in Fig. 5 implies that at low LPW concentrations the addition of MAS

16   in larger quantities should be avoided. It may be explained by the competition between

17   bulk phase and anodic biofilm, when the degradation of organic matter in bulk phase is

18   more notable and therefore only a significantly reduced amount of substrate is able to

19   reach the bioactive layer on the anode surface. In other words, most of the chemical

20   energy bound in the feed material is lost via an undesired bioprocess. Recently, Chae

21   et al. (2008) demonstrated that competitive degradation processes may occur in the

22   anodic side of two-chamber bioelectrochemical systems, which can cause lower

23   performance. It has turned out from the results that the gas evolved in the anode

compartment contained a considerable amount of methane besides carbon dioxide, which implies that methanogenic strains were able to utilize the organic substrate present via electrochemically-inactive reactions, leading eventually to a depressed energy efficacy. On the other hand, when LPW concentration increases in the anode cell (higher substrate loading), the phenomena taking place due to fresh inoculum injection (MAS content) has less negative effect on the anode-surface reaction and hence bioelectricity generation is not limited so strongly. Overall, it can be concluded that outsider microbes entering the MFC anode chamber can influence the substrate utilization and related bioelectricity production, although it seems to be dependent on substrate to inoculum ratio.

3.2. Effect of LPW and MAS concentrations on energy yield and COD removal

In Fig. 6, the dependency of energy yield on LPW and MAS concentrations is depicted. The results show that significantly higher energy yields (up to 8-9 J g^{-1} ΔCOD d^{-1}) were achieved in case of low COD loadings that occur with low LPW and MAS concentrations. As elucidated above, the addition of MAS ("reinoculation") could pose a threat on reliable MFC operation since microorganisms in the bulk phase compete for the nutrients and hence, reduce their amount being available for the electrochemically active anode-surface consortia, inherently causing depressed energy yields. Moreover, it can be observed that improved energy valorization of LPW favours its low concentrations. LPW is a complex organic matter and therefore, its bioelectrochemical decomposition has significant time-demand, which, in addition, is

1   likely influenced by the diffusion rate of substances into the biofilm, as well. Besides,

2   the biofilm can be characterized by limited substrate processing capacity. Thus, the

3   phenomenon of substrate saturation may occur, when the ability of microbes in the

4   biofilm is fully exploited, they are unable to take up more compounds and thereby

5   different bulky phase processes could take place in parallel that do not support

6   electricity generation. From the point of view of energy-realization, it means an

7   unbeneficial utilization of the substrate. Therefore, if the substrate level is maintained

8   sufficiently low or in other word, if the influent is properly diluted (Cercado-Quezada

9   et al., 2010) then the anode-living strains can have the possibility to convert bigger

10   portions of LPW into desired electricity more efficiently.

11   The COD removal capacity of an MFC is an important parameter to evaluate

12   the attractiveness of MFC technology from environmental perspective. COD removal

13   data are summarized in Table 5, where it can be seen that remarkable COD

14   degradation was possible from a complex substrate such as LPW, which is a notable

15   advantage of the MFCs. According to Table 5, the average COD removal was 87.3 %

16   and in case of MFC 1 it exceeded even 94 %. These values can be considered high in

17   the view of other literature data (Gálvez et al., 2009; Mohan et al., 2007; Rengasamy

18   and Berchmans, 2012) as well as in comparison with our previous accomplishments on

19   sugar industry wastewater (Bélafi-Bakó et al., 2014). The organic-removal efficiency

20   of the cells can be described well by the average COD removal rates, which varied

21   between 1.2 and 1.9 kg COD d^{-1} values, assuming 1 m^3 anode chamber volume.

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4. Conclusions

Based on the results, it was concluded that the wastewater (LPW)-based microbial fuel cell system operated successfully and it proved to be suitable for power generation with simultaneous COD removal. It was found that reinoculation of MFC by mesophilic sludge influenced the effectiveness of the preliminary adapted biofilm on the anode surface. In addition, it was observed that energy recovery was strongly dependent on LPW (substrate) concentration in the anode chamber and improved MFC performance favoured low organic material loadings.

Acknowledgements

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Figure Captions

Fig. 1: The simplified overview of WTP process and LPW generation

Fig. 2: The scheme of the experimental dual-chamber MFC

Fig. 3: Layout of the experimental design

Fig. 4: Voltage output as a function of time (in case of 5 mL:9 mL LPW-MAS ratio).
1: inoculation with raw MAS; 2,3: first and second adaptation steps with Na-acetate
addition, respectively; 4: addition of LPW-MAS mixture

Fig. 5: Effect of MAS-LPW mixture feeding on energy production

Fig. 6: Changes in energy yield to initial MAS and LPW concentrations

1 Fig.1

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1   Fig.2

Feed

e⁻   e⁻

Organic waste

$CO_2 + H^+$

H^+

H^+

H_2O

O_2

Aeration

Anode chamber   PEM   Cathode chamber

2

3

Feed

e⁻   e⁻

Organic waste

$CO_2 + H^+$

H^+

H^+

H_2O

O_2

Aeration

Anode chamber   PEM   Cathode chamber
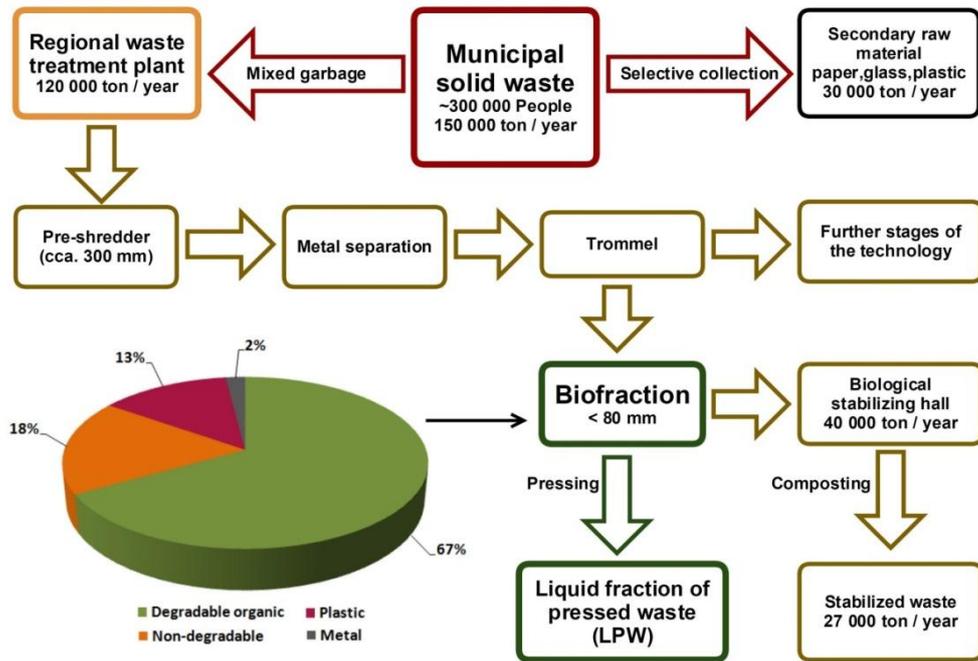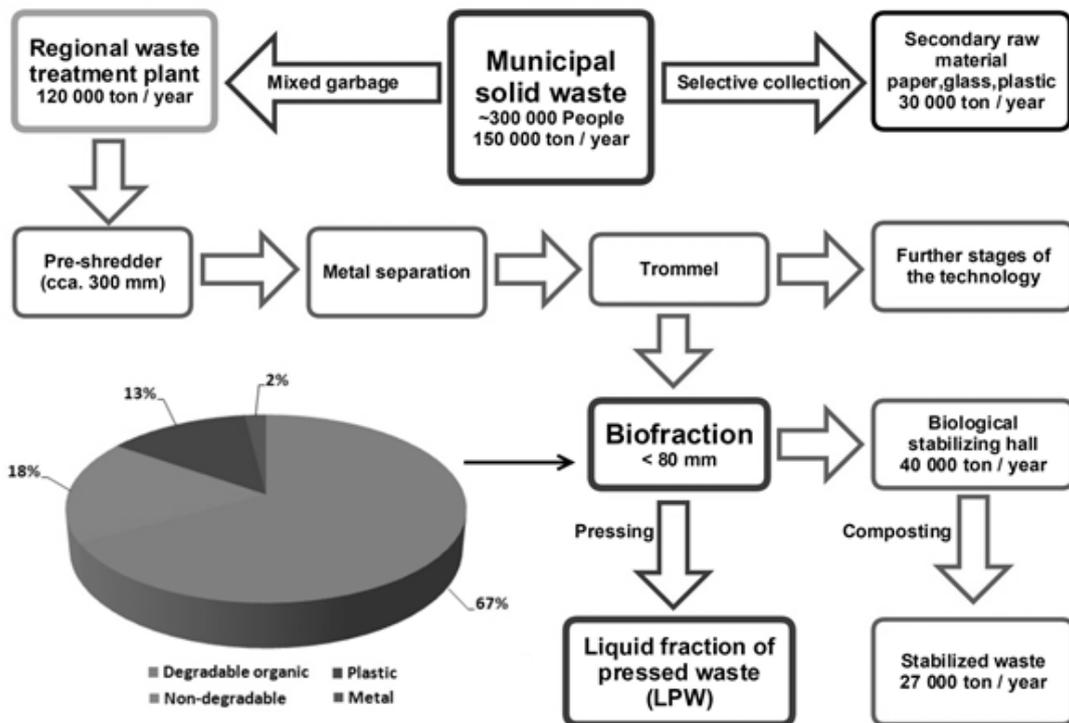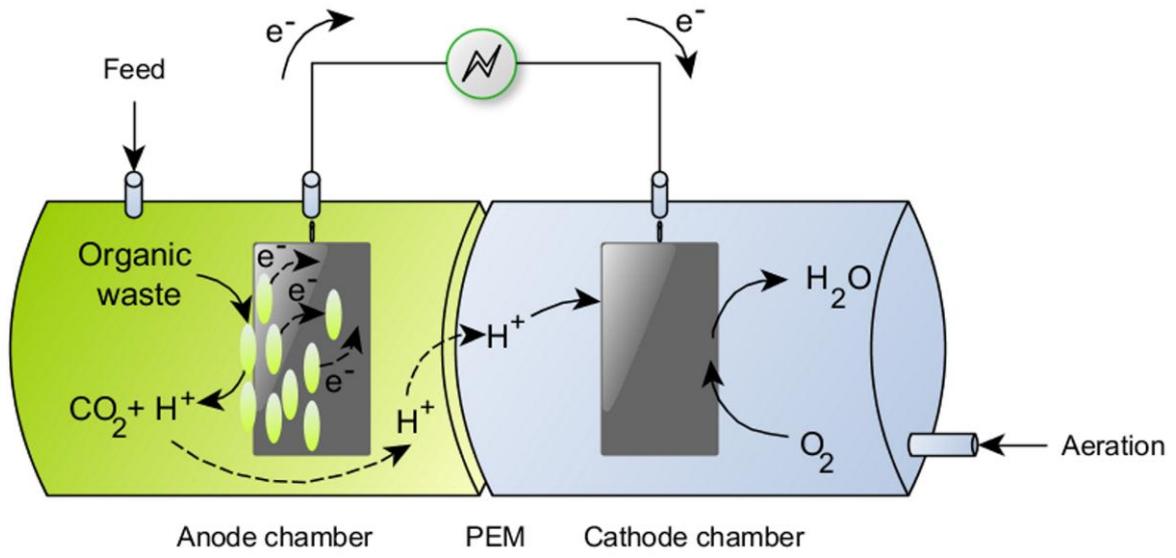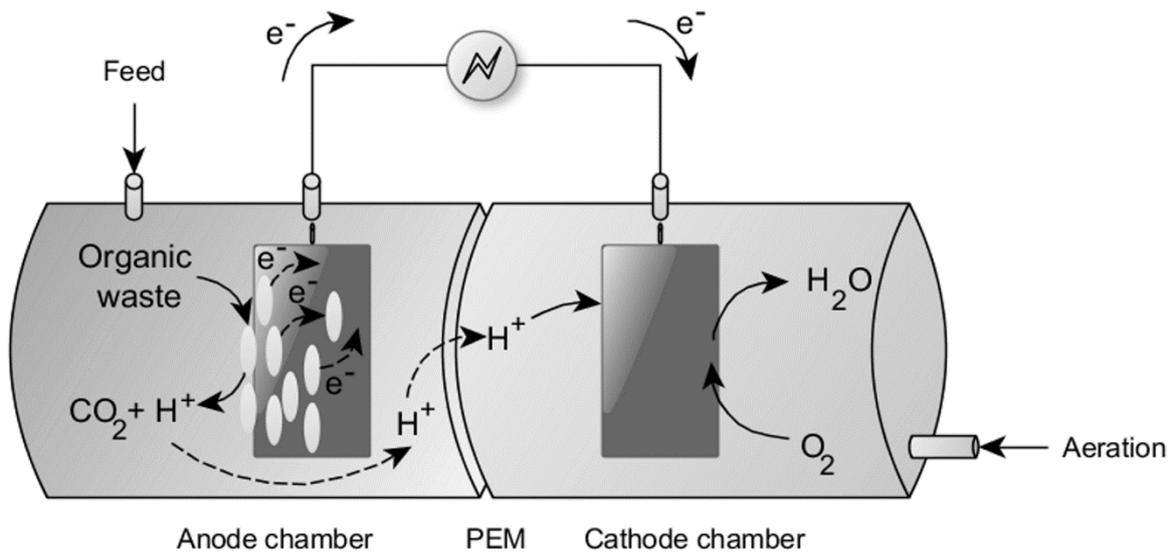
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1   Fig.3
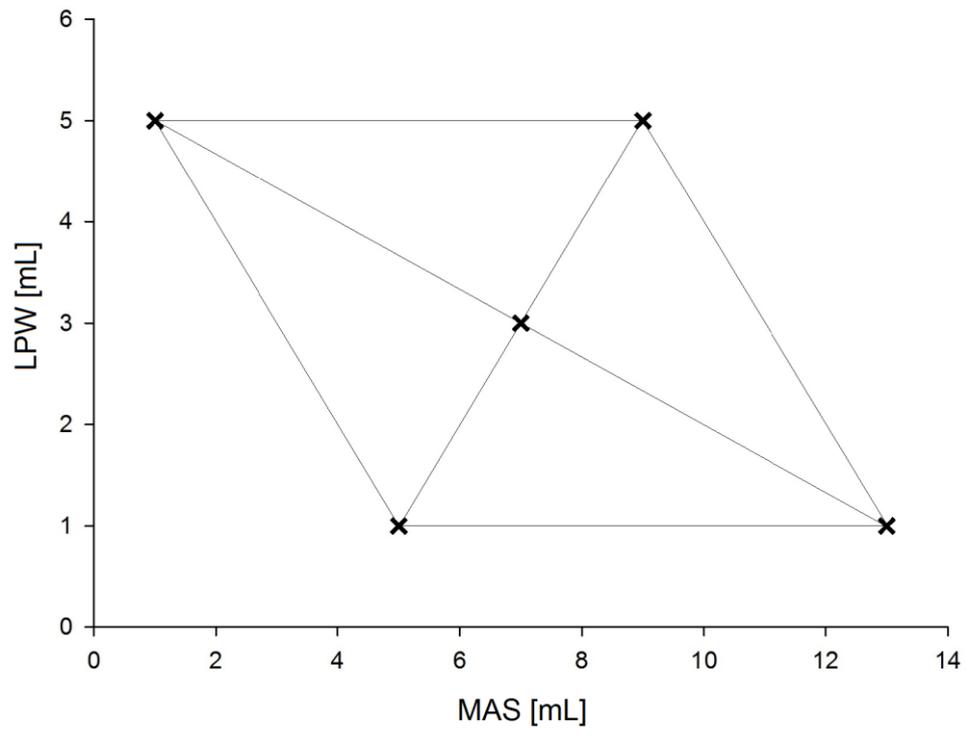

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1   Fig. 4
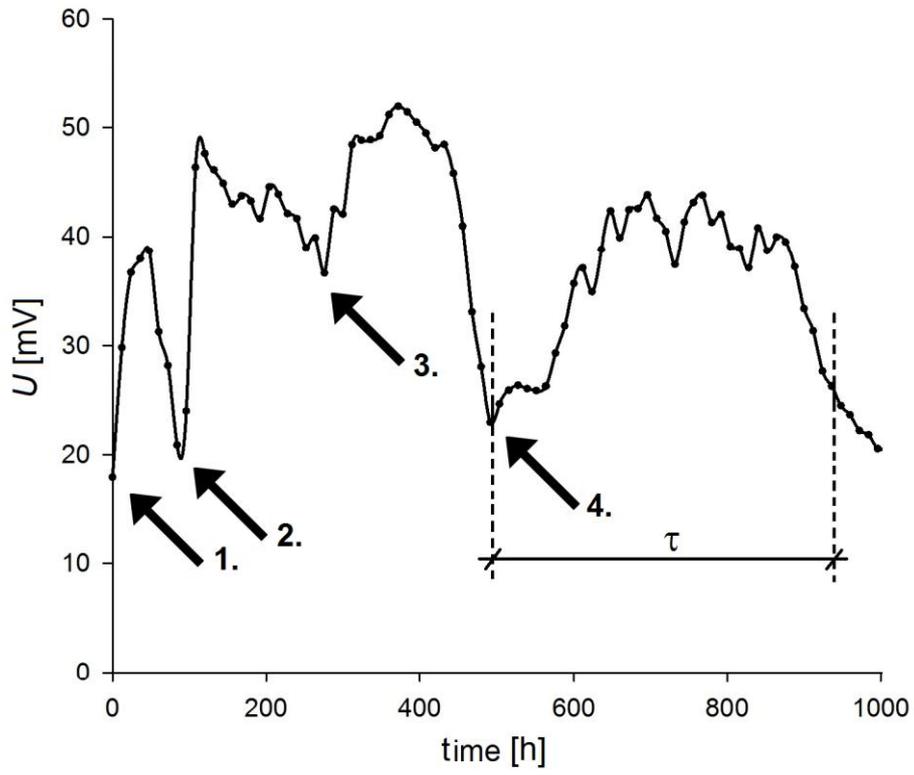
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1   Fig. 5
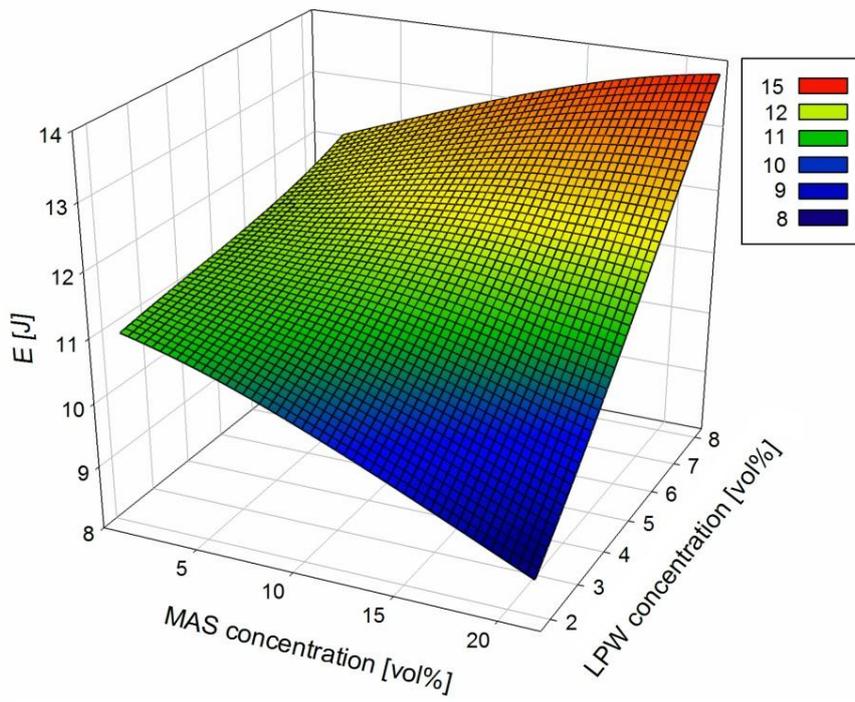
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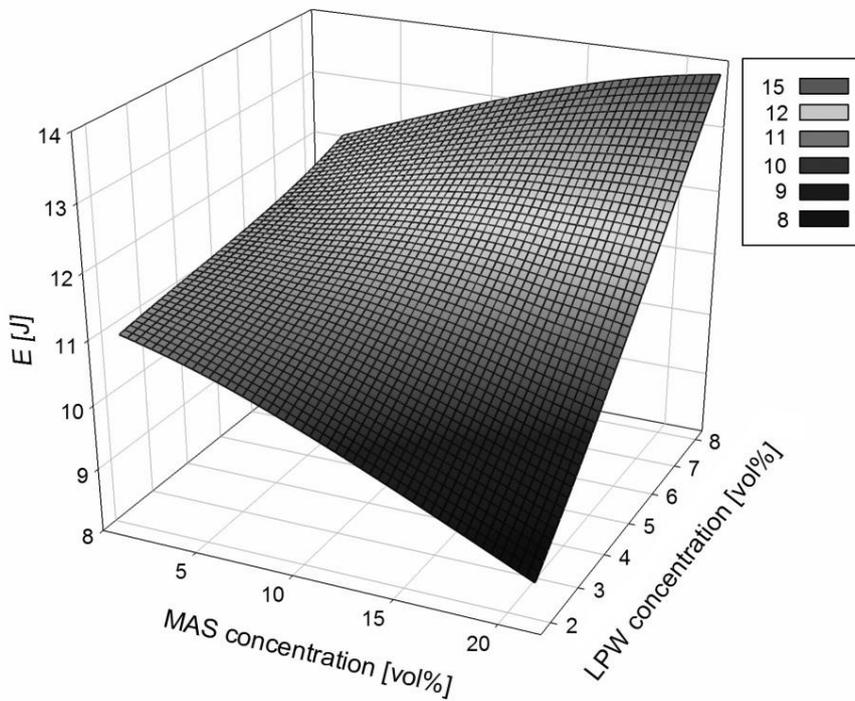
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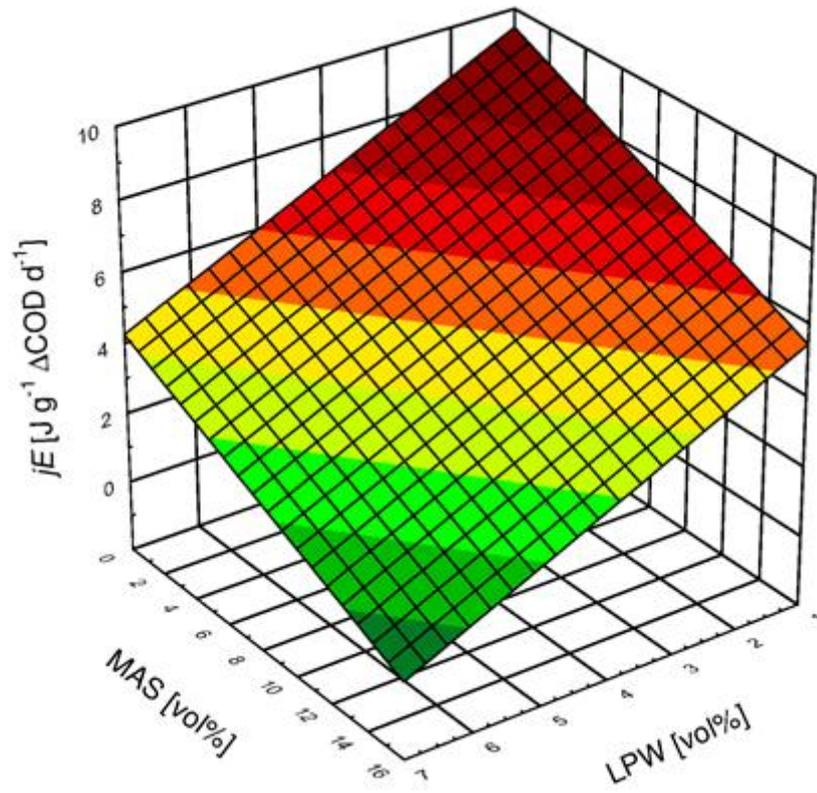
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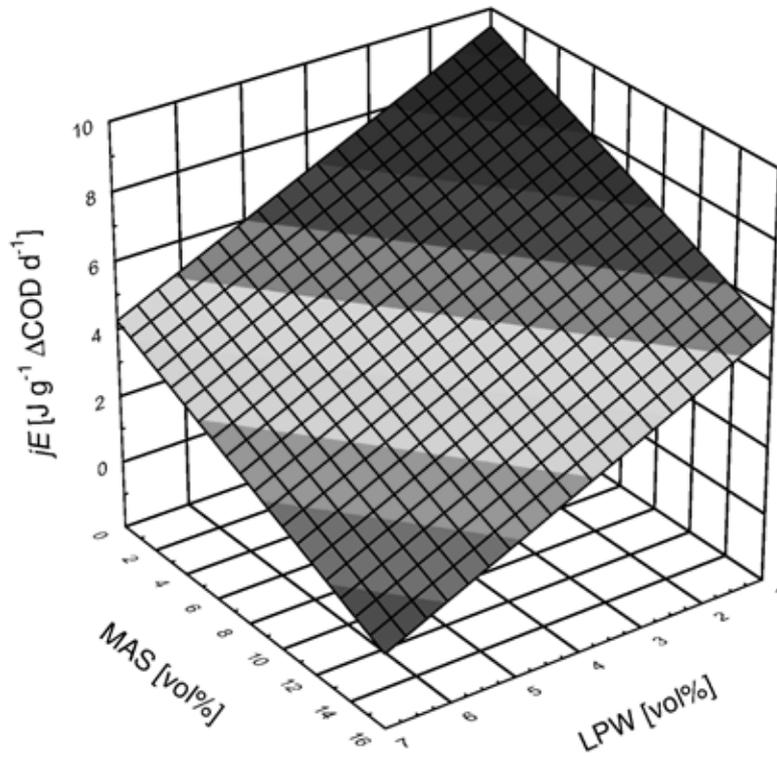
3

1   Table 1 – The main characteristics of LPW

2

Parameter	Value
total COD	111.6 g L^{-1}
BOD$_5$	50.8 g L^{-1}
pH	4.7
Protein content	42.5 g L^{-1}
TS	7.3 %
Reducing sugar content	1.6 g L^{-1}
TOC	35.9 g L^{-1}

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1   Table 2 – Parameters of the experimental design

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MFC ID	Added LPW [mL]	Added MAS [mL]	LPW concentration [vol%]	MAS[a] concentration [vol%]
1.	5.0	9.0	8.33	15.00
2.	1.0	5.0	1.67	8.33
3.	1.0	13.0	1.67	21.67
4.	3.0	7.0	5.00	11.67
5.	5.0	1.0	8.33	1.67

3   [a]: the term MAS concentration is identical to "reinoculation ratio"

4

1   Table 3 – Values of energy parameters obtained from the results of the experimental

2   design

3

MFC ID	LPW:MAS [mL:mL]	$U_{\max}$ [mV]	jI_{max} [mA m^{-2}]	jP_{max} [mW m^{-2}]	E [J]	CE [%]
1.	5:9	43.8	175	7.7	13.4	1.95
2.	1:5	54.6	218	11.9	10.6	1.86
3.	1:13	47.8	191	9.2	8.8	1.25
4.	3:7	49.9	200	10.0	11.6	1.57
5.	5:1	38.0	152	5.8	12.0	1.41

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1   Table 4 – Comparison of current density values

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MFC type	Feed	Current density [mA m^{-2}]	Reference
Dual-chamber MFC	Food industry wastes, compost leachate	232	Cercado-Quezada et al. (2010)
Single-chamber MFC	leachate	114	Ganesh and Jambeck (2013)
Dual-chamber MFC	pre-digested landfill leachate	418-548	Tugtas et al. (2013)
Dual-chamber MFC	Liquid fraction of pressed solid waste	152-218	This work

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4

1   Table 5 – COD parameters of each MFCs

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MFC ID	COD input [g]	Initial COD [g L^{-1}]	Final COD [g L^{-1}]	COD removal [%]
1.	0.855	44.67	2.31	94.8
2.	0.268	34.88	5.11	85.4
3.	0.512	38.94	4.11	89.5
4.	0.562	39.77	8.52	78.6
5.	0.612	40.61	4.84	88.1

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4

1   List of abbreviations

Abbreviation	Full name
BOD	Biological oxygen demand
CE	Coulombic efficiency
COD	Chemical oxygen demand
LPW	Liquid fraction of pressed solid waste
MAS	Mesophilic anaerobic sludge
MFC	Microbial fuel cell
TOC	Total organic carbon
TS	Total solids

2