BCSIR

Available online at www.banglajol.info/index.php/BJSIR

Bangladesh J. Sci. Ind. Res. **59(4)**, 249-260, 2024

Short Communication

BANGLADESH JOURNAL OF SCIENTIFIC AND INDUSTRIAL RESEARCH

ISSN. 0304-9809

Effect of electrode combinations on bioelectricity generation in microbial fuel cell using wastewater from pulp and paper industry

H. Iqbal[1,2], A. Bhowmik[1,2], J. Uddin[1], S. Yasmin[3], D. Islam[4], S. M. T. Abedin[2], M. S. Jamal[1]* and M. Hossain[1]*

[1]*Institute of Energy Research and Development, Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka 1205, Bangladesh*

[2]*Department of Chemistry, Jahangirnagar University, Savar, Dhaka 1342, Bangladesh*

[3]*Institute of National Analytical Research & Services, Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka 1205, Bangladesh*

[4]*Biomedical and Toxicological Research Institute, Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka 1205, Bangladesh*

ARTICLE INFO

Received: 28 August 2024

Revised: 04 December 2024

Accepted: 09 December 2024

eISSN 2224-7157/

DOI: https://doi.org/10.3329/bjsir.v59i4.75663

Abstract

Microbial fuel cell (MFC) technology is an innovative approach to sustainable power generation from wastewater. This study aims to use non-toxic metal (Al, Zn, and Cu) and carbonaceous electrode sets with reduced internal resistance in the six MFCs to reduce the cost and toxicity of electrodes and produce electricity along with wastewater management in the pulp and paper industry. Among them, MFC-5 (Zn as an anode, Cu as cathode material, and $CuSO_4.5H_2O$ as electrolyte) shows maximum voltage, current, and power density at 1185 mV, 4.79 mA, and 939 mW/m^2 respectively. Another cell (MFC-2) with carbonaceous cathodic material and containing no electrolyte solution, exhibits the second highest output voltage, current, and power density at 1193 mV, 2.69 mA, and 533 mW/m^2 respectively. This research also demonstrated that MFC-5 and MFC-2 effectively removed 94.8±1% and 93.6±0.5% of biochemical oxygen demand (BOD) after 15 days, highlighting their potential for wastewater treatment.

Keywords: Wastewater; Pulp and paper; Microbes; Secondary energy; MFC

Introduction

Environmental-friendly alternative energy sources and bioenergy production are gaining more interest due to increasing global energy demand and depletion of fossil resources (Guo *et al.* 2015; Mandley *et al.* 2020; Popp *et al.* 2014). The possibility of bioenergy generation through microbial metabolism has opened new opportunities to replace conventional fossil fuel-based energy because of its renewable and ecological character (Chaturvedi and Verma, 2016). On the other hand, industries are starting to focus on wastewater recycling as part of sustainable development and improved wastewater management (Chen *et al.* 2020; Palanisamy *et al.* 2019). Therefore, the reuse and recycling of water have become crucial to reducing the overuse of freshwater in natural ecosystems. Therefore, in the current context, energy generation and wastewater management are two major challenges that researchers have to address (Gude, 2015). Bioelectrochemical systems like microbial fuel cell (MFC) technology are one of the possible graceful solutions that can integrate these two issues (Davis, 1967; Logan *et al.* 2015).

MFC is an innovative microbial electrochemical technology with the potential to extract chemical energy and release resource recovery from organic wastewater contaminants via biocatalytic reactions (Gajda *et al.* 2018; Omine *et al.* 2018; Sahu, 2019). The bacteria in the MFC anode chamber can catalyze biodegradable organic materials and generate electrons which subsequently pass through the external circuit, are received by the electron acceptor at the cathode,

*Corresponding author's e-mail: mosharof@bcsir.gov.bd, msjdubd@gmail.com

and generate electricity (Prasad and Tripathi, 2017; Timmis, 1995). Traditional fuel cells, such as hydrogen fuel cells, require energy to generate hydrogen, while MFCs do not require any external energy source, produce no additional greenhouse gases, and are self-contained and biodegradable (Iigatani *et al.* 2019; Jayashree *et al.* 2019; Subha *et al.* 2020). These key advantages over conventional fuel cells make MFC a potential contender for upcoming sustainable green energy sources (M. Li *et al.* 2018).

The MFCs are typically designed as a two-chamber system with the bacterial biofilm placed at the anode chamber and kept isolated from the cathode chamber through a polymeric proton exchange membrane (PEM) (Chakraborty *et al.* 2020; Das, 2020). Air cathodes, in which air bubbles are fed into the cathode chamber to saturate the dissolved oxygen to the electrode, are one of the common tactics utilized in MFCs (Lawson *et al.* 2020). However, exo-electrogenic bacteria at the anode chamber oxidize fuel to produce CO_2, and release electrons, and protons (Sonawane *et al.* 2020). Protons flow from the anode to the cathode chamber through the PEM, and the electrons flow to the external circuit. The protons recombine with electrons at the cathode and form water in the presence of free oxygen (Katal and Pahlavanzadeh, 2011; Ratheesh *et al.* 2021). However, the use of PEM is very expensive. For this reason, in this study, a salt bridge is used as a substitute for PEM. The movement of electrons from the anode to the cathode is a favorable and spontaneous process that leads to energy production in the MFCs. The general reaction involving the MFCs is (Uddin *et al.* 2021):

$$C_6H_{12}O_6 + 6H_2O \longrightarrow 6CO_2 + 24H^+ + 24e^- \quad \text{.....(1)}$$

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

The oxygen reduction reaction is crucial for determining MFC efficiency, involving the transfer of four electrons to form H_2O under acidic conditions (Harnisch and Schröder, 2010). The standard reduction potential of O_2 in this reaction is +1.23 V at pH 0 and 1 atm, as verified by thermochemical calculations and empirical data (Pegis *et al.* 2015). However, such conditions are rarely achievable in MFCs since bacteria, predominantly neutrophils, grow optimally near neutral pH (~7.0), where the reduction potential decreases significantly (S. Li *et al.* 2017). For effective operation, high current density and a positive onset potential are critical. While noble metal electrocatalysts like platinum meet these criteria, their high costs and scarcity hinder large-scale application. Cheaper alternatives, such as copper and carbon-based materials, offer higher reduction potentials and cost-efficiency, making them promising substitutes (Prasad and Tripathi, 2017; Wang *et al.* 2013).

Several attempts have been made at MFC application using synthetic and raw wastewater such as paper industry wastewater, brewer wastewater, tannery wastewater, domestic wastewater, food processing wastewater, and starch processing wastewater for treatment and power generation (Huang *et al.* 2021; Senthilkumar and Naveenkumar, 2023; Subha *et al.* 2020). The composition of wastewater from the pulp and paper industry varies significantly between mills. Wastewater typically has a wide range of biochemical oxygen demand (BOD) values ranging from 5000 to 15000 mg/L (Simate, 2015). More than 250 distinct inorganic and organic compounds have been found in the pulp and paper industries' effluents, and MFCs can effectively eliminate these contaminants (Lebeer *et al.* 2011). A few studies have been conducted using water samples from pulp and paper industries worldwide, but no such operation has been developed in Bangladesh. In Bangladesh, the per capita consumption of paper and cardboard is approximately 3.5 to 4 kg, compared to the global average of around 50 kg. The primary pulp and paper industries in Bangladesh are located in the Dhaka and Chittagong regions, with a total of 80 paper mills (Quader, 2012). In Bangladesh, the pulp and paper industries generate a large amount of wastewater because of the low percentage of reused water and poorly organized wastewater treatment (Karmaker *et al.* 2022).

To solve the above issues, in this study, we have treated the pulp and paper industry wastewater samples with MFCs, placed different non-toxic, inexpressible metals i.e. Zn, Al, and additionally, carbon electrode combinations, recorded the continuous voltage, current, and power output for 15 days and checked the parameters of the treated water. Initially, we had used aerial oxygen flow as an oxidizing species at the cathode chamber but the pH condition of the cell and the nature of the electrodes subdued the efficiency in some cases. However, the electrolyte solution has far-reaching effects, expanding power yield multiple times. Finally, after 15 days of treatment water parameters were measured and Field emission scanning electron microscopy (FESEM) was carried out on bacterial biofilms for identification and investigation of their adhesion properties.

Materials and method

Collection of Wastewater Samples and Characterization

The pulp and paper industry wastewater sample was collected from a Meghna pulp and paper mill, in Dhaka, Bangladesh. The grab sampling technique was applied by dipping down a 30-liter gallon bottle into the water reservoir and placed at ambient temperature for 6 hours to settle down solid particulates (Barrows *et al.* 2017). The initial pH of the water sample was then measured by using a

digital pH meter (Radiometer PHM 240). A conductivity meter (CDM 230) was employed to measure the conductivity. Total suspended solids (TSS), and total dissolved solids (TDS) were measured by using ASTM D5907 methods. The turbidity of the water sample was measured by a digital turbidity meter (Super Scientific 860040 Turbidity Meter) (Gaikwad and Munavalli, 2019).

The standard BOD_5 technique is used to quantify the biodegradable component of sewage and wastewater. The BOD_5 is a gauge of the amount of dissolved oxygen needed for the biochemical oxidation of organic components of an effluent sample seeded with a microbial culture for over 5 days at 20 degrees Celsius (Kim *et al.* 2003). To Measure BOD_5, the effluent sample is poured completely into a special BOD bottle: a glass bottle with a "turtleneck" and a ground glass stopper. The initial and final DO is measured with a V-TECH BOD meter and BOD_5 is calculated by the conventional method.

Preparation of Electrode Materials

In this experiment, carbon, zinc, and copper plate electrodes with 5 cm width and 12 cm length are used (Fig. 1). Because of the very negative reduction potential, the non-toxic transition metal zinc and its grid form as well as the most widely abundant metal aluminum have been chosen for the anode. This was done to produce a higher potential difference between the two sides of the cell. Zn has a reduction potential of -0.76 volts, whereas aluminum has a reduction potential of -1.66 volts. The carbonaceous electrode served as the benchmark by which correlations were made between the metal electrodes and the various MFCs that were found in the literature review.

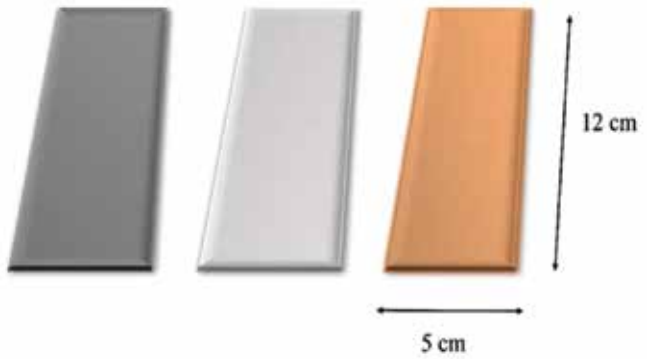

Fig. 1. Pictorial representation of carbon, zinc, and copper plate electrode

Due to the higher positive reduction potential ($E^0 = +0.34V$), the cheaper unprecedented transition metal copper has been employed as cathodes in two out of six cells which my expected to improve the potential difference from the concept of the traditional fuel cell mechanism (Yang and Reddy, 2014). Albeit, areal oxygen at atmospheric pressure was adopted at the cathode chamber of the first four MFCs. The later two cathode chambers were treated with electrolyte solutions. The potential difference and power generation would greatly rely on the electrode surface-oxidant coalescence behavior. The combination of anode and cathode materials of six MFCs is shown in Table I.

Table I. MFC IDs and related electrode materials

MFC ID	Anode materials	Cathode materials	Cathodic electrolyte
MFC -1	Zn plate	Cu plate	--
MFC -2	Zn grid	Carbon plate	--
MFC -3	Al grid	Carbon plate	--
MFC -4	Carbon plate	Carbon plate	--
MFC -5	Zn plate	Cu plate	$CuSO_4.5H_2O$
MFC -6	Carbon plate	Carbon plate	$Fe_2(SO_4)_3$

Construction of MFC

The most fundamental part of MFCs was assembled using a lightweight plastic container (Fig. 2) that was divided into an anode compartment and a cathode compartment (each container has a volume of 4L). A piece of plastic tubing measuring 12 centimeters in length and 1.5 centimeters in diameter ran between the two different chambers to join them. At a temperature of 75°C 5g of agar powder were combined with 100 m of a 1M KCl solution, and the mixture was agitated for 2 h. After that, the viscous solution was transferred into the plastic tube, where it was later cooled and dried to become the salt bridge for that particular MFC. While the cathode was being replenished with distilled water, industrial muck and bacterial inoculum were poured into the anode chamber. Electrode pairs were then set in position, and the anode chamber was enclosed with a protective sheet to prolong the anaerobic environment.

To determine the current generated by the fuel cell, the current and voltage were measured using a digital multimeter (Model UT136B+, UNI-T, USA). The data were carefully recorded at 24-h intervals. The power density was calculated from the experimental current value and electrode area.

Fig. 2. (a) Schematic diagram of MFCs (b) Experimental setup of MFCs

FESEM Analysis of bacterial film

Bacterial morphological structures were analyzed by field scanning electron microscope (FESEM; JSM-7610F, JEOL, Japan (accelerating voltage: 5 kV, magnification: 10000X and 20000X) used to take a photo. After 15 days of water sample treatment, the anode was carefully recovered from the water sample. The bacterial membrane attached to the electrode was then treated with 5% formalin solution for more than 2 h to immobilize the bacterial cell wall. The sample was then dried for more than 6 h at 55 °C and the morphology of biofilm was analyzed by FESEM (Koivuluoto *et al.* 2010; Lebeer *et al.* 2011).

Results and discussion

Properties of wastewater

The initial parameters of the wastewater sample were reported. The sample was determined to have a high BOD content (12,853 mg/L) and to be somewhat acidic, with a pH value of 5.9. The presence of metal ions and anions in the sample contributed to the sample's higher value of conductivity, which was measured at 965 µS/cm. The total dissolved solids (TDS) and the total suspended solids (TSS) measured at 3958 and 2369 mg/L. The sample had a very turbid physical appearance since it included a high concentration of dissolved organic components as well as inorganic metal salts. A comprehensive listing of all of the values is shown in Table II.

Table II. Properties of wastewater

Parameter	Value
pH	5.9±0.05
BOD_5 (mg/L)	12853±9.8
Conductivity (µS/cm)	965±4.5
TSS (mg/L)	2369±9.6
TDS (mg/L)	3958±10.5
Turbidity (NTU)	601±3.7

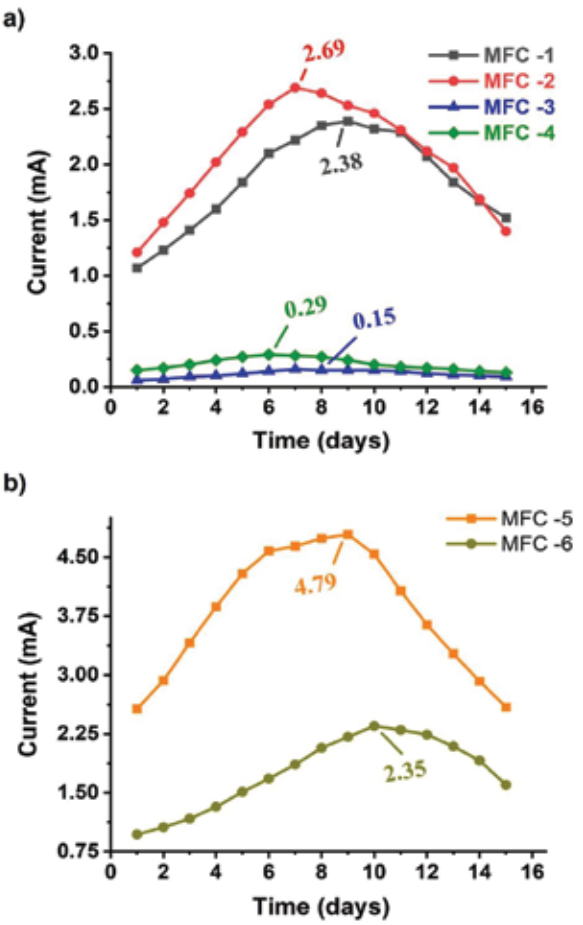

Fig. 3. Variation of output current in different MFCs with time (a) MFC-1, 2, 3, 4 and (b) MFC-5, 6

Electricity generation

The comparison of output current and output voltage of MFC-1, 2, 3, 4, and MFC-5, 6 are shown in Fig. 3(a-b) and Fig. 4(a-b). These figures depict that the output voltage and current patterns for MFC-1 and 2 are almost similar. The MFC-1 was constructed with zinc and copper plate and in MFC-2, zinc grid, and carbon plate were used as the electrodes. The maximum output voltage and current for MFC-1 were 1152 mV and 2.38 mA while for MFC-2, it was 1193 mV, and 2.69 mA respectively. The use of different electrodes as cathodes, in the absence of an electrolytic solution hasn't produced any significant impact on electricity generation. However, the use of the electrolytic solution in the cathode compartment in MFC-5, constructed with zinc and copper electrodes, causes a significant impact on the output current (4.79 mA). The output current is almost twice that of MFC-1 (2.38 mA) with an almost similar value of output voltage. The output voltage of MFC-1 is 1152 mV and for MFC-5, it is 1185 mV. Furthermore, the decrease in the current value of MFC-4 which contains carbon material both as anode and cathode, is due to the breakdown of microbial food nutrients during the MFC process and an increase in side products in the form of CO_2 which causes a toxic effect on cells. Another reason is the absence of an electrolyte solution (Obileke *et al.* 2021).

The power density of prepared MFCs is shown in Fig. 5(a-b). Though Al has the least value of reduction potential (E^0 = -1.66V), the MFC-3 cell, fabricated with the aluminum grid and carbon electrode, was found to have the least amount of power density (7 mW/m^2 of peak value). This value is approximately one-fifth of the MFC-4 counterpart, where carbonaceous electrodes were placed in both

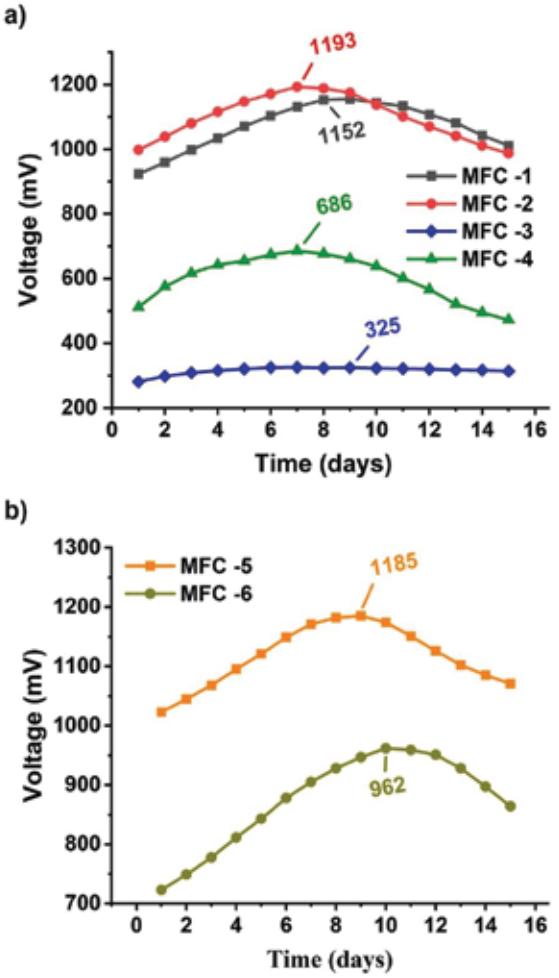

Fig. 4. Variation of output voltage in different MFCs with time (a) MFC-1, 2, 3, 4 and (b) MFC-5, 6

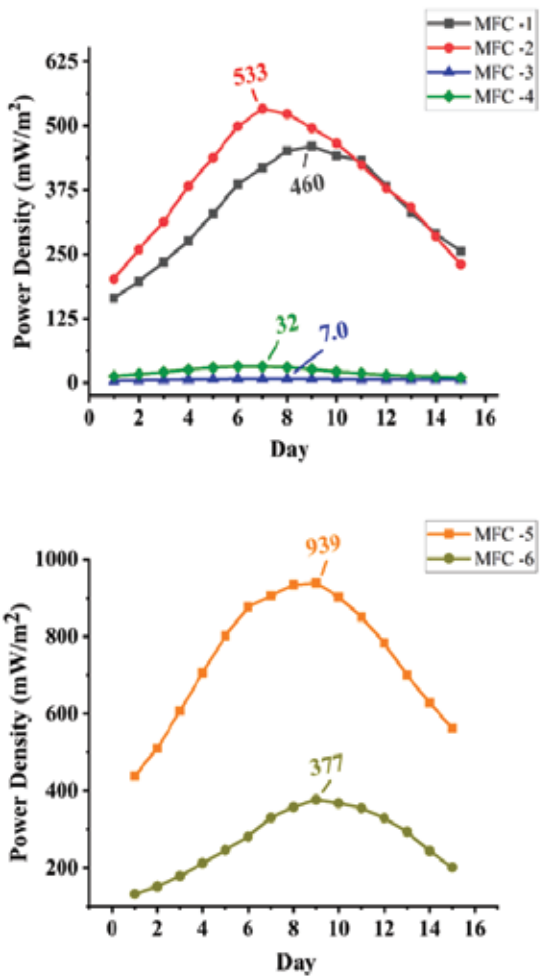

Fig. 5. Variation of power density in different MFCs with time: (a) MFC-1, 2, 3, 4 and (b) MFC-5, 6

compartments. This discrepancy is due to the difficulty of microorganisms to adhere to the surface of aluminum, a finding that was verified further by the fact that bacterial biofilms could not be examined by scanning electron microscopy (SEM). Moreover, the energy density pattern of MFC-3 tended to reluctantly decrease over 15 days of treatment. MFC-1 (Zn anode) and MFC-2 (Zn grid) showed sufficient good results since they reached the tip value of 460 mW/m^2 and 533 mW/m^2 which is almost 14th and 17th greater than the carbonaceous counterparts respectively. MFC-4 and MFC-6 are equivalent since both have carbon cathode and anode electrodes. In MFC-4, atmospheric oxygen served as the electron acceptor while 1.0M $Fe_2(SO_4)_3$ solution was used as the electron acceptor in the cathode chamber of MFC-6. MFC-4 has a maximum power density of 32 mW/m^2, whereas MFC-6 has a maximum power density of 377 mW/m^2. With an apex value of 939 mW/m^2, the MFC-5 cell with a 1M $CuSO_4.5H_2O$ solution had the highest power density among the six examined cells, and its power density was almost twice that of its MFC-1 counterpart. The highest power density is observed due to the use of $CuSO_4.5H_2O$ solution in MFC-5. Due to the higher standard reduction potential of $CuSO_4.5H_2O$ solution, Cu^{2+} can easily accept the electrons. Hence, maximum current is produced. Ultimately the power density increases in MFCs which contain electrolytes in a cathode chamber. Because of the augmentation of bacterial metabolism, the power density is maximum near 7-10 days in all MFCs. And then the gradual decline in power density has been attributed to bacterial nutrient depletion and toxic by-products (Obileke *et al.* 2021).

Decontamination of water sample

MFC treatment of wastewater samples significantly reduces the pollutants of the water sample, which can be clearly understood from the analysis of the final sample after treatment (Fig. 6). The initial BOD of the water sample was reported as 12,853 mg/L. The percentages of removal were calculated according to the following equation

$$\text{BOD removal (\%)} = \frac{(C_i - C_f)}{C_i} \times 100$$

Where, C_i = Initial BOD concentration (mg/L) and C_f = Final BOD concentration (mg/L). The maximum 94.8±**1**% BOD reduction is shown by MFC-5 (Fig. 6). MFC-2, 1, and 6 also showed better performance with percentages of 93.6±0.5%, 91±0.9%, and 89.7±0.5% respectively. MFC-3 showed the lowest value with a BOD

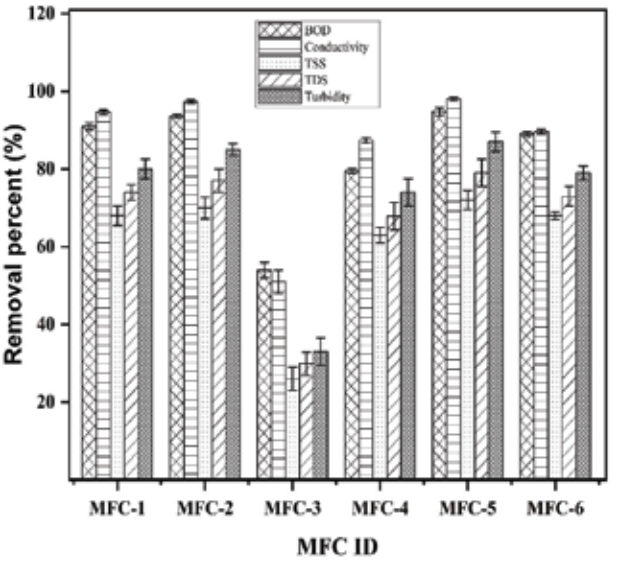

Fig. 6. Variation in removal efficiency of water parameters across different MFC configurations after 15 days of treatment

recovery of 54±2%. The results of MFC-3 were also consistent with the power generation as it shows the lowest power generation. Conductivity is also significantly reduced from the initial value after treatment. Samples taken from MFC-2, 5, and 6 experienced a reduction of conductivity at 97.3±0.5%, 98±0.5%, and 89.6±0.6% respectively, while MFC-1 decreased by almost 94.6±0.6%. MFC-3 also kept consistency with the lowest conductivity reduction value (51±3%). However, the decrease in conductivity also has a significant effect as it is a means of removing ionic compounds and heavy metals from the water sample. Total Suspended Solids (TSS) and Total Dissolved Solids (TDS) are also important indicators to measure pollutant recovery which is also related to water turbidity. As BOD and conductivity decreased after MFC treatment, TSS, TDS, and turbidity followed the same trend. These values decrease in sequence MFC-5, 6, 2, 1, 4, and 3 (Table III).

FESEM Analysis of bacterial biofilm

The microbial populace in the anode compartment is a crucial biological factor for determining the effectiveness of electron transport. Particularly, organic substrates provide carbon sources and electron donors associated with the outnumber of processes (Xiao and He, 2014). The substrates enter the glycolysis pathway, undergo a series of reactions, and then enter the tricarboxylic acid cycle and electron transport cycle to generate adenosine triphosphate

Table III. Wastewater samples after fifteen days of treatment with different MFCs

Sample	BOD_5 (mg/L)	Conductivity (μS/cm)	TSS (mg/L)	TDS (mg/L)	Turbidity (NTU)
MFC-1	1032±8.5	52±0.45	758±1.5	1029±6.5	120±3.7
MFC-2	751±4.2	26±1	711±1	910±4.2	90±1.5
MFC-3	5913±10.5	467±4.3	1753±8.5	2771±7.5	403±1
MFC-4	2628±6.8	121±3.6	877±3.6	1267±4	156±0.87
MFC-5	657±2.5	17±0.06	663±2.8	831±2.5	78±2
MFC-6	1314±4.3	100±6.8	758±0.8	1069±7.5	125±3.5

(ATP). In this synthesis mechanism, a progression of phases is engaged with electron transport e.g. NADH coenzyme Q reductase, nicotinamide adenine dinucleotide (NADH), ubiquinone, succinate dehydrogenase, cytochrome bc1, cytochrome c, cytochrome oxidase from microbial to the cell layer to electron acceptor employing a redox reaction and the prospective microorganisms with the capability of external electron transfer are being used for MFC (Konovalova *et al.* 2018; Kumar *et al.* 2013).

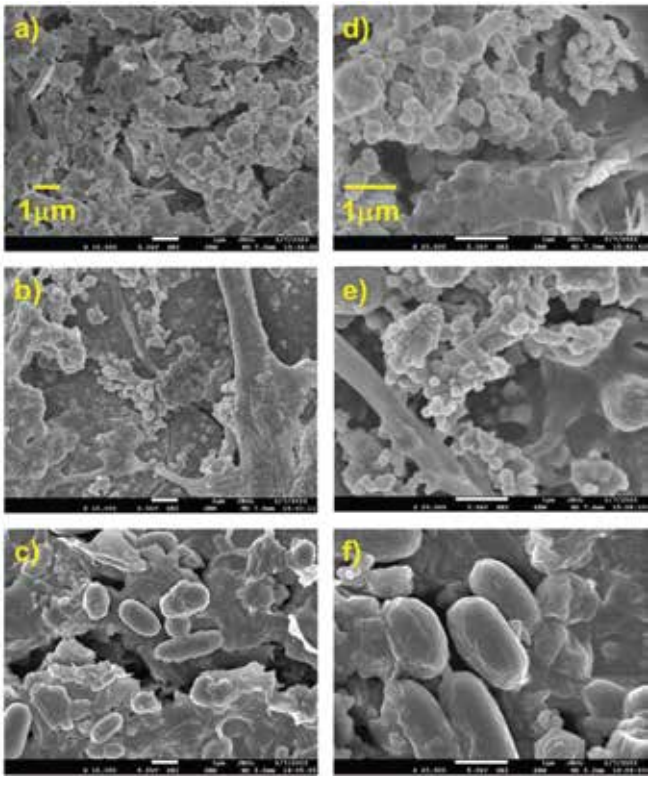

Fig. 7. FESEM image of the bacterial layer to the anode of (a) MFC -1, (b) MFC -2, (c) MFC -4 for low magnification, and (d) MFC -1, (e) MFC -2, (f) MFC -4 for high magnification

There is a significant amount of lignocellulose-containing material content in the pulp and paper industry. Lignin, hemicellulose, and cellulose are the components that makeup lignocellulose material. These constituents may be hydrolyzed into their smaller components and utilized as feedstocks in processes that aim to valorize lignocellulose (Brown *et al.* 2021). Microorganisms such as Bacillus sp., Paenibacillus sp., Geobacter sp., Clostridium sp., and Citrobacter sp. make up a significant portion of the collection. It has been demonstrated that these bacterial species can break a range of pollutants, such as adsorbable organic halides and substances with high chemical oxygen demand (COD), and release electrons to the anode via a technique known as extracellular electron transfer (EET) (Xiao and He, 2014). In this study, the compacted biofilm on the anode was examined with FESEM at two magnifications powers i.e. at 10,000X and 20,000X (Fig. 7(a-f)). Similar types of bacterial nano-threads (Geobacter) were found for MFC-1 and MFC-2 but on carbon plate, rod-shaped bacteria (Bacillus) predominate. This phenomenon is also consistent with the electricity generation plot. MFC-1 and MFC-2 have an almost similar pattern of output voltage, current, and power density. Whereas the voltage for MFC-4 is almost half and the output current and power density are almost 10 times lower than the former. However, the bacterial layer couldn't be possibly isolated for MFC-3 where the Al grid was utilized as an anode electrode.

Performance analysis

A substantial number of MFC evaluations were reported to be based on sodium acetate as an anodic electrolyte or a synthetic wastewater solution; nevertheless, in our investigation, we exploited wastewater directly from the paper industry to assess the feasibility. Both the initial water parameters, such as BOD, Conductivity, TDS, and TSS, and the final water

parameters were recorded for each cell as a whole. Only a handful of the studies that were looked at indicated a COD recovery value of less than 70%. We have reported the BOD levels after treatment and discovered a maximum value of 94.8±1% BOD eradication after 15 days of treatment. This finding is consistent with the findings of previous research and is thus plausible.

A combination of zinc and copper electrodes in the presence of a copper sulfate solution as a catholyte was discovered to have the highest power density (939 mW/m^2) among the various metallic electrode material combinations that were used in this study (Table IV). These combinations were both inexpensive and readily available. In the end, the bacterial morphology was investigated using FESEM, and the results

Table IV. Comparative study of MFC performance

Sample	Running Period	Electrode Materials	Cathodic Electron Acceptor	Maximum Power Density (mW/m^2)	FESEM Image	BOD / COD Removal	Reference
Industrial Wastewater (Sugar, Leather, pulp & paper Industry)	9 days	Carbon paper, carbon plate, graphite		5.1	Reported	51% COD removal	(Sahu, 2019)
Synthetic wastewater, Industrial Wastewater	7 days	Iron plate, Graphite plate	$KMnO_4$, K_3FeCN_6	Not reported	Reported	84% BOD removal	(Jayashree *et al.* 2019)
Sodium Acetate		Graphite fiber brush, Pt plate	Ferricyanide	2460	Not Available	Not reported	(Lawson *et al.* 2020)
Synthetic lactate wastewater	21 days	Carbon cloth, $MnFe_2O_4$/PANI	Air		Reported	86% BOD removal	(Khilari *et al.* 2015)
Sodium acetate	6 days	Geopolymer graphite, Portland cement graphite	Fumarate	Not reported	Reported	Not Reported	(S. Zhang *et al.* 2021)
Pulp & paper industry wastewater		Biochar with metal	Potassium ferric solution	47	Reported	64% COD removal	(Senthilkumar and Naveenkumar, 2023)
Sodium Acetate	1 year	Activated Carbon: Carbon Black	Air	1560	Not Available	Not reported	(X. Zhang et al., 2014)
Pulp & paper industry wastewater	15 days	Zinc, Carbon plate	Air	533	Reported	92% BOD removal	This study
Pulp & paper industry wastewater	15 days	Zinc, Copper	$CuSO_4.5H_2O$	939		93% BOD removal	This study

showed that the development of bacteria is heavily reliant on the electrode material. On the other hand, comparison information from other research could not be located.

Conclusion

In conclusion, non-toxic metal (Al, Zn, and Cu) and carbonaceous electrode sets with reduced internal resistance were applied in the six MFCs to reduce the cost and toxicity of electrodes and produce electricity using wastewater from the pulp and paper industry. The investigation revealed that certain electrode combinations may successfully outperform the currently available data sets. Surprisingly, the power production and performance of MFCs depended mostly on bacterial growth at the negative electrode. Geobacter effectively flourished on the Zn and its grid electrode, which provided the maximum power density, while considerable bacterial biofilm could not be collected for Al, even though it was predicted to yield the highest power density due to its very negative reduction potential. Bacillus proliferated well on carbonaceous electrodes but did not generate substantial power under cathodic areal conditions. Finally, MFC-5 (Zn as the anode, Cu as cathode material, and $CuSO_4.5H_2O$ as electrolyte) shows maximum voltage, current, and power density such as 1185 mV, 4.79 mA, and 939 mW/m^2 respectively. Furthermore, the cell MFC-2 with carbon plate as cathodic material and containing no electrolyte solution exhibits the second highest output voltage, current, and power density at 1193 mV, 2.69 mA, and 533 mW/m^2 respectively. The characteristics of the water after 15 days of treatment were also verified. MFC-5 and MFC-2 exhibited the highest level of rectification with 94.8±1% and 93.6±0.5% of BOD removal.

Acknowledgment

The authors are thankful to the Institute of Energy Research and Development (IERD), Bangladesh Council of Scientific and Industrial Research (BCSIR) for laboratory support. This research is conducted under the R&D project entitled "Production of bioenergy from waste materials through Microbial Fuel Cell".

References

Barrows APW, Neumann CA, Berger ML and Shaw SD (2017), Grab: Vs. neuston tow net: A microplastic sampling performance comparison and possible advances in the field, *Anal Methods.* **9**(9): 1446–1453. https://doi.org/10.1039/c6ay02387h

Brown DM, Pawlak J and Grunden AM (2021), Bacterial valorization of pulp and paper industry process streams and waste, *Appl Microbiol Biotechnol.* **105**(4): 1345–1363. https://doi.org/ 10.1007/ s00253- 021- 11107-2

Chakraborty I, Das S, Dubey BK and Ghangrekar MM (2020), Novel low cost proton exchange membrane made from sulphonated biochar for application in microbial fuel cells, *Mater Chem Phys.* **239**: 122025. https://doi.org/10.1016/j.matchemphys. 2019.122025

Chaturvedi V and Verma P (2016), Microbial fuel cell: a green approach for the utilization of waste for the generation of bioelectricity, *Bioresour Bioprocess.* **3**(1): 38. https:// doi.org/10.1186/ s40643-016-0116-6

Chen TL, Kim H, Pan SY, Tseng PC, Lin YP and Chiang PC (2020), Implementation of green chemistry principles in circular economy system towards sustainable development goals: Challenges and perspectives, *Sci Total Environ.* **716**: 136998. https://doi.org/10.1016/j.scitotenv.2020.136998

Das KS (2020), Microbial Fuel Cells: A Path to Green, Renewable Energy, pp 195–206. https://doi.org/ 10.1007/978-81-322-3965-9_9

Davis JB (1967), Microbial process of producing electricity. 3. https://www.google.com/patents/US3305399

Gaikwad VT and Munavalli GR (2019), Turbidity removal by conventional and ballasted coagulation with natural coagulants, *Appl Water Sci.* **9**(5): 1–9. https://doi.org/10.1007/s13201-019-1009-6

Gajda I, Greenman J and Ieropoulos IA (2018), Recent advancements in real-world microbial fuel cell applications, *Curr Opin Electrochem.* **11**: 78–83. https://doi.org/10.1016/j.coelec.2018.09.006

Gude VG (2015), Energy and water autarky of wastewater treatment and power generation systems, *Renew Sustain Energy Rev.* **45**: 52–68. https://-doi.org/10.1016/j.rser.2015.01.055

Guo M, Song W and Buhain J (2015), Bioenergy and biofuels: History, status, and perspective, *Renew Sustain Energy Rev.* **42**: 712–725. https://-doi.org/10.1016/j.rser.2014.10.013

Harnisch F and Schröder U (2010), From MFC to MXC: Chemical and biological cathodes and their potential for microbial bioelectrochemical systems, *Chem Soc Rev.* **39**(11): 4433–4448. https://-doi.org/10.1039/ c003068f

Huang X, Duan C, Duan W, Sun F, Cui H, Zhang S and Chen X (2021), Role of electrode materials on performance and microbial characteristics in the constructed wetland coupled microbial fuel cell (CW-MFC): A review, *J Clean Prod.* **301:** 126951. https://doi.org/10.1016/j.jclepro.2021.126951

Iigatani R, Ito T, Watanabe F, Nagamine M, Suzuki Y and Inoue K (2019), Electricity generation from sweet potato-shochu waste using microbial fuel cells, *J Biosci Bioeng.* **128**(1): 56–63. https://doi.org/ 10.1016/ j.jbiosc.2018.12.015

Jayashree S, Ramesh ST, Lavanya A, Gandhimathi R and Nidheesh P V (2019), Wastewater treatment by microbial fuel cell coupled with peroxicoagulation process, *Clean Technol Environ Policy*. **21**(10): 2033–2045. https:// doi.org/ 10.1007/ s10098- 019-01759-0

Karmaker A, Hasan M and Ahmed S (2022), A modified approach to Industrial Pollution Projection System for the assessment of sectoral pollution loads in Bangladesh, *Environ Monit Assess.* **194**(6): 406. https://doi.org/10.1007/s10661-022-10073-0

Katal R and Pahlavanzadeh H (2011), Influence of different combinations of aluminum and iron electrode on electrocoagulation efficiency: Application to the treatment of paper mill wastewater, *Desalination* **265**(1-3): 199–205. https:// doi.org/ 10.1016/ j.desal.2010.07.052

Khilari S, Pandit S, Varanasi JL, Das D and Pradhan D (2015), Bifunctional Manganese Ferrite/Polyaniline Hybrid as Electrode Material for Enhanced Energy Recovery in Microbial Fuel Cell, *ACS Appl Mater Interfaces*. **7**(37): 20657–20666. https://doi.org/ 10.1021/acsami.5b05273

Kim M, Youn SM, Shin SH, Jang JG, Han SH, Hyun MS, Gadd GM and Kim HJ (2003), Practical field application of a novel BOD monitoring system, *J Environ Monit.* **5**(4): 640–643. https://doi.org/10.1039/ b304583h

Koivuluoto H, Honkanen M and Vuoristo P (2010), Cold-sprayed copper and tantalum coatings - Detailed FESEM and TEM analysis, *Surf Coatings Technol.* **204**(15): 2353–2361. https:// doi.org/ 10.1016/ j.surfcoat.2010.01.001

Konovalova EY, Stom DI, Zhdanova GO, Yuriev DA, Li Y, Barbora L and Goswami P (2018), The microorganisms used for working in microbial fuel cells, *AIP Conf Proc.* 1952. https://doi.org/10.1063/ 1.5031979

Kumar GG, Sarathi VGS and Nahm KS (2013), Recent advances and challenges in the anode architecture and their modifications for the applications of microbial fuel cells, *Biosens Bioelectron.* **43**(1): 461–475. https://doi.org/ 10.1016/ j.bios. 2012.12.048

Lawson K, Rossi R, Regan JM and Logan BE (2020), Impact of cathodic electron acceptor on microbial fuel cell internal resistance, *Bioresour Technol.* **316**: 123919. https://doi.org/ 10.1016/ j.biortech. 2020.123919

Lebeer S, Verhoeven TLA, Claes IJJ, De Hertogh G, Vermeire S, Buyse J, Van Immerseel F, Vanderleyden J and De Keersmaecker SCJ (2011), FISH analysis of Lactobacillus biofilms in the gastrointestinal tract of different hosts, *Letters Appl Microbiol.* **52**(3): 220–226. https:// doi.org/ 10.1111/ j.1472-765X. 2010.02994.x

Li M, Zhou M, Tian X, Tan C, McDaniel CT, Hassett DJ and Gu T (2018), Microbial fuel cell (MFC) power performance improvement through enhanced microbial electrogenicity, *Biotechnol Adv.* **36**(4): 1316–1327. https:// doi.org/ 10.1016/ j.biotechadv.2018.04.010

Li S, Cheng C and Thomas A (2017), Carbon-Based Microbial-Fuel-Cell Electrodes: From Conductive Supports to Active Catalysts, *Adv Mater.* **29(**8): 1–30. https://doi.org/10.1002/adma.201602547

Logan BE, Wallack MJ, Kim K-Y, He W, Feng Y and Saikaly PE (2015), Assessment of Microbial Fuel Cell Configurations and Power Densities, *Environ Sci Technol Lett.* **2**(8): 206–214. https://doi.org/ 10.1021/acs.estlett.5b00180

Mandley SJ, Daioglou V, Junginger HM, van Vuuren DP and Wicke B (2020), EU bioenergy development to 2050, *Renew Sustain Energy Rev.* **127**: 109858. https://doi.org/10.1016/j.rser.2020.109858

Obileke K, Onyeaka H, Meyer EL and Nwokolo N (2021), Microbial fuel cells, a renewable energy technology for bio-electricity generation: A mini-review, *Electrochem Commun.* **125**: 107003. https://doi.org/ 10.1016/ j.elecom.2021.107003

Omine K, Sivasankar V and Chicas SD (2018), Bioelectricity Generation in Soil Microbial Fuel Cells Using Organic Waste ***In:*** Microbial Fuel Cell Technology for Bioelectricity, Springer International Publishing, pp 137–150. https:// doi.org/ 10.1007/ 978-3-319-92904-0_7

Palanisamy G, Jung H-Y, Sadhasivam T, Kurkuri MD, Kim SC and Roh S-H (2019), A comprehensive review on microbial fuel cell technologies: Processes, utilization, and advanced developments in electrodes and membranes, *J Clean Prod.* **221**: 598–621. https://doi.org/ 10.1016/ j.jclepro.2019. 02.172

Pegis ML, Roberts JAS, Wasylenko DJ, Mader EA, Appel AM and Mayer JM (2015), Standard Reduction Potentials for Oxygen and Carbon Dioxide Couples in Acetonitrile and N,N-Dimethylformamide, *Inorg Chem.* **54**(24): 11883–11888. https:// doi.org/ 10.1021/ acs.inorgchem.5b02136

Popp J, Lakner Z, Harangi-Rákos M and Fári M (2014), The effect of bioenergy expansion: Food, energy, and environment, *Renew Sustain Energy Rev.* **32**: 559–578. https://doi.org/10.1016/j.rser.2014.01.056

Prasad J and Tripathi RK (2017), Maximum electricity generation from low cost sediment microbial fuel cell using copper and zinc electrodes, *International Conference on Information, Communication, Instrumentation and Control (ICICIC)*, pp 1–4. https://-doi.org/10.1109/ ICOMICON.2017.8279078

Quader M (2012), Paper Sector in Bangladesh: Challenges and Scope of Development, *J Chem Eng.* **26**: 41–46. https://doi.org/10.3329/jce.v26i1.10181

Ratheesh A, Elias L and Aboobakar Shibli SM (2021), Tuning of Electrode Surface for Enhanced Bacterial Adhesion and Reactions: A Review on Recent Approaches, *ACS Appl Bio Mater.* **4**(8): 5809–5838. https://doi.org/10.1021/acsabm.1c00362

Sahu O (2019), Sustainable and clean treatment of industrial wastewater with microbial fuel cell, *Results Eng.* **4:** 100053. https://doi.org/ 10.1016/ j.rineng. 2019.100053

Senthilkumar K and Naveenkumar M (2023), Enhanced performance study of microbial fuel cell using waste biomass-derived carbon electrode, *Biomass Convers Biorefinery* **13**(7): 5921–5929. https://doi.org/ 10.1007/s13399-021-01505-x

Simate GS (2015), Water treatment and reuse in breweries. ***In:*** Brewing Microbiology, Elsevier, pp 425–456. https://doi.org/10.1016/ B978-1-78242 -331-7.00020-4

Sonawane JM, Ezugwu CI and Ghosh PC (2020), Microbial Fuel Cell-Based Biological Oxygen Demand Sensors for Monitoring Wastewater: State-of-the-Art and Practical Applications, *ACS Sensors* **5**(8): 2297–2316. https://doi.org/10.1021/ acssensors.0c01299

Subha C, Kumar MD, Kannah RY, Kavitha S, Gunasekaran M and Banu JR (2020), Bioenergy recovery from food processing wastewater-Microbial fuel cell, *Food Waste to Valuable Resources: Applications and Management*, INC. https://doi.org/ 10.1016/ B978 -0-12-818353- 3.00012-2

Timmis KM (1995), Environmental biotechnology, *Bio Technology.* **13:** 2. https://doi.org/10.1038/ nbt0295-105a

Uddin MJ, Jeong YK and Lee W (2021), Microbial fuel cells for bioelectricity generation through reduction of hexavalent chromium in wastewater: A review, *Int. J. Hydrogen Energy.* **46**(20): 11458–11481. https://doi.org/10.1016/j.ijhydene.2020.06.134

Wang VB, Du J, Chen X, Thomas, AW, Kirchhofer ND, Garner LE, Maw MT, Poh WH, Hinks J, Wuertz S, Kjelleberg S, Zhang Q, Loo JSC, and Bazan GC (2013), Improving charge collection in Escherichia coli-carbon electrode devices with conjugated oligoelectrolytes, *Phys Chem Chem Phys*. **15**(16): 5867–5872. https://doi.org/10.1039/c3cp50437a

Xiao L and He Z (2014), Applications and perspectives of phototrophic microorganisms for electricity generation from organic compounds in microbial fuel cells, *Renew Sustain Energy Rev.* **37**: 550–559. https://-doi.org/10.1016/j.rser.2014.05.066

Yang H and Reddy RG (2014), Electrochemical deposition of zinc from zinc oxide in 2:1 urea/choline chloride ionic liquid, *Electrochim Acta*. **147:** 513–519. https:// doi.org/ 10.1016/ j.electacta.2014.09.137

Zhang S, Schuster J, Frühauf-Wyllie H, Arat S, Yadav S, Schneider JJ, Stöckl M, Ukrainczyk N and Koenders E (2021), Conductive Geopolymers as Low-Cost Electrode Materials for Microbial Fuel Cells, *ACS Omega*. **6**(43): 28859–28870. https://-doi.org/10.1021/ acsomega.1c03805

Zhang X, Xia X, Ivanov I, Huang X and Logan BE (2014), Enhanced activated carbon cathode performance for microbial fuel cell by blending carbon black, *Environ Sci Technol.* **48**(3): 2075–2081. https://doi.org/ 10.1021/es405029y