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Research Article

Degradation of Metal Ions with Electricity Generation by Using Fruit Waste as an Organic Substrate in the Microbial Fuel Cell

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A potential and developing green technology for producing renewable energy and treating wastewater is the microbial fuel cell (MFC). Despite several advancements, there are still several serious problems with this approach. In the present work, we addressed the problem of the organic substrate in MFC, which is necessary for the degradation of metal ions in conjunction with the production of energy. The utilization of fruit waste as a carbon source was strongly suggested in earlier research. Hence, the mango peel was used as a substrate in the current study. Within 25 days of operation, a 102-mV voltage was achieved in 13 days, while the degradation efficiency of Cr^{3+} was 69.21%, Co^{2+} was 72%, and Ni^{2+} was 70.11%. The procedure is carried out in the batch mode, and there is no continuous feeding of the organic substrate. In addition, a detailed explanation of the hypothesized mechanism for this investigation is provided, which focuses on the process of metal ion degradation. Lastly, future and concluding remarks are also enclosed.

1. Introduction

Heavy metals endanger the environment. Heavy metals' extremely toxic and nonbiodegradable qualities affect human health and ecosystems, particularly because they are used in industries and produce harmful effluent [1, 2]. When these effluents bioaccumulate in the food chain, humans will be exposed to the toxic materials, and subsequently, the effects can be grave [3, 4]. The discharge of these metals through contaminated effluent into bodies of water causes water pollution. Thus, treatment of heavy metal-based wastewater is required [5]. However, the technology of wastewater treatment also needs to overcome certain issues. Conventional methods of remediating heavy metals contained in waste require high operating costs and are not environment friendly [6–8]. As clean water is becoming an increasingly crucial need of today, the development of new approaches to wastewater treatment that are both sustainable and economical is receiving some attention [9]. A technique that is displaying promising results in recovering heavy metals from wastewater and generating energy simultaneously is called a microbial fuel cell (MFC) [10, 11]. The MFC technology could overcome the weaknesses of conventional treatment methods owing to its requirement for a low operating cost and eco-friendly nature [12]. An MFC is a technique that can convert the chemical energy of basic organic compounds such as sodium acetate and glucose into electrical energy while removing pollutants at the same time [13]. The simplest MFC has anode and cathode compartments, normally separated by a membrane for proton exchange to limit the electrolyte movement [14]. The bacteria in the anode chamber assist the activity of electron transportation to an electrode in the organic , and this is paired with the reduction of oxygen at the cathodic chamber, resulting in power production [15, 16]. The presence of waste-based materials from plants aids in the formation of electrons and protons in MFC. However, there is an issue regarding the poor performance of the organic substrate used in the fuel cell towards the microbial community because of its instability [17]. This problem is a major setback for the system and thus requires further attention to provide an efficient organic-based waste that can facilitate the efficient supply of energy for the bacteria present in cells for the process of electrogenesis. The greatest value of a MFC system is the ability of the microbes associated with the electrode to degrade wastes and harmful chemicals [18]. In this study, the plant-based waste used for the MFC system is collected from the mango (*Mangifera indica*) fruit, specifically its peels. Based on the previous research by Ajila et al. [19], the content of carbohydrates in the mango peel ranged from 20.8% to 28.2%. The percentage of the fiber substance in the peel is in the range of 3.28% to 7.40%, with a higher amount found in ripe peels. The peel has a moisture content of 66% to 75%, a protein content of 1.76% to 2.05%, and an ash content ranging from 1.16% to 3.0% [20]. The mango peel has the potential to be a beneficial organic waste with a high nutrient content. This study utilizes mango peels as plant-based organic waste in the MFC system in domestic wastewater (with Cr^{3+}, Co^{2+}, and Ni^{2+}). The efficiency of organic waste in facilitating the recovery of heavy metals and the simultaneous generation of energy using MFC.

2. Experiment Details

2.1. Materials and Reagents. The mango peel waste (obtained from a local fruit market), tape water, chromium nitrate nonahydrate, cobalt nitrate hexahydrate, nickel nitrate hexahydrate (all chemicals were purchased from Sigma–Aldrich), and distilled water (DI) was used in this research.

2.2. Microbial Fuel Cell Assembly and Operation. Tap water was received, and 100 ppm of chromium, cobalt, and nickel ions were introduced into the wastewater. After the supplementation, the tap water is now recognized as synthetic water for this study. The physicochemical properties of both the tap water and the synthetic water are summarized in Table 1. The instruments utilized for the analysis of the temperature, pH, and conductivity of the tap and synthetic waters were a thermometer, a pH meter, and an electrical meter, respectively. The mango peels were obtained from a nearby regional fruit marketplace and were then cut into small pieces after washing. A 500-mL mixture of synthetic water and 0.5 kg of mango peel waste was placed in the single-chamber MFC. The chamber volume used was 600 mL. This study used one electrode to act as the cathode, with a height of 10 cm and a radius of 1 cm. Two electrodes with a dimension of 10 cm × 1 cm (h × r) each to act as multiple anodes. The calculated surface area of the electrode was 69.13 cm^2. The distance between the anode and cathode electrodes was 12 cm. The electrodes were linked to each other using copper wire to help in electron transportation from anode to cathode, and 500 Ω of external resistance was provided after 3 days of open-circuit voltage. A decrease in the voltage value was observed after external resistance was implemented, but it slowly recovered. A large resistance should be used when there is an absence of voltage recovery, whereas smaller values of external resistance should be chosen when the value of the voltage shows no significant change [21]. For this work, the MFC was operated at room temperature for 25 days.

Table 1: Measured characteristics of the tap and synthetic water.

Parameters	Tap water	Synthetic water
pH	6.97	6.20
Odour	Unpleasant smell	Unpleasant smell
Electrical conductivity	110 μs	459 μs
Temperature	Room	Room
Cr^{3+}	0 ppm	100 ppm
Co^{2+}	0 ppm	100 ppm
Ni^{2+}	0 ppm	100 ppm

2.3. Electrochemical Test. Every day, the voltage of the cell was assessed using a multimeter. The efficiency of the current was measured through calculation of power density (PD) and current density (CD). Calculations of CD, PD, and internal resistance were carried out using the following equations [22]:

$$V = IR, \tag{1}$$

$$PD = \frac{V^2}{RA}, \tag{2}$$

$$CD = \frac{1}{A}, \tag{3}$$

$$r = \left(\frac{E - V}{V}\right)R, \tag{4}$$

where r denotes internal resistance, I denotes current, A denotes surface area, V denotes voltage output, R denotes external resistance, and E denotes electromotive force (emf). The emf value was obtained from the calculation through the measurement of the voltage for the open circuit by connecting with a voltmeter in the presence of resistance [23]. The redox reaction was examined using cyclic voltammetry. For every 10 days of the reaction, CV was recorded using a potentiostat, and the scan rate used was 10 mV/s with potential ranging from +0.8 V to −0.8 V. By using the "Rext-variation" scheme, the polarization behavior of the setup was studied, where a variable resistance box of 5000-100 Ω was applied to the external resistance control. Polarization curve analysis was carried out after the pseudosteady state of the operation. Approximately 30 min of variation time was required to observe the polarization behavior of the system.

The impact of anode resistance on voltage was additionally examined using electrochemical impedance spectroscopy (EIS). The frequency used ranged from 100 kHz to 100 mHz.

2.4. Metal Degradation and the Biofilm Study. The atomic absorption spectrometer (AAS) instrument is used to investigate metal recovery from wastewater. This instrument was used for the determination of the concentration of metal ions. After every 5 days, about 2 mL of the sample was collected from the MFC setup for analysis. Based on the AAS analysis results, the degradation efficiency (DE%) was calculated using the following equation:

$$DE\,\% = \frac{Ti - Tf}{Ti} \times 100, \tag{5}$$

where *Ti* denotes the initial concentration and *Tf* denotes the final concentration. The growth of a biofilm on the electrode indicates effective metal reprocessing and energy production. A scanning electron microscope (SEM) was used to investigate the morphology of electrode biofilms.

3. Results and Discussion

3.1. Voltage Trend. The generation of voltage that occurred during the metal degradation process that took place in the MFC operation is shown in Figure 1. The operation lasted 25 days and used the same external resistance (500 Ω). On day 13 of the operation, a voltage reading of 102 mV was reported as having reached its peak point. The findings indicated that the voltage production started out at a low value and steadily climbed until it reached its peak point on the 13^{th} day. After 14 days of continuous operation, a decreasing trend in the voltage begins to manifest itself. The decrease in voltage production is a sign that the bacterial species are reaching the phase of death. After a few days had passed, the observation revealed that there was still a tendency toward a lower voltage. It indicates that the exoelectrogens are unable to regain control of the oxidation process of the organic substrate, and as a result, the process is moving forward to its conclusion. According to this research, the highest voltage was attained on day 13. Despite this, a few studies have shown that the point where the voltage is the highest is also a good indicator of a significant change in the state of the metal from a soluble to an insoluble state [21, 23, 24].

3.2. Polarization Behavior. In addition, the polarization performance was investigated by using various external resistances to compare the CD, PD, and voltage relations. It has been determined, using Figure 2, that the voltage and CD have a relationship that is inversely proportional. When the voltage drops, the current density (CD) goes up. At 500 Ω, the PD was at a maximum of 0.099 mW/m^2, but at 5000 Ω, it only provided 0.060 mW/m^2. It is important for successful electron transfer that the internal resistance and the external resistance should be equal. The greater the resistance of the external environment, the lower the electron transit. When the external resistance is low, the potential does not stabilize as quickly as it would otherwise, but electrons are still made and moved at a high enough level. The high electron movement causes an instability in the voltage. The maximum CD that was ever recorded was 31.57 mA/m^2. The cathodic reaction rate was further boosted by the external supply of oxygen, which contributed to the stabilization of voltage generation despite the increased resistance. The internal resistance was 734.0 Ω. Few studies used comparable methodologies to describe how energy is produced in electrochemical fuel systems [24–27].

3.3. Electrochemical Impedance Spectroscopy. AnEIS-Nyquist curve plot was drawn to assess the charge transfer resistance provided by the fuel circuit model. The EIS measurement that was performed after the procedure was finished and can be seen in Figure 3. This measurement was included in this research. According to the research, a straight line with a high Z'_{image} (Ohm) suggests a low electron transportation rate, whereas a semicircle or semibent line shows a high rate despite a lower Z'_{image} (Ohm) [28]. The current research demonstrates that the semibent lines at the start of the Z'_{real} (Ohm) suggest that there was electronic movement, but afterwards the straight line indicates that there is no high electronic movement. This was found by comparing the two types of lines. In most cases, a high level of electron mobility may be inferred from a reduced internal resistance, as well as from the form of a semicircle. There is a possibility that poor electronic mobility will result from higher internal resistance than external resistance.

3.4. Cyclic Voltammetry and Specific Capacitance. During the operation, the CV study was taped at a variety of different times. The charts illustrate the propensity of the metals to undergo oxidation and reduction at a variety of different time periods. As can be seen in Figure 4(a), the MFC operation displayed the maximum current throughout the forward and reverse scan speeds. The forward scan revealed that the current was 0.00001 mA on day 5, 0.00002 mA on day 10, and 0.000045 mA on day 25 of the cycle. In a manner analogous, the reverse scan revealed that the current was −0.0000 mA on day 5, −0.00001 mA on day 10, and −0.000025 mA on day 25 of the cycle. On day 25, both the forward and backward scans showed that the current was at its highest possible level. Both the forward scan and the reverse scan provide information on the rate at which the metal ions in the wastewater are being oxidized or reduced, respectively. Both the oxidation and reduction peaks reached their highest point on day 25, with the oxidation peak reaching 0.08 V and the reduction peak reaching −0.8 V. Additionally, the CV curves are utilized to calculate the Cp values at various time intervals during the process. During the operation of the MFC, the Cp values demonstrate the rate of biofilm production as well as the stability rate. This is shown in Figure 4(b). The high Cp rates demonstrate that the maturation of the biofilm development is becoming nearer on a step-by-step basis. The Cp readings have been on a decreasing trend, which indicates that the formation of the

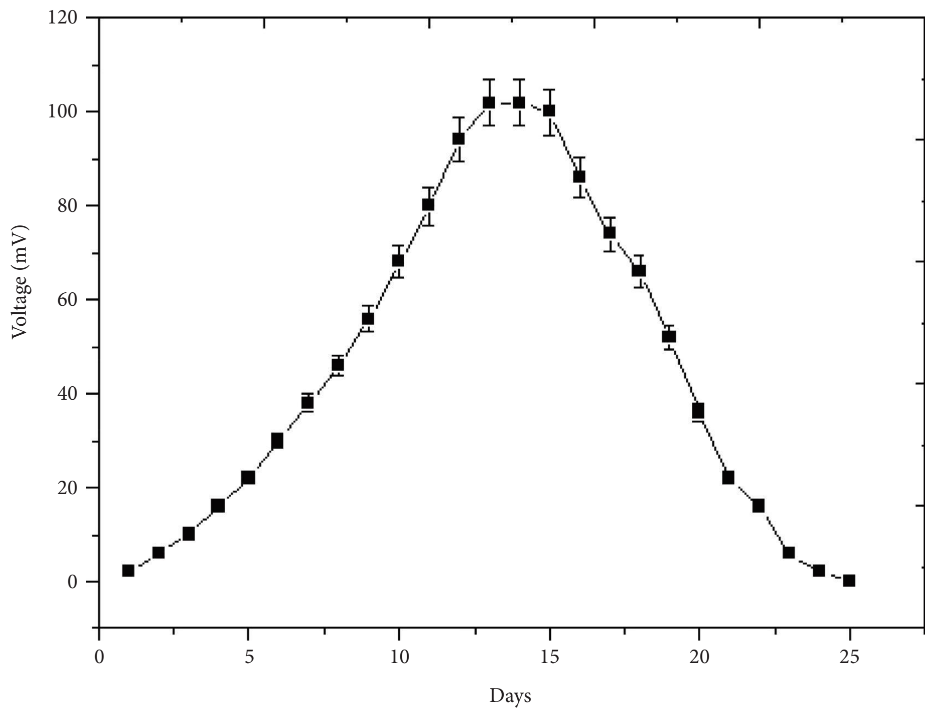

Figure 1: Voltage trend during the MFC operation for 25 days.

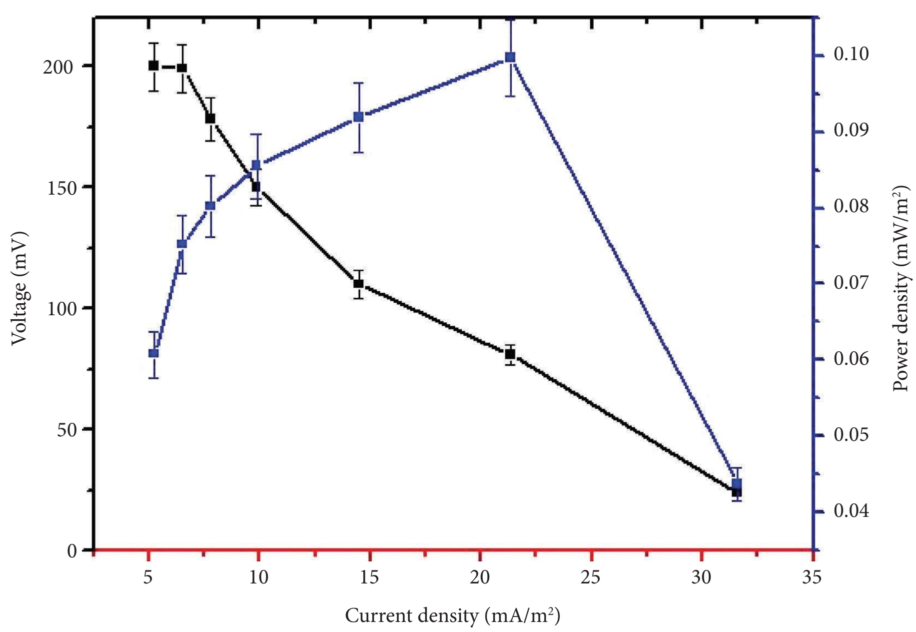

Figure 2: Polarization behaviour of operation.

biofilm was disrupted, and thus, the biofilm is not functioning as it should. According to the findings of this investigation, the concentration of Cp increased from 0.00002 on day 5 to 0.00014 on the following day 25. It was evident that the biofilm, after some time had passed, had achieved stability and had developed without being disrupted on the anode surface. A similar line of reasoning was followed by Hong et al. [29] in order to justify the production and the constancy rate of biofilm.

3.5. Degradation of Metals. The results of the investigation concerning the degradation of metal ions are shown in Table 2. The degradation of metals via the use of a bioelectrochemical system is emerging as one of the most promising and cutting-edge methods, especially in relation to MFC. The current research presents novel findings about the degradation of metal ions in synthetic water. As the process gets closer to being finished, there is a progressive improvement in the efficiency of the metal degradation.

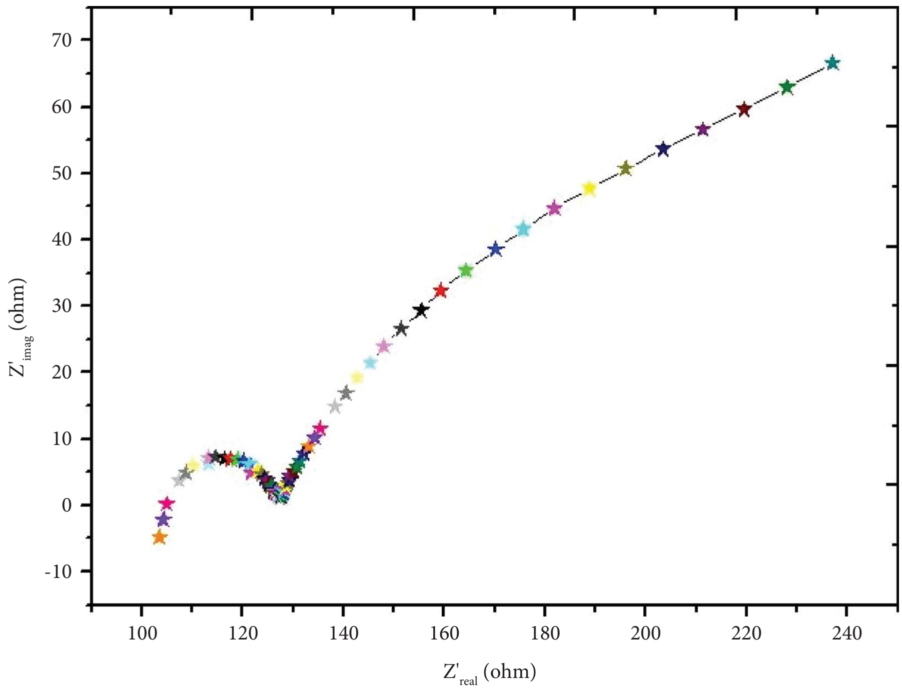

Figure 3: Electrochemical impedance spectroscopy study of the system.

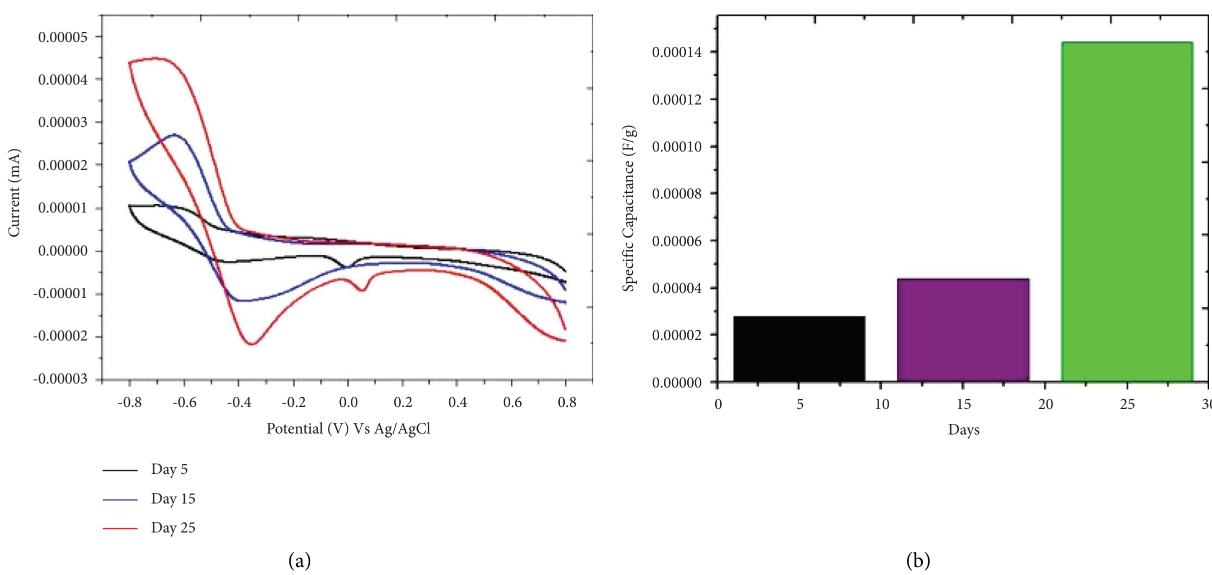

Figure 4: (a) CV at different days (b) specific capacitance values.

Table 2: Recorded degradation efficiency of metal ions in MFC.

Organic substrate	Concentration of metal in chamber (ppm)	Operational days	Degradation percentage of Cr^{3+}	Degradation percentage of Co^{2+}	Degradation percentage of Ni^{2+}
Mango peel extract	100	0	0.00	0.00	0.00
		5	13.53	17.20	15.84
		10	33.45	36.89	35.27
		15	51.00	57.10	58.35
		20	64.82	66.90	67.49
		25	69.21	72.00	70.11

When compared to the findings of the previous research, the ones presented here are rather intriguing. In this research effort, an efficiency of degradation of more than 70% is achieved. The presented data suggest that the metal ion concentration was reduced in the synthetic water that had been treated.

3.6. Biofilm Study via SEM. The collection of different bacterial species in and around the anode electrode is what makes up the biofilm. The biofilm oversees energy generation and transmission, as well as metal degradation. During the procedure, the biofilm was produced in a natural way without any assistance from the outside. About 97% of biofilm is made up of water, 3%–6% extracellular polymeric materials (EPSs), and 2%-2% bacterial species [21, 30]. The extracellular polymeric substance is the primary component of the biofilm and is responsible for the bacterial activities that simultaneously generate electrons and breakdown metals [31]. It is the region of the biofilm that contains the most water content. The biofilm's age is determined by the EPS, and the effectiveness of the EPS depends on the availability of the organic substrate. In addition, the components of EPS compositions include 40–95% polysaccharides, 1–60% proteins, 40% lipids, and more than 5% nucleic acids [32, 33]. The organic substrate improves the functioning of the EPS, which ultimately results in a biofilm that is durable and robust. According to the CV data shown previously, the existing biofilm seemed to have a high degree of stability. On the other hand, the SEM images of the electrode exhibiting biofilm are shown in Figures 5(a) and 5(b). Images taken using a scanning electron microscope (SEM) revealed that the anode electrode containing the biofilm demonstrated the normal proliferation of bacterial species (Figure 5(a)). Therefore, bacterial behavior and stability are very useful tools in environmental remediation. In addition, scanning electron microscopy (SEM) inspection of the anode biofilm revealed a surface with a similar shape. The presence of conductive pili-type bacterial species is indicated by the presence of the rod filament structure in the anode electrode biofilm. According to the findings of a number of studies, the rod-shaped and filamentous appendage structures are characteristic of bacterial species that are of the conductive pili type [21, 24, 27]. The most frequently mentioned conductive pili species are *Klebsiella pneumoniae*, *Acinetobacter*, *Escherichia*, *Bacillus*, and *Lysinibacillus* [24, 34].

4. Mechanism of the Present Study

MFC rely on bacterial activity for metal ion production, transport, and reduction. In the literature, numerous bacterial species, including *Bacillus*, *Klebsiella*, *Escherichia*, and *Actinobacillus*, are well-known exoelectrogens [21, 24]. In the current investigation, the waste from mango peels was used as an organic substrate for several kinds of bacteria. The mango peel waste started off as polysaccharides, but it will eventually be converted into simple glucose. This glucose will then be oxidized further by bacterial species, which will result in the production of electrons and protons. The oxidation and reduction reactions that were seen in this investigation may be expressed in the following way:

$$\begin{aligned}
\text{Oxidation: mango peel waste} &\longrightarrow C_6H_{12}O_6 + 6H_2O \longrightarrow 6CO_2 + 24H^+ + 24e^-, \\
\text{Reduction: } 24H^+ + 24e^- + 6O_2 &\longrightarrow 12H_2O, \\
\text{General reaction: } C_6H_{12}O_6 + 6O_2 &\longrightarrow 6CO_2 + 6H_2O + \text{Electricity}.
\end{aligned} \tag{6}$$

After some time has passed, the electrons and protons that have been produced are moved from an anode electrode to a cathode electrode during the oxidation process. Because there was just one chamber in MFC, the protons were able to pass through with no obstruction from the anode to the cathode chamber. Before electrons can be sent to the cathode from the anode electrode, they must first go through a series of steps that include many processes. Bacterial cells are the source of these electrons. The mechanism that has received the most attention is discussed in the following paragraphs and is also shown in Figure 6.

(a) In order to move electrons from the bacterium cell to the anode, redox-active proteins such as OmcS, OmcZ, OmcB, OmcT, and OmcE are used. These proteins are responsible for the transmission of electrons. This mechanism is being used by the families of *Geobacter* to transfer electrons.

(b) Short-range electron transfer is an additional method that may be used to successfully transfer electrons. To carry out this procedure, the bacteria made use of their own self-produced reduced shuttles as well as oxidized shuttles. Both the *Desulfuromonadaceae* and *Geobacteraceae* families can produce their own electron shuttles. Components of the electron shuttle include certain molecules such as OmcA, MtrF, MtrE, and MtrC.

(c) The ability of a bacterial population to transmit electrons over long distances relies on the bacteria's conductive pili. It has the appearance of a conducting metal, but in reality, it is a component of the bacterial body that was used to carry electrons straight from the cell of the bacterium to the anode [35].

In addition, after the biofilm research and the bacterial analyses, it was discovered that the current study followed the long-range mechanism for electron transfer. This was

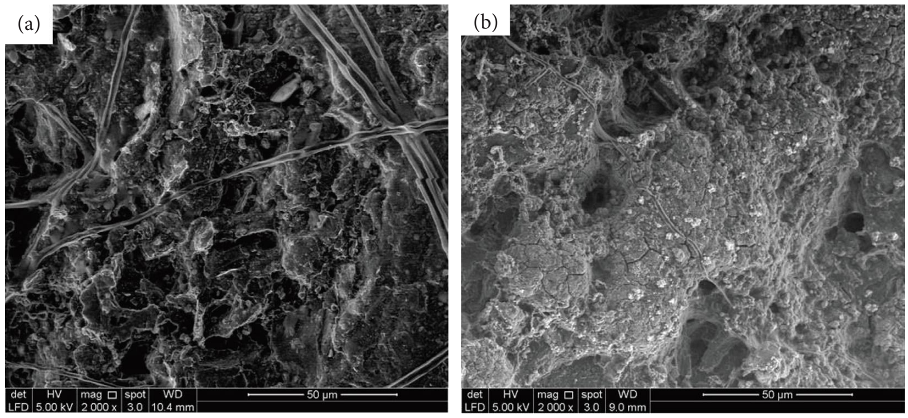

Figure 5: SEM images of (a) treated anode and (b) treated cathode.

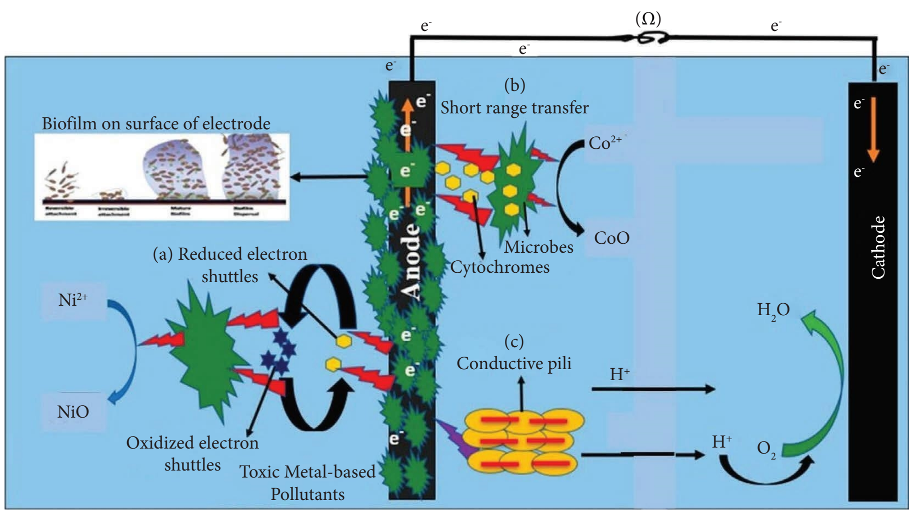

Figure 6: Mechanism of the present study (modified from reference [35], open access MDPI publisher).

discovered after analyzing the biological tests. In addition, scanning electron micrographs of the biofilm revealed the existence of a rod-shaped structure, indicating that all the detected species are of the conductive pili type of bacterial species. According to all the evidence (biological testing and earlier published research), the conductive pili of bacteria species are to blame for the electron transfer that was seen in this investigation. During a redox reaction, the soluble metal ions are effectively transformed into a state that is insoluble. The insoluble condition was discovered in the form of sludge, as was mentioned earlier in the sentence. In addition, the findings of the AAS test only indicated the amounts of metal ions that were still present in the water samples. The metal ions that were removed were changed to oxides, and a sludge-like substance was seen in the MFC because of this transformation. According to the previous literature, the metal ions that are extracted are changed into their oxide forms, and the sludge that is produced contains the metals in their oxide forms [21, 24, 36]. The following is a mechanism of the biological events that occur during metal reduction:

(i) Reduction of Cr^{3+}

$$\begin{aligned} Cr^{3+} + 3e^- &\longrightarrow Cr_{(s)}, \\ 2Cr^{3+} + 3H_2O &\longrightarrow Cr_2O_3 + 6H^+, \\ Cr_2O_3 + 6e^- + 6H^+ &\longrightarrow 2Cr_{(s)} + 3H_2O. \end{aligned} \tag{7}$$

(ii) Reduction of Ni^{2+}

$$\begin{aligned} Ni^{2+} + 2e^- &\longrightarrow Ni_{(s)}, \\ 2Ni^{2+} + 2H_2O &\longrightarrow 2NiO + 4H^+, \\ NiO + 2e^- + 2H^+ &\longrightarrow Ni_{(s)} + H_2O. \end{aligned} \tag{8}$$

(iii) Reduction of Co^{2+}

$$\begin{aligned} Co^{2+} + 2e^- &\longrightarrow Co_{(s)}, \\ 2Co^{2+} + 2H_2O &\longrightarrow 2CoO + 4H^+, \\ CoO + 2e^- + 2H^+ &\longrightarrow Co_{(s)} + H_2O. \end{aligned} \tag{9}$$

5. Concluding Remarks and Future Recommendations

The present research focused on the usage of the residual mango peel in MFC to generate energy and degrade metal ions. The voltage that could be measured to the greatest level was 102 mV. The presence of many bacterial species on the anode surface suggests that the metal degrading process has been effective. In addition, according to the data, well-known species of exoelectrogenic bacteria were found in the MFC operation that is now underway. The favorable bacterial activities that take place because of the oxidation of the organic substrate are responsible for the high efficiency with which metals are degraded. It indicates that the organic substrate underwent a vigorous oxidation process, which led to an abundant production of electrons as a byproduct. The current electrochemical measurements, on the other hand, demonstrate an energy efficiency that is much lower than that of prior studies. After conducting in-depth research and analysis, the conclusion was reached that, despite all the contributing elements, there has been a decline in energy efficiency. The investigation revealed that there was a fault with electron transportation, namely, that electrons were not delivered to the cathode in an efficient manner. It is because of the quality of the electrode that the current investigation utilized the commercial graphite electrode, which was not successful in properly transferring the electron. Electrodes that are based on derivatives of graphene should be used since they are highly conductive and modern materials. This will result in improved electron transport. For the electrode material to be effective over the long term, it must be biocompatible, chemically stable, ultraconductive, and thermally balanced. The problems that need to be overcome to get the MFC to the level of commercial viability may be tackled with the collaboration of professionals from various sectors, including microbiology, material science, and bioelectrochemistry.

Data Availability

All the data used to support the findings of the study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

Ghada Mohamed Aleid and Anoud Saud Alshammari equally contributed to this work.

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