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Article

Electricity Generation from Single Chamber Microbial Fuel Cells using Granular Activated Carbon as Anode for Wastewater Treatment

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Abstract. Locally sourced granule-activated carbon (GAC) has been used in wastewater treatment systems, but it is not widely used in single chamber microbial fuel cells (SCMFCs). The GAC in an anode coupled with a carbon cloth was used in the SCMFCs in this research, with 1.00-L of the GAC added to the carbon cloth in the anode. Three SCMFCs were operated in continuous flow mode. The synthetic wastewater was prepared from rice flour dissolved in tap water, with a chemical oxygen demand (COD) value of 1,816.32 ± 204.78 mg/L. The organic loading rates (OLRs) in the study were 0.91, 1.82, and 7.27 kgCOD/m^3-d. The maximum power density levels generated by the three SCMFCs were 13.16, 7.96 and 4.11 mW/m^2 from OLRs of 0.91, 1.82, and 7.27 kgCOD/m^3-d, respectively, and the efficiency rate of COD removal were 24.96%, 24.85%, and 20.75%, respectively. This research also presented a voltage boost converter circuit for charging a 0.25F storage supercapacitor. Only two OLRs provided an electric voltage that could run the circuit (0.49V and 0.45V generated by OLRs of 0.91 and 1.82 kgCOD/m^3-d, respectively.) The supercapacitor set was charged to reach the maximum output of 5.25 V taking 19.5 hr and 37.5 hr, respectively.

Keywords: Single chamber microbial fuel cells, granule-activated carbon, COD removal, voltage boost converter circuit.

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1. Introduction

There is a growing demand for energy from the agricultural industry due to urbanization and economic development. Some processing factories using rice flour starch as an ingredient release high levels of chemical oxygen demand (COD) wastewater. However, bacteria can easily reduce starch-based wastewater. The bacteria use the nutrients in wastewater and during nutrient digestion, electron exchange occurs in a process which is called the oxidation-reduction process. Using this principle, a microbial fuel cell (MFC) can be utilized for wastewater treatment and electrical power generation. In Thailand, MFC is currently only at the laboratory scale due to the high cost of the electrode so, the electricity output is quite low. Anaerobic wastewater treatment is popular for the first stage of processing a high COD loading rate. A fuel cell could be used to produce power by generating H_2 [1] or directly generating electricity [2].

In an MFC, electricity is produced by the electrochemical processes of microorganisms oxidizing organic compounds. The electrons produced by the oxidation processes are released outside the cell and are accepted by the anode electrode as the electron acceptor in anaerobic conditions. Many studies have found that electricity could be generated from various sources of organic matter, such as domestic waste [3], composite vegetable waste [4], various food industries [5], starch processing [6, 7], brewery wastewater [8, 9], cheese whey [10], palm oil mill effluent [11], sewage sludge [12], decolorization in wastewater treatment [13], and leachate wastewater [14]. Such reports have also proved that an MFC could be used for wastewater treatment while simultaneously producing electricity under various conditions. The optimized conditions depend on the characteristics of the wastewater, the architectures of the reactor (including the types of cathode, anode, and connection wire) and environmental conditions (such as the pH, temperature, conductivity, quantity, and source of the sludge).

However, MFCs cannot produce sufficient voltage to drive a small electronic device. Various methods have been developed to increase the voltage level in a single chamber microbial fuel cells (SCMFC). Examples include: using a boost converter circuit to extend the voltage from 0.3V to 2V with 74% efficiency [15], using a self-powered ultra-low power DC-DC converter circuit [16], designing an on-chip boost converter circuit [17], connecting SCMFCs in series and parallel to increase the voltage and power [18], using an LTC3108 IC as a boost converter circuit to increase the voltage [19], or using the ultra-low-power energy harvester circuit [20].

This research aimed to design a new SCMFC for wastewater treatment and simultaneously generate electricity. We used a voltage boost converter circuit to charge supercapacitors for use as the power source of small electronic devices. The contributions of this research can be summarized as:

- This research developed a new rice flour wastewater treatment process that could simultaneously generate electricity. We used microbial action with rice flour wastewater to generate electricity in an SCMFC in continuous mode flow operation. The rice flour wastewater treatment process involved the loss of electrons using an anaerobic process at the anode, with the electrons induced to the cathode via copper wires. The advantage of using rice flour wastewater treatment was that it is environmentally friendly without requiring any power input. Furthermore, the power output from the SCMFC could be used directly, in contrast to other wastewater treatment processes that produce biogas that is then converted to electricity in a lather.
- We increased the area of the anode electrode by using granule-activated carbon (GAC) to increase the electrical power of the SCMFC. The high surface area of the GAC promoted rice flour wastewater treatment by increasing the number of micro-organisms attached to the surface of the GAC. In addition, the GAC acted as a filtering medium for suspended solids in the wastewater influent. Furthermore, GAC has a lower cost and is more easily obtained than other anode types, such as carbon cloth. A pilot SCMFC was set up to treat real industrial wastewater, such as from palm oil, sugar, and rubber production.

The paper is organized as follows: Part 1 provides the introduction, followed by the methodology in part 2, with the results in part 3, along with a discussion. Lastly, part 4 provides the conclusion.

2. Methodology

2.1. Wastewater

The synthesized wastewater used in the experiments was made by mixing rice flour with tap water. The general characteristics of the raw wastewater were: Total chemical oxygen demand (TCOD) 14,500-21,800 mg/L, BOD5 10,000-12,500 mg/L, pH 3.84-3.92, TP 54-60 mgP/L, TKN 360-400 mg/L, sulfate 18,000-20,000 mg/L, and conductivity 2.77 mS/cm.

2.2. Single Chamber Microbial Fuel Cell

The anode consisted of 1.00-L of GAC placed on carbon fiber (456 cm^2, made in Thailand) at the bottom of the SCMFC reactor chamber. The chamber was made from clear acrylic (20 cm width, 25 cm length, and 15 cm height) and had an empty volume of 6,000 mL. The cathode was made from carbon cloth (B-1 from Clean Fuel Cell Energy LLC) with a total area of 150 cm^2. The cathode was coated with Teflon on the side exposed to air. The cathode and anode were 15 cm apart and were connected by an external copper wire. Wastewater was fed into the bottom of the SCMFC reactor and the effluent of treated wastewater was removed from the top of the reactor. A schematic SCMFC is shown in Fig. 1 and 2.

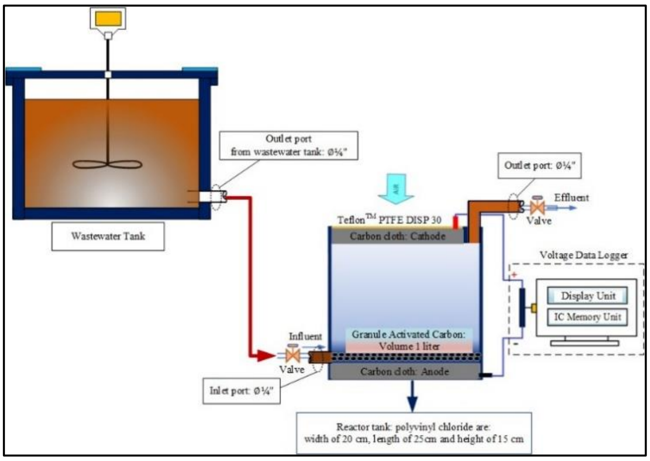

Fig. 1. Components of SCMFC used in this research.

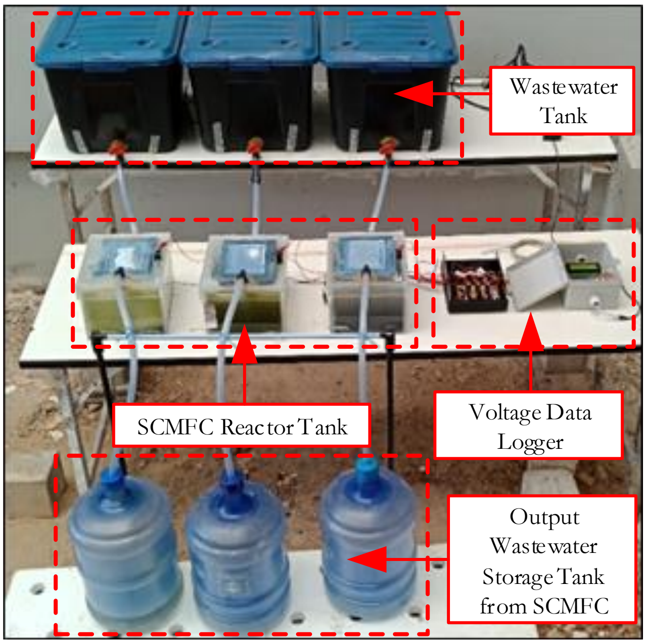

Fig. 2. Constructed SCMFC reactor used in this research.

2.3. Inoculum and Operation

The inoculum was powdered bacteria (Bactocel™) purchased from a convenience store and was used without any additional processing. The initial volume of mixed liquor suspended solids (MLSS) in the chamber was kept at 20,000 mg/L. The wastewater was fed continuously into the SCMFC reactor. The flow rate was controlled using a gate valve. Synthesized wastewater was prepared from 100 g of rice flour diluted with 20 L of tap water. The wastewater storage tank was installed higher than the SCMFC reactor to provide the head. A motor was used for mixing and preventing flour sedimentation in the storage tank. The OLRs were controlled at 0.91, 1.82, and 7.27 kgCOD/m^3-d. The hydraulic detention times (HRTs) were adjusted to 48, 24, and 6 hr, respectively. COD and SS were analyzed according to APHA standard methods for the examination of water and wastewater [21]. Total dissolved solids (TDS) and pH were measured using a Hach HQ40D Portable Meter. The circuit voltage was measured and stored using a data logger every 30 minutes. Polarization curves were obtained by varying the external resistance over the range from 150 to 15,000 Ohms at the end of each experiment.

2.4. Calculations and Analyses

The power from the SCMFC was calculated according to Eqs. (1)-(3):

$$P_{SCMFC} = I_{SCMFC} \times V_{SCMFC} \quad (1)$$

where P_{SCMFC} = Output power (W) of SCMFC,
I_{SCMFC} = Output current (A) of SCMFC,
V_{SCMFC} = Output voltage(V) of SCMFC
and

$$V_{SCMFC} = I_{SCMFC} \times R_{in} \quad (2)$$

where R_{in} = Internal resistance (Ohm) of SCMFC

$$P_{SCMFC} = \frac{(V_{SCMFC})^2}{R_{in}} \quad (3)$$

The coulombic efficiency (CE) was estimated by measuring the current relative to the theoretical current on the basis of consumed COD as show in Eq. (4) [28].

$$CE = \frac{8 \times I_{(max)}}{((F) \times (\Delta COD)} \quad (4)$$

where $I_{(max)}$ = Maximum current (A/s)
F = Faraday's constant (96,485 C/mol)
ΔCOD = COD removal (g/s)
Q = Wastewater flowrate (m^3/s)

The internal resistance of the SCMFC was calculated using the polarization slope method (the slope in a plot of current density and voltage versus internal resistance).

2.5. Proposed Circuits

As illustrated in Fig. 3, the components of the voltage boost converter circuit consisted of three subsections: the DC input voltage from the SCMFC, the self-oscillator circuit, and the charging supercapacitors circuit using the energy harvester LTC3588-1 IC module. The self-oscillator circuit boosted the energy harvester via an autonomous start-up voltage oscillator. The circuit had a configuration of discrete electronic components that could produce a free-running signal. The circuit required a resistor, a capacitor, a transformer, and only one transistor. The comprehensive mathematical analysis and experimental validation method can be referenced in [22].

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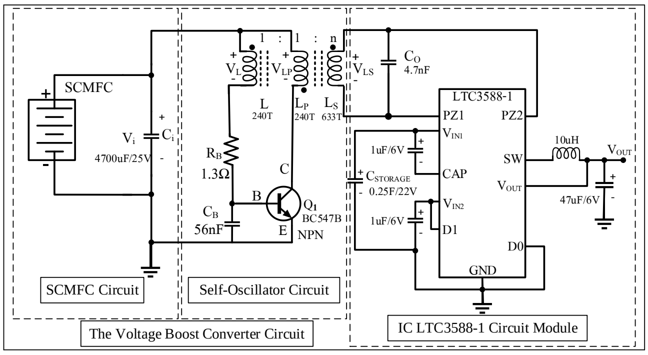


Fig. 3. Circuit diagram of proposed components in this research.

An energy harvester LTC3588-1 IC module was used as the supercapacitor charging circuit, as shown in Fig. 3. This module provided a complete energy harvesting solution optimized for high impedance voltage sources. It contained a low loss full wave bridge rectifier and a high efficiency synchronous buck converter to transfer energy from an input storage device to an output at a regulated voltage capable of supporting loads up to 100mA. According to the features of the LTC3588-1 IC module, this circuit was connected to the output of the self-oscillator circuit at the P_{Z1} and P_{Z2} input pins. A C_o 4.7nF capacitor, which was connected between the P_{Z1} and P_{Z2} input pins, was used to prevent reverse current flow in each element. The LTC3588-1 IC module rectified the voltage waveform and stored the harvested energy in an external storage supercapacitor of 0.25F/22V (by connecting 4 supercapacitors of 1F/5.5V in series), which was connected to the V_{IN1} and V_{IN2} pins of the LTC3588-1 IC module. An ultralow quiescent current undervoltage lockout (UVLO) mode with a wide hysteresis window allowed charges to accumulate on the 0.25F/22V supercapacitor until the voltage across the supercapacitor reached 5.25V, so that the buck converter could efficiently transfer a portion of the stored charge to the 47uF output capacitor. The features of the LTC3588-1 IC module can be referenced in [23], [24].

The harvester efficiency was calculated using Eq. (5):

$$\eta = \frac{P_{out}}{P_{in}} \times 100\% \tag{5}$$

where P_{in} and P_{out} are the input power of the SCMFC and the output power of the boost converter, respectively.

The self-oscillator circuits were implemented using the circuitry from [22]. The efficiency of the voltage boost converter circuit was affected by some parameters, such as parasitic losses of the transformer, transistor, and winding. On the other hand, the efficiency of the LTC3588-1 IC module was affected by just only a few power losses.

3. Experimental Results and Discussion

3.1. Power Generation

In the first stage of the study, there were fluctuating values in the electrical voltage because the microorganism had not acclimatized to the wastewater. Initially the microorganism preferred to grow at low rather than high OLRs, as indicated by the initial electrical voltages of 0.12 V, 0.20 V, and 0.28 V for OLRs of 7.27, 1.82, and 0.91 kgCOD/m^3-d, respectively. Later, the values of electrical voltage values continued to fluctuate. Green bacterial growth spread in the reactors because the reactor was made from clear acrylic. Furthermore, the greatest quantity of green bacteria was recorded at the lowest OLR and similarly, the electrical voltage from the lowest OLR (at 230 hours) fluctuated more than for the highest OLR. Green bacterial growth in the SCMFC reactor inhibited power generation because oxygen was produced by the green bacteria during the daytime due to photosynthesis, which then inhibited the process of electron transport to the cathode. This represented electron loss regarding electricity generation because free oxygen in the reactor was the most effective as an electron acceptor. After 230 hours, the reactor was covered with a black board panel to block sunlight and this made the electrical voltage more stable, as shown in Fig 4. When the bacteria were acclimated to the wastewater, the electrical voltage was stable and the SCMFC produced electrical power continuously if wastewater was fed into the reactor chamber.

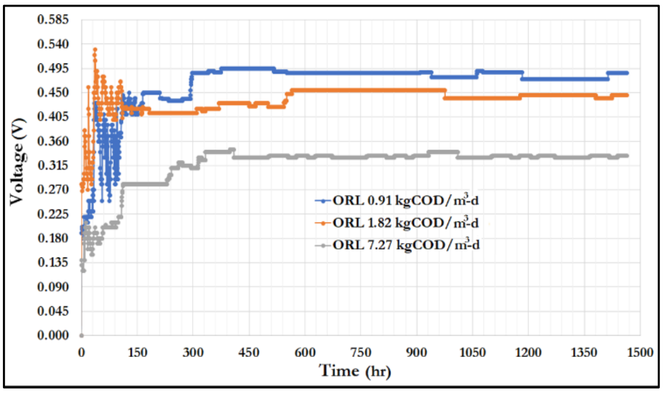


Fig. 4. Voltage generated by SCMFC during this study.

The electrical voltage of the OLR of 0.91 kgCOD/m^3-d was stable from 230 hours until the end of the experiment and was the highest obtained from this study. The electrical voltage tended to increase with time; however, increasing the OLR decreased the efficiency of wastewater treatment.

The power density achieved from the study is shown in Fig. 5. The maximum power density obtained from the OLR of 0.91 kg COD/m^3-d was 13.16 mW/m^2 and the next highest was 7.96 mW/m^2 from the OLR of 1.82 kg COD/m^3-d. When OLR increased to 7.27 kg COD/m^3-d power density was decreased to be 4.11 mW/m^2. A high

OLR generated a lower power density because the power output was from the combination of the biochemical reaction, the type of microorganism, and the ability of electron transfer from the anode to the cathode.

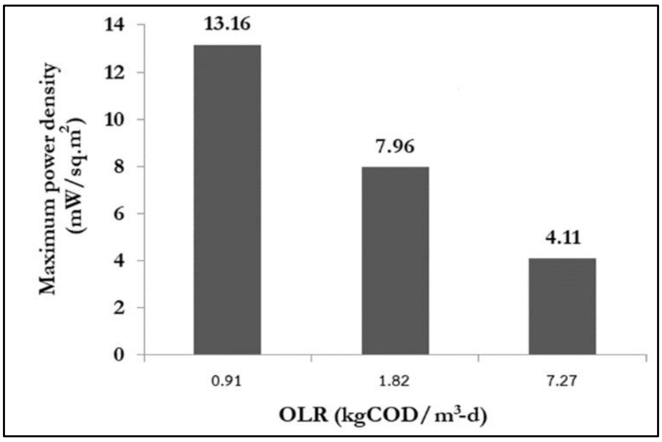


Fig. 5. Power density.

3.2. Wastewater Treatment

This research operated in continuous flow mode for the wastewater infeed from the bottom of the SCMFC reactor, with effluent at the top. The wastewater treatment in the chamber operated under anaerobic conditions. The COD removal efficiency percentages were 24.96%, 24.85%, and 20.75% from the OLRs of 0.91, 1.82, and 7.27 kg COD/m^3-d, respectively (see Fig. 7). The efficiency of COD removal from the OLRs of 0.91 and 1.82 kg COD/m^3-d were not significantly different because the detention time for all the SCMFCs were lower for the anaerobic treatment process (48, 24, and 6 hr). Normally the HRT for anaerobic conditions is longer for high load wastewater treatment to minimize the effect of the growth rate of anaerobic microbes. The growth rate of the anaerobic bacteria was quite low. Furthermore, the pH of the influent wastewater was not made more alkalinity before feeding into the reactor, with both the influent and effluent values being below 7 (acidic), as shown in Fig. 10. An acidic pH is not favored by microorganisms used to treat wastewater under anaerobic conditions. This was reflected in the lower COD removal efficiency. Thus in anaerobic wastewater treatment, alkaline conditions (6.5-8.2) are very important to maintain a suitable pH for microbial growth [25, 26, 27] by using acidogenic bioconversion of rice mill wastewater before feeding it into a two-chambered MFC, as this could improve the efficiency of COD removal to more than 60%. The SS removal efficiency using the SCMFC in the current study was very high compared to other anaerobic treatments. This highlighted the benefit of the GAC for filtration at the inlet of the reactor. The concentration influent, effluent, and efficiency of SS removal are shown in Figs. 8 and 9. We found the efficiency percentages of SS removal were 96.14%, 95.58%, and 95.49% for the OLRs of 0.91, 1.82, and 7.27 kg COD/m^3-d, respectively. SS in the synthesized wastewater settled readily because the rice flour powder was not very soluble; therefore, the wastewater influent contained a high level of suspended solids.

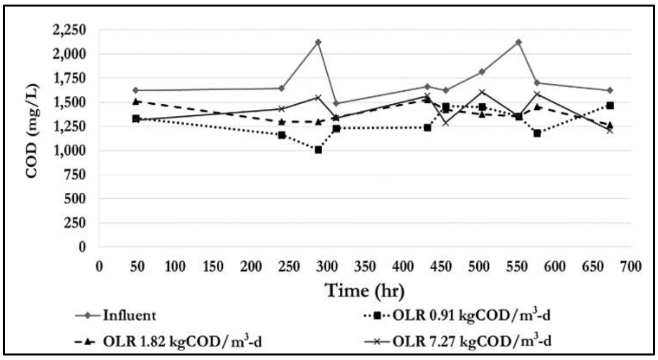


Fig. 6. COD concentration influent and effluent.

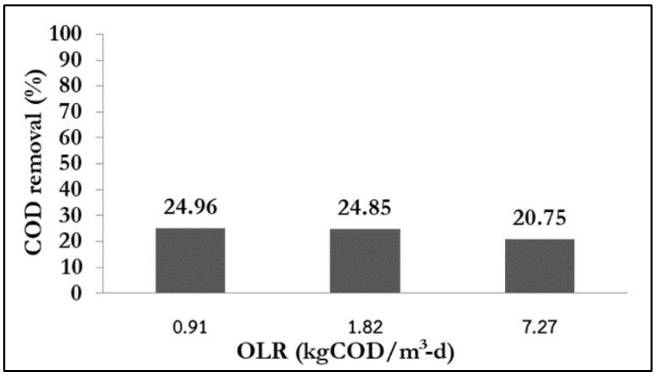


Fig. 7. Efficiency of COD removal.

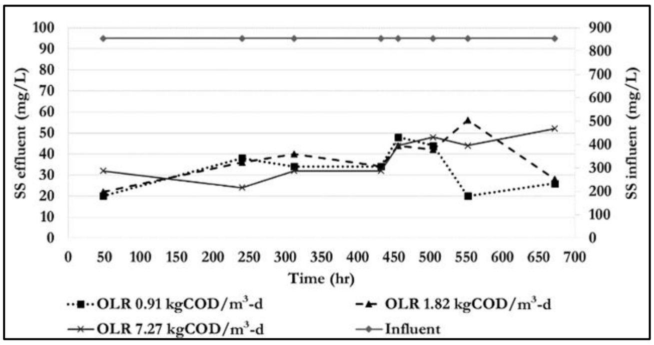


Fig. 8. SS concentration influent and effluent.

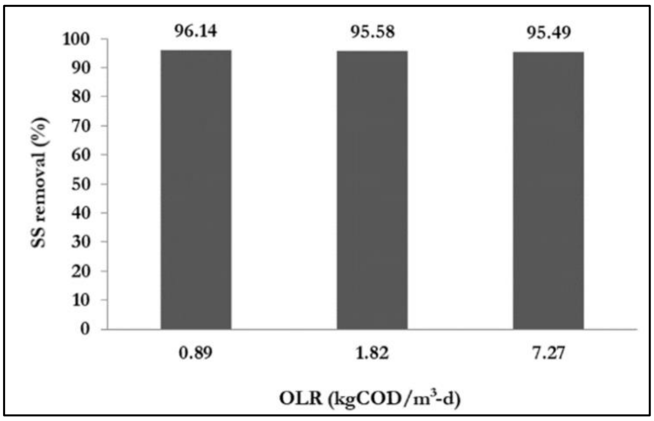


Fig. 9. Efficiency of SS removal.

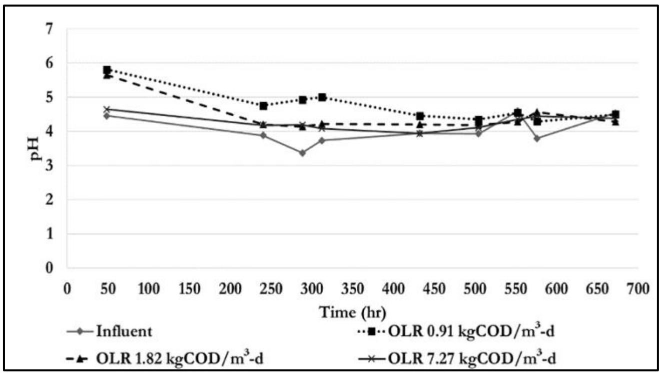


Fig. 10. pH in SCMFC.

3.3. Coulombic Efficiency

The ratio of power generation in the SCMFC to COD reduction is represented by the coulombic efficiency (CE). The result of CE is shown in Fig. 11. The maximum CE was obtained from the lowest OLR. The CE was very low at 0.277%, 0.122%, and 0.018% for OLRs of 0.91, 1.82, and 7.27 kg COD/m^3-d. The quantity of power generation depended on the electron flow from the anode to the cathode, with the electrons produced from COD utilization through the bacterial activity. However, the lower CE might have been due to some COD loss by other processes, such as absorption by GAC, or perhaps COD was removed by the growth of methanogens, subsequently reducing the electron yield [27]. The lower OLR but higher CE because of the lower OLR is more appropriate for COD reduction through bacterial activity (longer detention time), so the bacteria can produce a greater electron flow. Compared to other research, the detention times of our study were lower (48, 24, and 6 hr), so the COD removal was quite low.

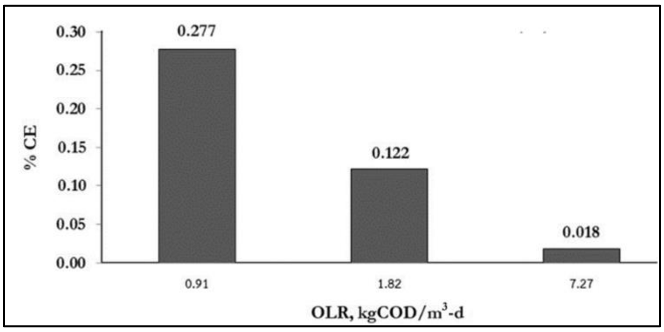
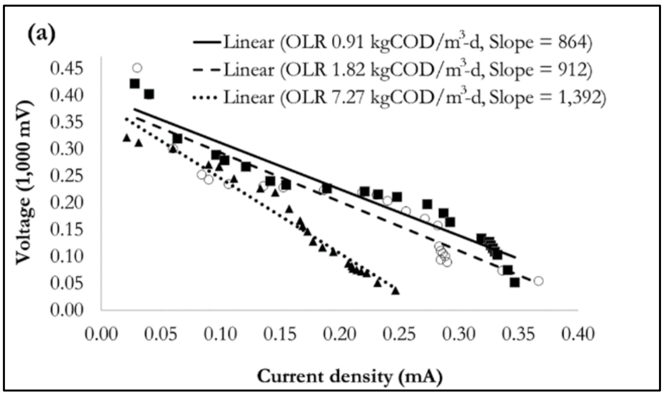


Fig. 11. Coulombic efficiency.

3.4. Polarization Curve and Internal Resistance

The polarization slope method using a plot of the electrical current and the electrical voltage curves was applied to examine the internal resistance and maximum power density by varying the external resistance from 150 to 15,000 Ohms. Fig. 12(a) and 12(b) show the slope from the polarization method and the polarization curve, respectively. The internal resistance of the OLRs of 0.91, 1.82, and 7.27 kg COD/m^3-d were 864 Ohms, 912 Ohms, and 1,392 Ohms, respectively.

The internal resistance increased with increasing OLR. The internal resistance depends on the distance between the anode and the cathode and was higher than in another study because the reactors in the current study were greater than in the other study [26]. The internal resistance results varied inversely with the power density generated (Fig. 5). This was evident the higher the OLR and the greater the distance between the anode and cathode, the greater the decrease in the efficiency of electricity generation.



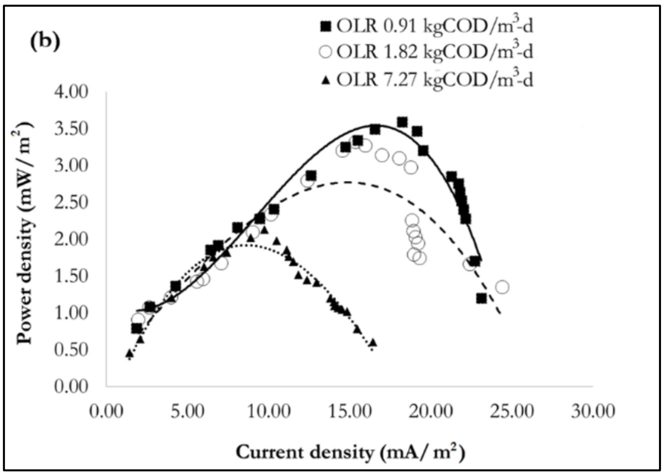


Fig. 12. (a) Slope from polarization method; (b) polarization curve.

3.5. Start-up and Operation of Circuit Converter

The prototype of the voltage boost converter for this research is shown in Fig. 13(a), (b). The transformer was built using a ferrite ring toroid core with a primary inductance (L) of 240 turns, a secondary inductance (L_p) of 240 turns, and couple inductance (L_s) of 633 turns. The input of this voltage boost converter circuit was applied from the SCMFC, while the internal resistance series with the internal voltage source of the SCMFC was 864 Ω, for 0.49V at an OLR of 0.91kgCOD/m^3-d and 912.4Ω for 0.45V at an OLR of 1.82kgCOD/m^3-d.

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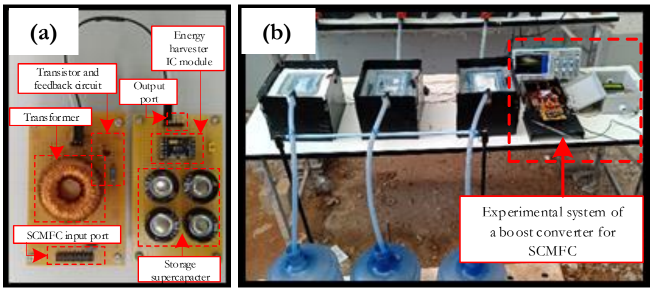

Fig. 13. (a) Photograph of circuit board; (b) experimental system of voltage boost converter circuit for SCMFC.

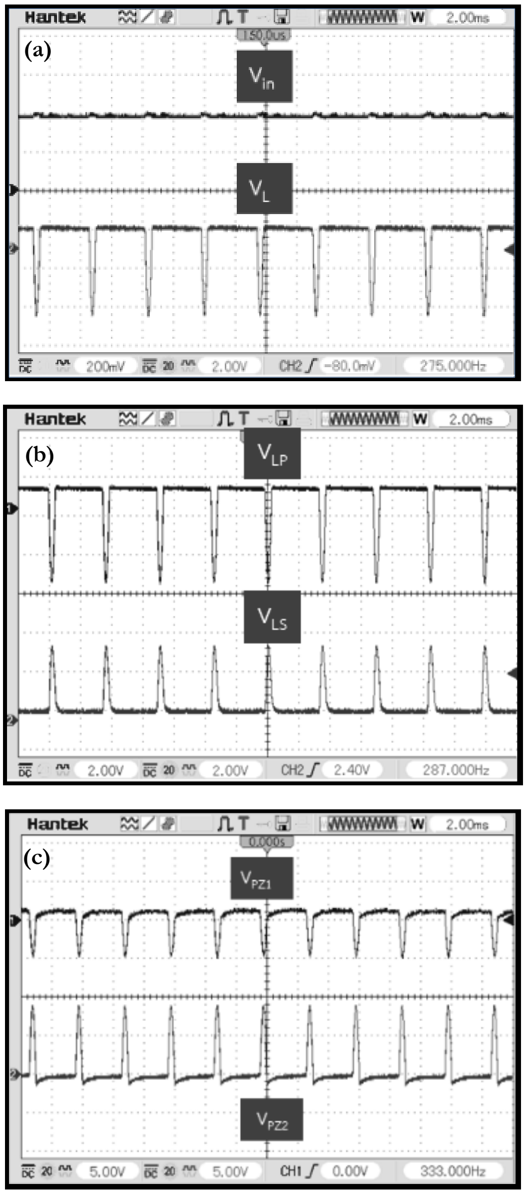

Fig. 14. Experiment: (a), (b), (c) voltage waveforms from SCMFC in switching operation during energy harvesting.

From Figs. 14 (a) and (b), the peak voltage levels of V_{in}, V_L, V_{Lp}, and V_{Ls} in the self-oscillator circuit (Fig. 3) were 0.49V (OLR 0.91kgCOD/m^3-d), 4.21V, 5.04V, and 3.2V, respectively after the output of ORL 0.91kgCOD/m^3-d of the SCMFC became stable at 1,463 hr. From Fig. 13 (c), the peak voltage of V_{PZ1} and V_{PZ2}, in the IC LTC3588-1 circuit module (Fig.3) had peak to peak values of 5.12V and 8.93V, respectively.

3.6. Experimental Energy Storage Results

The stored energy in the 0.25F/22V supercapacitors is shown in Fig. 15. When the bacteria had acclimated to the rice flour wastewater, the electrical voltage was stable for a continuous 1,463 hours during the experiment. The SCMFC could produce electrical power continuously as long as the rice flour wastewater was fed into the reactor chamber. While the SCMFC was connected to the voltage boost converter circuit, it provided electrical power to charge the 0.25F/22V supercapacitors. The electrical voltage of the supercapacitors 0.25F/22V reached the maximum value of 5.25 V(VC storage) at 19.5hrs for the OLR of 0.91kgCOD/m^3-d (Fig. 15 (a)). At the same time, the output pin of the LTC3588-1 IC module was selected and produced the output voltage (Vout_LTC3588) at 3.3V. After that, at about 42.5hr for the OLR 1.82kgCOD/m^3-d (Fig. 15 (b)), the electrical voltage of both SCMFCs decreased to 0.235V and the voltage boost converter stopped working. The voltage boost converter could not be used to store energy in the supercapacitors, due to insufficient electrical power to drive the voltage boost converter circuit. Therefore, to get the voltage boost converter circuit working again, we temporarily disconnected the SCMFC from the voltage boost converter circuit. After that, the electrical voltage of the SCMFC increased continuously and reached the maximum electrical voltage again in 240 sec. Then, when the SCMFC was reconnected to the voltage boost converter circuit, the energy from the voltage boost converter circuit could be stored in the supercapacitors again.

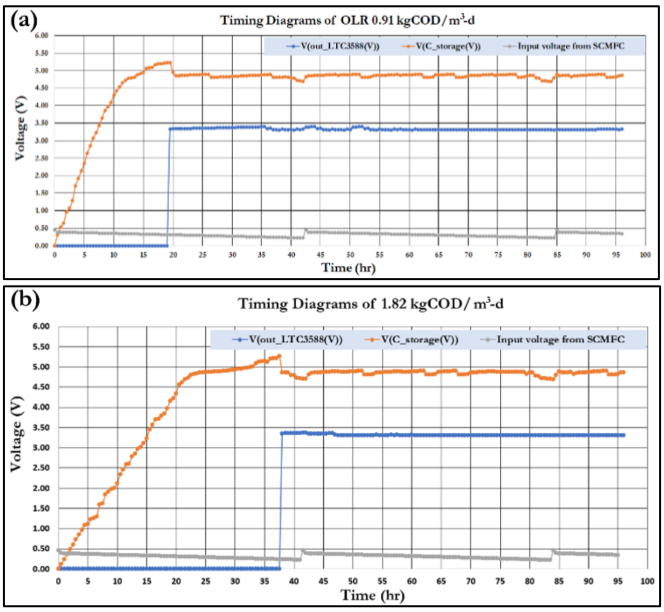

Fig. 15. Timing diagrams of charging voltage across supercapacitor (V(C_storage(V)) and output voltage selection from LTC3588-1 module (Vout_LTC3588(V)).

The voltage boost converter circuits had maximum efficiency percentages of 21.39% and 15.51% for OLRs of 0.91 and 1.82 kgCOD/m^3-d, respectively. The power dissipation of the voltage boost converter circuit was 42.45μW. In addition, we found that the OLR of 7.27 kgCOD/m^3-d of SCMFC could not be used for energy storage in the supercapacitors, due to insufficient electrical power to drive the voltage boost converter circuit in the second stage.

4. Conclusion

Rice flour wastewater could be treated using SCMFC technology in the continuous flow mode of operation. Suspended solids from rice flour wastewater could be removed effectively (96.14%, 95.58%, and 95.49) for OLRs of 0.91, 1.82, and 7.27 kg COD/m^3-d, respectively, with COD removal efficiency percentages 24.3%, 24.84%, and 20.95%, respectively. The minimum internal resistance for the microbial fuel cell was observed for the OLR of 0.91 kgCOD/m^3-d. The maximum power of 13.16 mW/m^2 was generated from the OLR of 0.91 kg COD/m^3-d. This study successfully enhanced a single chamber microbial fuel cell for suspended solid removal from wastewater and electricity production using GAC as the anode. Advantages of this new method were it is cheaper than using carbon cloth and it increases the surface area for the filtration of suspended solids and microorganisms can attach to it for growth. The SCMFC could treat the rice flour wastewater and produce electricity at the same time. Using the supplied voltage source of the SCMFC in continuous flow mode operation, the SCMFC was sustainable power source that did not generate any type of pollution. The research successfully achieved energy storage from rice flour wastewater using an SCMFC in continuous flow mode operation with a voltage boost converter circuit. This research could be applied as a pollution-free and renewable power source for some electronic devices.

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