

Optimization of Microbial Fuel Cells Operating Parameters for Better Removal of Organic Matter and Higher Energy Production from Wastewater

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Abstract: In this study, four double-chambered Microbial fuel cells (MFCs) operated by primary effluent wastewater mixed with anaerobic sludge as substrate, was designed, built, and optimized for better higher energy production and subsequently better removal of organic matter. Optimized MFCs operating parameters as a function of energy produced include electrode material type, electrode size, salt bridge diameter, type of salt solution that used in salt bridge, and concentration of the salt solution used in the salt bridge. Three duplicates-MFCs for each parameter value were used. Output open-circuit voltage (OCV) was measured for each MFC one time daily and for one week for each tested operating parameter. Data obtained showed that (i) MFCs with copper electrodes produce output voltage significantly higher than MFCs with carbon brushes electrodes which, in turn, achieved output voltage significantly higher than both that achieved by MFCs with zinc electrodes and MFCs with manufactured carbon electrodes, (ii) MFCs with 10 mm salt bridge shown significantly higher output voltage than MFCs with both 16 and 24 mm salt bridges, (iii) KCl salt bridge in MFCs is significantly more efficient than NaCl salt bridges, and (iv) MFCs with 1M KCl salt bridges can produce output voltage significantly higher than that produced by MFCs with 3M KCl salt bridges.

Keywords: Microbial fuel cells, Wastewater treatment, Energy production, Electrode material type and size, Salt bridge diameter, Type and concentration of salt solution.

1. INTRODUCTION

Non-renewable sources of energy including fossil fuels are dramatically decreasing worldwide; due to the heavily increasing energy consumption. Moreover, increasing carbon emissions from fossils fuels resources, followed by a change in carbon percentage in the atmosphere have a great input into global climate change; so extensive research was started globally to find cheap, environmentally friendly, and sustainable sources of renewable energy [1]. The research was devoted also to energy demand reduction through better socioeconomic demand management [2].

Renewable energy is the energy created by naturally replenished sources such as winds, sunlight, tides, etc. Other energy sources such as biomass, anaerobic digestion, and biofuel are widely considered renewable energy [3].

With increasing population, urbanization, and industrialization, the generation of wastewater was increased with composite content and negative impacts on the land and water environment. In addition, wastewater treatment plants worldwide are a big consumer of electrical energy [4-6]. Consequently, the

use of alternative and renewable energy sources in wastewater treatment such as MFCs is important for developed as well as developing countries to reduce dependence on fossil fuel use and meet the increasing energy demands.

MFCs are important, reliable, and effective devices that directly convert the chemical energy stored in organic matters into electrical energy using microorganisms, organic matter, and anaerobic conditions [7, 8]. Historically, the concept of electricity production using microorganisms was discovered in 1911 [9, 10]. Then it was proven that batch biological fuel cells could produce more than 35 volts [11, 12]. Clear principles for MFCs were identified in 1976 [13, 14]. Extraction of electrical current from MFCs that were operated using wastewater was conducted in the 1980s using pure cultures and artificial electron mediators [15, 16]. Later in the 1980s, it was discovered that the generated electricity could be significantly increased if electron mediators were added [17].

Typical MFC used in testing microbial activity and optimizing materials consists of two compartments: anodic chamber and cathodic chamber, separated by a salt bridge or proton exchange membrane (PEM) [18-21]. Substrate (organic-rich matter) is added to the anodic chamber, where conversion of organic matter

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occurs by anaerobic microorganisms [22, 23]. In addition to energy production and organic matter removal from wastewater, MFCs as an anaerobic process simultaneously reduce or have minimal or no net-CO₂ emission contributing to climate change reduction [24]. Electrons and protons are produced from oxidation half-reaction of the substrate, electrons transfer from the anode to the cathode using external electrical load (resistance); where protons transfer to the cathode was done through salt bridge [25, 26].

As a comparison between MFCs and conventional aeration treatment of wastewater, it was found that both processes were able to remove more than 90% of COD; but with a shorter time (8 days) in conventional aeration than (10 days) in MFCs [27]. In terms of operating temperature, MFCs have the advantage of working at lower temperatures than conventional systems [28]. In term of power production by MFCs systems there are difficulties encountered including anode limitations and electrochemical losses that impedes their commercial applications in wastewater treatment [29]. It was reported that the combination of the two technologies (conventional wastewater treatment and MFC) allows for broadening the spectrum of bioconversion technology [30].

It was found that different aspects of MFC design and function were heavily investigated and reported in the literature. Particular interest has been devoted, among others; to the choice of electrode material, the type of microorganism, or the electron transfer mechanism [31, 32]. However, there is only an incomplete knowledge of how different MFC design and operating parameters influence the energy output of the cell [33-38].

MFCs application as an alternative energy source is presently narrowed to certain areas such as wastewater treatment, powering underwater monitoring devices, power supply to remote sensors, BOD sensing, biosensors for online monitoring of treatment processes, nitrification-denitrification, and hydrogen and methane production. In addition, MFCs could be used to treat biomass, algae, landfill leachate, agricultural runoff, and industrial waste [39]. With further improvements in design, cost-effectiveness, and performance efficiency, it would be possible to scale up and use MFCs as a renewable energy resource [40]. It was reported that MFCs as an energy source and wastewater treatment method can be used in remote communities and also as a desalination plant [41-43]. Apart from generation electricity, the sediment MFCs

have been used for successful in situ bioremediation [44, 45].

Multiple modules of large-scale MFC system development and applications are currently of high demand but these modules may present new challenges and limitations associated with energy extraction such as the limited effectivity of the open-air cathodes which are still unresolved and need to be addressed in a systematic manner [46-50].

The relations and effects of flow rate and substrate supply rate, in terms of power output and chemical oxygen demand (COD) removal efficiency were investigated by changing anode distance to the membrane or anodic volume. Results suggested a trade-off correlation between these two parameters [51].

This paper describes the optimization of several operating parameters that affect the design, operation, and performance of an MFC including anode electrode material type, electrode size, and type and concentration of the salt solution that is used in salt bridge.

2. MATERIALS AND METHODS

2.1. Design and Assembly of MFC Model

The MFC system consists of a double-chambered-MFCs with different types of electrodes, salt bridges for protons exchange, primary effluent wastewater, and anaerobic sludge as substrate. See Figure 1. Particulars of MFC chambers, electrodes, salt bridges, mixing system, cathodic chamber, substrate composition and sampling, electrical panel, temperature control system, and voltage and power measurement are given below.

2.1.1. Chambers

MFCs were constructed using glass jars for both anode and cathode chambers. The volume of the used jars was 1 liter, whereas the used volume was 800 mL; this is to prevent substrate and anodic chamber solution from dropping outside the jars during mixing/shaking.

2.1.2. Anode Electrodes

Four types of electrodes (Zinc plate, Copperplate, Carbon brushes, and pre-treated Carbon electrodes) were prepared and inserted into twelve different WW-MFCs, three duplicates for each one of the four electrodes, dimensions of all electrodes were 7 cm X

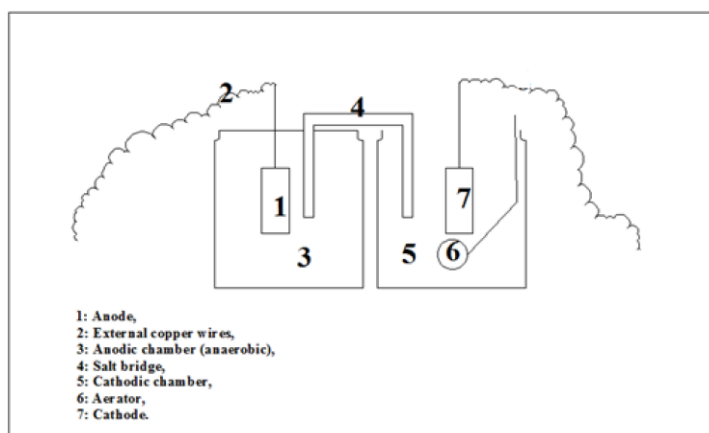


Figure 1: Schematic diagram of DS-MFC components.

24 cm. The area of anodes was equal in all MFCs and equal to 28 cm².

All Electrodes were soaked into a mixed solution of anaerobic sludge and primary effluent wastewater (20% sludge in terms of volume) at 35 °C for 3 days; to allow culture to form on the surface of the anode.

2.1.3. Salt Bridges

Salt bridges were used as proton exchange media due to their low cost and availability comparing to PEMs. Pyrex Glass tubing was used as the structure of the bridges; because it is an inert material chemically and electrically. U-shaped salt bridges were constructed from straight glass tubes and using Bunsen flame for bending to form U-shaped tubes. Salt bridges of 16 mm diameter and filled with 1 M concentration KCl salt solution used.

2.1.4. Mixing System

A mechanical mixing system consisting of a shaking plate was used. The mechanical shaker consists of geared motor, transition mechanism, bearing, caster wheel, and movable box. All MFCs were fixed inside the built shaker during all experiments. See Figure 2.



Figure 2: Employed mixing system.

2.1.5. Cathodic Chamber

Oxygen is the best efficient electron acceptor as mentioned before. In this experiment, the used cathodic solution consists of aerated distilled water. Distilled water was used, 800 mL distilled water was used in each cathodic chamber. Evaporation of distilled water was noticed due to the heater and aeration effect; so cathodic chambers were refilled on daily basis.

Aerators used in fish tanks were applied to aerate the cathodic chambers. Aerators were operated for 15 minutes intervals; each operating interval was followed by 15 minutes break to prevent the exhausting of the aerators. It was assured that the amount of aeration is approximately equal in all cathodes.

2.1.6. Substrate Composition and Sampling

Primary effluent wastewater mixed with anaerobic sludge was used as a substrate for all of the performed experiments in this study. A sampling of the substrate was done from the weirs of the primary sedimentation tank in the Nablus wastewater treatment plant (WWTP) and various locations along the weir, and then a composite sample was prepared by merging 5 different samples from five different locations. Because Nablus wastewater system is combined with the sewer system, sampling of the substrate was done in non-rainy days and at least after 72 hours of any raining storm [52].

The substrate was collected in 10 liters plastic container, and then stored at 35 °C until use within 1-2 days, to assure keeping microorganisms active.

In addition, sludge was collected in a glass container from the anaerobic digester in the same WWTP and stored at 35 °C until use within 1-2 days.

Used Substrate in all experiments was a mixed solution of anaerobic sludge and primary effluent wastewater (20% sludge in terms of volume).

2.1.7. Electrical Panel

To ease the voltage and power measurements, and electrical panel was prepared.

For each MFC, a 1000-ohm resistance was fixed at the electrical panel and connected with the electrodes with copper wires, for all MFCs the length of the copper wires was equal (1 meter). See Figure 3.

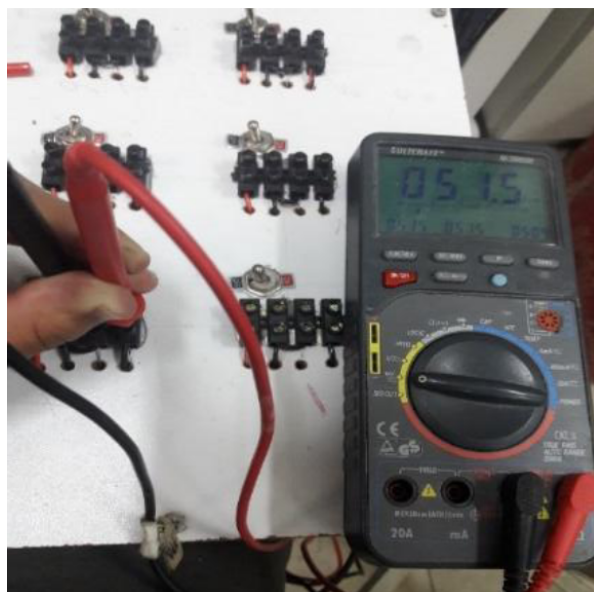


Figure 3: electrical panel.

2.1.8. Temperature Control System

The temperature control system was designed, built, and applied to the hood where MFCs were operated in, to keep the temperature within 34-36 °C during the operation of the MFCs. The temperature control system consists of Arduino, two waterproof temperature sensors, two air sensors, a heater, a Bluetooth device for monitoring, and a microcontroller.

2.1.9. Voltage and Power Measurement

Voltage measurements were made with Voltcraft M-3860M multi meter. Recording voltage measurements were performed after the reading stabilized (became constant) on the multi meter screen.

2.2. Optimization Plan

The optimization plan of DS-MFCs operating parameters consists of two parts, optimization of salt bridge material and optimization of salt bridge characteristics.

2.2.1. Effect of Anode Electrode Material

MFCs with different anode electrode materials (Zinc, copper, manufactured carbon, and carbon brushes), 3 duplicates for each, were operated for 7 days, and the behavior of each MFC was inspected in terms of output voltage. Voltcraft M-3860M multi meter (manufacturer: METEX) was used to measure output open circuit voltage on daily basis.

2.2.2. Effect of Salt Bridge Characteristics

Effect of salt bridge characteristics was conducted as follows:

- Three different diameters salt bridges (10, 16, and 24 mm) were studied in DS-MFCs using carbon brushes electrodes and 1M-KCl salt. They were compared to each other in terms of output power.
- The copper electrode was used based on the results of electrode optimization experiments.
- Two types of salt solutions, 1 M KCl and 1 M NaCl, were used in WW-MFC and compared each to the other in terms of output voltage.
- Then, three different concentrations of KCl solutions (1M, 2M, and 3M) were used in WW-MFCs, three duplicates for each concentration, and compared each to the other in terms of output voltage.
- The efficiency of salt bridges depends on their diameter, length, and type of salt used, and concentration of the salt solution. Salt bridges length was specified practically, in a way that the ends of the bridges were approximately reached the middle of the chambers. For the selection of the other characteristics of the salt bridges, three experiments were performed. All output voltage measurements were performed on daily basis for all salt bridge experiments.

2.3. Data Management and Analysis

One-way ANOVA was used as a tool for statistical analysis to find the significant differences between different conditions investigated. Linearization using EXCEL was utilized to characterize COD kinetic model. Excel was used also to find the relationship between COD of the substrate in MFC and output voltage of the same MFC.

No COD samples were taken through this part.

2. RESULTS AND DISCUSSION

The effect of four parameters involved in the operation and design of an MFC and their optimization including the effect on the output voltage of electrode material, effect of salt bridge diameter, effect of salt bridge solution, the effect of salt concentration has been studied separately and listed below.

3.1. Substrate and Sludge Quality

Influent wastewater to West Nablus WWTP is mostly domestic wastewater with few slaughters' houses wastewater and three sesame pastes and sweets factories. Average influent characteristics as reported by Nablus Municipality are: COD=990 mg/L, BOD=400 mg/L, TSS=410 mg/L, pH=7.8 and conductivity=1500 μ S/cm [53]. Samples were collected from the effluent of the primary sedimentation tank; to get rid of unnecessary solids.

COD for the anaerobic sludge and the used substrate was found to be 48,000 mg/L and 547 mg/L respectively. The volume of substrate used for each MFC is 800 mL, COD for the mixed sludge-wastewater was directed to be within 300-1700 mg/L.

3.2. Effect of Anode Electrode Material on the Output Voltage

The four different electrode materials used in MFCs with three duplicates were: Zinc sheets, Copper sheets, manufactured pretreated carbon electrodes, and Carbon brushes. In these experiments, all experimental conditions and variables were fixed except the electrode material. All used electrodes have the same

dimensions 7 cm X 4 cm (14.28 cm³ substrate/1 cm² electrode). Figure 4 includes the measurements of the output voltage for each electrode material during 7 days of operation.

As shown in Figure 4 and in terms of output voltage, Copper sheets electrode-MFC is the most efficient electrode, followed by Carbon brushes-MFC, then Zinc sheets-MFC, and finally manufactured Carbon electrodes-MFC. The decreasing trend of the output voltage with time was linear.

Statistical analysis is used to approve or to disclaim the initial conclusion. One-way ANOVA and post hoc tests are used using the seven days data applying IBM SPSS statistics 2.0 software.

One-way ANOVA was used and checked as follows:

- Continuous dependent variable: Output voltage data can be considered as a continuous data-interval continuous variable.
- The independent variable should consist of two or more groups: here have 4 groups (4 electrode materials).
- Independence of observations, which means that there is no relationship between the observations in each group or between the groups themselves: this condition is satisfied here.
- There should be no significant outliers: SPSS was used to check outliers and found that there are no outliers in the data.

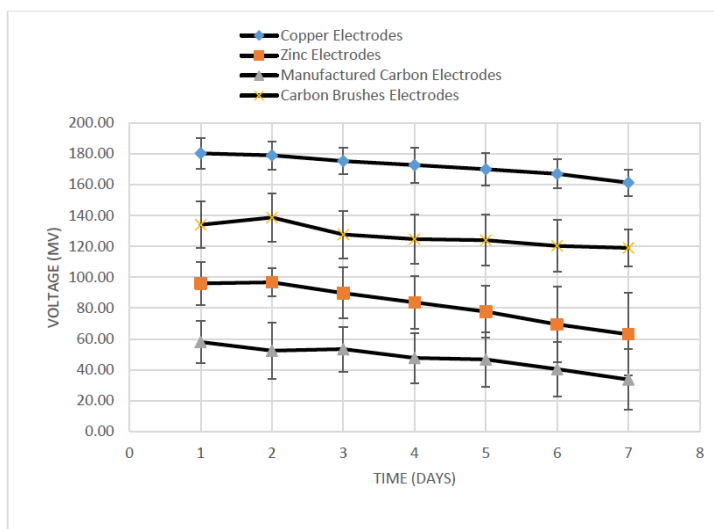


Figure 4: Output voltage in MFCs of different electrode materials.

- Dependent variable should be approximately normally distributed: Shapiro-Wilk test was used to check normality and it was found that all the distributions are approximately normal distributed (All sig. values were more than 0.05).
- Homogeneity of variances: Levine's test for homogeneity of variances was used in SPSS to check for homogeneity and it was found that this condition was satisfied

As a result, it was found that there is a significant difference between electrode materials used in terms of output voltage (significance less than 0.05).

To allocate which electrode materials have significant difference between them, Post Hoc tests was performed on SPSS. It can be noticed that Copper electrodes-MFCs are significantly more efficient than Carbon brushes-MFCs and Zinc electrodes-MFCs. Carbon brushes-MFCs are significantly more efficient than zinc electrodes-MFCs; whereas Zinc-MFCs are significantly more efficient than manufactured carbon-MFCs in the first two days only.

3.3. Effect of Salt Bridge Diameter on the Output Voltage

Three salt bridges diameters of, 10 mm, 16 mm, and 24 mm, were used in 3 different MFCs, three MFC-duplicates for each diameter. Accordingly, nine DS-MFCs were constructed, 1M-KCl salt was used, Copper electrodes were used (based on the results of anode electrode experiments). The area of each electrode was 28 cm² and the temperature was

maintained to 35 °C. In these experiments, all conditions were fixed except the salt bridge diameter; Primary effluent wastewater was used as substrate (790 mL) in addition to anaerobic sludge (10 mL). Dissolved oxygen in the water was used as a cathodic chamber and this experiment was running for one week. Figure 5 includes the three salt bridge results.

As shown in Figure 5, and based on statistical analysis performed, the best output voltage is achieved by the 10-mm salt bridge-MFC, followed by 16 mm and then the 24-mm salt bridge-MFC.

One Way ANOVA test was used and checked as follows:

- Continuous dependent variable: Output voltage data can be considered as a continuous data-interval continuous variable.
- The independent variable should consist of two or more groups: 3 groups (3 salt bridge diameters).
- Independence of observations, which means that there is no relationship between the observations in each group or between the groups themselves: this condition is satisfied here.
- There should be no significant outliers: SPSS was used to check outliers and found that there are no outliers in the data.
- Dependent variable should be approximately normally distributed: Shapiro-Wilk test was used

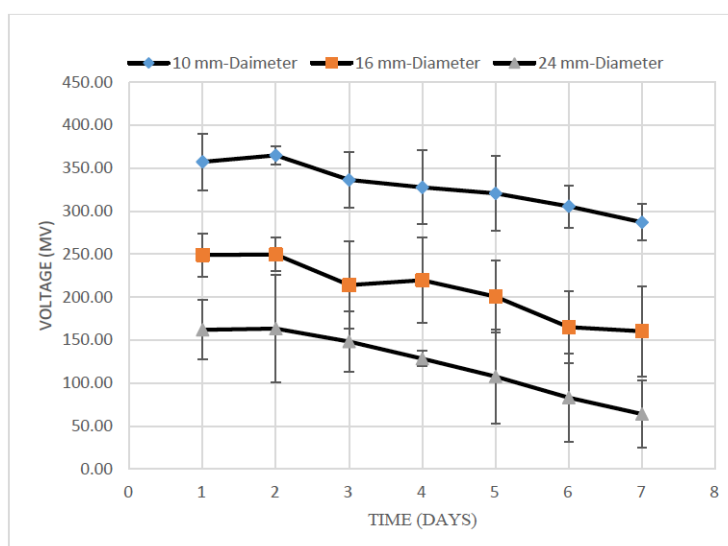


Figure 5: Output voltage of MFCs with different salt bridge diameters.

to check normality and it was found that all the distributions are approximately normal distributed (All sig. values were more than 0.05 obtained from SPSS output).

- Homogeneity of variances: Levine's test for homogeneity of variances was used in SPSS to check for homogeneity and it was found that this condition was satisfied.

As a result, it was found that there is a significant difference between salt bridges diameters used in terms of output voltage (significance less than 0.05).

To allocate which diameters have a significant difference between them, Post Hoc tests was performed on SPSS. It can be noticed that 10 mm salt bridges-MFCs are significantly more efficient than 16 mm salt bridges-MFCs; whereas 16 mm salt bridges-MFCs are significantly more efficient than 24 mm salt bridges-MFCs in the first one days only.

3.4. Effect of Salt Bridge Solution on the output Voltage

Two salt solutions were used in salt bridge preparation, KCl, and NaCl. All experimental conditions were fixed except the salt type. Six DS-MFCs were constructed, 1M-KCl and 1M-NaCl salts were used in 10 mm-salt bridges (based on the results of salt bridge experiments), Copper electrodes were used(based on the results of anode electrode experiments), area of each electrode was 28 cm², and the temperature was maintained at 35 °C. Primary effluent wastewater was used as substrate (790 mL) in addition to anaerobic sludge (10 mL). Dissolved oxygen in the water was used as a cathodic chamber and this experiment was

running for one week. Figure 6 includes the results of this experiment.

As shown in Figure 6 the best output voltage looks to be achieved by the KCl salt bridge-MFC.

One Way ANOVA test was used and checked as follows:

- Continuous dependent variable: Output voltage data can be considered as a continuous data-interval continuous variable.
- The independent variable should consist of two or more groups: 2 groups (2 salt types).
- Independence of observations, which means that there is no relationship between the observations in each group or between the groups themselves: this condition is satisfied here.
- There should be no significant outliers: SPSS was used to check outliers and found that there are no outliers in the data.
- Dependent variable should be approximately normally distributed: Shapiro-Wilk test was used to check normality and it was found that all the distributions are approximately normal distributed (All sig. values were more than 0.05).
- Homogeneity of variances: Levine's test for homogeneity of variances was used in SPSS to check for homogeneity and it was found that this condition was satisfied.

As a result, it was found that there is a significant difference between both salts and KCl is more efficient than NaCl.

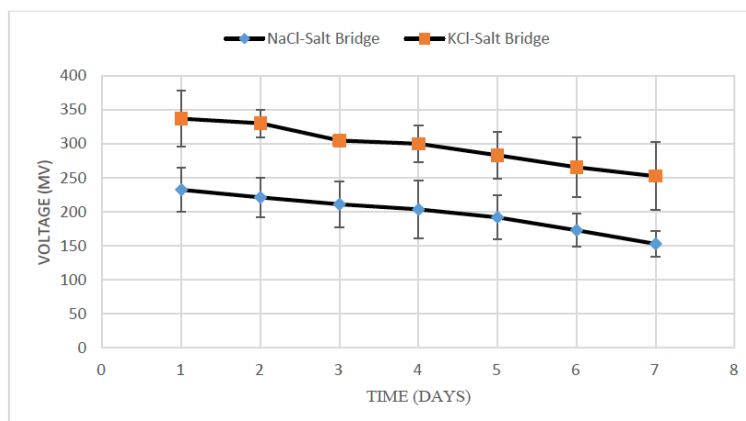


Figure 6: Output voltage for KCl and NaCl salt bridges.

3.5. Effect of Salt Concentration on the output Voltage

Three KCl salt concentrations, 1M, 2M, and 3M, were used in salt bridge preparation. All experimental conditions were fixed except the salt concentration in the salt bridges; output voltage was monitored for seven days. Nine DS-MFCs were constructed, 10 mm-salt bridges were used, Copper was used, the area of each electrode was 28 cm^2 , and the temperature was maintained to 35°C . Primary effluent wastewater was used as substrate (790 mL) in addition to anaerobic sludge (10 mL). Dissolved oxygen in the water was used as a cathodic chamber and this experiment was running for one week. Figure 7 illustrates the results of this experiment.

One Way ANOVA test was used and checked as follows:

- Continuous dependent variable: Output voltage data can be considered as a continuous data-interval continuous variable.
- The independent variable should consist of two or more groups: 3 groups (3 concentrations).
- Independence of observations, which means that there is no relationship between the observations in each group or between the groups themselves: this condition is satisfied here.
- There should be no significant outliers: SPSS was used to check outliers and found that there are no outliers in the data.
- Dependent variable should be approximately normally distributed: Shapiro-Wilk test was used to check normality and it was found that all the

distributions are approximately normal distributed (All sig. values were more than 0.05).

- Homogeneity of variances: Levine's test for homogeneity of variances was used in SPSS to check for homogeneity and it was found that this condition was satisfied.
- As a result, One Way ANOVA can be used here to compare between the 3 groups; 1M, 2M, and 3M KCl salt bridges-MFCs. Using ANOVA in SPSS it was found that there is a significant difference between salt concentrations used for the salt bridges in terms of output voltage (significance less than 0.05).

To allocate which groups have a significant difference between them, Post Hoc tests were performed on SPSS. It can be noticed that 1M KCl salt bridges-MFCs are significantly more efficient than 3M KCl salt bridges-MFCs. 1M KCl salt bridges-MFCs are significantly more efficient than 2M KCl salt bridges-MFCs in the first three days only; the same thing as in a comparison between 2M KCl salt bridges-MFCs and 3M KCl salt bridges-MFCs.

As shown in Figure 7 and based on the performed statistical analysis, the best output voltage is achieved by the KCl salt bridge-MFC.

It can be observed that, in general, the output voltage is decreasing with time; this can be justified by the decreasing of nutrients with time in the substrate. Variations in output voltage between duplicates for the same electrode material can be attributed to many causes: variation in substrate constituents, variation in microorganism's cultures, and variation in salt bridges efficiency, and variation in electrode position in the anode.

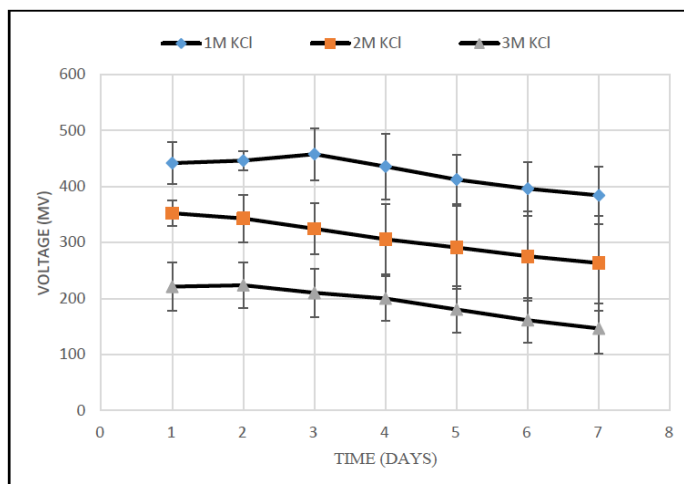


Figure 7: Output voltage for KCl and NaCl salt bridges.

4. CONCLUDING REMARKS

Based on the results obtained in this research work, the following concluding remarks were observed:

- The design and operation of MFCs is not a straightforward process and is determined by several interrelated parameters.
- An optimized design of MFCs could be achieved by optimizing its operating parameters, *i.e.*, using a copper electrode, a smaller size salt bridge made of potassium chloride, and a smaller salt concentration filled in the salt bridges.
- Out of the four-electrode material applied in MFCs, the copper electrode was the best followed by carbon brushes.
- The smaller the salt bridge diameter used in the MFC, the best output voltage achieved.
- Salt bridge made of potassium chloride salt used in MFC, proved to be more efficient than that made of sodium chloride salt in output voltage.
- The smaller the salt concentration filled in the salt bridges that are used in MFC the better output voltage achieved.
- Fixing of optimally obtained operating parameters and varying COD concentration of the substrate revealed that a relationship exists between the output voltage and COD value in MFC.

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