

Different Types of Microbial Fuel Cell (MFC) Systems for Simultaneous Electricity Generation and Pollutant Removal

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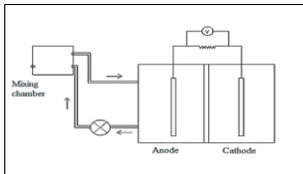
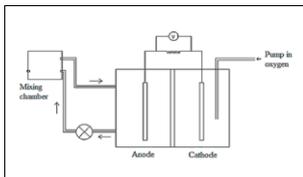
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Graphical abstract



Abstract

The aim of this study is to identify the relationship between the rate of electricity generation and the rate of carbon and nitrogen removal from wastewater using different MFC processes. Determining whether the generation of electricity using MFC process could be related to the rate of pollutant removal from wastewater is noteworthy. Three types of MFC process configurations include the batch mode (SS), a continuous flow of influent with ferricyanide (PF) as the oxidizing agent and a continuous flow of influent with oxygen (PU) as the oxidizing agent. The highest quantity of electricity generation was achieved using the continuous flow mode with ferricyanide (0.833 V), followed by the continuous flow mode with oxygen (0.589 V) and the batch mode (0.352 V). The highest efficiency of carbon removal is also achieved by the continuous flow mode with ferricyanide (87%), followed by the continuous flow mode with oxygen (51%) and the batch mode (46%). Moreover, the continuous flow mode with ferricyanide produced the highest efficiency for nitrogen removal (63%), followed by the continuous flow mode with oxygen (54%) and the batch mode (27%).

Keywords: Microbial fuel cell (MFC); electricity; pollutants; carbon; nitrogen

Abstrak

Tujuan kajian ini adalah untuk menentukan hubungan di antara kadar penjanaan elektrik dengan kadar penyingkiran karbon dan nitrogen daripada air sisa dengan menggunakan proses Sel Bahan Api Mikro (SBAM) yang berbeza. Perkara ini akan menjadi sangat menarik untuk mengetahui sama ada penjanaan tenaga elektrik yang menggunakan proses SBAM dapat dikaitkan dengan penyingkiran bahan pencemar daripada air sisa. Tiga jenis tetapan proses SBAM yang mana termasuk mod sesekumpul (SS), influen dengan ferisianida aliran berterusan (PF) sebagai ejen pengoksidaan dan influen dengan oksigen aliran berterusan (PU) sebagai ejen pengoksidaan. Kuantiti tertinggi penjanaan elektrik voltan yang dicapai oleh mod aliran berterusan dengan ferisianida ialah 0.833 V, diikuti oleh mod aliran berterusan dengan oksigen 0.589 V dan mod sesekumpul 0.352 V. Keberkesanan penyingkiran karbon yang tertinggi juga dicapai oleh mod aliran berterusan dengan ferisianida iaitu 87%, diikuti oleh mod aliran berterusan dengan oksigen 51% dan mod sesekumpul 46%. Selain itu, mod aliran berterusan dengan ferisianida juga telah ditunjukkan sebagai sistem yang telah menjana keberkesanan penyingkiran nitrogen yang tertinggi (63%) diikuti oleh mod aliran berterusan dengan oksigen (54%) dan mod sesekumpul (27%).

Kata kunci: Sel Bahan Api Mikro (SBAM); elektrik; bahan pencemar; karbon; nitrogen

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1.0 INTRODUCTION

The energy crisis, limitation of resources and environmental issues has prompted studies to discover diverse sources of alternative energy. One solution is the use of microbial fuel cells (MFC) in wastewater treatment processes to generate electricity generation and simultaneously remove pollutants. Wastewaters containing various sources of organic matter can be used as a substrate to generate electricity. The electricity generated will be used for the overall operation of the wastewater treatment plant.

This process potentially reduces the cost of treatment plant operation, notably for an activated sludge treatment plant that requires a high energy input for the aeration, sludge treatment and pumping processes.

Microbial fuel cells (MFCs) are an emerging technology. MFCs can generate bio-energy from organic matter and simultaneously treat the wastewater (1-5). The principle of the operation of MFCs refers to the fact that the organisms naturally generate electricity because they transfer electrons from an electron donor to an electron acceptor at a higher electrochemical

potential. The biological process performed by the microorganisms in the wastewater at the anodic compartment produces electrons and transfers the electrons to the anodic electrode. The electrons will then be transferred to the cathode by an external circuit that connects the two electrodes in the MFC. A current is generated when the electron is reduced by the electron acceptor in the cathode chamber, typically oxygen. Currently, ferricyanide is being introduced as the electron acceptor, enhancing the bio-energy generation in terms of the electrical voltage. In this study, the use of ferricyanide is introduced into the design, and the results are compared with a design that employs oxygen as the electron acceptor.

Carbon removal from wastewater is crucial because carbon compounds are the main energy source for the microorganisms. Therefore, decreasing the carbon contained in the wastewater reduces the concentration of microorganisms in the wastewater. Additionally, nitrogen removal from wastewater is an important component of treatment because high nitrogen contents in the wastewater can cause a bad smell. Nitrogen removal has been difficult to achieve in single-process systems; therefore, multiple reactors are recommended (6-7). Two stage processes and modified anoxic processes using sequencing batch reactors can be used to treat ammonia-rich wastewater, but these processes incur high operational costs (8). Whereas MFCs have been recently shown to generate power and treat wastewater, COD removal was observed to be accompanied by a high level of ammonia removal (9). Therefore, the relationship between the electricity generation of a MFC with the rate of nitrogen and carbon removal is being studied. The rate of bio-energy generation is highly affected by the biological activities of the microorganisms in the anodic chamber. In addition, the factors affecting the electron generation by the microorganism are being studied. Several other physical factors of the MFC design can also affect the rate of bio-energy generation such as the size of the electrode being used, the mode of the anodic process, the distance between the two electrodes and the components of the electrolyte in the cathodic chamber.

MFCs represent the latest study in alternative and renewable energy by generating electricity from wastewater using bacteria. The first observation of electrical current generated by bacteria was initiated by M. C. Potter (10), but few practical advances have been achieved in this field in the subsequent 55 years (11). A breakthrough for MFC applications occurred in 1999 when the process was found to not require mediators (12). Microorganisms oxidize organic matter in the MFC, producing electrons that travel through a series of respiratory enzymes in the cell to produce energy for the cell in the form of ATP (adenosine triphosphate). The electrons are then released to a terminal electron acceptor (TEA) which accepts the electrons and becomes reduced. However, certain bacteria can transfer electrons exogenously or out of the cell to a TEA such as a metal oxide like iron oxide. These bacteria that can exogenously transfer electrons, called exoelectrogens, can be used to produce power in a MFC. The mixed community in the activated sludge was shown to be the best community of bacteria for pollutant removal whereas simultaneously producing electricity in a MFC cell (13-14).

In 2004, electricity generation using a MFC was linked to wastewater treatment in a demonstration treating practical levels of domestic wastewater (2). In this current study, the efficiency of the electricity generation between the anodic batch mode and the anodic continuous mode is to be verified. The design displaying a higher electricity generation will then be modified with different oxidizing agents, i.e., ferricyanide, to enhance the generation of electricity. The results of the three designs are evaluated and compared for the generation of electricity and the removal of nitrogen compounds and carbon.

■2.0 EXPERIMENTAL

2.1 Wastewater Sampling

Domestic wastewater was obtained from the Indah Water Consortium (IWK) at Taman Mawar, Puchong. This wastewater is a raw sample without physical filtering or biological treatment. A combination of substrates was added as the nutrition supply for the microorganism in the wastewater. The trace element nutrition mix contained glucose 1 g/l, ammonium chloride 0.5 g/l, magnesium (II) chloride 0.3 g/l, disodium phosphate 0.87 g/l, sodium phosphate 0.68 g/l, cobalt (II) chloride 0.025 mg/l, manganese (II) chloride tetrahydrate 15 mg/l, zinc chloride dehydrate 11.5 mg/l, calcium chloride dehydrate 0.005 mg/l and manganese chloride 15 mg/l. The pH of the substrate is adjusted to a pH of 7 using an acidic solution and an alkaline solution during the operation of the MFC.

2.2 MFC Configurations and Operation

Three types of MFCs were designed to evaluate the efficiency of the electricity generation and the removal of nitrogen and carbon in wastewater: the batch mode (SS), anodic continuous mode (PU) and anodic continuous mode with ferricyanide (PF). The anodic electrolyte for the three designs was 180 ml of wastewater, 500 ml of the glucose substrate, 10 ml of the mineral mixture and 10 ml of the vitamin mixture. A carbon cloth (25 mm x 60 mm) was used as the electrode for the anode and cathode in all designs. The cloth was pre-treated with 1N hydrochloric acid (HCl) and 1N of sodium hydroxide (NaOH) to remove impurities (15). A PEM (proton exchange membrane), Nafion 117, was placed in the middle of the MFC, dividing the anodic compartment from the cathodic compartment. The membrane was soaked in 3 vol% hydrogen peroxide (H₂O₂) for 1 hour at 80°C and rinsed with distilled water. Then, the membrane was soaked in 0.5 M sulfuric acid (H₂SO₄) at 80°C for 1.5 hours before being rinsed with distilled water (16). These pre-treatment steps rid the membrane of impurities which may affect the results of the MFC. The cathode and anode was connected to a copper electrical wire by crocodile clips. The wire was 2 m in length, and a computerized digital voltmeter was connected to the wire in series to record the voltage passing through the wire. An epoxy material was used to seal the gap between the cover and the container of the anodic compartment to ensure that no air can enter or be released from the container, creating an anaerobic condition over the 10-day-operation.

For the SS system, both the anodic and cathodic compartments were tightly sealed, and no addition or removal of solution from the system occurred throughout the 10-day-operation. In total, 700 ml of phosphate buffer solution with a pH of 7 was used as the electrolyte for the cathode compartment. Only one sample was collected after the 10-day-operation. This sample was then compared with the composition of the wastewater before undergoing the MFC operation.

For the PU system, identical to the SS system, both the anodic and cathodic compartments were tightly sealed, but oxygen was continuously supplied to the cathodic electrolyte using an aquarium pump. Piping connected a pump to the mixing chamber at the anodic compartment as shown in Figure 1. The mixing chamber is a fabricated container used to control the regular flow of the solution being pumped out from the anodic compartment, and the flow was directed back into the anodic compartment. This anodic continuous flow condition can enhance the growth of microorganisms by improving the homogeneous mixing of the substrate with the wastewater. Therefore, each microorganism efficiently receives nutrients, enhancing the rate of

electrons being generated from the exoelectrogens and improving the electrical generation and the removal rate of nitrogen and carbon. The mixing chamber was pre-loaded with 500 ml of wastewater to monitor the continuous flow of the anodic electrolyte. In total, 200 ml of sample was withdrawn from the anodic compartment every 48 hours during the 10-day-operation. Therefore, 5 samples were collected during the operation. In total, 200 ml of the 1 g/l glucose substrate was added to the anodic compartment after each removal of 200 ml from the system.

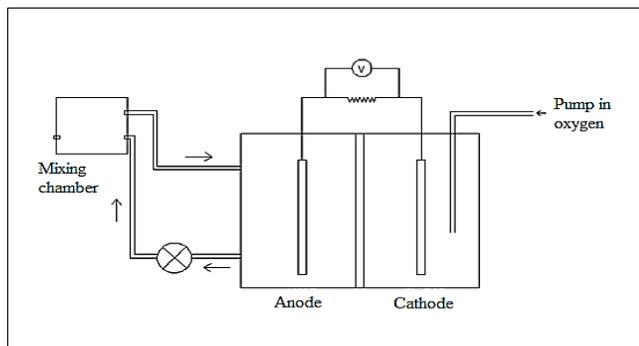


Figure 1 The arrangement and apparatus of the PU system

For the PF system, the arrangement and the material used for the anodic compartment of the system was similar to the PU system. A pump was not connected to the cathodic compartment to provide oxygen because ferricyanide was added to the electrolyte in the cathodic compartment. The ferricyanide acted as the oxidizing agent that received electrons instead of oxygen. In total, 50 mM of ferricyanide was to be added to the 700 ml phosphate buffer solution in the cathodic compartment at a pH of 7. Similarly, 200 ml of sample was withdrawn from the anodic compartment every 48 hours during the 10-day-operation. Therefore, 5 samples were collected. In total, 200 ml of the 1 g/l glucose substrate was added to the anodic compartment after each removal of 200 ml of sample from the system. Figure 2 shows the arrangement of the PF system design.

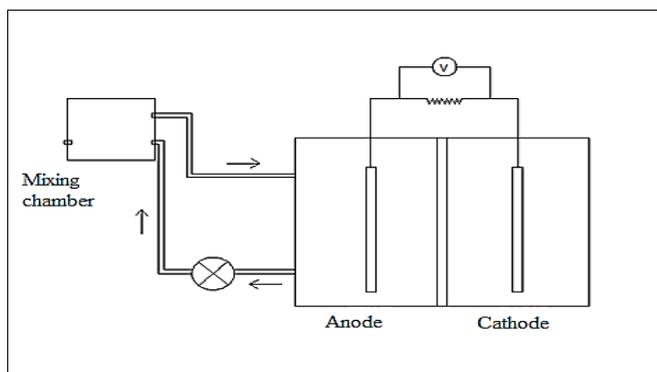


Figure 2 The arrangement and apparatus of the PF system

An analysis was conducted for COD (APHA 5220B), ammonia nitrogen, organic nitrogen and nitrate (APHA 4500C). The voltage generated was recorded by a computerized digital voltmeter as OCV (open circuit voltage) over the 10 days of operation. Meanwhile, a polarization was performed during the middle of operation to evaluate the voltage generated with different values of resistance. Resistors with various values of

resistance were used to evaluate the voltage differences: 10 Ω , 51 Ω , 100 Ω , 510 Ω , 1000 Ω , 2000 Ω , 3300 Ω , 5100 Ω , 10000 Ω and 20000 Ω . The digital voltmeter was connected with the circuit in parallel during the polarization to record the voltage across the resistor. The results of the voltage collected during the polarization with resistance and the values of the collected OCV were then analyzed.

3.0 RESULTS AND DISCUSSION

3.1 Voltage Generation

Based on the results recorded in other studies, the rate of the voltage generation is mainly affected by several factors:

- The rate of electron generation from the exoelectrogens which were grown on the electrode in a biofilm (17).
- The number of electrons that were transferred to the cathode from the anode (18).
- The efficiency of the oxidizing agent in accepting electrons transferred from the anode (19).
- The loss of electrons transferred from the diffusion of oxygen from the cathode to anode (20, 21).

The shape of the current-voltage curve is mainly determined by the overpotential in the electro-chemical cell: the activation overpotential, the ohmic overpotential and the concentration overpotential (22). After 240 hours of operation in the three systems, the voltage generated throughout the MFC operation was recorded as OCV and arranged accordingly for the comparisons (Figure 3).

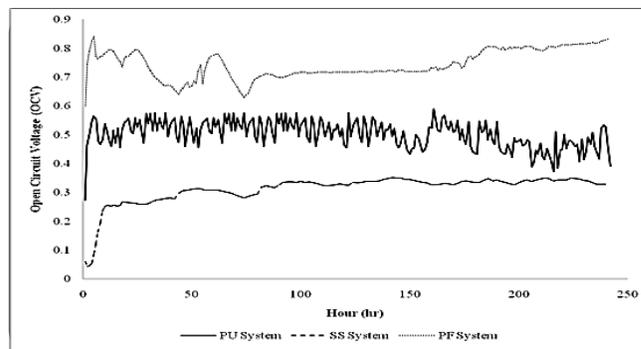


Figure 3 Open circuit voltage (OCV) recorded for the SS, PU and PF systems

Generally, select similarities are noted in the plots of the three systems. The graphs started with a sharp increase in voltage, related to the initiation of the MFC operation. This increase is called the 'biofilm formation phase' and occurred within the first 24 hours. This is the time required for the microorganism to form the biofilm on the surface of the anode. However, Lim *et al.* (20), Jiang *et al.* (23), and Rodrigo *et al.* (21) mentioned that the time for the biofilm to completely adapt to the surrounding was approximately 10 days to produce electrons. The voltage increased as the total number of microorganisms forming the biofilm on the surface increased. This process ended when the surface of the electrode was fully covered by the biofilm and no further increase in the voltage was noted. Comparing the three plots, the time required for the SS system to complete this phase was longer compared to the other two systems. This longer time

results from the anodic continuous flow process improving the activity of the microorganisms in the wastewater. After the formation of the biofilm on the electrode, the microorganisms compete with each other for nutrients in the substrate. This phase is called the ‘survival phase’. This phase stopped when the voltage generally increased, which was approximately 180 to 200 hours into the operation. The graph displayed fluctuations in this period because the microorganisms were competing for survival; only the surviving microorganisms can produce electrons used in the voltage generation. A more uniform value in the voltage then appears when a portion of the microorganisms were dead and a lower competition for the nutrient supply is noted. The surviving microorganisms have a more constant supply because of the competition, and the voltage generated was stable. Therefore, this phase is called the ‘stable phase’, occurring between 200 to 240 hours of MFC operation.

From the graph, the variation in voltage initiates from the beginning of operation for the three systems. The highest voltage was generated by the PF system, ranging from 0.6 to 0.8 V. For the PU system (which used oxygen as the oxidizing agent in the MFC operation), the voltage generated ranged from 0.28 to 0.578 V. The lowest range in the voltage generated was for the SS system (from 0.06 to 0.35 V). This shows that the PU and PF systems generated higher voltages than the SS system. Therefore, the anodic continuous flow process was effective in enhancing the voltage generation by mixing the substrate with the wastewater in the compartment. The rate of electron generation increased in the continuous flow because the percentage of microorganisms receiving sufficient nutrients from the electrolyte was higher, resulting in a higher efficiency of voltage generation from the continuous anodic flow mode compared to the batch mode.

Additionally, the voltage generated for the two anodic continuous flow mode PU and PF systems were compared. We found that the PF system with ferricyanide as the oxidizing agent in the cathodic compartment generated higher voltages (almost 50 % higher than the values generated by the PU system with oxygen as the oxidizing agent). This result shows that ferricyanide is a more suitable oxidizing agent, improving the voltage generation in MFC operation.

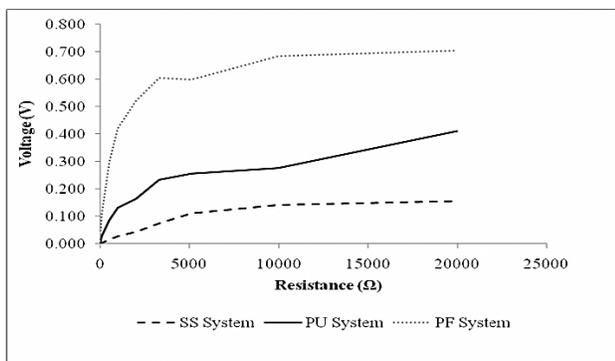


Figure 4 Voltage across different resistances for the SS, PU and PF systems

Figure 4 demonstrates the voltage generated during the polarization process by using resistors with different resistances in the three systems. Generally, the voltage generated increased as the resistance increased. However, the highest range of voltages was generated by the PF system, followed by the PU system and eventually the SS system. The efficiency of the voltage generation also improved in the anodic continuous flow process in a MFC,

and the use of ferricyanide as the oxidizing agent enhanced the voltage generated in the anodic continuous flow system.

3.2 The Analysis of COD

The carbon content in the wastewater is determined by the COD value in this study. The COD value of the wastewater sample before the MFC operation was 2028 mgO₂/l. The values of the COD after the MFC operation for the SS, PU, and PF systems are 1092, 988 and 270 mgO₂/l, respectively.

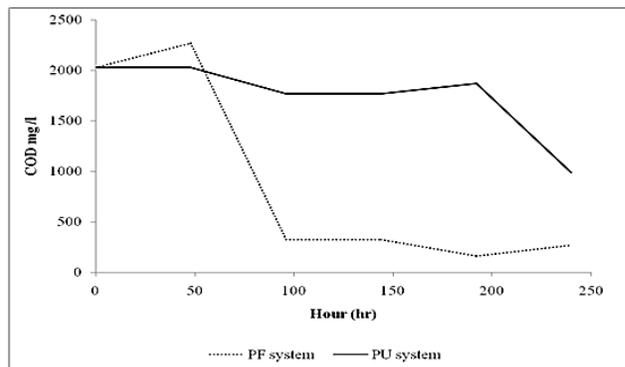


Figure 5 Results of the COD tests for the PU and PF systems

Figure 5 shows that the COD decreases throughout the 240-hour operation of the PU and PF systems. The graphs shown for the two systems are different in shape. For the PF system, the decreasing rate is the highest between the 48-hour to 96-hour in which the reduction of COD is more than 1500 mgO₂/l. The COD kept decreasing until 192 hours and displayed a slight increase during the last 48 hours of operation. The increase in COD is likely from a small amount of oxygen entering the system when the cover of the container was opened to collect the samples. In the PU system, the COD values decreased more uniformly throughout the 240 hours of operation when compared to the PF system.

Table 1 Results of the COD tests for the SS, PU and PF systems

MFC system	COD value before MFC operation (mg O ₂ /l)	COD value after MFC operation (mg O ₂ /l)	Efficiency in the reduction of COD (%)
SS	2028	1092	46.15
PU	2028	988	51.28
PF	2028	270	86.69

Table 1 shows that the efficiency of COD removal for the three systems. The most effective system is the PF system (86.69%), followed by the PU system (51.28%) and the SS system (46.15%). Based on these results, the anodic continuous flow process increased the rate of carbon removal from the wastewater as shown in the PU and PF systems. Moreover, the use of ferricyanide as the oxidizing agent improved the removal rate by 35.41 % for the PF system when compared to the PU system (24).

Generally, the use of ferricyanide as the oxidizing agent in the anodic continuous flow system (PF) simultaneously improves the voltage generation and the rate of COD removal (25) (Figure 6). The rate of voltage generation increased as the rate of COD removal increased throughout the 240 hours of operation.

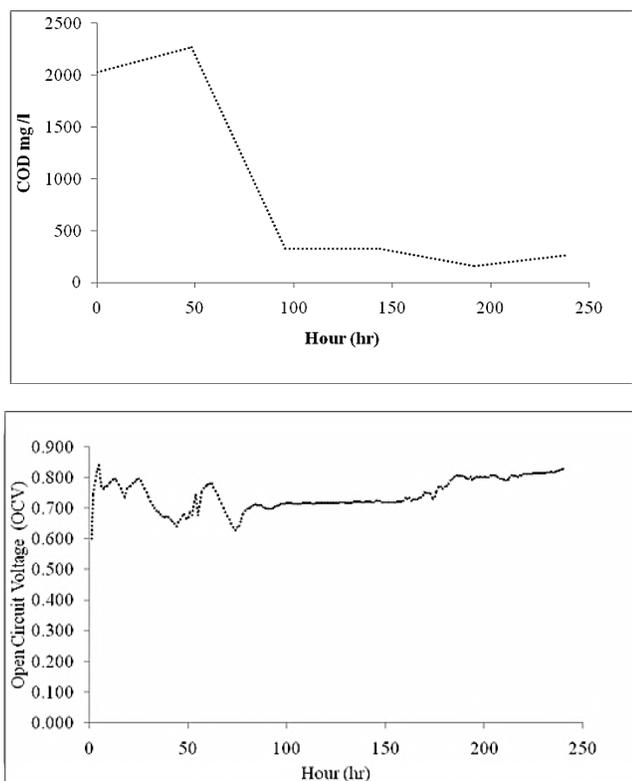


Figure 6 Comparison of the results of the COD tests to the OCV for the PF system

3.3 The Analysis of Ammonia Nitrogen and Organic Nitrogen

The compositions of nitrogen before the MFC operation were 4.48 mg/l ammonia nitrogen, 1.68 mg/l organic nitrogen and a TKN of 6.16 mg/l. The percentage of ammonia nitrogen removed after the 240-hr operation of the SS system is 49.1%; no change was noted in the organic nitrogen value after the operation. The TKN value decreased by 27.27%. For the PU system, the percentage of the ammonia removed after the 240-hr operation is 50%, the percentage of organic nitrogen removed is 66.67% and the TKN value decreased by 54.55%. These results show that the amount of nitrogen removed is double the removal rate of the SS system, likely because the anodic continuous flow in the PU system enhanced the activity of the microorganisms when decomposing the nutrients in the wastewater. Table 2 shows that the rate of the nitrogen components decreased during the entire 240-hr operation of the PU system.

Table 2 Amounts of nitrogen removed by the PU system during the 240-hr MFC operation

Hour (hr)	Ammonia Nitrogen, N-NH ₃ (mg/l)	Organic Nitrogen (mg/l)	TKN (mg/l)
0	4.48	1.68	6.16
48	4.48	1.12	5.6
96	3.92	1.12	5.04
144	3.92	0.56	4.48
192	2.8	0.56	3.36
240	2.24	0.56	2.8

The percentage of nitrogen components removed is higher for the PF system (50% of the ammonia nitrogen is removed; 100% of the organic nitrogen is removed; and the TKN is reduced by 63.63%). The removal rate is higher than the SS and PU systems, likely because the activity of the microorganisms was enhanced in the anodic continuous flow system by using ferricyanide as the oxidizing agent (5). Based on the values shown in Table 3, the rate of reduction is uniform throughout the operation.

Table 3 Amounts of nitrogen removed in the PF system during 240 hr of MFC operation

Hour (hr)	Ammonia Nitrogen, N-NH ₃ (mg/l)	Organic Nitrogen (mg/l)	TKN (mg/l)
0	4.48	1.68	6.16
48	3.92	1.12	5.04
96	2.8	0.56	3.36
144	2.24	0.28	2.52
192	2.24	0.168	2.41
240	2.24	0	2.24

Based on the results collected, the anodic continuous flow system using the ferricyanide as the oxidizing agent improved the rate of nitrogen removal from wastewater and the rate of voltage generation (Figure 7).

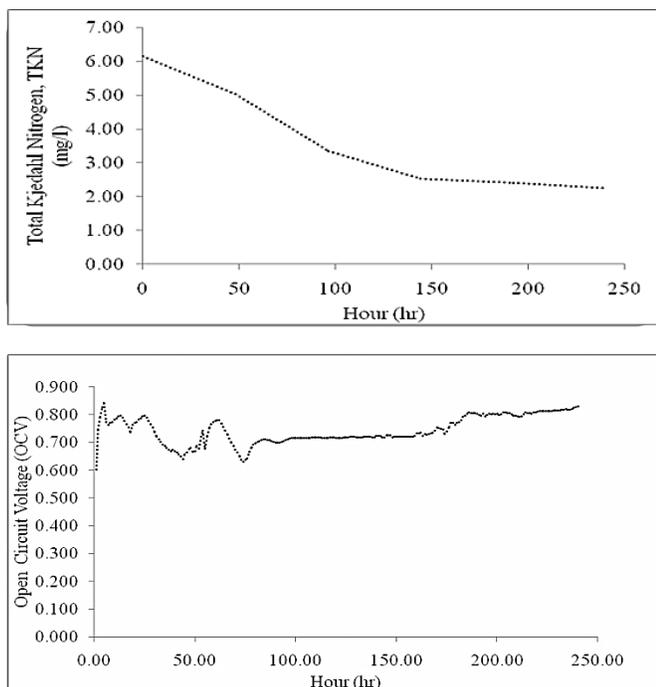


Figure 7 Comparison of the results of the TKN tests to the OCV for the PF system

3.4 The Analysis of Nitrate Nitrogen

The value of nitrate contained in the wastewater before undergoing the MFC operation was 28 mg/l. For the SS system, the reduction in the nitrate was approximately 5 mg/l or 17.85% after 240 hours of operation. For the PU system, the percentage of nitrate reduction is 58.96% after 240 hours of operation. The percentage of nitrate reduction for the PF system is 63.75%, which is the highest among the three systems. The difference in the nitrate value between the two continuous systems may be because of the oxygen supply in the PU system. Nitrite molecules are easily oxidized to nitrate molecules in aerated conditions. Figure 8 shows the slight differences between the PU and PF systems for the nitrate removal during the operation.

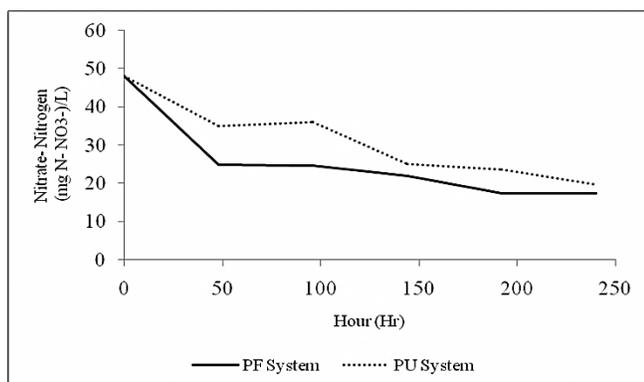


Figure 8 Comparison of the results of the nitrate tests for the PU and PF systems

3.5 pH Analysis

Overall, the pH did not change substantially in the anodic electrolyte after the MFC operation in all three systems; the anodic electrolyte consists of the wastewater and substrate. The pH of the sample was 6.88 before undergoing the MFC operation. The pH of the electrolyte dropped to 6.76 (or by 1.74%) in the SS system. For the PU system, the pH of the anodic electrolyte dropped by 1.88%, and PF system shows the highest drop of 5.81% in terms of pH after 240 hours of operation. The acidity of the anodic electrolyte increased because of the denitrification of nitrogen compounds in the wastewater (26). The maximum power generation could occur when a biofilm grows on both the cathode and anode electrode; this process might increase the pH to up to 9 (27).

4.0 CONCLUSIONS

Based on the results found in the study, the anodic continuous system could produce a higher voltage generation than the batch mode MFC design. The rate of carbon and nitrogen removal from the wastewater in the anodic continuous flow systems is higher than that in the batch mode. For the two anodic continuous flow systems, the PF system shows a rate of voltage generation higher than the PU system. The highest voltage generated by the PF system was 0.833 V, whereas the highest voltage was 0.589 V for the PU system. Moreover, higher rates of simultaneous carbon and nitrogen removal increased the power generation. Therefore, we conclude that using ferricyanide as the oxidizing agent is more efficient for the generation of voltage and also for the removal of nitrogen and carbon when compared to the design using oxygen as the oxidizing agent.

Acknowledgements

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