

See discussions, stats, and author profiles for this publication at: <http://www.researchgate.net/publication/276061421>

Comparison of Anaerobic Degradation Processes for Bioenergy Generation from Liquid Fraction of Pressed Solid Waste

ARTICLE *in* WASTE AND BIOMASS VALORIZATION · MAY 2015

DOI: 10.1007/s12649-015-9379-y

DOWNLOADS

16

VIEWS

25

8 AUTHORS, INCLUDING:



Nándor Nemestóthy

University of Pannonia, Veszprém

48 PUBLICATIONS 380 CITATIONS

SEE PROFILE



Katalin Bélafi-Bakó

University of Pannonia, Veszprém

90 PUBLICATIONS 920 CITATIONS

SEE PROFILE



Péter Bakonyi

University of Pannonia, Veszprém

26 PUBLICATIONS 98 CITATIONS

SEE PROFILE



Robert Kurdi

University of Pannonia, Veszprém

19 PUBLICATIONS 171 CITATIONS

SEE PROFILE

1 **Comparison of anaerobic degradation processes for bioenergy generation from liquid**
2 **fraction of pressed solid waste**

3
4 Rózsenszki T^a, Koók L^a, Hutvágner D^a, Nemestóthy N^a, Bélafi-Bakó K^a, Bakonyi P^{a,*}, Kurdi
5 R^b, Sarkady A^b

6
7 ^a: Research Institute on Bioengineering, Membrane Technology and Energetics

8 University of Pannonia

9 Egyetem ut 10, 8200 Veszprém, Hungary

10
11 ^b: Institute of Environmental Engineering

12 University of Pannonia

13 Egyetem ut 10, 8200 Veszprém, Hungary

14
15 *Corresponding Author: P. Bakonyi

16 Tel: +36 88 624385

17 Fax: +36 88 624292

18 E-mail: bakonyip@almos.uni-pannon.hu

19

20

1 **Abstract**

2

3 A novel substrate (obtained from biofraction of municipal solid waste by pressing and
4 called LPW) rich in organic substances was used in three anaerobic degradation processes
5 (biogas, biohydrogen fermentation and microbial fuels cells) to comparatively assess their
6 feasibility for energy recovery. It has turned out that all the processes have successfully degraded
7 that substrate and produced energy carriers (methane and hydrogen) as well as bioelectricity. The
8 maximum energy yields ($\text{J g}^{-1} \text{COD}_{\text{removed}} \text{d}^{-1}$) and associated COD removal capacities were 255,
9 200, 2.8 and 46 %, 52 % and 72 % for biohydrogen, biogas and microbial fuel cell, respectively.
10 The outcomes suggested the prominence of biohydrogen process for simultaneous waste
11 treatment and energy recovery from LPW under the test conditions ensured.

12

13 **Keywords:** organic waste, anaerobic degradation, biogas, biohydrogen, microbial fuel cell

14

1 **1. Introduction**

2
3 Nowadays, energy production and waste treatment are two crucial issues to maintain an
4 adequate level of sustainable and environmental-safe technologies [1-3]. Among the various
5 options to manage municipal garbage, deposition in landfills is the less attractive. Hence,
6 applications of methods that utilize organic waste streams and simultaneously produce
7 (renewable) energy are of particular interest and represent a hot research field currently.
8 Biological approaches – being either aerobic or anaerobic – have been proven to efficiently
9 manage various substances, including different fractions of municipal solid waste which is
10 formed in remarkable quantities globally.

11 The North-Balaton Waste Treatment Plant (WTP) was opened recently in
12 Királyszentistván, Hungary, which is one of the most up-to-date domestic solid waste treatment
13 plants. It is able to receive 120 000 t waste year⁻¹ from 158 villages, cities nearby in Veszprém
14 county. The processing steps of WTP are outlined in Fig. 1. First, the incoming wastes are
15 mechanically treated. After shredding, iron is removed and the remaining parts are consecutively
16 separated and sorted. As a result, an appropriate portion is sent for recycling, while others are
17 subjected to further techniques [4].

18 Approximately, one third of the total annual waste is of organic origin and referred as the
19 biofraction of municipal solid waste (BFMSW). It is drum-screened and exclusively treated
20 aerobically by composting, which does not provide any benefits from energy point of view.
21 However, BFMSW has notable energy potential. Therefore, in this study, it was aimed to test
22 alternative ways of utilization that make energy recovery from BFMSW possible. In accordance
23 with the interest of WTP, raw BFMSW has undergone pressing and a liquid fraction (liquid
24 pressed waste – LPW) rich in organic compounds was obtained. Subsequently, LPW was
25 employed as a substrate in our experiments (Fig. 2) for energy production in mature (biogas) as
26 well as emerging (biohydrogen, microbial fuel cell) anaerobic degradation technologies.

27 Anaerobic digestion and concomitant biogas formation is an industrially recognized
28 process [5-7], while biohydrogen is still an emerging energy carrier even though it has been
29 studied for a long time [8-10]. Biogas and biohydrogen fermentations involve the
30 microbiologically-assisted degradation of organic compounds and result in flammable gaseous
31 energy carriers, which should be collected and purified [11-15]. In our earlier projects biogas

1 [16,17] as well as biohydrogen [18-22] production from various substrates have been
2 investigated.

3 Microbial fuel cells (MFCs) are prosperous applications in the early-stage of research and
4 gained remarkable attention from the scientific community in the last decade [23-25]. The MFC
5 is a device that converts chemical energy into electricity through the biocatalytic activity of
6 electrochemically active microorganisms. The classical MFC system consists of two main
7 compartments: an anaerobic anode chamber and an air-sparged cathode compartment that are
8 physically separated by a proton-exchange membrane (PEM). In appropriate conditions, biofilm
9 can grow on the surface of the anode. Organic substances such as acetate, sugar or wastewater
10 can be used as feedstock [26,27] in the microbial degradation process and as a result biomass,
11 CO₂, electrons and protons are produced. The electrons are delivered from the anode to the
12 cathode through an external circuit, while protons move across the membrane to cathode to
13 maintain electroneutrality. Oxygen supplied in the cathode chamber reacts with the protons and
14 electrons to form water. The main process, which is crucial in terms of the electrical
15 performance, is the transport of electrons from the cells to the anode surface. Therefore the
16 properties and the surface area of the applied electrode material play an important role in the
17 performance of the MFC [28]. In our former research, mesophilic anaerobic sludge was used as
18 inocula in the MFCs seeking the operating stability and the effects of various substrates
19 (monosaccharides, organic acids, industrial wastes) on system performance [26,27].

20 In this work, driven by the request of WTP, alternative routes for the BFMSW treatment
21 were tested. Hence, LPW, a liquid derivative of BFMSW was provided by the WTP and applied
22 as a novel starting material in three anaerobic technologies, namely biogas, biohydrogen and
23 microbial fuel cell for energy extraction purposes. To our best knowledge, this is the first study
24 on the applicability of LPW in the three contestant application mentioned.

25
26

2. Materials and methods

2.1. Substrate (LPW) and inoculum

LPW, the liquid fraction of BFMSW gained by pressing, was used as a substrate in the experiments. The scheme of LPW production as well as the major LPW characteristics are presented by Fig. 2. Anaerobic sludge – taken from a domestic biogas plant operated in mesophilic conditions – with 30 g L⁻¹ total COD value and pH of 7.8 was applied as seeding source.

2.2. Biogas and biohydrogen experiments

Experiments for biogas (Fig. 3) and biohydrogen (Fig. 4) production were carried out in WTW OXITOP 100 manometric vessels having 500 mL total volume (Fig. 5). The reactors were filled with 50 mL substrate-sludge mixture in various ratios for biogas (Table 1) and in 1:1 ratio for biohydrogen (Table 2). The bioreactors were initially purged with high purity nitrogen (>99.9%) to ensure fully anaerobic conditions. Subsequently, the vessels were closed by special manometric caps which were able to record the increasing pressure of the evolving gas in well-defined time intervals. The sealed bottles were incubated at 37 °C and 220 rpm stirring rate was thoroughly applied using magnetic bar. The volume of the gas produced was calculated from the pressure data by using the ideal gas law. The given cumulative gas volumes in Table 1 and 2 are valid at 37 °C and 1 bar pressure. For the biohydrogen measurements, the initial pH values (Table 2) were adjusted by HCl solution and the mesophilic sludge was subjected to heat-pretreatment (75 °C, 45 min) to kill methanogenic organisms by adopting the method described in our previous research [21].

2.3. Microbial fuel cells

Dual-chamber MFCs were constructed (Fig. 6). Anode and cathode chambers had the same 60 mL capacity. Nafion 115 proton exchange membrane (7 cm² surface area) was employed in between anode and cathode compartments. The anode and cathode electrodes

1 possessed 25 cm² surface areas and were made of carbon cloth. In the anode cell, using untreated
2 mesophilic sludge was loaded (60 mL) as inoculum and anaerobic circumstances were provided,
3 while the cathode side was filled with distilled water (60 mL) and continuously purged with air.
4 The system was thermostated at 37 °C. The voltage generated was measured by using data logger
5 (National Instruments USB-6008) and fixed 100 Ω external resistors. The cells were started-up
6 using the sludge only to develop electrochemically active anode-surface biofilm (first step in Fig.
7 7). Afterwards, the anode chamber was fed with the LPW and the collection of data was started
8 (second step in Fig. 7) and initial COD values were measured (Table 3). Prior to the addition of
9 LPW (Table 3), equal amount of anode-side solution was withdrawn to keep constant working
10 volume.

11 **2.4. Analytical methods**

12
13
14 The gas compositions for biogas and biohydrogen were occasionally determined by
15 Hewlett Packard 5890 gas chromatograph equipped with thermal conductivity detector (TCD).
16 Capillary CarboPlot[®] column was employed (Agilent Technologies, length: 60 m, ID: 0.32 mm,
17 film thickness: 1.5 mm) with N₂ (99.9 %) as carrier gas at a flow rate of 30 mL min⁻¹. Split ratio
18 of 50:1 was applied. The temperature of injector, column oven and detector was 130°C, 90 °C
19 and 115 °C, respectively. Chemical oxygen demand (COD) was followed according to standard
20 methods [29]. The pH was measured by using a conventional pH glass electrode.

21 **2.5. Evaluation of experimental results**

22
23
24 The results presented throughout in this work are the mathematical averages of triplicates
25 and the standard deviations were below 5 %. Energy yields (J_E) (Table 4) were calculated by
26 considering the gravimetric energy content of methane (55.7 kJ g⁻¹) and hydrogen (142 kJ g⁻¹)
27 generated, the full operation times and the COD removed during the bioprocess. For MFCs,
28 current (*I*) data was computed according to Ohm's law in view of the voltage (*U*) and the applied
29 resistance (*R*). J_E for MFC was determined from voltage and current data, operating time and the
30 COD eliminated. The cumulated energy (*E*) data for MFC can be estimated on the basis of power

1 (from the voltage and current data) generated by integrating peak area (Fig. 7). This quantity
2 shows the usable electrical energy (Table 3).

3 The efficiency of the treatments was characterized by J_E and COD removal, which were
4 calculated by Eq. 1 and Eq. 2:

$$5 \quad J_E = E \Delta\text{COD}^{-1} t^{-1} \quad (1)$$

$$6 \quad \text{COD removal (\%)} = 100(\Delta\text{COD} \text{ COD}_0^{-1}) \quad (2)$$

7
8
9
10 where E is the produced energy in case of each processes, ΔCOD is the COD removed in the
11 course of the experiments, COD_0 is the initial total COD and t is the operation time.

12 13 **3. Results and discussion**

14 15 **3.1. Biogas production from LPW**

16
17 Since LPW had not been used in any microbial degradation process so far, experiences
18 with its utilization were not available. Therefore, experiments were carried out applying various
19 ratios of LPW and untreated sludge. It has turned out that high LPW/sludge ratios in the mixture
20 should be avoided because the pH will drop to the acidic range that inhibits methanogenic
21 organisms. Hence, initial LPW/sludge ratios were set to lower levels and it was found that the
22 1:1 ratio provided the highest cumulative methane formation (424 mL), as it can be observed in
23 Table 1. The trends of biogas and methane evolution are illustrated in the progress curve (Fig. 3)
24 for the case of using 1:1 LPW to sludge ratio. Besides gas production performance, the initial
25 and final COD values are also listed in Table 1. It can be seen that biogas fermentation efficiency
26 correlates well with COD removal since higher under high LPW/sludge ratio the low biogas
27 yield (65 mL) was accompanied by low COD reduction (14.4 %). On the other hand, under
28 optimal circumstances, quite good (52.3 %) COD removal could be attained where (Table 1).

29 Projecting the results with the literature is rather difficult due to the lack of studies with
30 LPW substrate. Nevertheless, landfill leachate may be taken into account as a material which has
31 more or less similar origin than LPW since it is also derived (at least in part) from solid wastes.

1 Oz and Yarimtepe [30] investigated the utilization of raw landfill leachate ($28.5 \text{ g COD L}^{-1}$, 0.35
2 BOD COD^{-1}) for biogas evolution in batch reactors under mesophilic temperature ($35 \text{ }^\circ\text{C}$) and
3 $1.13 \text{ L biogas L}^{-1} \text{ d}^{-1}$ yield accompanied by 44 % soluble COD reduction was achieved. These
4 research outcomes more or less agree with the ones in this study, where $0.31 \text{ L biogas L}^{-1} \text{ d}^{-1}$ gas
5 yields and 52.3 % COD removal efficiency could be reached. Furthermore, it was shown that
6 (ultrasound) pretreatment of leachate can significantly enhance the anaerobic decomposition
7 efficiency since it increases the soluble COD/total COD ratio and subsequently the amount of
8 accessible substances for the bacteria [30]. Hence, this strategy may be equally useful for the
9 management of LPW in the future. In another paper, Luo et al. [31] produced biogas from
10 leachate of solid municipal waste demonstrating high-strength organic pollutant content ($41\text{-}59 \text{ g}$
11 COD L^{-1}) and good biodegradability ($0.59\text{-}0.64 \text{ BOD COD}^{-1}$), which are comparable with our
12 LPW characteristics ($111.6 \text{ g COD L}^{-1}$, $0.45 \text{ BOD COD}^{-1}$). The measurements were conducted in
13 continuous expanded granular sludge bed reactor at $35 \text{ }^\circ\text{C}$. The COD removal exceeded 80%,
14 while $6 \text{ L}_{\text{STP}} \text{ biogas L}^{-1} \text{ d}^{-1}$ yield was obtained under $15 \text{ kg COD m}^{-3} \text{ d}^{-1}$ organic loading rate and
15 3 d hydraulic retention time. In our batch system, $0.27 \text{ L}_{\text{STP}} \text{ biogas L}^{-1} \text{ d}^{-1}$ and 52.3 % COD
16 removal efficiency could be attained under $1.75 \text{ kg COD m}^{-3} \text{ d}^{-1}$ OLR (considering 40 days of
17 fermentation and 3.5 g of total COD initially loaded to the batch bottles with 25 mL of LPW and
18 25 of mL sludge, as appears in Table 1). Presumably, the differences of the systems are
19 accountable for the altered performances in terms of COD removal and biogas yield and suppose
20 that continuous applications could be more productive and feasible to convert liquid landfill
21 wastes into bioenergy carrier, methane.

22

23 **3.2. Biohydrogen evolution from LPW**

24

25 Pre-experiments were carried out to estimate the proper ratio for LPW and pretreated
26 sludge. It was found that their equal portions (1:1) in the mixture are beneficial for our purpose
27 (similarly to the biogas measurements). It is known that the initial pH can have a marked impact
28 on biohydrogen formation process and acidic levels are favored by H_2 -producing strains [8].
29 Consequently, a wide range of pH (4.5-7.0) was screened to find the optimal value. The time
30 profiles of the cumulated gas formations are depicted in Fig. 4, meanwhile other experimental
31 results are summarized in Table 2. It can be seen in Fig. 4 that remarkable gas volumes were

1 formed, except at pH=4.5. The H₂ content, however, was surprisingly low in the cases of pH 5.5-
2 7.0 since vast majority of the gas was CO₂. Nonetheless, even though a longer lag (adaption)
3 phase was observed with pH=5.0 finally it resulted in the highest gas volume produced (161
4 mL), which contained high amount (69.9 %) of H₂. Pertaining COD, as it can be extracted from
5 Table 2, removal efficiency was lower than in case of biogas but it is still adequate. The best
6 COD reduction (46.3 %) was coupled to the highest H₂ production at pH of 5, which was
7 assigned as the optimum value and should be applied in the further experiments.

8 Similarly to the case of biogas, comparison of results from LPW is attempted with
9 landfill leachate. Liu et al. [32] optimized batch hydrogen generation from compost leachate and
10 juice wastewater. As they concluded, initial pH of 5.5 was the most preferred by the H₂-
11 producing cells at 35 °C. These findings nicely confirm ours in this study with LPW, since
12 optimum pH of 5 was identified at 37 °C, where the highest H₂ yield of 2.7 mol H₂ kg⁻¹
13 COD_{removed} was attained. It was also demonstrated by Liu et al. [32] that the co-digestion of juice
14 wastewater and compost leachate has led to a 28 % increment (2.06 mol H₂ mol⁻¹ glucose instead
15 of 1.6 mol H₂ mol⁻¹ glucose) in the hydrogen yield, which indicate well that liquid derivative of
16 solid biodegradable waste can serve as a potential nutrient source. In contrast, Watanabe and
17 Yoshino [33] conducted batch biohydrogen production research using landfill leachate derived
18 from waste glass, waste plastics and construction waste. In their study, maximum biohydrogen
19 yield (2.67 mol H₂ mol⁻¹ carbohydrate added) could be reached at pH=7 and 30 °C. The
20 disagreement of optimum can be attributed to unstandardized experimental conditions e.g.
21 inocula origin and substrate composition. Nevertheless, the gas mixture formed in all cases was
22 free of methane, even though the liquid waste substrates (leachates and LPW) were not treated
23 before use to suppress possible methanogenic activity.

24 25 **3.3.Bioelectricity generation from LPW**

26
27 In this study five MFCs were operated in parallel with different LPW supplementation
28 (Table 3). The data showed that the degradation of organic substances in the LPW and
29 bioelectricity production took place in parallel. A typical voltage vs. time diagram is
30 demonstrated in Fig. 7.

1 MFCs can be characterized well in terms of substrate utilization by the calculation of the
2 cumulated energy yield (Table 3) which ranged between 2.55 J and 18.25 J depending on the
3 amount of LPW added. It can be pointed out that COD removal was excellent, mostly exceeding
4 80 %, which shows that MFCs are useful in wastewater treatment. Fig. 8 plots the cumulated
5 energy vs. LPW COD input where a linear relationship could be observed between the two
6 variables within the experimental borders. However, it should also be noted from Fig. 7 that
7 higher doses of LPW demanded significantly more operating to be bioconverted, which can play
8 a key role in process planning and changes the picture when calculating time-dependent energy
9 yields.

10 In agreement with biogas and biohydrogen related section above, MFC performance from
11 LPW is discussed comparatively with leachate. Leachate, as a landfill-originated liquid waste has
12 been shown as a feasible starting material in microbial fuel cells that are able to sufficiently
13 reduce the COD and BOD contents and simultaneously generate electricity. For example, Gálvez
14 and co-workers [34] established a cascade system encompassing of 3 MFCs connected in series
15 to treat leachate ($12.9 \text{ g COD L}^{-1}$, $0.49 \text{ BOD COD}^{-1}$) and reported 79.4% COD removal
16 efficiency in 4 days with continuous recirculation. Ganesh and Jambeck [35] designed MFCs
17 where leachate was used not only as a substrate but also as an inoculum source. In their study,
18 COD could be removed with a 75 % efficiency using activated carbon anode in batch reactor,
19 which is slightly lower than in this work from LPW (mostly >80%). The current density reported
20 (114 mA m^{-2}) was fairly in par with our values ranging between 128-176 mA m^{-2} . Tugtas et al.
21 [36] used MFC to post-treat anaerobically pre-digested landfill leachate. In their study, stabilized
22 current densities varied from 418 to 548 mA m^{-2} and 90 % of the volatile fatty acids were
23 removed in the course of 12 days batch operation. As it can be deducted, not only current
24 densities but also COD elimination efficacies were higher than in this work (72-85 %). In
25 addition, The MFC performances obtained by Tugtas et al. [36] were better in comparison with
26 other literature studies applying untreated leachate and hence, it would appear that MFC could
27 serve as promising auxiliary equipment for partially digested liquid-form landfill wastes. It is
28 also to note that their MFC devices were continuously stirred which could have a notable
29 contribution to the attractiveness of the system, since it facilitates the mass (nutrient) transfer
30 between anodic biofilm and bulk phases.

31

3.4. Comparison of the processes

The three processes can be compared on the basis of the products (regarding their energy content) generation in microbial fuel cell under optimal conditions. In Table 4, the performances of biogas, biohydrogen fermentation and bioelectricity are shown. For comparison purposes, maximum energy yields ($\text{J g}^{-1} \Delta\text{COD d}^{-1}$) and respective COD removal efficiencies were chosen as primarily process indicators.

Regardless of the technology, it can be seen that COD elimination capacities were found rather comparable. However, MFC provided the best one with 71.6 %, followed by biomethane (52.3 %) and biohydrogen (46.3 %). Considering J_E , it has turned out that biohydrogen took the lead ($255 \text{ J g}^{-1} \Delta\text{COD d}^{-1}$), followed by biomethane ($200 \text{ J g}^{-1} \Delta\text{COD d}^{-1}$) meanwhile MFC remained order of magnitude behind them with $2.8 \text{ J g}^{-1} \Delta\text{COD d}^{-1}$.

4. Conclusions

Three anaerobic degradation processes were applied to test a new substrate, the liquid fraction of BFMSW (LPW) for biogas, biohydrogen formation and for power generation in microbial fuel cells. Successful experiments were carried out in all the processes, the unconventional substrate – rich in organic substances – was microbiologically transformed into methane, hydrogen and power. It has turned out that all the processes degraded the waste effectively, the COD removal efficiencies for MFC measurements exceeded even 70 %, while in case of biogas and biohydrogen it was found below. Biogas formation provided the highest energy output, but the highest energy yield (energy value normalized by the COD degradation value and operating time) was found in case of biohydrogen production. Thus, from the energy point of view, biohydrogen seems the most promising way to obtain energy from the particular waste material, LPW.

In the future, measurements will be continued with all the three processes to further optimize the bioprocess conditions before final recommendation since not only energy, but other aspects can be taken into account e.g. reliability of the process, pre- or post-treatment is necessary or not. For example, the gaseous energy carriers (CH_4 or H_2) needs a transformation step for utilization (e.g. combustion of methane in a co-generation process, oxidation of

1 hydrogen in a PEMFC with preliminary gas purification), while MFCs provide directly electric
2 energy. As a result, combined applications (i.e. biohydrogen integrated with MFC) might be
3 better solutions. In addition, the energy investment of waste processing to obtain LPW should
4 also be considered when selecting the most feasible technology for the management of municipal
5 solid waste. It is likely that the final system should involve the treatment of residual solid waste
6 (secondary product of pressing LPW substrate). For this purpose, anaerobic-phased solids
7 digesters could be good candidates to produce additional biogas and compensate the energy
8 consumption of pressing [37].

10 **Acknowledgements**

11
12 This work was supported by the European Union and co-financed by the European Social
13 Fund in the frame of the TAMOP-4.2.2.A-11/1/KONV-2012-0071 and TAMOP-4.2.2/A-
14 11/1/KONV-2012-0038 projects. Nándor Nemestóthy acknowledges the János Bolyai Research
15 Scholarship of the Hungarian Academy of Sciences.

1 **References**

- 2
- 3 [1] Fino, D., Conti, E., Martini, R., Conti, R.: Future Perspectives for Bio-Energy Production in
4 the Province of Turin. *Waste Biomass Valor.* 2, 59-64 (2011)
- 5 [2] Savva, P.G., Costa, C.N., Charalambides, A.G.: Environmental, Economical and Marketing
6 Aspects of the Operation of a Waste-to-Energy Plant in the Kotsiatis Landfill in Cyprus. *Waste*
7 *Biomass Valor.* 4, 259-269 (2013)
- 8 [3] Zis, T., Bell, M.G.H., Tolis, A., Aravossis, K.: Economic Evaluation of Alternative Options
9 for Municipal Solid Waste Management in Remote Locations. *Waste Biomass Valor.* 4, 287-296
10 (2013)
- 11 [4] Sarkady, A., Kurdi, R., Rédey, A.: RDF – Refuse derived fuel, possibilities in the North-
12 Balaton Regional waste management system. *Pollack Periodica.* 9, 23-30 (2014)
- 13 [5] Gerardi, H.M.: *The microbiology of anaerobic digesters*, John Wiley & Sons, Hoboken, New
14 Jersey (2003)
- 15 [6] Mata-Alvarez, J., Macé, S., Llabrés, P.: Anaerobic digestion of organic solid wastes. An
16 overview of research achievements and perspectives. *Bioresour. Technol.* 74, 3-16 (2000)
- 17 [7] Speece, R.E.: *Anaerobic biotechnology for industrial wastewaters*, Archae Press, Tennessee,
18 USA (1996)
- 19 [8] Bakonyi, P., Nemestóthy, N., Simon, V., Bélafi-Bakó, K.: Review on the start-up experiences
20 of continuous fermentive hydrogen producing bioreactors. *Renew. Sust. Energ. Rev.* 40, 806-813
21 (2014)
- 22 [9] Bakonyi, P., Nemestóthy, N., Simon, V., Bélafi-Bakó, K.: Fermentative hydrogen production
23 in anaerobic membrane bioreactors: A review. *Bioresour. Technol.* 156, 357-363 (2014)
- 24 [10] Kumar, G., Bakonyi, P., Periyasamy, S., Kim, S.H., Nemestóthy, N., Bélafi-Bakó, K.:
25 Lignocellulose biohydrogen: practical challenges and recent progress. *Renew. Sust. Energ. Rev.*
26 (2015). <http://dx.doi.org/10.1016/j.rser.2015.01.042>

27

28

- 1 [11] Bakonyi, P., Nemestóthy, N., Bélafi-Bakó, K.: Biohydrogen purification by membranes: An
2 overview on the operational conditions affecting the performance of non-porous, polymeric and
3 ionic liquid based gas separation membranes. *Int. J. Hydrogen Energ.* 38, 9673-9687 (2013)
- 4 [12] Bakonyi, P., Kumar, G., Nemestóthy, N., Lin, C.Y., Bélafi-Bakó, K.: Biohydrogen
5 purification using a commercial polyimide membrane module: studying the effects of some
6 process variables. *Int. J. Hydrogen Energ.* 38, 15092-15099 (2013)
- 7 [13] Ramírez-Morales, J.E., Tapia-Venegas, E., Nemestóthy, N., Bakonyi, P., Bélafi-Bakó, K.,
8 Ruiz-Filippi, G.: Evaluation of two gas membrane modules for fermentative hydrogen
9 separation. *Int. J. Hydrogen Energ.* 38, 14042-14052 (2013)
- 10 [14] Bauer, F., Persson, T., Hulteberg, C., Tamm, D.: Biogas upgrading - technology overview,
11 comparison and perspectives for the future. *Biofuels Bioprod. Biorefin.* 7, 499-511 (2013)
- 12 [15] Chmielewski, A.G., Urbaniak, A., Wawryniuk, K.: Membrane enrichment of biogas from
13 two-stage pilot plant using agricultural waste as a substrate. *Biomass Bioenergy* 58, 219-228
14 (2013)
- 15 [16] Szentgyörgyi, E., Nemestóthy, N., Bélafi-Bakó, K.: Anaerobic moving bed biofilm
16 fermenter for biogas production. *Env. Prot. Eng.* 36, 117-125 (2010)
- 17 [17] Szentgyörgyi, E., Nemestóthy, N., Bélafi-Bakó, K.: Application of membranes in biogas
18 production. *Desalin. Water Treat.* 14, 112-115 (2010)
- 19 [18] Bélafi-Bakó, K., Búcsú, D., Pientka, Z., Bálint, B., Herbel Z., Kovács, K.L., Wessling, M.:
20 Integration of biohydrogen fermentation and gas separation processes to recover and enrich
21 hydrogen. *Int. J. Hydrogen Energ.* 31, 1490-1495 (2006)
- 22 [19] Bakonyi, P., Nemestóthy, N., Lövitusz, É., Bélafi-Bakó, K.: Application of Plackett-
23 Burman experimental design to optimize biohydrogen fermentation by *E. coli* (XL1-BLUE). *Int.*
24 *J. Hydrogen Energ.* 36, 13949-13954 (2011)
- 25 [20] Bakonyi, P., Nemestóthy, N., Ramirez, J., Ruiz-Filippi, G., Bélafi-Bakó, K.: *E. coli* (XL1-
26 BLUE) for continuous fermentation of bioH₂ and its separation by polyimide membrane. *Int. J.*
27 *Hydrogen Energ.* 37, 5623-5630 (2012)

- 1 [21] Bakonyi, P., Borza, B., Orlovits, K., Simon, V., Nemestóthy, N., Bélafi-Bakó, K.:
2 Fermentative hydrogen production by conventionally and unconventionally heat pretreated seed
3 cultures: a comparative assessment. *Int. J. Hydrogen Energ.* 39, 5589-5596 (2014)
- 4 [22] Bakonyi, P., Nemestóthy, N., Lankó, J., Rivera, I., Buitrón, G., Bélafi-Bakó, K.:
5 Simultaneous biohydrogen production and purification in a double-membrane bioreactor system.
6 *Int. J. Hydrogen Energ.* 40, 1690-1697 (2015)
- 7 [23] Logan, B.E.: *Microbial fuel cells*. John Wiley & Sons, New York (2008)
- 8 [24] Logan, B.E., Hamelers, B., Rozendal, R., Schröder, U., Keller, J., Freguia, S., Aelterman,
9 P., Verstraete, W., Rabaey, K.: *Microbial fuel cells: methodology and technology*. *Environ. Sci.*
10 *Technol.* 40, 5181-5192 (2006)
- 11 [25] Lovley, D.R.: *Microbial fuel cells: novel microbial physiologies and engineering*
12 *approaches*. *Curr. Opin. Biotechnol.* 17, 327-332 (2006)
- 13 [26] Bélafi-Bakó, K., Vajda, B., Nemestóthy, N.: Study on operation of a microbial fuel cell
14 using mesophilic anaerobic sludge. *Desalin. Water Treat.* 35, 222-226 (2011)
- 15 [27] Vajda, B., Nemestóthy, N., Bakonyi, P., Belafi-Bakó, K.: Xylose substrate as the only
16 nutrient in the operation of microbial fuel cells. *Env. Prot. Eng.* 40, 132-141 (2014)
- 17 [28] Wei, J., Liang, P., Huang, X.: Recent progress in electrodes for microbial fuel
18 cells. *Bioresour. Technol.* 102, 9335-9344 (2011)
- 19 [29] APHA, 1995. *Standard methods for the examination of water and wastewater*. 19th ed. New
20 York, USA: American Public Health Association.
- 21 [30] Oz, N.A., Yarıntepe, C.C.: Ultrasound assisted biogas production from landfill leachate.
22 *Waste Manage.* 34, 1165-1170 (2014)
- 23 [31] Luo, J., Lu, X., Liu, J., Qian, G., Lu, Y.: Biogas recirculation for simultaneous calcium
24 removal and biogas purification within an expanded granular sludge bed system treating
25 leachate. *Bioresour. Technol.* 173, 317-323 (2014)
- 26 [32] Liu, Q., Zhang, X., Zhou, Y., Zhao, A., Chen, S., Qian, G., Xu, Z.P.: Optimization of
27 fermentative biohydrogen production by response surface methodology using fresh leachate as
28 nutrient supplement. *Bioresour. Technol.* 102, 8661-8668 (2011)

1 [33] Watanabe, H., Yoshino, H.: Biohydrogen using leachate from an industrial waste landfill as
2 inoculum. *Renew. Energ.* 35, 921-924 (2010)

3 [34] Gálvez A, Greenman J, Ieropoulos I. Landfill leachate treatment with microbial fuel cells;
4 scale-up through plurality. *Bioresour. Technol.* 100, 5085-5091 (2009)

5 [35] Ganesh, K., Jambeck, J.R.: Treatment of landfill leachate using microbial fuel cells:
6 Alternative anodes and semi-continuous operation. *Bioresour. Technol.* 139, 383-387 (2013)

7 [36] Tugtas, A.E., Cavdar, P., Calli, B.: Bio-electrochemical post-treatment of anaerobically
8 treated landfill leachate. *Bioresour. Technol.* 139, 266-272 (2013)

9 [37] Zhu, B., Zhang, R., Gikas, P., Rapport, J., Jenkins, B., Li., X.: Biogas production from
10 municipal solid wastes using an integrated rotary drum and anaerobic-phased solids digester
11 system. *Bioresour. Technol.* 101, 6374-6380 (2010)

12

1 **List of tables and figures**

2

3 **Table 1** – Results of the biogas experiments

4

5 **Table 2** – Results of the biohydrogen experiments

6

7 **Table 3** – Experimental and calculated results of the MFC measurements

8

9 **Table 4** – Comparison of energy yields and COD removal efficiencies

10

11 **Fig. 1** – Scheme of the processes in the waste treatment plant

12

13 **Fig. 2** – LPW production and characteristics

14

15 **Fig. 3** – Biogas and biomethane formation as a function of time for 1:1 LPW to sludge ratio

16 Biogas: triangle; Methane: square

17

18 **Fig. 4** – Gas formation curves of the biohydrogen experiments

19 dark blue: pH=7; red: pH=4.5; green: pH=5; purple: pH=5.5; light blue: pH=6; orange: pH=6.5

20

21 **Fig. 5** – The experimental set-up for biogas and biohydrogen production

22 1 – manometric cap; 2: septum for gas sampling

23

24 **Fig. 6** – Illustration of the dual chamber MFC

25

26 **Fig. 7** – Effect of substrate (9 mL LPW) feeding on voltage output. 1: Inoculation 2: Substrate
27 (LPW) addition. (t represents the time interval that was considered for energy yield calculation)

28

29 **Fig. 8** – Cumulated energy output as a linear function of COD input ($R^2 = 0.989$) and required
30 operation time for the various cases (dotted line)

31

32

1 **Table 1**

LPW (mL)	Sludge (mL)	COD* _{in}	COD* _f	COD removal (%)	Biogas (mL)	Methane (mL)
15	35	2.7	1.18	56.5	515	355
25	25	3.5	1.65	52.3	622	424
35	15	4.4	3.7	14.4	369	65

*: initial (COD_{in}) and final (COD_f) COD values are in [g COD 50 mL⁻¹ mixture] unit

2

1 **Table 2**

Initial pH	COD [*] _{in}	COD [*] _f	COD removal (%)	Gas formed (mL)	H ₂ (%)	CO ₂ (%)
4.5		3.02	13.7	22	66.5	33.5
5		1.88	46.3	161	69.9	30.1
5.5	3.5	2.18	37.7	149	11.3	88.7
6		2.27	35.1	132	9	91
6.5		2.61	25.4	122	8	92
7		2.39	31.7	138	7	93

*: initial (COD_{in}) and final (COD_f) COD values are in [g COD 50 mL⁻¹ mixture] unit

2

3

1 **Table 3**

LPW feed (mL)	COD* _{in}	COD* _f	COD removal (%)	Current density (mA m ⁻²)	Operation time (d)	Cumulated energy (J)	J _E (J g ⁻¹ ΔCOD d ⁻¹)
1	0.36	0.1	72.2	160	3.5	2.55	2.80
3	0.64	0.11	82.8	128	12	6.32	0.99
5	1.07	0.2	81.3	140	14.5	10.71	0.85
7	0.92	0.18	80.4	152	15.5	15.93	1.39
9	1.29	0.26	79.8	176	16	18.25	1.11

*: initial (COD_{in}) and final (COD_f) COD values are in [g COD 60 mL⁻¹ mixture] unit

2

3

1 **Table 4**

	BioCH ₄	BioH ₂	MFC
Energy obtained (J)	14611	1 240	2.55
Time period (d)	40	3	3.5
COD removal (%)	52.3	46.3	72.2
J _E (J g ⁻¹ ΔCOD d ⁻¹)	200	255	2.8

2

3

Fig. 1

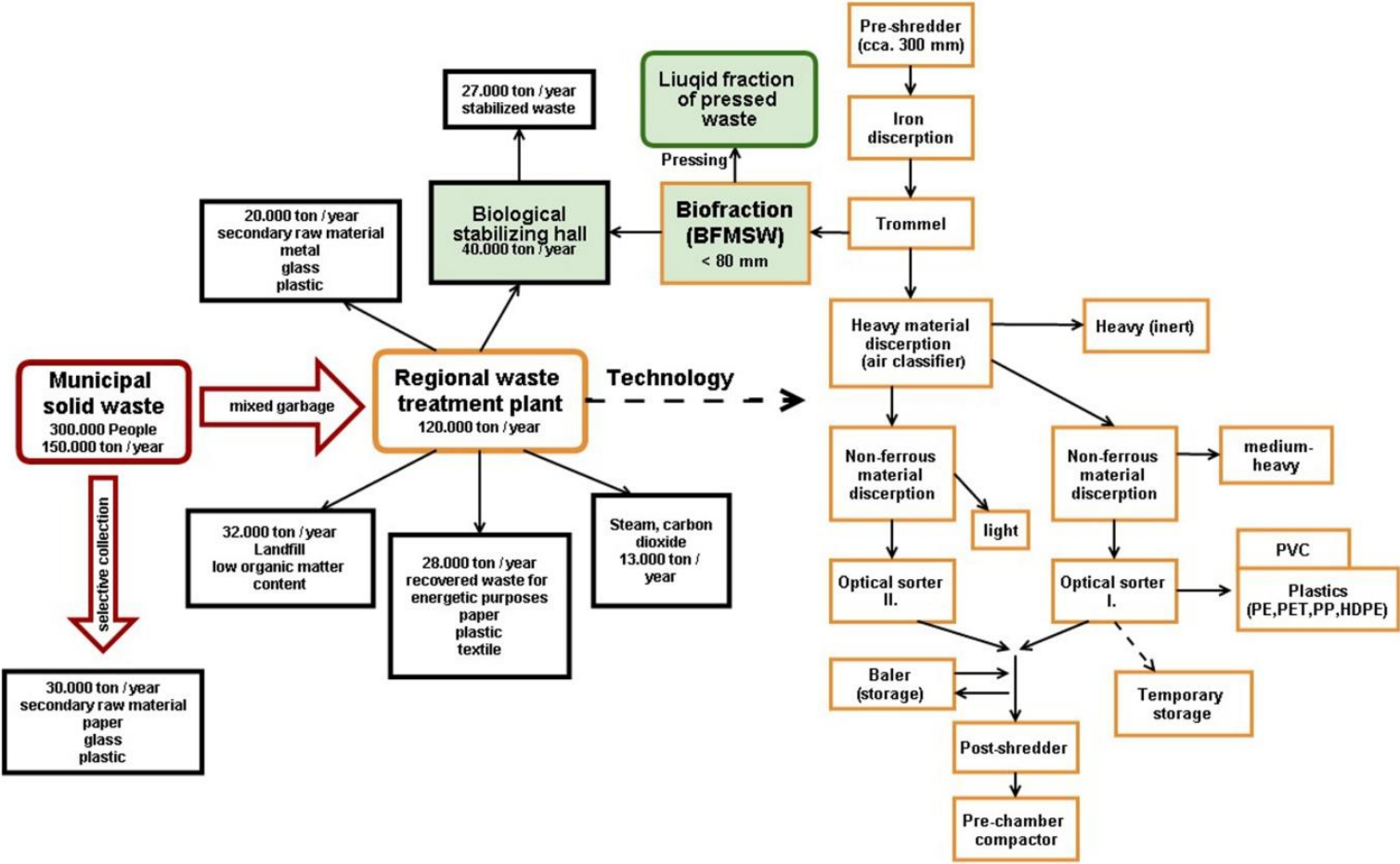


Fig. 2

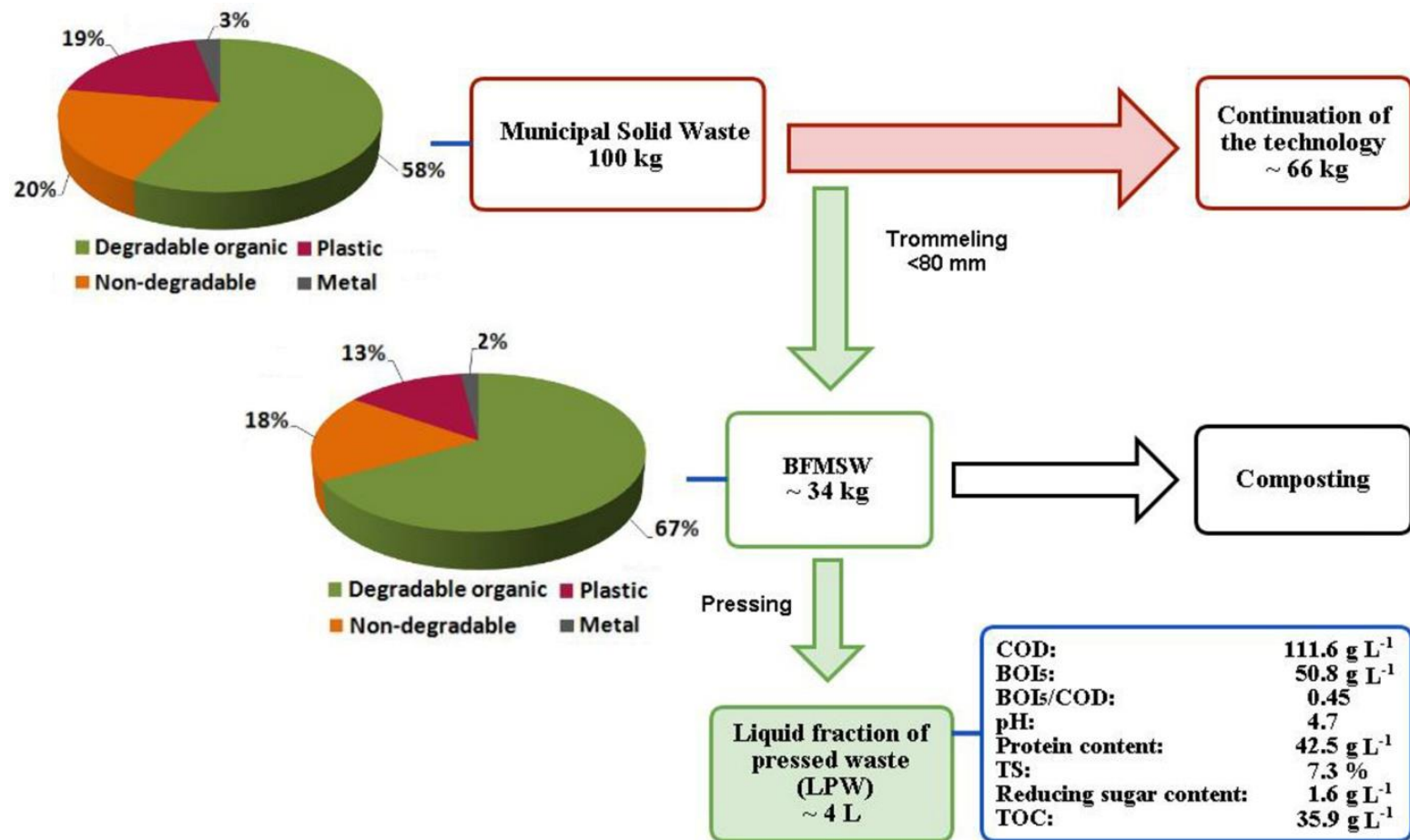


Fig. 3

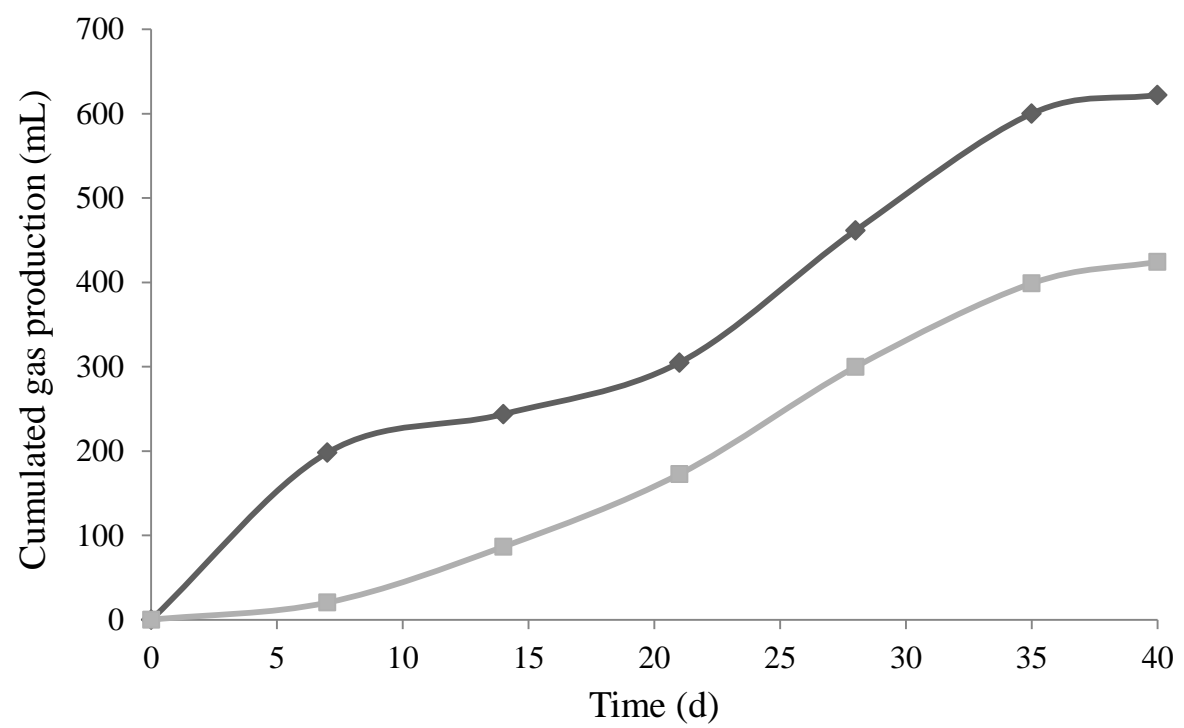


Fig. 4

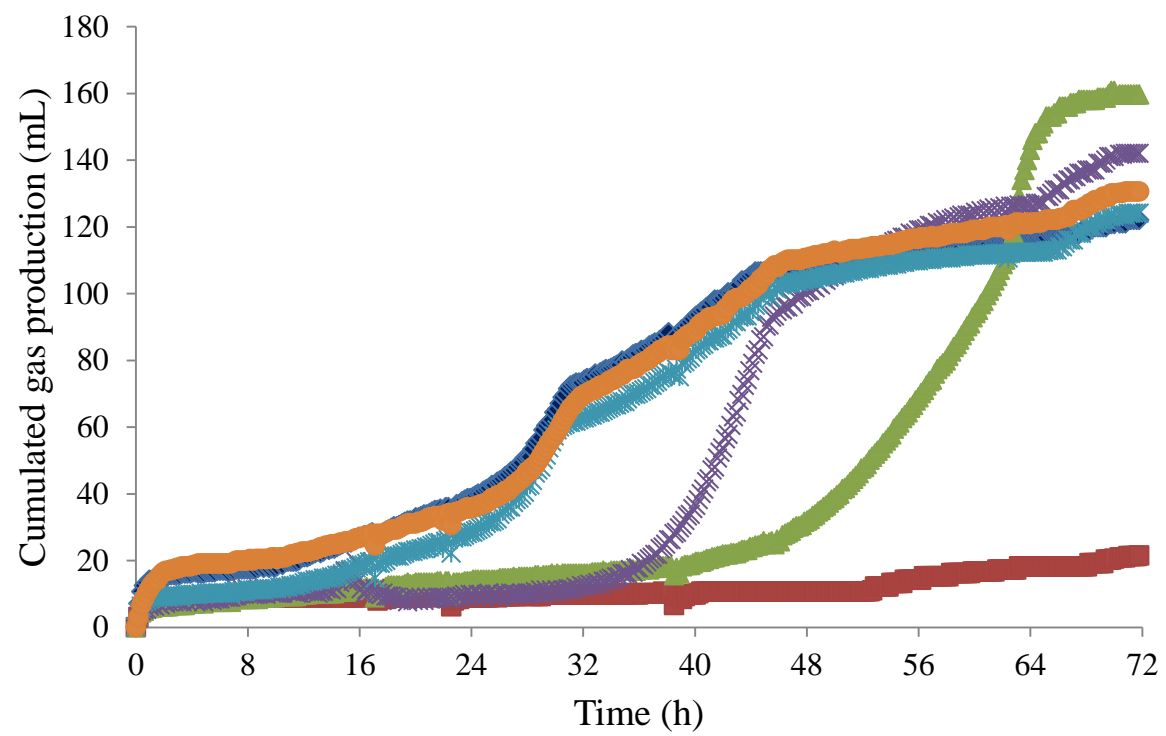


Fig. 5



Fig. 6

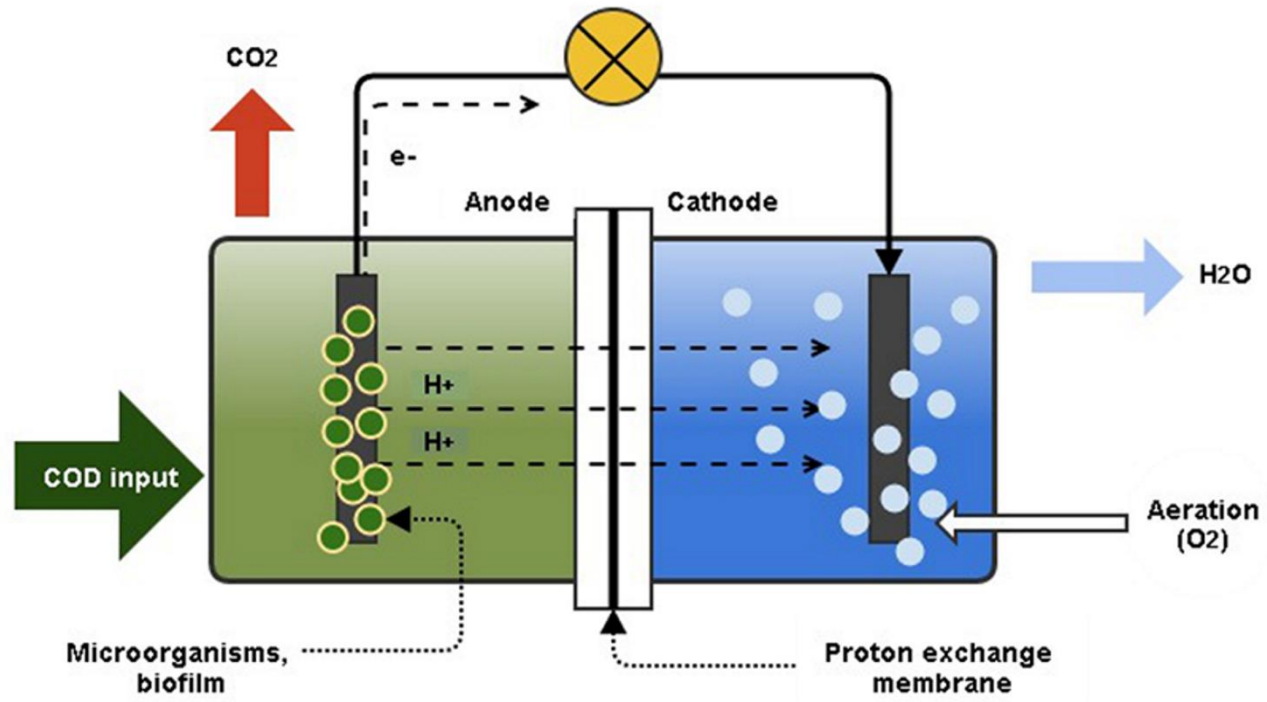


Fig. 7

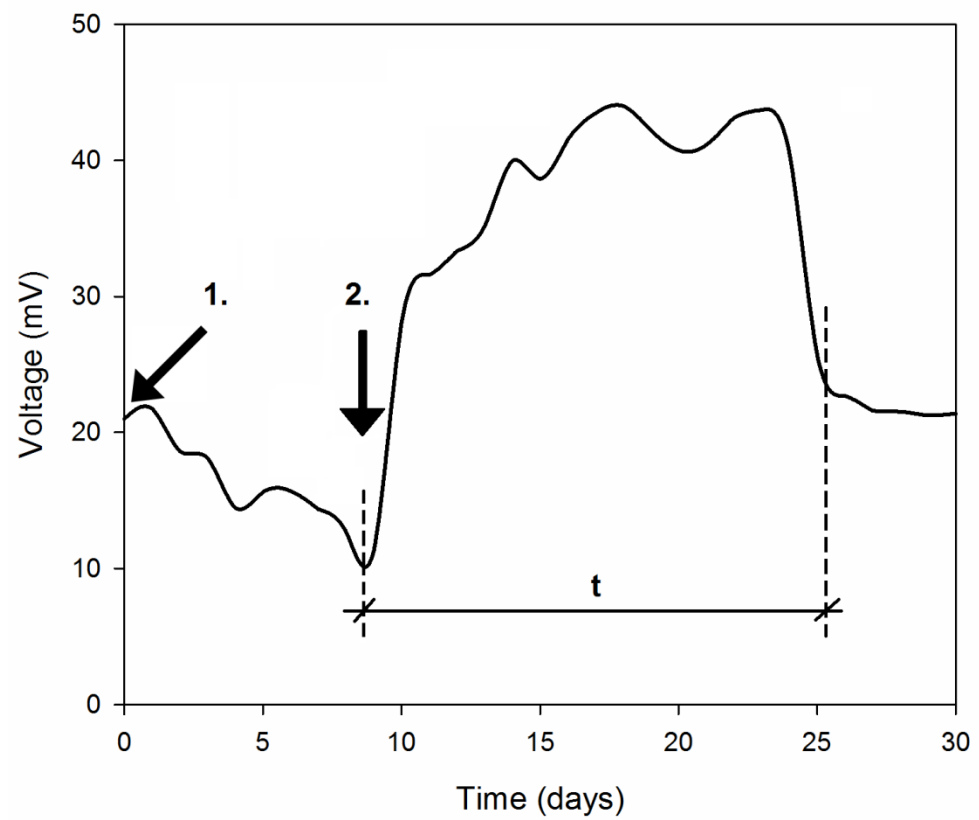


Fig. 8

