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Artikel riset

Compost Solid-phase Microbial Fuel Cell (CSMFC) Performance using Graphene and Graphite as Electrodes

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Abstract

Organic waste is a type of waste produced by many sector, which need to managed appropriately. During its development, composting is one of the organic waste management efforts that is often be applied, Another alternative organic waste management in the form of Microbial Fuel Cell (MFC) has emerged. Several researchers conducted studies on MFC performance which was influenced by many factors, especially the electrode which contributes to the electron transfer process. This study has a concern about energy optimization through CSMFC technology using different electrode's material. Electrode materials from Graphene and Graphit has good electro-conductivity and has a large surface area, making it suitable for bacteria to adhere. The sampled reactors are consists of two types of electrodes in the form of graphite and graphene. Each materials has anode and cathode ratio of 1:1, 2:1, and 3:1. The samples measured into three kinds, which called a mature compost measurement, electrochemical measurement, and biochemical measurement. Some collected sampling data were then processed and analyzed statistically using SPSS software. The processed and analyzed data included the calculation of power density, total N, C/N ratio, and moisture content. Any data like voltage (V) and electric current (I) are needed to obtain a power density. The highest average voltage, current, power and power density are produced by the N₃ reactor (graphene 3:1) that is 269 x 10⁻³ V, 163 x 10⁻⁶ A, 56 x 10⁻⁶ Watt and 1.914 x 10⁻³ W / m^2 . There is no significant effect of variations in the type of electrode (graphite and graphene) on CSMFC performances.

Keywords : Compost Solid-phase Microbial Fuel Cell (CSMFC); Graphene; Graphite.

1. Introduction

Organic waste is a type of waste produced by many people. Some of the contributors to organic waste include leaves, food scraps, animal waste, and so on. In Semarang, the composition of organic waste even reaches 43.96% of the total waste production (KLHK, 2019). The existence of organic waste that has not appropriately managed can cause various problems.

During its development, composting is one of the organic waste management efforts that is often be applied. Furthermore, another alternative organic waste management in the form of Microbial Fuel Cell (MFC) has emerged. According to Rabaey and Verstraete (2005), the application of MFC in organic waste management provides the possibility of sustainable energy production by utilizing the activity of bacteria that decompose organic material. MFC technology is known as environmentally friendly technology. MFC converts chemical energy in organic waste into electrical energy with the help of biocatalysts in the form of decomposing bacteria (Ci et al., 2015).

MFC has considerable potential to be applied in organic waste management. To obtain better MFC performance, several researchers conducted studies on MFC performance which was influenced by variations in pH, electron acceptors, electrode surface area, decomposing bacteria, and C / N ratio (Logan et al., 2006). One of the main performance optimization factors of MFC is the electrode which contributes to the electron transfer process. The electrode materials in MFC must have a specific characteristic to support MFC performance. Therefore, the electrical conductivity of an electrode material must be well cared for. The electrode material has at least biocompatibility, high conductivity, and having an affordable price.

Most of the electricity production technologies use carbon as an electrode. Carbon is available in various forms such as slabs, bars, sheets and powders. Electrodes in the form of plates or rods are relatively cheap, easy to use, and have a definable area (Logan et al., 2006). One type of carbon is graphene. According to Ci et al. (2015), electrode material made from graphene not only increases the electrocatalyst but also as a metal-free catalyst. Graphene also has good electro-conductivity and has a large surface area, making it suitable for bacteria to adhere. In addition to the graphene carbon material that is often used as an electrode, graphite carbon is also used.

In line with this study, previous research about the Performance of Microbial Fuel Cell Producing Bioelectricity with Different Types of Electrodes in Fishery Industry Liquid Waste has been conducted by Bustami, et al. (2017). Those studies analysed the optimum production of electricities using aluminium and carbon graphite as electrodes. It was indicated any effects of variation electrode material towards the electrical products generated by CSMFC technology. The results suggest the possibility of using aluminium and graphite carbon as the electrode to produce the highest electricity (Bustami, et al., 2017).

As the results of previous research, it is essential to obtain a study related to energy optimization through CSMFC technology using different electrode's material. This study aims to observe any possible impacts affected by other electrodes utilization in CSMFC technology. This study compares the use of graphite and graphene carbon to energy produced in CSMFC. Any factors associated with CSMFC performance, such as pH, temperature, C/N ratio, electron acceptors, electrode surface area, and other factors will be investigated.

2. Methodology

This research was conducted from July 2019 to October 2019 at the Environmental Engineering Laboratory of Universitas Diponegoro. This research was carried out using Lab-scale reactor with single-chamber type, which has a varied number and type electrodes. There are six reactors with two types of electrodes used in the form of graphite carbon and graphene carbon. Each class has anode and cathode ratio of 1:1, 2:1, and 3:1, which can be seen in Fig. 1 below.

The reactor is made from plastic material with a total volume of 2.5 L in the form of a cylinder. The cylinder shape is chosen because it has no angle so that the compost will be flat and easier to observe.

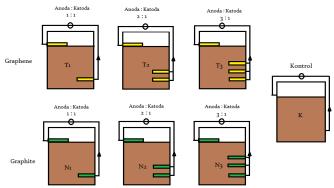


Figure 1. Scheme-view of reactors

The compost base material used is in accordance with Ariyanti et al. (2019), namely the mixture of leaf waste and food waste with a ratio of 50:50 is better than a combination of leaf waste and food waste and food waste with a ratio of 40:60 or 60:40. The combination of leaf waste and food waste was chosen based on previous research from Groud et al. (2011), which states that a mixture of leaves and food waste can be a good substrate for processing in anaerobic digester because the mixture has very high organic content contained in it. The planned water content is 70% referring to Wang et al. (2014) and Wang et al. (2015).

At the beginning of the research, the compost solid-phase microbial fuel cell (CSMFC) reactor was made with one compartment (single chamber) measuring 2.5 litres. The anode is keeping under anaerobic conditions to get optimal CSMFC process. After that, the electrodes are installed in parallel by adjusting the predetermined variations.

In the next process, a preliminary test is carried out in the form of a waste characteristic test and an electrode characteristic test. A solid-waste characteristic test were consists of moisture content test, C-Organic test, N-total test, P-total test, and the COD test. Meanwhile, the electrode characteristic test were consist of voltage test, resistance test, and electrical current test.

The implementation stage of this research started from the running reactor until data collection. The implementation stage was carried out for 30 days with seven times of parameter testing, at Ho, H5, H10, H15, H20, H25 and H30. There are six reactors and one control reactor to compare the final results. Each composting reactor has been treated by adding EM4 activator for the starter and stirring every five days for homogeneous final compost.

The sampled parameters are measured into three types, which called a mature compost measurement, electrochemical measurement, and biochemical measurement that can be seen more clearly in the following table 1.

| Parameter | Sampling Frequency | Methods |
|-----------------------------|-----------------------------|-------------------|
| Mature compost measurement | | |
| - C-Organic | Ho, H5, H10, H15, H20, H25, | Spectrophotometry |
| - N- Total | and H30 | |
| Electrochemical measurement | | |
| - Voltage | Every day | Multimeter |
| - Current | | |
| Biochemical measurement | | |
| - pH | Every day | SNI 6989.11-2004 |
| - Temperature | | |

Table 1. List of sampling test

Some collected sampling data were then processed and analyzed statistically using SPSS software. The processed and analyzed data included the calculation of power density, total N, C/N ratio, and moisture content. Any data like voltage (V) and electric current (I) are needed to obtain a power density. The formula below shows a power density calculation used in this study.

| PD = | $\frac{V \times I}{A}$ (1) |
|------|---|
| Wher | e : |
| PD | = Power Density |
| Ι | = Electric current (A) |
| V | = Voltage (V) |
| А | = The surface area used by microorganisms growing at anode (m2) which has a 0.05 m2 |
| | of area. |

3. Results and Discussions

3.1 Preliminary Results

3.1.1 Solid-waste Characteristic

The initial waste obtained from food scraps and dry leaves is tested to get the initial character of the waste. Food waste which is taken weighs 20 kg, and leaf waste weights 10 kg. The characteristic waste results can be seen in Table 2.

| Table 2. The Preliminary solid-waste characteristic | | | | | |
|---|----------------------|------------|------------|--|--|
| No. | Parameter | Food waste | Leaf waste | | |
| 1 | C Organic (%) | 27.5 | 13.2 | | |
| 2 | N Organic (%) | 0,59 | 0,43 | | |
| 3 | Rasio C/N | 48,59 | 32,1 | | |
| 4 | Moisture content (%) | 55 | 15 | | |
| 5 | pН | 6.1 | 5,9 | | |

| Before the running stage, it is necessary to measure the moisture content of the solid waste. |
|---|
| Water additions, both leaf and food waste was 70% -80% reached, then it has mixed before filling the |
| reactor. Solid-waste ratio of 1: 1 containing 300 gram leaf waste and 300 gram food waste. 20 ml MOL |
| was added as a compost starter for each reactor and then stirred until blended. Thereafter, putting the |

3.1.2 Electrodes Characteristic

waste into the reactor along with arranging the electrodes is necessary.

The electrodes used in this research are graphite carbon and graphene carbon because they have high conductivity and are easy to obtain. The dimensions used are 12.5 cm long, 4 cm wide, and 1 cm thick. The surface area of each electrode is 0.0133 m2.

The test is carried out by measuring the internal resistance on the electrodes. From the test results of 9 graphite electrodes and nine graphene electrodes, the resistance value of the graphite electrode is 6 ohms while the graphene electrode is 3 ohms. Graphene tends to be more porous than graphite. The graphenes porous would increase the biocompatibility so that microorganisms can attach to the electrodes as many as possible.

3.2 Sampling Results

3.2.1 CSMFC Composting Performance

a. Temperature

Temperature is a parameter that affects composting performance. According to (Damanhuri and Padmi, 2016), a low temperature would affect a long time composting process. Whereas, if the temperature is too high, the oxygen content will decrease. The carried test is using a soil-meter, plugged into the middle of the reactor, and repeated in 2 times. Measurement results at each reactor can be seen in Figure 2.

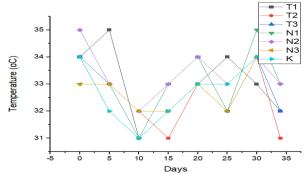


Figure 2. Daily temperature measurements

Figure 2 illustrates a fluctuating temperature with a 32 °C lowest value and 36 °C highest value. The highest temperature occurs on the first day of testing, which shows there is an increase due to the start of the composting process. Furthermore, the mesophilic process occurs with a temperature between 30-37 °C which lasts until the end of the study.

Regarding the composting process, according to Sánchez et al. (2017), the mesophilic stage takes place at a temperature of 15-35 $^{\circ}$ C. In this study, the peak temperature occurred on day 5 of 35 $^{\circ}$ C. Exothermic processes cause temperature increase by microorganisms that produce heat.

b. Moisture content

The moisture content in compost affects the activity of microorganisms and conductivity in the compost (Liang et al., 2003). The too high water content will block oxygen and carbon dioxide production so that harmful compounds accumulate in the compost and cause an anaerobic compost (Hamoda et al., 1998). Still, if the water content is less than 40%, the activity of microorganisms will decrease. Ideal water content for composting ranges from 50-70% (Liang et al., 2003). Measurement of water content is collected in every five days, each day 0, 5, 10, 15, 20, 25 and 30, as shown in Figure 3 below.

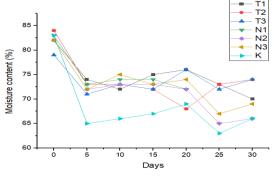


Figure 3. Moisture content measurements

From the results in Figure 3, the average initial water content (day o) is above 80% due to the initial waste which contains a high water content. In the next day, the water content ranged from 65% to 74%. This figure is in line with the preliminary test results to keep the water content between 60-80 %, avoiding evaporation. That 60-80 % of water content is categorized into optimal water content for composting (Liang et al., 2003).

c. pH

Acid-base conditions describe the process of organic compounds degradation by microorganisms. At the beginning of composting, there will be a decrease in pH due to the emergence of simple organic acids by microorganisms. Then the pH will gradually increase until the compost is settled at a pH of 7-8. The pH also influences the potential electrode in the system (Logan et al., 2006).. The pH measurements for 30 days can be seen in Figure 4.

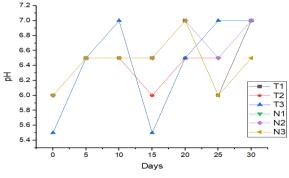


Figure 4. pH measurements

The decrease in pH is caused by the emergence of organic acids produced by microorganisms. This organic acid is not only produced but also used by microorganisms as a substrate. In low oxygen conditions, the production of organic acids is higher, resulting in a lower pH. In this study, the lowest pH showed a value of 5.5; this was caused by the condition of the compost, which received enough oxygen, so that little fermentation occurred. The pH value and also the acid content affect the toxicity effect of organic acids. (Sundberg, 2003).

The increase in pH occurs after touching the value of 5.5, then slowly the pH rises to approach the neutral value between 6 to 7. This phenomenon affects the activity of microorganisms which can grow optimally at neutral pH.

d. Organic Carbon and N-total

The carbon and nitrogen content in compost is needed by microorganisms to produce energy and also to build new cells. Bacteria break down organic compounds in complex form into simpler compounds (Findlay, 2013). The optimal C/N ratio value was different for aerobic and anaerobic microbe types. The recommended C/N ratio for composting is between 25-35 compared to anaerobic digestion, which is 20-30 (Torres-Climent et al., 2015). Low C/N conditions will produce high ammonia and Volatile Fatty Acids, which can cause an odour in composting.

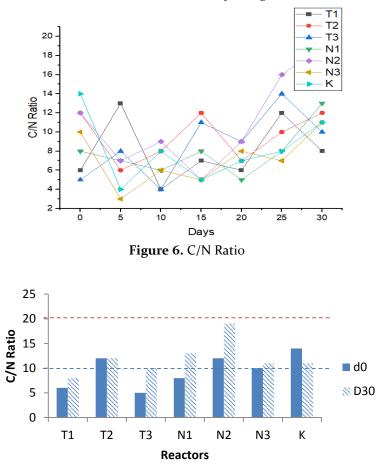


Figure 7. Intial C/N ratio and final C/N ratio

In Figure 6, it can be seen that there is a change in the value of the C/N ratio. The lowest value obtained at N₃ on day 5 with 3.6, while the highest obtained at N₂ on day 30 with 19.1. The C/N comparison ratio can be seen in figure 7. From the whole reactor, the comparison between the values of day 0 and day 30 shows that the C/N value is getting closer to the quality standard. This value proves that the composting process that takes place causes the compost to ripen even though the T₁ and T₃

reactors have not met the criteria for mature compost yet. In this case, it shows that the C/N ratio in the reactor with graphene has an optimum value that is better than the reactor with graphite.

3.2.2 CSMFC Electricity Production a. Electrical Voltage

The electric voltage in MFC occurs due to the transfer of electrons released by bacteria when breaking down organic compounds. The highest voltage is N₃ compared to N₁ and N₂; this is in accordance with Al-Fetlawi (2016), which states that the surface area of the electrodes affects the performance of MFC. In N₁ and N₂ the number of electrodes is less than N₃ so that the amount of electrode surface area is smaller. Also, compared to graphite, graphene electrodes have a more porous surface. According to Hou et al. (2014), surface area and biocompatibility can increase the interaction between the biofilm and the electrode, so that the colonization of bacteria results in a higher reactive surface.

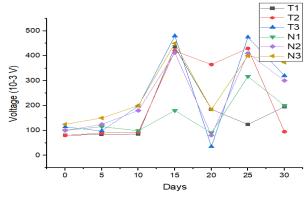


Figure 8. Electrical voltage measurements

b. Electrical Current

Electric current is the movement of electrons caught by the anode, then flows according to the potential difference towards the cathode. The electric current test can be seen in Figure 9.

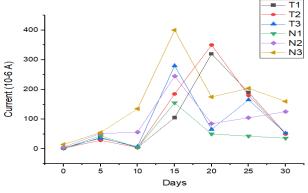


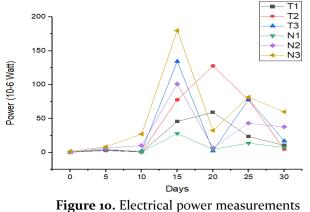
Figure 9. Electrical current measurements

In the graph, it can be seen that there is a fluctuation in the value of electric current. The highest value was N₃ on day 13 with a value of 401 μ A, while the lowest value was N₁ on day 10 with a value of 7 μ A. The average value of electric current obtained by reactor 3 is 176.35 μ A.

c. Electrical Power

Electric power is the result of electrical measurements for each reactor regardless of the number of electrodes. The amount of electrical power is expressed in 10⁻⁶ Watts, which is obtained from the multiplication of current and voltage. The results of the observations can be seen in Figure 10. The

maximum value obtained by the N₃ reactor is 180×10^{-6} Watt, and the T₃ reactor obtains the minimum value with a value of 0.456 x 10^{-6} Watt. The N₃ reactor shows the largest average value with a value of 60.46 x 10^{-6} Watt, and the lowest average is obtained for the N₁ reactor with a value of 5.4×10^{-6} Watt.



d. Power Density

Power density (PD) is the amount of power generated by the MFC divided by the total surface area of the electrodes. PD is expressed in units of 10^{-3} W / m2. PD is calculated using the total area of the anode in which biological reactions occur (Logan et al., 2006). The maximum and minimum PD values occur in the T1 reactor with a value of 11.12×10^{-3} W / m2 and T3 0.04 x 10^{-3} W / m2. The average PD in graphene is 1.36×10^{-3} mW / m2 while in graphite it is 1.63×10^{-3} mW / m2. This shows that the PD interval on the graphite electrode is larger, so it tends to be unstable compared to graphene even though it has a larger average value. The graph of the PD value for 30 days can be seen in Figure 11.

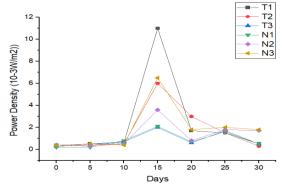


Figure 11. The power density measurements of all CSMFC reactors

3.3. Stastical Anaysis

Statistical analysis was conducted to prove the hypothesis whether there was an effect of the independent variables, namely the number and type of electrodes on the dependent variable, namely composting performance. The independent variables measured are the C / N ratio as an illustration of the level of compost maturity and power density as an illustration of the CSMFC electricity production level. The statistical analysis used for the parameters of the C / N and PD ratio is Two Way ANOVA.

Two Way ANOVA test was used because in this study tested the C/N ratio based on two independent variables, namely the number of electrodes (1: 1; 2: 1; 3: 1; and Control) and the type of electrodes (Graphite, Graphene and Control) which were divided into seven reactors (T1, T2, T3, N1, N2, N3, and K). For testing on PD using two factors or independent variables, namely the number of electrodes (1: 1; 2: 1 and 3: 1) and the type of electrodes (Graphite and Graphene). Four requirements must be met before the ANOVA test, which:

- 1. The sample comes from an independent group.
- 2. The variance between groups must be homogeneous.

- 3. Residual values are normally distributed.
- 4. The dependent variable data must be in the form of an interval/ratio, and the independent variable must be a category.

Two Way ANOVA test on C/N and PD ratio variables are using seven different and unrelated categories namely T1, T2, T3, N1, N2, N3, and K. Number 1 and 4 requirement have been fulfilled as the value of C/ N variable showing a ratio and the value of the PD variable showing an interval. After requirement 1 and 4 are met, the variance uniformity test is carried out to determine whether all samples have the same variance (homogeneity). Based on the results of the homogeneity test, the significance obtained by the C/ N and PD variables is 0.299 and 0.140 or greater than 0.05. So that all population variants are said to be homogeneous. With these results, the second requirement for the Two Way ANOVA test was fulfilled.

The last requirement is the normality test value. The normality test is carried out to determine whether the sample residual standard value is normally distributed. If the data normality test is not normally distributed, a transformation or an outlier elimination is required. Based on the results of the SPSS 25 test, the significance value of each sample is greater than 0.05, so the data comes from the normal population. Because condition three has been fulfilled, the statistical test can be continued to the next stage, namely the Two Way ANOVA test.

Two Way ANOVA was carried out on a CN ratio, also on PD ratio based on seven different and unrelated categories, namely T1, T2, T3, N1, N2, N3, and K. The interaction test between independent variables was carried out to determine whether these variables had a significant effect or not.

The significance value of the type of electrode * The number of electrodes towards CN ratio is 0.618 or greater than 0.05, so it can be said that there is no interaction between the number and type of electrodes with the CN ratio. It also exists in the significance value of the type of electrode * the number of electrodes towards PD ratio. The significance value is 0.152 or greater than 0.05, so it can be said that there is no interaction between the number and type of electrodes with the power density.

Based on the statistical analysis, the number and type of electrodes do not significantly affect both composting performance and electricity production performance. This refers to the process of compost degradation in which bacteria do not depend on electrodes for metabolic processes. The anode is used to help bacteria as an electron acceptor which will flow electrons to the cathode to react with oxygen. The electrodes do not directly affect the production of electricity but only act as a bridge for electrons to get the final acceptor in the form of oxygen. From the power density statistical test, it can also be seen that the number and type of electrodes do not have a significant effect on PD production. These results indicate that the power density (power per anode surface area) is not affected by the type of electrode (graphene and graphite) and the number of electrodes (anode: cathode 1: 1, 2: 1, 3: 1). It can be seen also that in the type of graphene electrode, the greater the number of electrodes, the greater the current, voltage and power. The performance of the graphene electrode is more stable than that of the graphite electrode.

4. Conclusions

In overall, the results show that CSMFC can be used as an alternative to organic waste management. Based on the results of the research that has been obtained, it is known that there is no significant effect of variations in the number of electrodes (anode: cathode 1: 1; 2: 1 and 3: 1) on the composting performance and electricity production of Compost Solid Microbial Fuel Cell. In addition, there is no significant effect of variations in electrode types (graphite and graphene) on composting performance and electricity production of Compost Solid Microbial Fuel Cell. The average value of electric voltage, electric current, electric power and the highest power density is produced by the N₃ reactor (graphene with anode: 3: 1 cathode), which is 269 x 10⁻³ V, 163 x 10⁻⁶ A, 56 x 10⁻⁶ Watt and 1.914 x 10⁻³ W/m².

Should be noted that the electrical installation in the CSMFC reactor is designed to be tidier and stronger and made of materials that are not easy to react. This is intended, in order to survive in various conditions to minimize any disturbance when retrieving data. Also, sample testing is carried out as soon as possible to avoid changing sample conditions.

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