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Enhancing Energy Production and Pollutants Removal in Landfill Leachate Using Anode-Modified Sediment Microbial Fuel Cells

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ABSTRACT

The utilization of sediment microbial fuel cell (SMFC) technology presents a paradigm-shifting method for converting the chemical energy obtained from organic and inorganic compounds found in sediment and wastewater substrates into electrical energy. This concept exhibits potential as an environmentally sustainable solution within the future energy sector and presents opportunities for wastewater remediation. This study aims to investigate the influence of anode modification in the SMFC system on generating electrical energy and removing pollutants in landfill leachate. The modification entails synthesizing a nanostructured copper layer on stainless steel (Cu-SS), subsequently compared to the conventional copper (Cu) anode. Results underscore the effectiveness of anode modification, as SMFCs featuring modified anodes exhibit twice the electrical output compared to unmodified counterparts. Modified anode SMFCs yield voltage and current density readings of 615 mV and 17 mA/m^2, respectively. In addition to electricity generation, the study delves into the SMFC's efficacy in nitrogen compound removal. Experimental results unveil the impressive capability of modified anode SMFCs, achieving 81.02% removal of Biological Oxygen Demand (BOD), while unmodified counterparts reach 76.64%. Furthermore, the removal percentages for ammonia, nitrate, and nitrite compounds within SMFCs equipped with modified anodes are 88%, 51%, and 13%, respectively. This comprehensive analysis underscores the multifaceted benefits of anode modification, amplifying electrical output and enhancing the SMFC's proficiency in nitrogen compound removal, thereby contributing to its potential applications in developing sustainable wastewater treatment and energy generation systems.

Keywords: sediment microbial fuel cell, electroplating, modified anode, leachate, BOD, nitrogen.

INTRODUCTION

The sediment microbial fuel cell (SMFC) technology represents a transformative approach to converting chemical energy derived from organic and inorganic substances within sediment and wastewater substrates into electrical energy. This innovation holds promise as an environmentally friendly solution within the future energy sector and offers potential for wastewater remediation. The performance of SMFCs is contingent upon numerous factors, encompassing reactor configuration, operational conditions, electrode surface area, and electrode material (Abbas et al., 2017; Zhao et al., 2020; Goleij et al., 2021). Among these factors, selecting the proper electrode material, particularly for the anode, emerges as a paramount consideration (Al-Dawery et al., 2021; Liang et al., 2020). The choice of electrode material significantly influences the growth of microorganisms and the efficiency of electron transfer processes. Critical prerequisites for materials serving as electrodes in this context include robust conductivity, biocompatibility, chemical stability, resistance to corrosion, and cost-effectiveness (Emalya et al., 2021). Conventionally, carbon-based anodes have been a prevalent choice in SMFC systems (Santoro et al., 2017; Tran et al., 2019). Nevertheless, alternative investigations explore the utilization of metal-based anodes (Haque et al., 2015; Emalya et al., 2022; Prasad & Tripathi, 2017) primarily due to their heightened conductivity.

Recent reports show that the voltage and power outputs achieved through SMFC have remained relatively modest (Apollon et al., 2022; Taşkan et al., 2021). A prominent enhancement avenue involves modifying the anode, as demonstrated by Sayed et al. (2020), who coated carbon-cloth material with iron nanostructures to address domestic sewage treatment. Their findings revealed that the power density of the modified anode doubled, reaching 80 mW/m², while the generated voltage increased from 600 mV to 800 mV in the open circuit. Another approach to voltage improvement through anode modification was explored by Yang et al. (2022). Their study involved modifying carbon felt through chemical oxidation and physical coating, with the anode featuring a 5% (w/w) physical coating of graphene oxide displaying heightened electricity generation (maximum power density of 132 mW/m²) compared to both the unmodified anode and the chemically oxidized modified counterpart.

Numerous studies have delved into anode modification, yielding promising results. Notably, Tominaga et al. (2022), Liang et al. (2020), and Zhang et al. (2018) have each reported instances where modified anodes yielded electricity outputs up to tenfold greater than unmodified counterparts. In light of these advancements, this study seeks to delve into the impact of anode modification in the SMFC system on electrical energy generation and pollutant removal in landfill leachate. The anode modification process involves a straightforward electroplating technique, wherein copper nanostructures are synthesized on stainless steel. The outcomes are subsequently compared with those derived from the utilization of conventional copper. This investigation aims to contribute to the growing body of knowledge on anode modification's potential for enhancing energy output within SMFC systems, with implications for future sustainable waste treatment and energy generation practices.

MATERIALS AND METHODS

Leachate and sediment collection

The leachate and sediment samples employed in this study were collected from the stabilization pond at the Aceh Regional Waste Management Agency's leachate treatment facility in Blang Bintang, Aceh Besar, Indonesia. A total of 20 liters of leachate was gathered directly from the pond using sample bottles and transported to the laboratory for further examination. Sediment samples were also taken from the bottom of the stabilization pond. The sediment underwent screening via an eight-mesh sieve to eliminate gravel components, while the leachate underwent pre-filtration to isolate dry leaves. Before utilization, the leachate and sediment were stored within a temperature-controlled refrigerator, maintained at 3–5°C.

Sediment microbial fuel cell setup

This study employed two SMFCs, referred explicitly to as SMFC-cs and SMFC-c, respectively. The reactors were carefully fabricated using transparent acrylic, with an impressive effective volume of 4.0 L. The SMFC reactor was fitted with a fluorescent bulb, guaranteeing a light level of 305 lux at the surface of the leachate. The lighting schedule followed an automated cycle of 12 hours of illumination followed by 12 hours of darkness. A data-gathering system was smoothly included in the SMFC reactor to simplify voltage readings. Both reactors utilized a cathode made of graphite, but they differed in terms of their anodes. The SMFC-cs reactor had a modified anode made of stainless steel that was electroplated with copper, while the SMFC-c reactor had a conventional anode made of copper. It is worth mentioning that both the cathode and anode have the same surface area, which is 67 cm². The anode was positioned 2 cm above the reactor base and maintained a constant distance of 4 cm from the cathode. The electrodes were connected using copper wire, which was linked with an external resistance (R) of 5.6 kΩ. Figure 1 provides a visual depiction of the arrangement of the SMFC reactor used in this investigation.

Modified anode preparation

The preparation of the modified anode followed a procedure inspired by the methodology outlined in the experiment conducted by Sayed

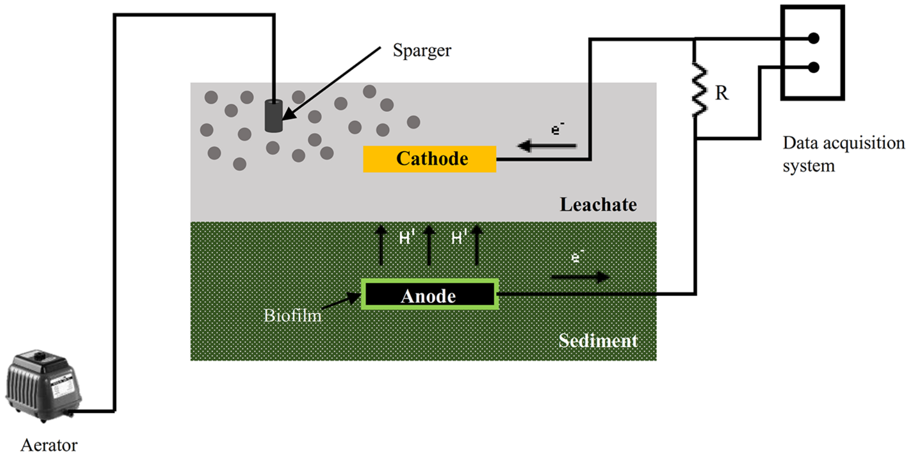

Figure 1. Schematic diagram of the SMFC reactor

et al. (2020), with certain adjustments. Initially, stainless steel (SS) plates underwent sanding, followed by a 5-minute immersion in ethanol (Merck, 99.5% purity). Subsequently, the SS plates were meticulously washed with distilled water and then subjected to an additional five-minute immersion in a 17% HCl (Merck) solution. After drying, the stainless steel plates underwent another round of washing with distilled water and were selected to serve as the cathode in the electroplating cell. In contrast, the anode was crafted from copper. Both electrodes were submerged in a 0.2 M $CuSO_4$ (Merck) solution, connected to a power source, and positioned at a distance of 1 cm. Employing a 3 V voltage, the electroplating process extended over an hour. The resulting copper-modified stainless steel (Cu-SS) anode underwent a finishing treatment, followed by thorough washing and air-drying throughout the night.

Analysis and calculation

Temperature (MAX6675 Thermocouple Temperature Sensor), pH (DFRobot Gravity pH Sensor kit V1), and voltage (INA219 Current and Voltage Sensor) measurements were taken daily using sensors connected to a data acquisition system (Arduino Mega 2560). Current and power were calculated following references from Sahu (2019) as written in Eqs. 1 and 2.

$$I = V/R \quad (1)$$

$$P = I \times V \quad (2)$$

where: I – electric current (mA), V – electric voltage (mV), R – external resistance (Ω), P – power (mW).

The effectiveness of pollutant removal was evaluated by measuring changes in biological oxygen demand (BOD) as well as ammonia, nitrate, and nitrite concentrations. BOD levels were monitored every five days using a BOD sensor analysis (VELP Scientifica). A 25 mL sample from the reactor was diluted with distilled water to a total volume of 100 mL and placed into the analysis bottle. The bottle was then sealed with the BOD sensor and incubated at 20 ± 2°C for five days. After the incubation period, the BOD value of the sample from the reactor was recorded and displayed by the sensor. Ammonia, nitrate, and nitrite concentrations were analyzed every two days using a UV-Vis spectrophotometer (Shimadzu Spectrophotometer UV-1800) and the optical density method. Standard HACH procedures were followed for all nitrogen tests: ammonia was measured using the salicylate method (HACH Method 10031), nitrate via the cadmium reduction method (HACH Method 8029), and nitrite using the ferrous sulfate method (HACH Method 8153).

RESULTS AND DISCUSSIONS

Bioelectricity production

The SMFCs use the redox gradients naturally occurring in aquatic sediments to produce energy. by moving electrons to the anode, bacteria that decompose organic materials in the sediment create a negative charge at the anode. Both direct electron transfer through the bacterial membrane's cytochromes and mediated electron transfer through the use of electron shuttles cause this. As electron acceptors are diminished at the cathode, a positive charge forms. Although oxygen is the most frequently utilized electron acceptor, other materials that can also receive electrons include nitrate, iron, manganese, and sulfate. A voltage potential is created as the electrons go from the anode to the cathode via the circuit.

Figure 2a presents the voltage generated by SMFCs using leachate effluent from the landfill leachate treatment unit as substrates. Notably, the SMFC-cs utilizing stainless steel anodes modified with copper manifest substantially higher voltages compared to SMFCs relying solely on pure copper anodes. This distinction is highlighted by the highest voltage recorded from the Cu-SS anode at 615 mV, in contrast to the pure Cu anode registering at 306 mV. This finding aligns with what has been reported by Sayed et al. (2020), who also modified the anode in an SMFC by electroplating stainless steel on carbon cloth. Their results showed that modified anodes provide higher power density and voltage ratings than unmodified ones. A study conducted by Nosek & Cydzik-Kwiatkowska (2022) yielded similar effects on the use of reduced graphene oxide (rGO) as a microbial fuel cell (MFC) anode. They explored utilizing an rGO anode and a combination rGO-iron (rGO-Fe) anode in MFCs fueled by wastewater. The MFC with the modified rGO-Fe anode produced a much higher voltage compared to the other configurations, 109.4±75.1 mV specifically. Furthermore, the power generated by the rGO-Fe MFC was 3.9 times greater than that of the MFC without any anode modification. Likewise, Fig. 2b illustrates the power density of the modified anode SMFCs. The power density of the modified anode exhibits a similar trend as the voltage, and its value is double that of the unmodified Cu anode, precisely measuring 4.8 mW/m^2 and 2.5 mW/m^2, respectively. Similar results were reported by Song et al. (2013), who examined coating stainless steel net electrodes with carbon nanotubes in SMFCs. Without any modification, the plain stainless steel net electrodes yielded a power density 3.2 times lower than that achieved with the carbon nanotube-coated electrodes. This significant improvement demonstrates the potential benefits of modifying the anode to enhance the performance in SMFC systems.

The electricity generation process is influenced by various factors, particularly from the perspective of the anode. These encompass the electrical conductivity and resistivity of the material deployed alongside the material's surface area. Notably, the modified anode's surface exhibits a rougher texture upon microscopic examination, as illustrated in Figure 3. The surface roughness or textural irregularity of the anode in the SMFCs can have a significant impact. An uneven surface can offer supplementary sites for microbial populations to establish colonization. The presence of irregularities on a surface might lead to an increase in the total surface area, hence providing more opportunities for microbial growth compared to a relatively smooth surface, as observed in the work of Sayed et al. (2020). Luo et al. (2013) also agreed that anode surface properties impact MFC performance. They demonstrated this by investigating how the electrochemical oxidation of carbon mesh anodes affects MFCs. Their results conveyed that modifications to create more favorable surface conditions on MFC anodes can lead to notable performance improvements.

Microbial populations have the ability to establish biofilms as they colonize the irregular

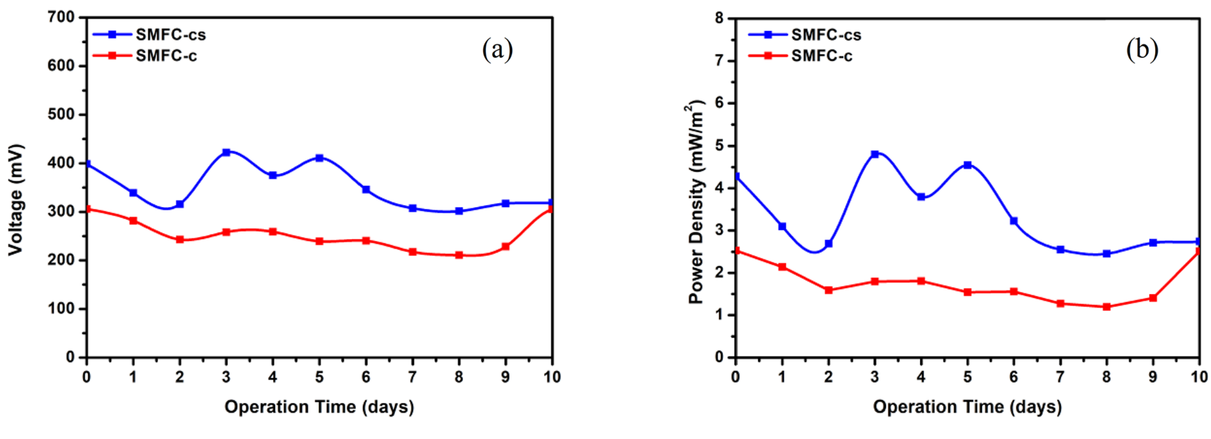

Figure 2. Voltage (a) and current density (b) generated using pure Cu anode and Cu-modified stainless steel anode (Cu-SS)

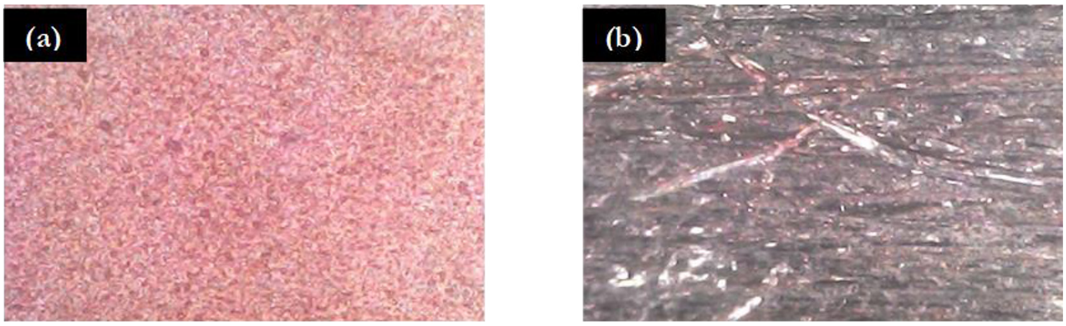

Figure 3. Morphological view of (a) copper-modified stainless steel (Cu-SS) and (b) copper (Cu) anodes using a microscope

surface of the anode. Biofilms contain diverse microorganisms, including electroactive bacteria, which are crucial for facilitating electron transfer processes. As a result, the existence of textural irregularities can stimulate heightened microbial growth by virtue of their expanded surface area. The presence of a varied microbial community on the anode's surface can improve electron transport. Certain bacteria in the biofilm can assist the flow of electrons from organic matter in the sediment to the anode, which is required for SMFCs to generate electrical current. Besides, factors beyond surface morphology, such as material electrical conductivity and resistivity, significantly impact the electron transfer process (Emalya et al., 2022). The comprehensive electrical conductivity and resistivity values for copper and stainless steel are detailed in Table 1. Although stainless steel's electrical conductivity is inferior to copper, incorporating copper nanoparticles effectively enhances its electrical conductivity. This multifaceted interaction underscores the intricate interplay of material properties that influence electricity generation within the SMFC system.

Operating parameters

The operational parameters of SMFCs play a pivotal role in shaping the metabolic activity of microorganisms, thereby exerting a profound impact on the electricity generation process (Logan & Rabaey, 2012). The SMFCs were operated without explicit control over temperature and pH conditions in this specific experiment. Consequently, no significant divergence emerged between the temperature and pH parameters observed in modified and unmodified anode SMFCs. The temperature dynamics inside the SMFCs are shown graphically in Figure 4a. At the beginning of the experiment, there was a discernible drop in temperature, followed by an upward trend until the fourth day. Then, the trend changed, and the temperatures began to drop until they stabilized on days nine and ten. The temperatures for the modified and unmodified anode SMFCs were 26.82°C and 26.97°C, respectively, at the end of the experiment. It's noteworthy that aside from the metabolic activity of microorganisms, temperature fluctuations within SMFCs can also be

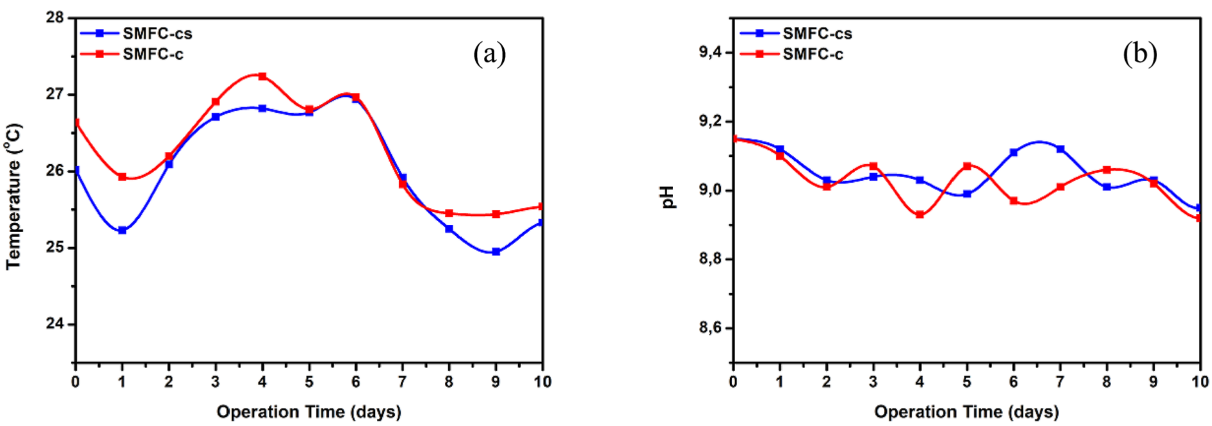

Figure 4. Changes in (a) temperature and (b) pH in the SMFC reactors

Table 1. Electrical conductivity and resistivity of copper and stainless steel materials

Materials	Electrical conductivity (S/m)	Electrical resistivity (Ω m)
Copper	5.85×10^{7}	1.7×10^{-8}
Stainless steel	0.14×10^{7}	7.2×10^{-5}

influenced by the ambient temperature surrounding the system (Emalya et al., 2022). This interplay highlights the complex interdependence between internal and external factors affecting the SMFC's temperature profile.

In the initial days of operation, microorganisms in the sediment of the SMFC undergo a critical adaptation phase. During this period, fluctuations in temperature and pH may be observed as the microbes accustom themselves to the new system (Emalya et al., 2023). Complex microbial, environmental, and electrochemical interactions drive changes as the microorganisms evolve to suit their habitat. This crucial adjustment stage can significantly transform the anodic environment, impacting overall SMFC performance. Figure 4b effectively demonstrates the observed variations in pH levels within the SMFCs during the duration of the experiment. Significantly, the pH fluctuations detected between the modified and unmodified anode SMFC were relatively small, exhibiting a continuous oscillation throughout the investigation. The average pH value for the modified anode SMFC was approximately 9.05±0.06, whereas the SMFCs of unmodified anode had a pH value of 9.03±0.07. The data provide evidence of a gradual yet persistent alteration in pH levels, highlighting the complex dynamics of maintaining pH homeostasis inside the SMFC setting.

The fluctuations in pH observed in the SMFCs are intricately linked to the many reactions occurring in the anodic and cathodic sections and the intricate metabolic activities of microorganisms. The pH profile seen throughout the experimental time is influenced by the equilibrium of these processes, as well as the general microbial dynamics of the system. The research conducted by Ghimire et al. (2019) and Emalya et al. (2022) highlights the crucial functions performed by microbial metabolism and electrochemical reactions in influencing pH fluctuations within SMFC. The complex interrelationship of various components highlights pH regulation's diverse and nuanced nature in these systems, enhancing our comprehension of the interaction between microbial activity and electrochemical processes.

Pollutant removal

Previous studies (Gul et al., 2021; Shen et al., 2022; Lawan et al., 2022) have demonstrated the potential of SMFC systems to address pollutant remediation in wastewater effectively. These findings highlight the theoretical and practical viability of SMFC systems for this purpose. These systems present a promising opportunity for mitigating the issue of wastewater contamination, serving as a means to reconcile the conflict between energy production and ecological preservation.

The Minister of Environment and Forestry Regulation No. P.59 of 2016 delineates the regulatory framework governing leachate waste disposal. This framework emphasizes the significant criteria that require attention, such as pH, COD, BOD, TSS, N-total, mercury (Hg), and cadmium (Cd). Adherence to these quality requirements is imperative due to its significant impact on human health and the preservation of aquatic habitats. In this experiment, the effectiveness of SMFC in reducing organic compounds was measured in terms of BOD. The leachate substrate's initial BOD concentration was recorded as 1096 mg/L in the experimental setting. Nevertheless, the BOD value in both reactors rose on the fifth day, possibly attributed to the dissolution of organic compounds from the sediment into the bulk of leachate above it. However, as the microorganisms acclimated, the BOD value subsequently declined until the conclusion of the experiment. At the completion of the trial, significant reductions in both SMFC-cs (modified anode) and SMFC-c (unmodified) were seen, with BOD concentrations reaching 208 mg/L and 256 mg/L, respectively. The BOD removal on the modified anode reached 81.02%, while that of the unmodified anode was 76.64%. This confirmed that SMFC-cs with modified anodes improved the BOD removal efficiency compared to unmodified SMFC-cs. The research results published by Yang et al. (2022) also demonstrated that anode modification in SMFC systems affects the electron transfer process at the electrodes, thereby improving their electrochemical performance. The removal of organic compounds reported in their study reached 82.1% on the modified anode. Similar positive effects from modified anodes have been demonstrated by others as well. Ajab et al. (2020) successfully

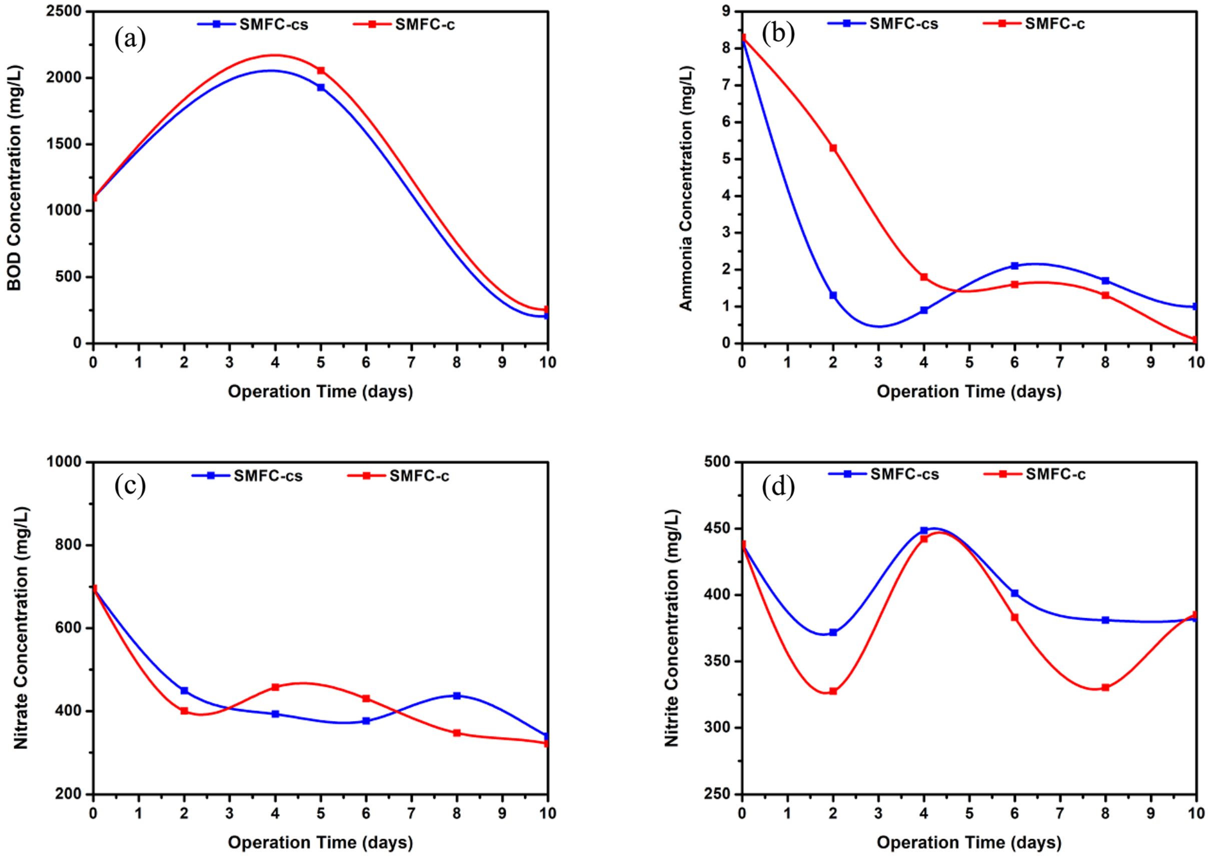

Figure 5. Changes in (a) BOD, (b) ammonia, (c) nitrate, and (d) nitrite concentrations

recovered approximately 61% of organic compounds from aqueous solutions using Ti/RuO_2 anodes coated with varying concentrations of $RuCl_3 \cdot H_2O$ solution. Morovati et al. (2022) presented comparable findings in their analysis of carbon felt anodes coated with cobalt manganese oxide ($MnCo_2O_4$) for COD and diclofenac sodium removal. Their results exhibited higher COD reduction with the $MnCo_2O_4$-coated anodes versus plain carbon felt. These studies further illustrate the potential of tailoring anode materials and surface properties to achieve enhanced output in applications like wastewater treatment. The nature of the modifications may vary, but properly engineered anodes appear widely effective at elevating system performance.

The elimination of nitrogen pollution is inherently interconnected with the complex nitrogen cycle. The biogeochemical process described above is a complex mechanism that facilitates the conversion of inert atmospheric nitrogen into diverse chemical forms vital for living sustenance. The process entails the assimilation of atmospheric nitrogen (N_2) by diazotroph bacteria, its subsequent transformation into ammonia, the oxidation of ammonium by Nitrosomonas bacteria resulting in the formation of nitrite, and the subsequent conversion of nitrite into nitrate facilitated by Nitrobacter bacteria. The denitrification process involves converting nitrate to nitrogen (N_2), resulting in its subsequent release into the atmosphere. Furthermore, the process of ammonification involves the decomposition of organic matter, resulting in the production of ammonium ions that can subsequently undergo conversion into nitrite.

The initial concentrations of ammonia, nitrate, and nitrite were equivalent for both reactors in the conducted experiment, measuring 8.3 mg/L, 696 mg/L, and 438 mg/L, respectively. The process of eliminating nitrogen molecules, specifically ammonia, nitrate, and nitrite, is depicted in Figure 5(b) to Figure 5(d). The ammonia concentrations for SMFC-cs and SMFC-c were observed to be 1.0 mg/L and 0.1 mg/L, respectively. The removal efficiencies for these systems were determined to be 88% and 99%, respectively. The experimental results demonstrated a 50% reduction in nitrate concentration, with levels decreasing from an initial value of 696 mg/L to 339 mg/L in the SMFC-cs system and 321 mg/L in the SMFC-c system. The nitrite concentrations in the SMFC-cs and SMFC-c samples were found to be 382 mg/L and 385 mg/L, respectively. Compared to the study conducted by Yang et al. (2022), this study showed relatively similar effectiveness in removing ammonia. Yang et al. (2022) reported research results in the field of marine aquaculture wastewater. Their findings showed that the ammonia removal efficiency of the modified anode reached 95.8%.

The study conducted by Ghimire et al. (2019) sheds light on a significant correlation between the elimination of nitrogen compounds and voltage. The findings indicate that higher voltage inputs lead to increased removal efficiency. Nevertheless, the results of this particular experiment did not reveal any statistically significant disparities in the elimination of nitrogen molecules between SMFCs equipped with modified and unmodified anodes. As mentioned above, the result highlights the necessity for additional research to clarify the complex interaction of variables that impact the elimination of nitrogen compounds in SMFCs.

CONCLUSIONS

The electroplating process, which introduced copper particles onto stainless steel, yielded remarkable enhancements in energy production within the SMFC system. Specifically, the SMFC equipped with the modified anode exhibited a substantial boost in electricity generation, approximately doubling the output compared to its unmodified counterpart. This adaptation played a pivotal role in elevating power generation while crucial operational parameters such as temperature and pH remained relatively stable. The anode SMFCs, whether modified or unmodified, maintained consistent temperatures at 26.82 °C and 26.97 °C, respectively. Similarly, the mean pH of the modified anode SMFC slightly exceeded that of the unmodified version, recording values of 9.05±0.06 and 9.03±0.07, respectively. These fluctuations in temperature and pH during SMFC operation result from a complex interplay of factors, including ambient conditions, reactions in the anodic and cathodic chambers, and the dynamic metabolic processes of microorganisms.

In addition to the noticeable increase in energy generation, the modified anode SMFC demonstrated a notable improvement in reducing BOD levels. Specifically, it achieved a 4% enhancement compared to the unmodified anode-equipped SMFC. This multifaceted performance improvement underscores the potential of anode modification to comprehensively enhance the functionality of an SMFC, addressing both energy generation and pollutant removal. Further exploring the intricate relationships among the altered anode, microbial behavior, and operational parameters could offer valuable insights into the fundamental mechanisms driving these enhancements. These findings can potentially advance the SMFC technology for sustainable energy production and wastewater treatment, contributing to environmental and energy sustainability goals.

Acknowledgments

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