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Research Paper

Generation of Electricity and Sludge Reduction in a Microbial Fuel Cell

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Abstract: In the last few years, the attractiveness of renewable energy production from waste has increased attention for the use of microbial fuel cells. Microbial fuel cell (MFC) has a set-up which generates electrical energy from the biochemical energy released by the catabolic reactions of microbial growth. An MFC system, equipped with two chambers having chromium-nickel plate electrodes, was used to investigate the electricity generation potential in parallel to sludge reduction and carbon removal. In the first stage, activated sludge was cultivated for 1 month in a batch reactor prior to seeding into MFC. In the second stage, a lab-scale two- chambered MFC system was constructed. In the monitoring stage, the operation of MFC was examined in 2 different set of experiments, where MFC voltage generation (V) and digestion of sludge were recorded. Experimental results showed higher SCOD removal efficiency in the aerobic batch reactor (41.2% and 42.7%) compared to MFC system (32% and 32%) during Run I and Run II, respectively. The observed decrease in VSS was 31.5% and 30.7% in the MFC system, 51.8% and 53.9% in the batch aerobic reactor during Run I and Run II, respectively. The last stage was conducted to observe electrical parameters. Experimental findings show that, MFC performance is comparable to that of an aerobic sludge digester with the additional benefit of electrical energy generation.

Keywords*: Electricity generation, COD removal, microbial fuel cells, sewage sludge, sludge reduction.*

Introduction

At present, sewage sludge production is increasingly growing worldwide therefore sustainable and innovative solutions for sludge management has gained a lot of importance. The treatment and disposal of excess sludge represents 25-65 % of the total operating costs of the wastewater treatment plant (WWTP) (Wei *et al*., 2003). Digestion is a widely used method for sludge stabilization generated in WWTPs but it is a high energy demand and therefore is an expensive process. It has been demonstrated that conventional ways of disposing of excess sludge are not anymore economically and environmentally feasible, therefore cleaner, environmentally sound and more feasible alternatives are explored in many research studies.

Moreover, along with human society's growth and development, energy is also becoming more important, and since non-renewable energy sources are running out and causing high capital losses, alternative renewable energy sources are urgently needed (Twidel and Weir, 2015). The current practices in the management of sludge consider energy recovery as a key issue together with strict regulatory obligations of disposal and the necessities of energy sustainability. Sewage sludge is a complex mixture of organics including proteins, carbohydrates, oil and fats and inorganic (metals) matter, together with a rich composition of dead and alive microorganisms (Magdziarz *et al*., 2016; Manara *et al*., 2012; Harrison *et al*.,2006). Sewage sludge may be considered as a mixture of multi - substances and it is very difficult to determine a typical composition for sewage sludge. The conversion of sewage sludge is a complex and difficult application due to the differences from other solid fuels, such as lignocellulosic biomass and coal (Shatir *et al*., 2017). However, it has been clearly proved sludge is a valuable resource due to its high organic content (Appels *et al*., 2008). Sewage sludge generated in WWTPs is commonly used in various applications such as: catalyst for chemical reactions (Yuan *et al*., 2013; Zhou *et al.,* 2014; Ma *et al*., 2016), as an adsorbent for a wide range of gaseous and liquid-phase contaminants (Athalathil *et al*., 2014; Leng *et al*., 2015; Chen *et al*., 2014;

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Hadi *et al*.,2015; Chen *et al*.,2002).

In recent years, the attractiveness of waste - generated renewable energy has increased research into microbial fuel cells. Microorganisms can be used for electricity generation. Microbial fuel cell (MFC) technology is a rapidly emerging renewable energy source and a very promising one. MFCs have the ability to be used to produce bioenergy, treat wastewater and generate useful products (Ryu *et al*., 2013; ElMekawy *et al*., 2015; Nikhil *et al*., 2018). Microbial fuel cells (MFCs) use whole micro-organisms as catalysts (Du *et al*., 2007). A diverse range of materials and designs have been applied to produce MFCs. A typical MFC contains anode (an anaerobic compartment) and a cathode (an aerobic chamber, that is separated by a proton membrane such as NAFION ® (Manara and Zabaniotou, 2012). Anodic compartment is fed with an organic substrate. Microbial degradation the organic matter generates electrons which are passed from the cells to the electrode either directly or indirectly by the mediators. The electrons are carried to the cathodic electrode through the electrical circuit (Kumar *et al*., 2017). In the cathode, electrons are combined with the protons with the help oxidants, typically oxygen (Catal *et al*., 2008; Logan, 2007). MFC is stated to be more efficient in terms of conversion and to have less sludge generation compared to conventional wastewater treatment technologies (Manara and Zabaniotou, 2012). Substrate is considered as a significant biological factor in terms of electricity generation in MFCs (Liu *et al*., 2009). As an alternative technology for wastewater treatment and energy production, MFCs have attracted many researchers (Liu *et al.,* 2005; Ogugbue *et al*., 2015; Sonawane *et al*., 2014). A variety of wastewaters: from domestic to more complex ones, such as brewery (Kim et al., 2017; Wen *et al.,* 2009) or swine wastewater (Du *et al*., 2007), pulping wastewater (Haavisto et al., 2019) as well as compounds such as glucose/ sucrose (Kim., 2010) have been used to obtain energy using MFC technology. It was previously reported that less power is generated using substrates such as peptone or meat processing wastewater than with a single compound like bovine serum albumin (BSA) (Heilmann and Logan, 2006).

For instance, a maximum power density of 8.5 W / m^3 and stable voltage of 0.687 V was accomplished using sewage sludge as fuel for MFC in previous research by Jiang *et al*. (2009) whereas voltage of 440.7mV and 220.7 mW/ m^2 was achieved by Liu *et al*. (2009) using surplus sludge as a fuel using single chamber MFC. Low-strenght wastewaters that are unsuitable for anaerobic digestion can be treated by the help of microbial fuel cells (Rittman *et al*., 2008; Watanabe, 2008). Additionally, it is capable of operating at ambient temperature (Mathuriya *et al*., 2009).

Although the key parameter for MFC evaluation is energy production, this technology can be an important and effective method for the treatment of excess sludge as well as the removal of organic pollutants. Microbial fuel cells can extract biomass energy directly from excess sludge and reduce the sludge yield (Wang *et al*., 2018). For example, Su *et al*. (2013) demonstrated that sludge production in a system combining an MFC and a membrane bioreactor could be reduced by about 61%. According to Gajaraj and Hu, (2014) combining activated sludge processes with MFCs could reduce the production of sludge by about 6–11%. Later, Cai *et al*. (2018) added scrap iron to anode sludge MFC to investigate the effect of zero valent iron (ZVI) on sludge decrement. The volatile removal efficiency of suspended solids in FeCSMFC (the FeC package attached to the anodic bottom and closed-circuit), was 66.2%, 24.48% higher than that of SMFC without ZVI.

This study aims to investigate the potential for generating electricity and removing carbon from two-chamber MFC fed with sewage sludge as the substrate source. In addition to that, given that MFC system has never been reported in comparison with aerobic digestion, to estimate and compare the impact of sludge digestion and carbon removal in the MFC system with sludge digestion conducted in an aerobic batch reactor.

Materials and Methods Activated sludge cultivation

Raw sewage sludge sample was collected from Bahcesehir Domestic Wastewater Treatment Plant, Istanbul, Turkey. During the preliminary experimental period, cylindrical glass batch reactor ($V_{effective}$= 4L) was inoculated with a seed sludge (6345 mg/L MLVSS) mixed liquor volatile suspended solids. The properties of active sludge samples, such as levels of COD (mg / L) may differ slightly. After being inoculated with a seed sludge, the reactor was operated over the next 30 days at a steady 24h hydraulic retention time (HRT). The sludge produced in the batch fill and draw reactor was used in MFC studies carried out for electricity generation and sludge digestion. Acclimation was performed by feeding the activated sludge with 1000 mg COD/L sodium acetate. Macro - and micronutrients for biological growth were added in 20 ml / L. The macronutrient solution contained (in g/l): 120 g/l NH_4Cl, 160 g/l KH_2PO_4, 320 g/l K_2HPO_4. The micronutrient solution included 15 g/l $MgSO_4 \cdot 7H_2O$, 0.5 g/l $FeSO_4 \cdot 7H_2O$, 1.51 g/l $CaCl_2 \cdot 2H_2O$, 0.606 g/l $MnSO_4 \cdot H_2O$ and 0.5 g/l $ZnSO_4 \cdot 7H_2O$. The excess amount of biomass was calculated and sufficient amount of sludge was wasted manually to keep the concentration of MLVSS in the reactor steady at around 6000 mgVSS/L. The batch reactor was operated at room temperature (24± 4°C).

Two-chambered MFC configuration and operation

Laboratory scale MFC comprised two plexiglass chambers (anodic and cathodic) separated by proton exchange membrane (Nafion 117). Initially, this membrane was held in distilled water for 12 h before installation, then suppressed between two compartments and sealed with screws. Each uniform cube chamber with dimensions of 15cm x15cm x 15cm had a total effective volume of 2L. Metallic based Chromium - Nickel plates (195 cm^2) were used as electrodes in both anodic and cathodic compartments which enabled indefinite use without corrosion or fouling. The electrodes were paired with a digital voltmeter (UT60F) which was connected via cable to a personal computer to transmit and record the data (Fig.1). The anodic chamber was operated under anaerobic conditions, while 2 L of tap water was poured into the cathode compartment and air-aerated through an air diffuser. The mixed sludge was agitated by magnetic stirring in the anode chamber.

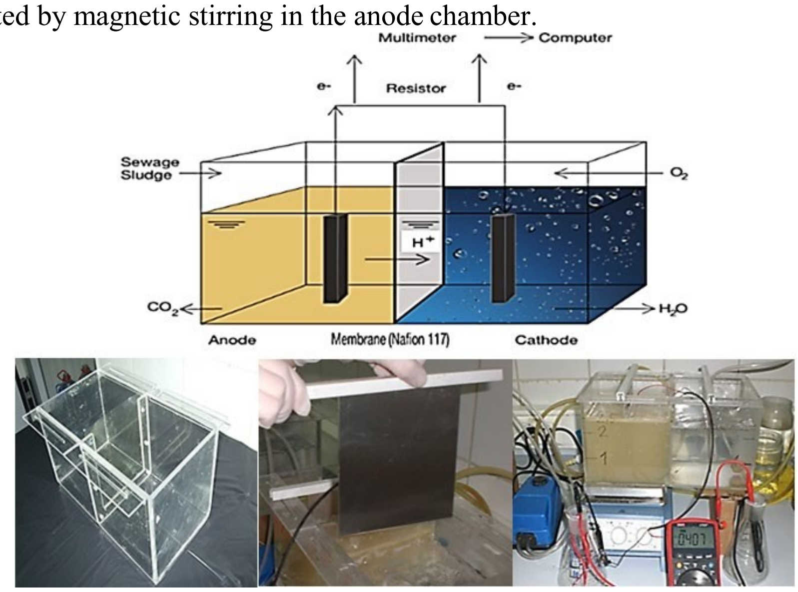

Figure 1. Experimental set- up of two-chambered MFC

The biomass used in MFC operation was the acclimated sludge withdrawn from 4L batch reactor. The anodic chamber was inoculated with 2 liters of acclimated sewage sludge. No substrates or artificial wastewater was fed to MFC system. Macro - and micronutrients were added to anode compartment. Operation of this current study was split into 2 stages. The 1st stage consisted of 2 experimental sets named Run I and Run II. These runs were performed under the same conditions for 240 h under open circuit (infinite resistance, zero current) to test MFC performance via voltage (V) measurement. A cylindrical batch reactor was set up with the aim of investigating and comparing the efficiency of aerobic digestion with sludge digestion in MFC in terms of sludge reduction and carbon removal. To test these impacts, the MFC anode chamber was introduced with 2 liters of sewage sludge (5430 mg / L VSS) and 1 liter was carried out to a batch reactor during Run I and 5240 mg/L VSS at Run II.

The MFC system was monitored in terms of electricity and power generation during the 2nd stage of the experimental sets. This stage was started by feeding the MFC system with an excess sludge of 5640 mg VSS/L. Voltage output was measured for 120 hours while varying external resistances. Electrochemical parameters were determined under 1000 Ω - 4000 Ω. For all experimental runs the same MFC setup has been used. The change in voltage of the fuel cells under various external resistance was recorded daily. MFC system was cleaned with water and distilled water after each experimental run, before using new acclimated sludge from batch reactor. The experimental runs were carried out at room temperature (24 ± 4 °C).

Analytical methods

Soluble chemical oxygen demand (SCOD) parameter has been evaluated according to the International Organization for Standardization, ISO 6060. Total (TSS) and volatile (VSS) suspended solids were conducted using standard methods outlined in Standard Methods (APHA et al., 1998). The pH analysis was determined using a calibrated pH meter (520Aplus pH meter, Orion) in accordance with Standard Methods (APHA, 1998). Samples were filtered through Millipore membrane filters with a pore size of 0.45 μm for the SCOD and through Millipore AP40 glass fiber filters for VSS analysis. All the measurements were done in dublicate.

Electrochemical monitoring

The cell voltages of the MFC was recorded using a precision digital voltmeter (UT60F, UNI-T) linked up to a computer for the continuous collection of data. Voltage was tracked down with 4 minute intervals and transmitted via cable from voltmeter to computer. Open circuit voltage was measured by removing external load (1st experimental stage), whereas external resistances (1000 Ω -4000 Ω) were applied during the 2nd stage. According to Ohm's law, current was deducted from the measured voltage drop across the resistor as $I = V/R_{ex}$ where I (A) is current, V (V) is voltage, Rex (Ω) is external resistance. Current density was estimated as $I_{an} = I/A_{an}$ where Ian ($a*cm^{-2}$) is the current density and A (cm^2) the projected surface area of the studied electrode. Power output of the cells was determined as P = I*V where P (W) is power. Power density produced by MFC was determined as Pan=P/Aan where Pan ($W*cm^{-2}$) is power density. Coulombic efficiency was obtained as $CE = C_p/C_{th} \bullet 100\%$, where C_p is the total coulombs calculated by integrating the current over time, and C_{th} is the theoretical amount of coulombs available based on the COD removed in the MFC (Logan et. al., 2006).

$$C_E = \frac{M_S \int_0^{tb} Idt}{Fb_{es}\, v_{An}\, \Delta c} \quad (1)$$

where Δc is the substrate concentration change over the batch cycle over a time tb, M is the molecular weight of the oxygen (32 g/mole), F is Faraday's constant (96.485 C/mol e^-), and v_{An} is the volume of liquid in the anode compartment, b_{es} is the number of electrons exchanged per mole oxygen (4 e^-).

Results and Discussion Biomass cultivation

It is favored to set the anode to a more negative potential (-0.2V) or to use pre-acclimated culture for cell acclimatization (Zhang et al., 2013), therefore activated sludge was cultivated for in a batch reactor prior to seeding into MFC. The initial sludge seeded into the batch reactor had a TSS value of 14085 mg/L and SCOD concentrations was 1132 mg/L. Acetate, which is readily biodegradable (Henze, 1992), was used as the source of organic matter. Culture stabilized within 30 days after the beginning to reach steady-state conditions. The effluent concentrations of SCOD decreased from 415 mg/L to 145 mg/L throughout the 30-day period, resulting in a removal efficiency of around 86 percent (data not shown). At the end of this period, it was targeted to obtain 5500 mg/ L of VSS in the reactor. The sludge age could not be determined due to the poor settling properties and sludge leakage during effluent withdrawal. By day 30 the concentration of VSS (mg / L) reached a value of 5430 mg / L and was used for continuous experiments in the MFC system.

Comparative evaluation of sludge digestion in MFC and aerobic batch reactor

The overall performance of MFC was analyzed in terms of COD removal efficiency, sludge reduction and electricity production. During Run I and Run II, sewage sludge was digested in two identically started systems: (1) in 1 L batch reactor with aerobic digestion of excess sewage sludge and (2) in MFC system with digestion of excess sewage sludge. The cellular voltage (OCV) profile was observed during digestion with MFC (Figure 2).

As sludge was introduced into MFC during Run I and Run II, an initial voltage output of 43 mV and 62.1mV was, respectively, instantly generated. No obvious electricity generation was observed and the voltage only reached 11.8±8.6 mV during first 96 hours in Run I, whereas a stable voltage generation of 60.4±3.3 mV was achieved during this period in Run II. After approximately 100 hours after the sludge adaptation in MFC, increment in voltage generation was observed in both experimental runs. For the next 140 hours stable voltage output of 112±16 mV was observed during Run I, producing a maximum voltage of 154 mV at 231st hour. At the same time during Run II, 119±12 mV, producing a maximum voltage of 146 mV at 240th hour. Considering Run I and Run II (OCV) during 240 hours, an average voltage of 115 mV of could be generated in a two microbial chamber fuel cells using oxygen as the electron acceptor. Oxygen is the most attractive end-electron acceptor in MFCs not only because of its high quality reduction potential but also because of practical environmental considerations (Wang, 2015). Moving average value of the data given in Figure 2 is best described by an exponential function however, this may not represent the exact function because of significant fluctuation in the data set. Nevertheless, the observed results explicitly demonstrate that the higher voltage is generated with the progress in sludge digestion (Figure 4). Jiang *et al.* (2009) used two-chambered MFC with potassium ferricyabine as its electron acceptor to generate electricity from sewage sludge. Stable voltage of 0.687 V (with 1000 resistor) was generated during the 250 h test. Later, Xie *et al.* (2016) integrated MFC into anaerobic-anoxic-oxic wastewater treatment process with real sewage as influent and obtained average voltage of 0.169±0.008 V (with 2000 Ω resistor). Findings in the present study are consistent with the findings of Cai *et al*., (2018) who obtained a stable electricity generation of 0.14 V after 5 days adaptation in Normal SMFC without the package of FeC (NSMFC) using residual sludge as fuel. The composition of excess sludge is complex, the hydrolysis rate of organics is slow, moreover it is difficult to use the refractory organics effectively (Ge et al., 2013). Sludge typically requires pretreatment to break down cells. It is ideal for sludge microbial fuel cell (SMFC) to efficiently remove biomass from pretreated sludge, to increase electricity generation efficiency, such as Mawioo *et al*. (2017) using microwave pretreatment in their study, Xie *et al.* (2016) embracing ultrasonic pretreatment, Yusoff *et al*. (2013) applying ozonation. However, such approaches simultaneously increase SMFC's energy usage and operational costs.

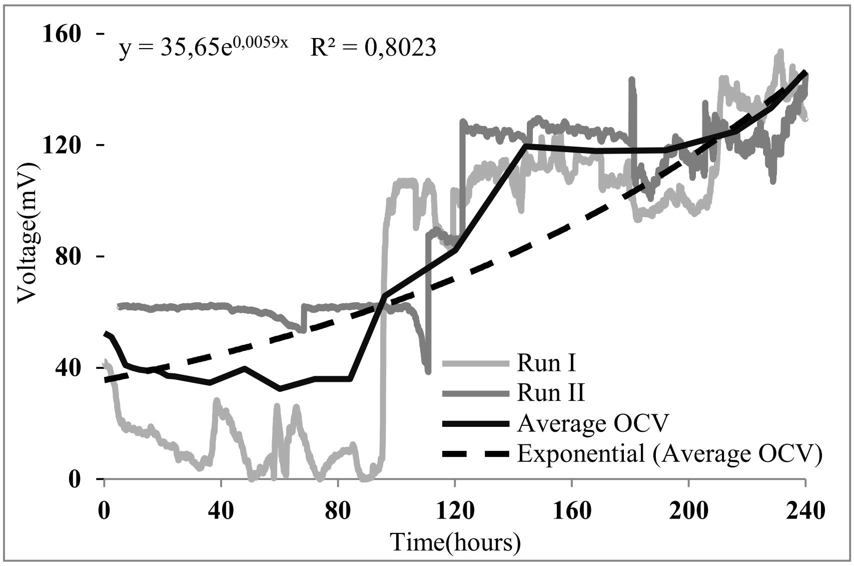

Figure 2. Monitored OCV values (Run I and Run II)

SCOD (mg/l) concentration gradually decreased (Run I) from 102.9 mg/l to 69.7mg/l in MFC system, started from 131.7 mg/l and became stable at the value of 77.4 mg/l in the aerated batch reactor. Fig.3 indicates that after 120 hours of digestion the solubilization process was slower than the COD removal processes (namely microbial growth) thus, the SCOD was kept constant at the levels which are apparently the soluble microbial products generated in endogenous respiration process. In Run II, SCOD removal efficiency was 41.2% in the aerobic batch system which was higher than that of 32% SCOD removal in the MFC system (Fig.3). During Run II, the concentration of SCOD (mg /L) gradually declined and became constant at 70.9 mg/L presumably the level of endogenous respiration due to the absence of organic matter in the batch system and at 78.8 mg/L in the MFC system. SCOD removal was stable after after 168 hours of operation in both aerobic and anaerobic digestion, with a higher efficiency in SCOD removal of 42.7 % the batch system compared to 32% SCOD removal efficiency achieved in MFC. The result is in the line with study by Cai *et al*. (2018) who found a slightly higher SCOD removal efficiency of 50.5% in SMFC system under open - circuit after 120 days. Moreover, Zhang *et al.* (2018) achieved TCOD removal of 20% in open circuit running SMFC using excess sludge as a fuel. SCOD results from both experimental runs revealed that aerobic digestion had higher digestion rate compared with anaerobic digestion. As it is reported by Buchanan and Seabloom (2004) anaerobic digestion is restricted to fermentation processes where higher organic compounds are reduced through this process to lower organic, whereas aerobic digestion undergoes additional respiration process instead of fermentation process. As these new cells will also undergo fermentation and respiration producing more cells, the digestion rate is also greater compared to anaerobic digestion.

The decrease in VSS reflects the effect of sludge decrement (Ge *et al.,* 2013). The VSS values obtained in the MFC system and aerobic batch system after 240 hours were 3630 and 2415 mgVSS/L, respectively (Run II). The reduction of sludge (VSS) had reached 31.5% (Run I) 30.7% (Run II) in the MFC system and 51.8% (Run I) 53.9% (Run II) in the batch aerobic digester. The efficiencies of carbon removal and the results of sludge reduction were closely related during both runs. Although Table 1 shows that higher VSS and carbon removal efficiencies removal (Figure 3) were achieved in aerobic batch digester, electricity was generated during anaerobic digestion in MFC. The generation of additional electrical power is the main advantage of MFC, where sludge reduction, carbon removal and electricity generation can be realized simultaneously. The MFC can be considered as an enhanced sludge digester with additional pathways for substrate hydrolysis and degradation. The sludge reduction observed in MFC was greater than that observed by Jia *et al.* (2009), who recorded reductions of 27.3% (TSS) and 28.7% (VSS) while using surplus sludge as a substrate for electricity generation and by Xiao *et al.* (2017), who integrated MFC into A/O process and showed the reduction of accumulative wasted activated sludge of approximately 24% at the end of 54th day.

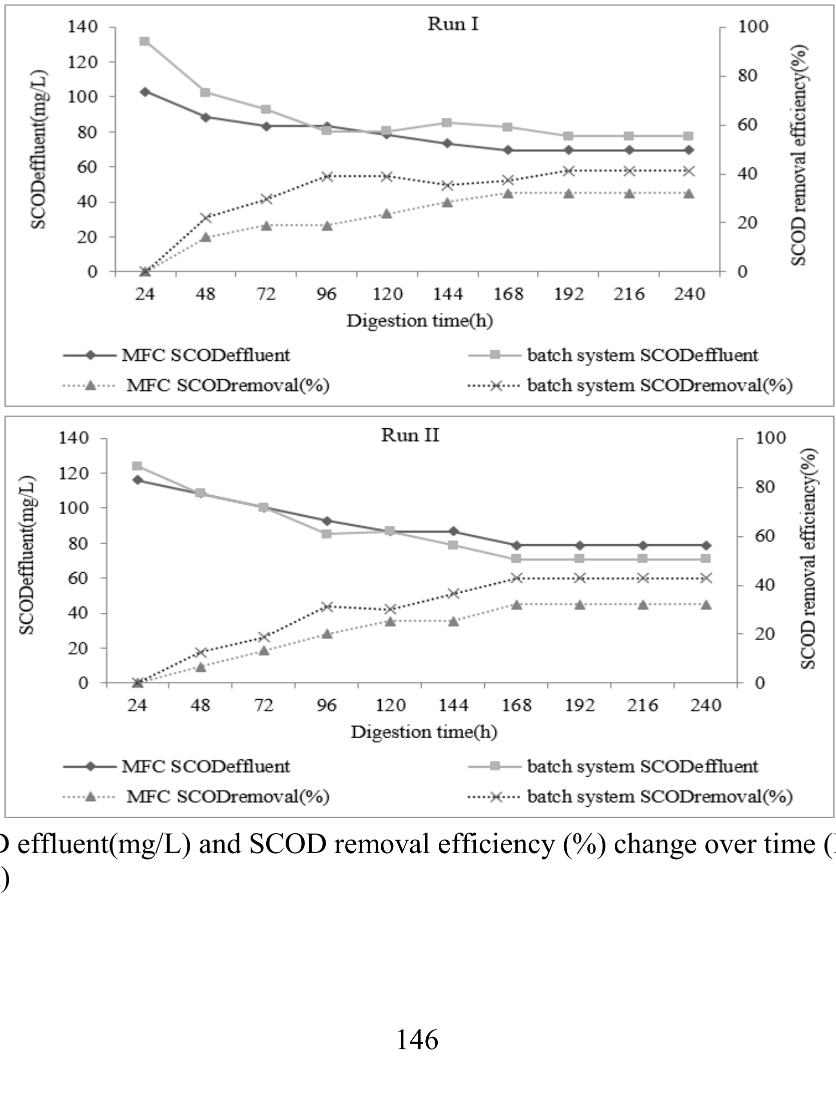

Figure 3. SCOD effluent(mg/L) and SCOD removal efficiency (%) change over time (Run I and Run II)

Table 1. MLVSS reduction (%) during Run I and Run II

	Run I		Run II	
	MFC	**batch system**	**MFC**	**batch system**
Initial TSS(mg/l) t_{0h}	7550	7550	7270	7270
Initial VSS(mg/l) t_{0h}	5430	5430	5240	5240
Effluent TSS(mg/l) t_{240h}	5200	3710	5070	3695
Effluent VSS(mg/l) t_{240h}	3720	2615	3630	2415
VSS removal efficiency(%)	31.5	51.8	30.7	53.9

Traditionally Microbial Fuel Cells (MFCs) operate at 6 to 8 pH values (Biffinger *et al.,* 2008). The pH values were in the range of 6 and 7.2 through experimental runs, which is the optimum pH level for MFC operation. Therefore, the impact of pH changes on the MFC's efficiency in electricity generation and sludge digestion was kept at minimum level.

MFC performance on electricity and power production

During this stage, the cell voltage was detected at an interval of 4 minutes with external resistance ranging from 1000 to 4000 Ω. After start - up of the system, each external resistance was applied for 24 hours of operation and the resistors were consequently applied (Figure 4).

The experimental data presented in Figure 4 reveals that when external resistances were connected to the circuit the cell voltage obtained in the MFC system has dropped significantly. This result is consistent with the general knowledge that the voltage should drop with decreasing resistance. The obtained OCV values were much higher (Figure 2), as the maximum voltage under infinite resistance is presented by OCV. Power and current generation in MFC system was observed under 4000Ω resistance (Figure 5).

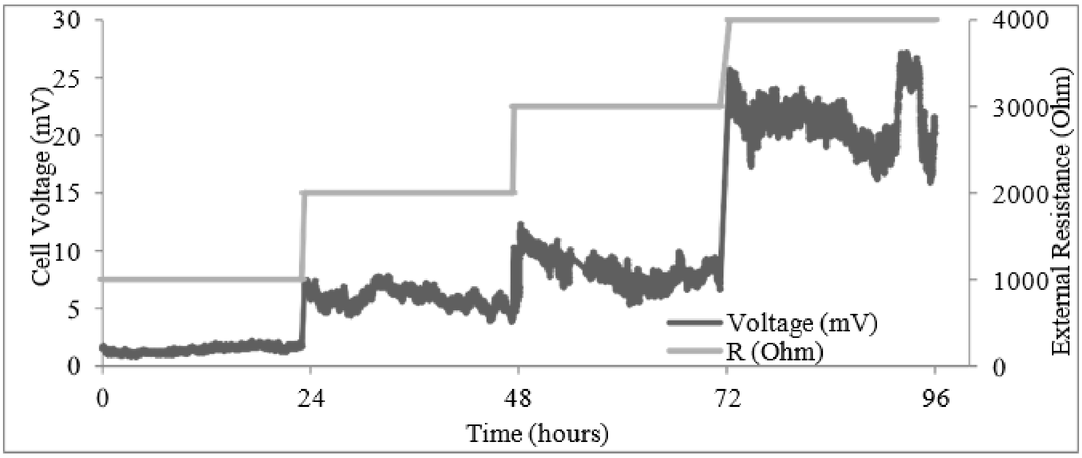

Figure 4. MFC cell voltage profile under various external resistance loads

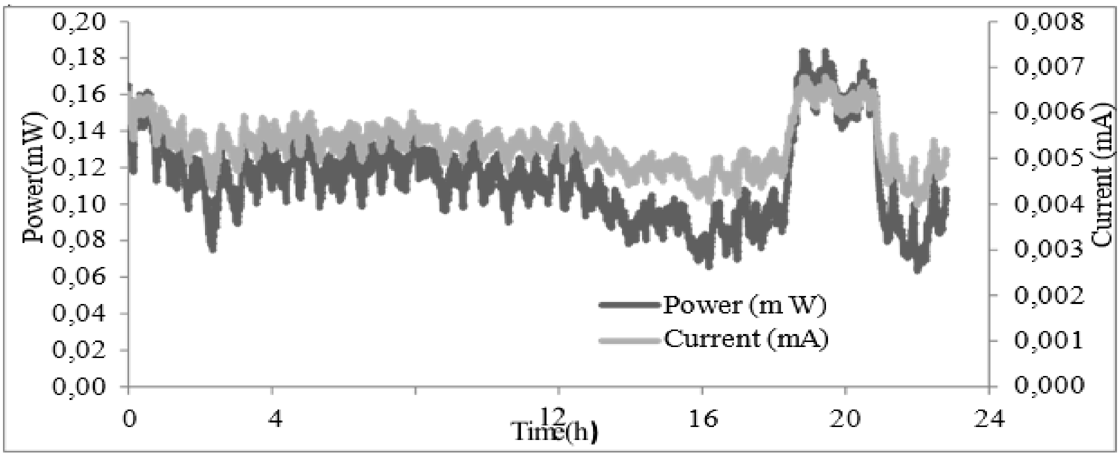

Figure 5. Current and Power generation (4000Ω resistance)

Figure 5 indicates maximum achieved of 0,0068 mA. At an external resistance of 4000 Ω, the cell had a peak power output of 0.184 mW. This power is significantly lower than that reported for other cells using sludge as fuel source with pretreatments (Yusoff *et al.,* 2013; Xiao *et al.,* 2011; Wang *et al.,* 2013; Abourached *et al.,*2014).

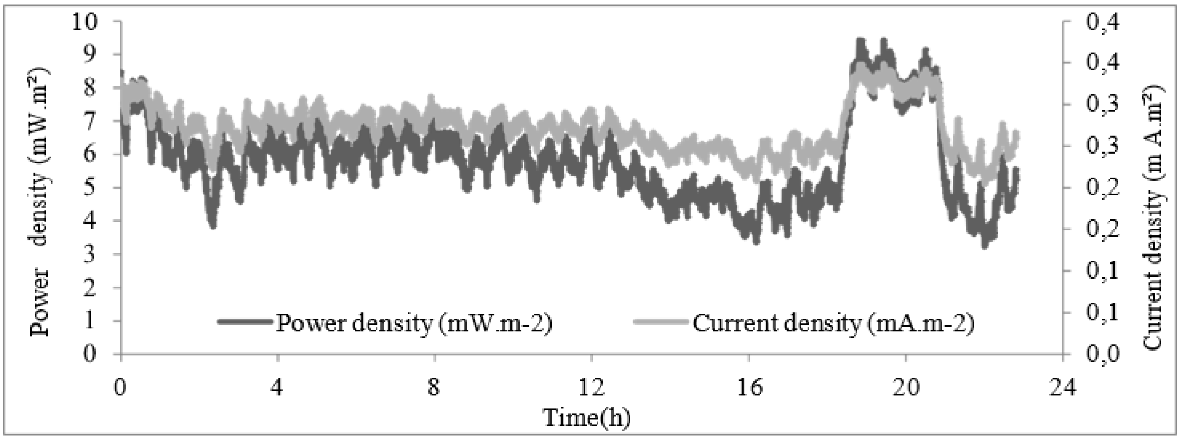

Figure 6. Current and Power density (4000Ω resistance)

Figure 6 shows maximum current density obtained of 0,35 mA.m^{-2}. and peak power density performance of 9.42 mW.m^{-2}. Hu *et al.* (2008) used a baffle- chamber membrane - less MFC to extract electricity from an anaerobic sludge but noticed that its electricity efficiency (0.3 mW / m^2) was significantly lower compared to tests where glucose was used as a substrate (161 mW / m^2). The system's coulumbic efficiency was calculated by integrating the current obtained in the final stage (Figure 7).

The coulombic efficiency calculated by integrating the current data in Figure 7 is found as 0.0068% which shows that the total number of coulombs that can be recovered can be recovered is very small compared with the total columns available in the sludge. However, this also shows that there is room for future studies to enhance the sludge digestion process along with significant electricity recovery.

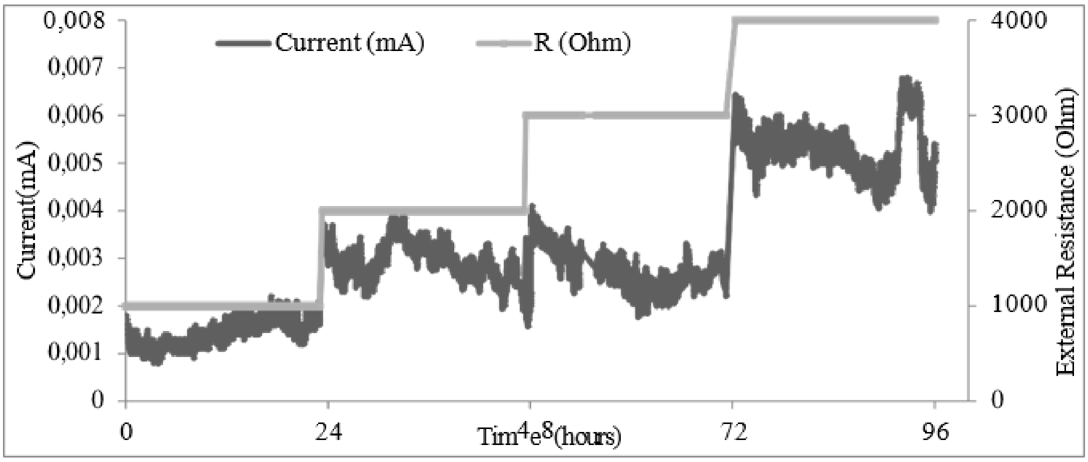

Figure 7. Current profile of MFC under different external resistance loads

Conclusion

The cell performance data in this study have shown that two-chamber microbial cell system with a simple configuration could be operated without the use of precious metal catalysts, single culture organisms or mediators for electricity generation and sludge reduction with a low cost fuel material - sewage sludge - without additional pretreatment and energy consumption. During the 240 h demonstration test, stable voltage output of 112±16 mV was observed during Run I, producing a maximum voltage of 154 mV and 119±12 mV with a maximum voltage of 146mV during Run II. Sludge disposal is a major issue that can be solved effectively by using sludge for different purposes, such as energy production, while reducing the excess amount of sludge. During Run I and Run II, the efficiency of SCOD removal in MFC system was ~32%, slightly lower than in aerobic digester. The results of this study indicate that when the organic matter in sewage sludge is used to generate electricity by MFC, it is possible to reduce the amount of excess sludge (up to 31.5% during this study) and lower the cost of sewage sludge treatment. Even though the reduction in VSS was lower than in aerobic batch digesters, more importantly, electricity was produced during anaerobic digestion in MFC. It is a field of work where a lot of research and development is required to be done in order to implement it on a large scale. Interfacing biology and electrochemistry may become of critical importance for the advancement of renewable energy principles as well as for coping with waste-use environmental problems.

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