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Research Article

Investigating the Activity of Carbon Fiber Electrode for Electricity Generation from Waste Potatoes in a Single-Chambered Microbial Fuel Cell

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Microbial fuel cells (MFCs), a technology that converts chemical energy into electrical energy, have been regarded as the most suitable method for sustainable energy production. However, most MFCs generate a low power output, limiting their large-scale industrial applications. Here, we introduce a high-performancesingle-chambered microbial fuel cell (SCMFC) based on carbon fiber anode and zinc oxide nanoparticle (ZnO NP)-fabricated copper cathode. Furthermore, best optimized conditions of temperature, pH, substrate, external resistance, and cathode fabrication were also considered to evaluate the performance of this SCMFC in treating potato wastewater along with bioenergy production. Results indicated that chemical oxygen demand (COD) could be effectively removed (80%) and maximum voltage, current density, and power density were 1.58 V, 0.235 mA/cm^2, and 0.3714 mW/cm^2, respectively. Hence, the SCMFC used in this study has a higher potential to treat potato wastewater along with high power production.

1. Introduction

In the modern world, research on green and sustainable fuels is needed to curtail the increasing environmental degradation and energy insufficiency caused by over-exploitation of fossil fuels, and fossil fuel stocks are increasingly strained due to their non-renewable nature [1]. Moreover, the practice of disposal and burning of solid waste has a direct impact on human survival as it becomes a source of water and environmental pollution due to emission of greenhouse gases [2].

Several studies concluded that to solve problems of energy crisis, water pollution, and solid waste management, microbial fuel cell technology has emerged as a preferable, clean, sustainable, and innovative technology as it uses microorganisms for bioenergy production along with the treatment of wastewater [3]. Microbial fuel cells (MFCs) are electrochemical devices that employ electro-active microbes to harvest chemical energy stored in the bonds of organic waste and convert it into electrical energy [4]. Among all MFC configurations, single-chambered microbial fuel cell (SCMFCs) has gained huge importance due to its simpler methodology and cost-effectiveness. However, MFC technology has not been preferred for large-scale industrial applications due to major limiting factors including expensive cathode catalysts and the unavailability of large-volume MFC reactors. The commonly used oxygen reduction reaction (ORR) catalyst, i.e., platinum metal, is not preferred on an industrial level due to the following limitations: it is rarely available, expensive, and easily deactivated in polluted environment [5, 6]. Hence, it is the need of the hour to find a suitable catalyst with high performance, excellent stability, and cost-effective properties. Recently, researchers suggested some substitutes including conducting polymers, carbon matrix materials, non-platinum metals, spinel compounds, and transition metal catalysts [7–9]. Among all materials, transition metal catalysts possess unique active sites for adsorbing oxygen and breaking O=O bonds during ORR. Transition metal (for example, Co, Zn, and Fe) nanocomposites have gained huge importance due to their better catalytic efficiency than precious platinum [10, 11]. Among these, ZnO nanocomposite catalysts could be used as an alternative to platinum because of high activity of oxygen reduction reaction and excellent stability. ZnO has been reported in different MFC studies to be used as anode or cathode catalyst. ZnO possesses high antibacterial performance that reduces the biofilm formation on electrode surface. Muthukumar et al. reported the preparation of iron-doped ZnO NPs on carbon anode with high surface area that showed high performance with maximum power density of 137 mW/m^2 through inhibition of biofilm production [12]. Yang et al. studied the preparation of graphene-doped ZnO or Co NPs as cathodic catalysts through a hydrothermal approach [13]. They found the reduction of biofilm formation during the operation of MFCs due to antimicrobial effect of ZnO. It has also been observed that reduction of biofilm formation depends on the concentration of ZnO added [14]. Kugarajah et al. reported the use of ZnO nanomaterials as fillers in cationic-exchange membranes for MFC operations to improve proton production and add antifouling features to the membrane [15]. However, to the best of the authors' knowledge, no one reported the synthesis of SCMFC having ZnO fabricated on Cu electrode treated as cathode while carbon fiber was treated as anode through the bacterial deposition. The ZnO NPs were fabricated on Cu electrode using the microemulsion method. Moreover, the performance of carbon fiber anode for extracting bioelectricity was analyzed with green synthesized ZnO NP fabrication and without ZnO fabrication. Our research group [16] used waste potatoes as substrate in a SCMFC and investigated the generation of electricity along with wastewater treatment and reported extraction of 1.12 V along with 40% COD removal. MFCs have also been investigated for the treatment of water effluents from potato chips processing plants [17]. Tian et al. [18] studied the effectiveness of single-chambered MFC in the degradation of potato pulp waste based on the eradication of organic matter and electricity generation.

In this study, novel anode and cathode electrodes have been developed to obtain high electrical conductivity and low resistance that will improve the efficiency of MFC. Various optimization parameters that significantly affect the performance of SCMFC also have been discussed to get the optimum results. Additionally, the COD experiment also has been conducted to check the efficiency of the proposed system.

2. Experimental

2.1. Collection and Preparation of Potato Extract. White potatoes, named locally "May Queen," which are harvested and consumed in Pakistan as major food type were used in this study. The waste potatoes were purchased from a local supermarket in Lahore, Pakistan, and cut into small pieces, then masticated by adding water with the help of an electrical blender, and filtered. The pH and conductivity of the prepared filtrate were checked by a pH meter (Hanna Instruments, USA) and conductometer (Thermo Scientific, UK), respectively.

2.2. Fabrication of Electrode with ZnO NPs. The microemulsion method was employed for the fabrication of cathode with nanoparticles. For this purpose, 50 mL of n-hexane and n-hexanol was magnetically stirred at 30°C to form a dispersion medium and 2 mL of Tween 20 as surfactant was added dropwise along with continuous stirring to get the mixture having oily appearance. At the same time, 0.1 M solution of $ZnSO_4$ and 9 M solution of HCl were mixed in a separate beaker to form a dispersed phase. Then, certain volume of dispersed phase was introduced to the dispersion medium (oil phase). Emulsion became transparent and crystal clear. Then, the electrode was placed in a chamber containing prepared emulsion, and external power supply of 0.3 V was applied and reaction was left undisturbed for 2 days. In this way, ZnO NPs were successfully fabricated on copper cathode.

2.3. Reactor Construction and Operation. The used MFC was a single-chambered reactor constructed from cylindrical glass having length of 13 cm and diameter of 9 cm with a working volume of 450 mL. Chamber packing has three openings: two openings are for electrodes while one is for thermometer connection at different stages. Both electrodes were made of different materials, anode is a carbon fiber having length of 16 cm and width of 3.5 cm while a copper cathode having length of 15.3 cm and width of 2.8 cm was employed in construction of microbial fuel cell. The distance between electrodes was 2 cm. Potato wastewater was introduced in SCMFC, and to check voltage, a multimeter (DT830D Digital Multimeter, Pakistan) was connected externally by adjusting it to voltage reading. The whole assembly was performed in an anaerobic environment with pH value of 7.8 at 30°C. The acetic acid substrate was added every 1 h to increase biofilm formation on anode surface, respectively. However, cell voltage was observed continuously after 30 min throughout the reaction through a digital multimeter.

2.4. Multiple Parameter Optimizations. Various factors affect the performance of microbial fuel cells as they have an effect on the kinetics of bacteria, rate of oxygen reaction, and rate of proton transfer through the solution and influence the metabolic process of bacteria. For obtaining high power density in SCMFC, ZnO-fabricated cathode, pH of the solution, temperature, substrate type, and external resistance were studied. These optimized process parameters were expected to give the maximum power density production by influencing the metabolic process of bacteria. The temperature affects bioenergy generation, COD, metabolic activity, and kinetics of bacteria, so the SCMFC was operated at different temperatures under optimized conditions (neutral pH, acetic acid substrate, and 80 Ω resistance) to get the maximum output [19]. These selected temperatures are 15°C, 30°C, 50°C, and 80°C. In every case, the voltage was recorded every 30 min, and a plot of voltage against time was plotted. Similar experiments were performed for pH, substrate, and resistance by keeping other factors constant. The pH was studied at seven different values ranging from 6–12 in the presence of 1 M NaOH and 1 M HCl solutions, respectively. Different substrates including ethyl acetate (1 mL), acetic acid (1 mL), sodium acetate (1.0 g/10 mL), glucose (1.0 g/10 mL), and sucrose (1.0 g/10 mL) were added continuously after 30 min in respective chambers. Four different SCMFCs with different resistances of 20 Ω, 40 Ω, 60 Ω, and 80 Ω were constructed, and the time-voltage graph was plotted by recording cell voltage every 30 min. Fabrication of cathode with metal nanoparticles has a great impact on bacterial biofilm formation on the anode in a microbial fuel cell. In this study, green synthesized ZnO nanoparticles were fabricated on the copper cathode to observe its effect on voltage and current extraction, so at optimized conditions such as 30°C, neutral pH, acetic acid substrate, and 80 Ω resistance, two MFC reactions were monitored with and without zinc oxide-fabricated cathode. After every 30 min, cell voltage was noted for plotting the graph between voltage and time.

2.5. Characterization. Scanning electron microscopy analysis was carried out using SEM (Hitachi S-4700) operating at 25 kV to confirm the successful formation and particle size investigation of synthesized ZnO NPs. The X-ray diffraction (XRD) analysis was performed by using XRD diffractometer (D8 Advance, Bruker Corporation, USA) operating at working conditions of 30 kV, 20 mA, and Cu-Kα radiations with wavelength of 1.54 A.

2.6. Chemical Oxygen Demand Determination. Standard closed reflux titrimetric method was used for the determination of COD. $HgSO_4$ was used for the removal of chlorides while H_2SO_4 of high purity was used for oxidation. 0.042 N solution was prepared from $K_2Cr_2O_7$ and solid Ag_2SO_4. Excess $Cr_2O_7^{-2}$ was titrated with 0.25 M solution of Mohr's salt using ferroin as an indicator. The change of color in solution from yellow to brown indicated the conversion of excess dichromate ion (Cr^{+6}) into trivalent form (Cr^{+3}). The amount of potassium dichromate consumed is equivalent to the organic matter of potato wastewater chemically oxidized. The following formula aids in estimation of COD in desired sample.

$$\text{COD}\left(\frac{\text{mg}}{L}\right) = \frac{(A - B)N \times 8000}{\text{mL sample}}, \quad (1)$$

where N is the normality of ferrous ammonium sulfate and A and B represent the volume of ferrous ammonium sulfate solution required for blank titration and sample titration, respectively.

3. Results and Discussion

3.1. Electricity Generation from SCMFC. MFC technology is used for biological treatment of solid organic waste along with electricity production. The basic reaction of a SCMFC was operated for 90 days at optimized conditions (30°C, pH 7, and addition of acetic acid, 80 Ω resistance, and fabricated cathode). Moreover, in this SCMFC, carbon fiber electrode was converted to anode through bacterial deposition and copper electrode was converted to cathode through fabrication of ZnO NPs on the surface of electrode. However, the presence of ZnO NPs was confirmed using SEM analysis. Figure 1(a) shows SEM micrographs which reveal that ZnO NPs are uniformly distributed with an average diameter of 43 ± 2 nm, respectively. XRD results also reveal the hexagonal wurtzite phase of ZnO that matched well with JCPDS file No. 36–1451. Figure 1(b) shows strong diffraction peaks at 31.1, 34.4, 37, 47.2, 56.5, 62.2, 67.8, and 69.4 degree of 2Θ which corresponds to the (1 0 0), (0 0 2), (1 0 1), (1 0 2), (1 1 0), (1 0 3), (1 1 2), and (2 0 1) crystal planes, respectively. The obtained XRD results were in good agreement with the results of Omri et al. [20] which confirm the presence of ZnO NPs.

Here, the reaction was proceeded through the oxidation of organic matter through bacteria that not only develop electric potential but also generate CO_2, protons, and electrons. These generated electrons and protons were further transferred to the cathode compartment where the combination of oxygen led to the formation of water. The general reactions are given below:

$$C_{12}H_{22}O_{11} + 13H_2O \longrightarrow 12CO_2 + 48H^+ + 48e^- \text{ (anodic.reaction)} \quad (2)$$

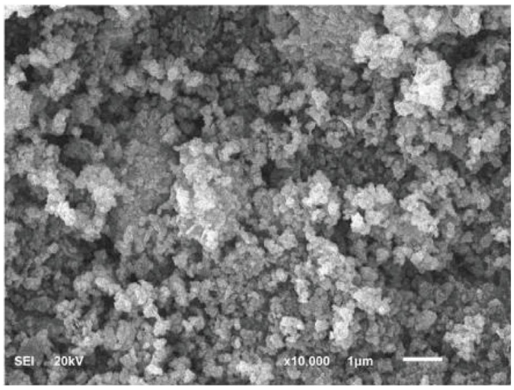
$$4H^+ + O_2 + 4e^- \longrightarrow 2H_2O \text{ (cathodic.reaction)} \quad (3)$$

The maximum value of open circuit voltage with optimized fabricated cathode was 1.58 V, current density obtained was 0.235 mA/cm^2, and power density was 0.3714 mW/cm^2. In case of optimized SCMFC without fabricated cathode, open circuit voltage obtained was 1.29 V, maximum output of power density was 0.212 mW/cm^2, and current density obtained was 0.165 mA/cm^2. The current density and power density were obtained using the following equation:

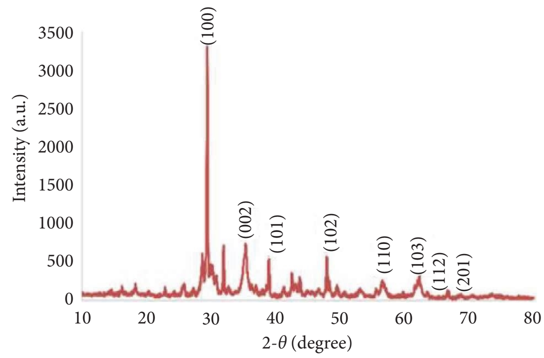
$$\text{current density} = \frac{\text{current}}{\text{surface area}}, \qquad \text{power density} = \frac{\text{power}}{\text{surface area}}. \quad (4)$$

In both cases, maximum voltage obtained was sufficient to turn on a LED bulb. We can assemble several such fuel cells in series to get any required output value from cheapest, eco-friendly, and greener technology. Figure 2 shows the maximum value of voltage obtained from fabricated optimized cell reaction and non-fabricated optimized cell reaction.

The cell voltage and current were observed from the very first day of operation and reached their peak value on the first day and then remained constant for the next 48 days.

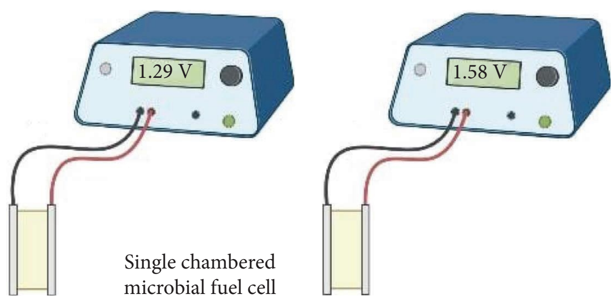


(a)



(b)

Figure 1: Confirmation for the presence of ZnO NPs in ZnO NPs-modified cathodes using (a) SEM analysis and (b) XRD analysis.



(a) (b)

Figure 2: Maximum voltage from SCMFC (a) without fabricated cathode and (b) with fabricated cathode.

But they started to decrease after 48 days. Figures 3(a)–3(c) show the time dependency of voltage, current, and power obtained on the first day of MFC operation.

Figure 3(a) shows that maximum voltage was obtained in a very short time of 210 min, and this increase in voltage to 1.58 V was due to greater activity of microbes and because of greater availability and movement of free ions. The decline in voltage value after 48 days was attributed to the smaller availability of ions and the rapid hydrolysis of potatoes which resulted in obvious decrease of pH in the MFC.

Figure 3(b) displays variation of current with time. In this case, sodium acetate was added as substrate to increase current output. Another advantage of using this substrate was that current attains stability and becomes constant for some time. As voltage and current are directly proportional to each other, the same trend is shown by current. Maximum current output obtained on first day is 13.17 mA, and this output is much greater than that obtained from non-fabricated cathode (9.26 mA). At starting time, current output was low because bacteria were not active at that time. Then, current value started increasing and gaining maximum value at 210 min. At this time, bacteria were showing their maximum activity. After 210 min, current intensity started decreasing because the maximum number of free ions has already been used, after which current became constant.

Figure 3(c) displays plot of power against time, and the data obtained from voltage and current were converted to power by using the formula $P = IV$ where P is power, I is current, and V is voltage. First, there is a slight increase in power, then it gained maximum value, and after that, it started decreasing and then became constant, and it is the real advantage of our research work. The highest value of power density obtained from this optimized reaction was 0.3714 mW/cm^2 which is a great achievement, and it was achieved at 210 min because at this time, microbes were highly active.

3.2. Optimization of Parameters. To get highest output from MFC, it is operated at optimized conditions, and these optimized conditions in this current study were ZnO-fabricated copper cathode, neutral pH, room temperature, acetic acid substrate, and 80 Ω resistance (higher external resistance, Figures 4 and 5). Comparison of fabricated and non-fabricated cathode voltage revealed that output is much greater in case of zinc oxide-fabricated cathode (Figure 4).

This increase in voltage was attributed to the microbes that started growing on the surface of the anode resulting in thicker biofilm formation on anode. Moreover, from comparison of different temperatures, it was observed that 30°C shows maximum voltage which means 30°C is the

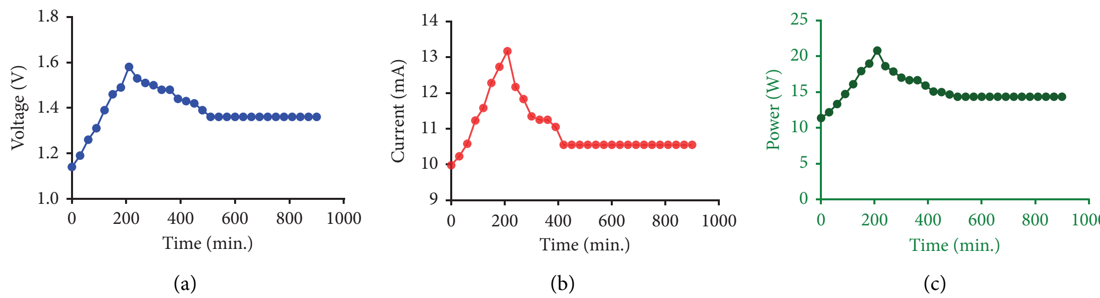


Figure 3: Time period variation of (a) voltage in V, (b) current in mA, and (c) power in W.

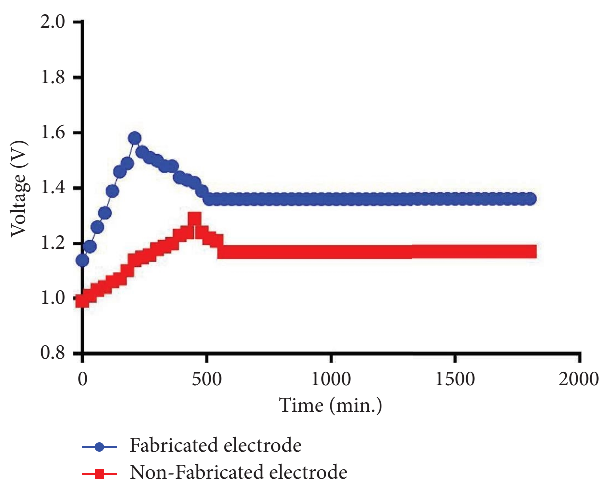


Figure 4: Comparison of voltage with fabricated and non-fabricated cathode.

optimum temperature at which microbes show their best efficiency to generate electricity. Experimental data indicated that performances of MFC slightly decreased with increase and decrease of temperature because the metabolic activity of bacteria was disturbed, and diffusion of substrate also limited the electricity production.

On comparing the voltage at different pH values, it was observed that pH 7 is the optimum pH value for bacterial activity which means natural environment is suitable for bacterial growth and their performance (Figure 5). At neutral pH, there is high rate of degradation of substrate providing more electrical energy. Other pH values close to neutral pH gave best results, and more alkalinity or acidity is unfavorable for bacterial growth affecting biofilm formation on anode, and oxygen reduction reaction was also changed; as a result, internal environment of reactor and wastewater treatment efficiency of MFC are affected. The electrochemical performance of the MFC declines at too acidic ($pH < 6$) and basic ($pH > 10$) conditions [21].

The substrate is a biological factor that plays a key role in affecting electricity production in MFCs as it is a source of carbon and energy [22]. Further comparison of voltage values with various substrates provided the results that when acetic acid was used as a substrate, SCMFC gave maximum output as compared to other substrates because acetic acid is a source of carbon which induces electro-active bacteria and acetate is the end product of various metabolic pathways for higher order carbon source. Deepika et al. used acetic acid as a substrate and found that it gave maximum voltage and current values of 0.47 V and 0.05 A, respectively [23]. In case of glucose, no remarkable increase in voltage was observed. First, voltage increased and then decreased, and after that, it became constant. At the initial stages, voltage increased because of higher concentration of electrogenic bacteria, but with the addition of more and more glucose, the concentration of fermentative bacteria increased which resulted in decrease of voltage. But all other substrates supported the electrogenic bacteria and leading to the higher voltage value because of thicker anodic biofilm formation. In the case of all these substrates, bacterial activity was lower at the start; hence, the value of voltage obtained was also low, but with the passage of time when the number of ions and bacterial activity increased up to a certain time and which is termed as optimum time. After an optimum time, there is no more release of ions, so no increase in microbial activity was observed resulting in a decrease of voltage. Figure 5(c) gives a comparison of different substrates. In one study, Chae et al. compared four different substrates for power output and found that acetate-fed MFCs showed the highest value than butyrate, glucose, and propionate [24]. Bond et al. reported that acetate is the simplest and extensively used substrate to be used as a carbon source for the induction of electro-active bacteria [25]. Liu et al. compared the use of acetate with butyrate and found that power generated by acetate in SCMFC was 66% higher than butyrate [26]. In a study, Liu et al. also compared the acetate substrate with protein-rich wastewater and observed twofold more electric power [27].

As external resistance was also an optimizing factor, the effect of various external resistances on efficiency of MFC showed that highest voltage value was recorded at 80 Ω which is the highest external resistance while all other resistances lower than 80 Ω yield low output because of limitations imposed on electrode reaction kinetics and mass transfer rate [28]. At low external resistance, the electron delivery to cathode is limited and production of electrical energy decreases, and microbes also produce extracellular polymeric substance (EPS) during their growth on the anode surface which consumes energy available for microbial growth [29]. Ghangrekar and Shinde carried out the study of resistance impact on MFC and they found that the higher the value of external resistance (0 to 4,000 Ω), the higher the cell voltage (358 mV) will be [30].

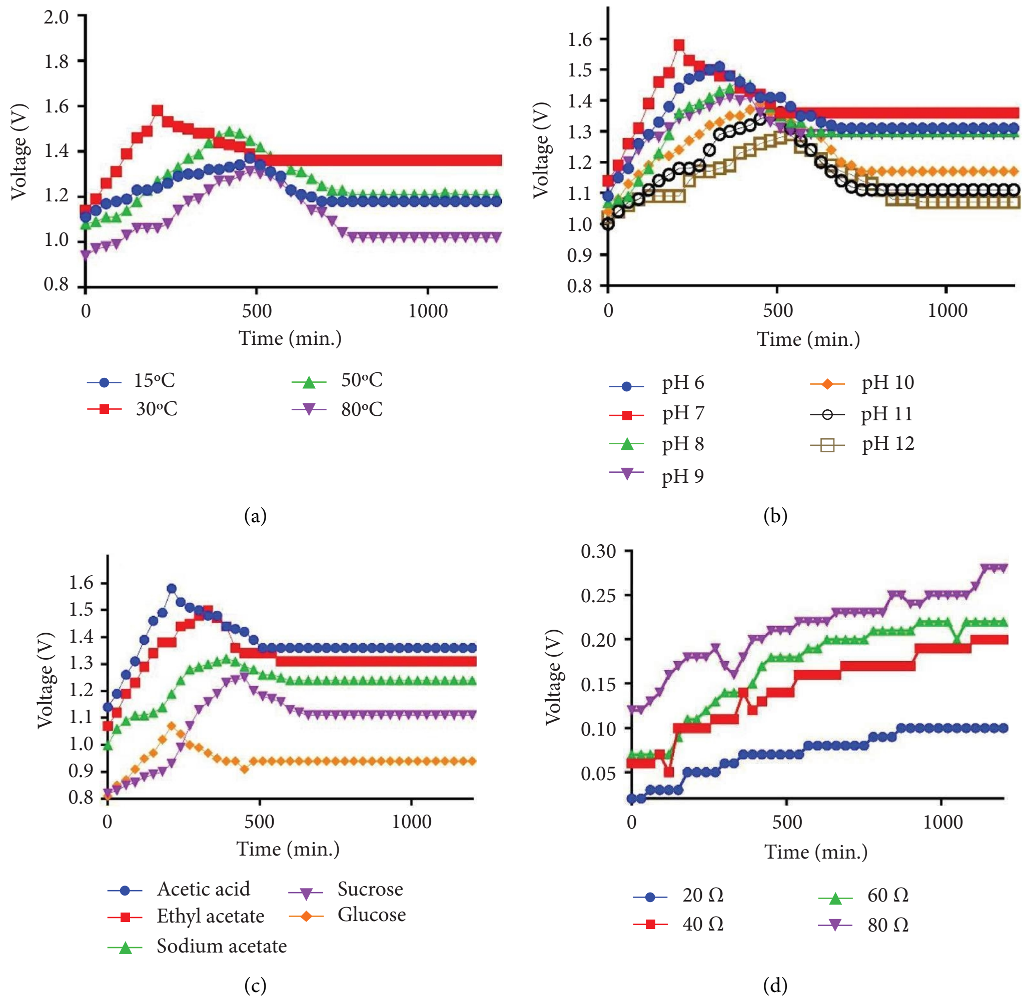


FIGURE 5: Comparison of voltage at various (a) temperatures, (b) pH values, (c) substrates, and (d) resistances.

3.3. COD Removal Efficiency. MFC technology is an effective method for energy production and treating wastewater by efficient degradation of organic compounds present in it, and thus the overall performance of MFC is calculated in terms of COD [31]. To analyze the performance of SCMFC in treating wastewater, COD of potato wastewater was calculated. The results showed that COD of potato wastewater was 84 mg/L while 67 mg/L was reported after COD removal which means there is 80% COD removal. The observed decrease in COD could be attributed to the fact that the acetic acid was favorable to the bacterial species on the anode [32]. Additionally, in the assembly, the substrate degradation rate is higher due to the active oxidation of acetic acid substrate by microbes present in MFC. The results indicated that SCMFC showed good COD removal efficiency at neutral pH and when carbon fiber anode is used. The results presented in graphs depicted that SCMFC possesses excellent COD removal efficiency (80%, 78.5%, and 77%, respectively) for the last three cycles for potato wastewater (Figure 6). Our research group [16] used copper anode in SCMFC and observed 40% COD removal of potato wastewater. COD was reduced up to 75% by using dual-chambered MFC in treating different types of waste water including dairy, sugar, and sewage [33]. Dong et al. [34] reported the preparation of five different MFC modules for treatment of brewery wastewater and eliminated COD up to 87.6%. Ma et al. [35] and Zoutberg and Eker [36] used up-flow anaerobic sludge blanket (UASB) reactors for the treatment of potato wastewater that showed 80–85% COD removal efficiency at 4.8 and 4.5 pH values.

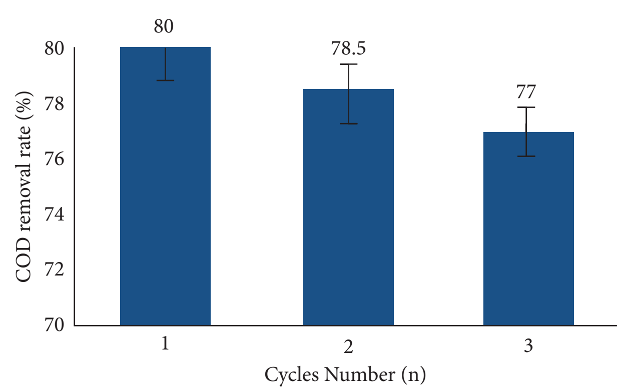


FIGURE 6: COD removal rate graph.

4. Conclusion

In this study, the efficiency of carbon fiber anode and electro-conductive performance of metal nanoparticle assembled Cu cathode in a SCMFC was investigated. Additionally, the factors significantly affecting the performance of SCMFC were studied and optimized for achieving targets. The experimental results of optimization demonstrated that bacteria required natural environmental conditions for their growth, biofilm formation, and for performing their metabolic activity. Further results indicated that metal nanoparticle assembled copper cathode and carbon fiber anode in an optimized microbial fuel cell-enhanced electro-conductive performance of bacteria. Therefore, MFCs are promising alternative sources of fossil fuels to generate electrical energy from waste. Even after an extensive research spanning over a period of more than 10 years, MFC technology has still not been commercialized due to some drawbacks such as expensive setup, and higher costs of membranes, electrode materials, and cathode catalysts. In addition, insufficient power output is another drawback linked to the mentioned technology. Thus, these drawbacks need a solution for a real-world applications of the MFCs. The MFCs used for wastewater treatment do not purify water completely. So, amendments are needed to make this technology more commercialized. The absence of polymer electrolyte membrane and provision of electrodes with high surface area can improve the power output in MFCs and make them more economical. In designing an MFC, when generation of electricity is desired, a higher potential can be yielded by increasing the bacterial metabolism rate which can be achieved by enhancing the cell potential and combining electrogenic bacteria and simple substrates such as acetate.

Data Availability

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

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

Muhammad Imran Din was responsible for supervision and review and editing. Mahroosh Iqbal was responsible for methodology and formal analysis. Zahoor Ahmad was responsible for review and analysis. Zaib Hussain was responsible for review. Rida Khalid was responsible for optimization and original draft preparation. Abdul Samad was responsible for formal analysis and review. Muhammad Ahmad was responsible for review and editing. Mahmood Ahmed was responsible for investigation, methodology, formal analysis, and review and editing. All authors have read and approved the final version of the manuscript.

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