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DESIGN OF CYLINDRICAL MICROBIAL FUEL CELL AND STUDY OF ANODE MODIFICATION USING NA Y ZEOLITE

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ABSTRACT

Microbial fuel cell (MFC) represents an eco-friendly approach to generate electricity and purifies wastewater simultaneously. As per this technology, the microorganisms are employed to convert the chemical energy stored in the biodegradable portion of organic matter into direct electric current by simultaneously treating the wastewater. In this study, dual chambered membrane microbial fuel cell was operated and was optimized using municipal wastewater as a substrate. Graphite rod and plate are widely used as anode material in microbial fuel cells (MFCs) because of its high specific surface area, low cost, good electrical conductivity. In this work, Nafion is used as membrane, platinum catalyst coated carbon cloth as cathode and it is open to air (aerobic process). Graphite rod and graphite plate were modified using the zeolite Na Y and used as anode. The maximum current produced by coated graphite plate is 0.82mA which was higher than coated graphite rod 0.72mA current.

Keywords: Microbial Fuel Cell, Electricity, Nafion Membrane, Graphite rod and plate, wastewater

INTRODUCTION

One of the most important environmental problems faced by the world is management of wastes. Extracting energy from organic and inorganic wastes can provide efficient means to solve the energy and environmental problems simultaneously. Microbial fuel cell represents a new method of renewable energy recovery. MFC technology has become an attractive technology today because of its capability to convert the chemical energy present in organic/ inorganic waste into electrical energy. MFC technology can be used for electricity generation, wastewater treatment and have many potential applications. MFCs are the bio electrochemical devices that typically consists of two chambers i.e the anode chamber (anaerobic: contains an electrode microorganisms and anolyte) and the cathode chamber (aerobic / anaerobic: contains an electrode, electron acceptor, and a catalyst) separated by a proton exchange membrane (PEM) e.g., nafion. The microorganisms used as biocatalysts to oxidize the substrate in the anode chamber and have been denoted as the power house of MFCs. Many researchers had worked on dual chamber MFC such as

Zhang et al., 2009 aimed to test wheat straw as a potential fuel in MFC for electricity production. From the findings, it was demonstrated that stable power could be generated from wheat straw. A power density of 123W/m^3 was obtained in the study.

Jafary et al., 2010 studied the effect of glucose concentration on the performance of two chambered microbial fuel cell was investigated using Saccharomyces cerevisiae as biocatalyst. During the experiment, optimum results of power and current density of 39.33 W/m^3 and 85.059 A/m^2 respectively obtained at glucose concentration of 5g/l.

Rodrigo et al., 2010 found that the wastewater treatment using anaerobic pre-treatment of activated sludge, electricity generation can be obtained within 8-10 days. The maximum power density reported in the study was 25 W/m^3 with a voltage of 0.23 V using domestic wastewater.

Kim et al. 2011 designed and constructed a double anode chambered MFC to test its performance which was inoculated with shewanella oneidesis MR-1. The MFC was fabricated using a transparent polycarbonate material as the anode and cathode while the Nafion material was used for the PEM. A maximum power and current density of 24 W/m^3 and 3.66 A/m^2 were obtained.

V. Vineetha, K.Shibu 2013 electricity generation was carried out with plain carbon rods and iron coated carbon rods as anodes. The maximum electricity generation (71mA) and maximum voltage production (351mA) was obtained MFC with heated iron coated carbon as anode.

Xia et al., 2013 used two different zeolites (mobil catalytic materials number 41 (MCM 41) and Na X), were used to modify graphite anodes and compared with unmodified anodes in dual chamber MFCs. It is found that Na X zeolite is a favourable, affordable material for anode modification of MFCs with maximum power density 215.33 ± 6.4 mW/m^2.

Miran et al., 2016 fabricated a rectangular double chamber. The PEM was made using Nafion, and the lemon peel substrate was agitated by a magnetic bar. It was revealed in the study that the voltage and power density generated were 0.58V and 371 W/m^3 respectively.

In the studies of Titon chandra Saha et al., 2019 double chamber MFCs were constructed for the generation of electricity from microorganisms present in organic waste samples. Generation of electricity (Maximum 5.78V and 5.03mA in multiple chamber MFC) was attained connecting multiple chambers containing MFCs and able to lid light.

One of the approaches to improving energy generation in MFCs is by modifying the existing anode materials to provide more electrochemically active sites and improve the adhesion of microorganisms. Anode modification is an alternative to improve electricity generation in Microbial Fuel Cells. The main limitations in up-scaling of MFCs include low power density and expensive electrode materials. Power density can be increased by ensuring that the anode is hydrophilic and has a high specific surface area. The surface roughness is of great importance for energy production in the MFCs as the anode morphology should facilitate the adherence of bacteria and subsequent biofilm formation. In this work an attempt was made to Design the cylindrical microbial fuel cell for the treatment of wastewater. Anode modification with using Na Y zeolite and generate electricity.

MATERIALS AND METHODS

Materials used

- Graphite rod and plate.
- Modified graphite rod and plate.
- Pt coated carbon cloth.
- Nafion membrane.

3.2 Chemicals used

- Acetate

- Ferricyanide
- Phosphate buffer solution.

3.9 Synthesis of Zeolite Na-Y

Zeolite Na-Y was synthesized by hydrothermal method which includes preparation of seed gel, feedstock gel and overall gel as given by (REF).

Seed gel:

(a) **(b)**

Figure 1: (a) The mixture of seed gel stirred at 600rpm (b) Seed gel formation

Feedstock gel:

(a) **(b)**

Figure 2: (a) The mixture of feedstock gel stirred at 600rpm (b) feedstock gel formation

Overall gel:

(a)

(b)

Figure 3: (a) The mixture of overall gel stirred at 600rpm (b) overall gel formation

PURIFICATION PROCESS

The product of hydrothermal process was filtered to separate supernatant. The supernatant was washed using demineralized water until the pH less than 9. The supernatant could be called as overall gel.

Modification of Anode

The bare graphite rod was placed into the Zeolite Na Y synthesis solution and hydrothermally synthesized at 100°c for 7 h in Hot Air Oven. The Na Y graphite rods were removed after hydrothermal synthesis, washed with deionized water, and dried overnight at 80°c. The Na Y zeolite-modified graphite rod anode was called Zeolite Na Y anode.

(a)

(b)

(c)

(d)

Figure 4: (a) Zeolite Na Y synthesis solution (b) Zeolite Na Y dried at 100°C (d) Graphite anodes dipped in Zeolite Na Y solution (e) Graphite anodes dried at 80°C.

MFC Setup

The MFC with 50 ml volumes for the anode and cathode chambers. Anode chamber containing 30 ml volume, cathode chamber contains 20 ml volume. Anode and cathode chambers were separated & placed into the MFC's and a proton exchange membrane (Nafion 117, "5cm x 5cm") was inserted between them MFC assembly's main goal is to avoid leakage in the cell and assemble all cells in an identical manner. In anode chamber with the help of copper wire graphite rod was placed and it is used as anode. On top of anode chamber cathode chamber is placed and platinum coated carbon cloth is inserted which is used as cathode and it is open to air. These copper wires are externally connected to 100ohm resistance to complete the circuit.

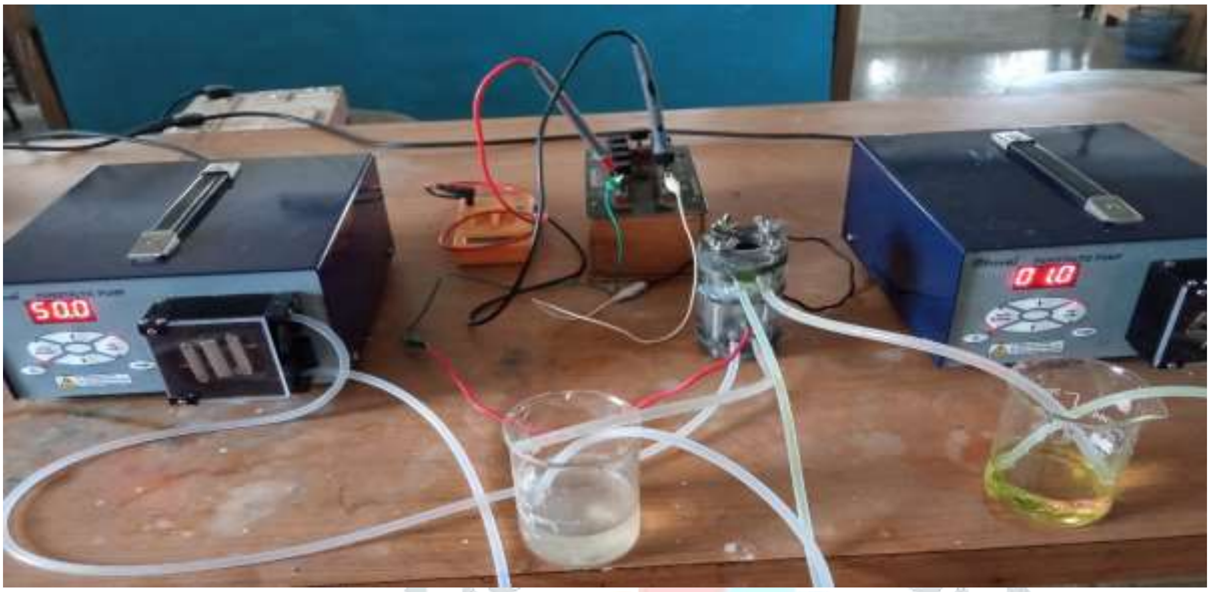


Figure 5: Experimental setup of MFC

Process

1. The present study was carried out to investigate the capability of biological process for the treatment of waste water. The experimental work was carried out wastewater treatment and produce electricity simultaneously.

2. A two chambered microbial fuel cell was constructed. In two chambers one is anode chamber and the other one is cathode chamber.

3. First we set the experimental set up of MFC. We connect the peristaltic pumps to anode and cathode chambers to maintain the flow between them. Anode chamber was maintained in anaerobic process.

4. In anode chamber wastewater and acetate used as anolyte solution. In cathode chamber ferricyanide, phosphate buffer solution used as catholyte solution. In this chamber graphite rod is taken as anode which is filled by anolyte solution.

5. Cathode chamber is filled by potassium ferricyanide and phosphate buffer solution by using peristaltic pumps. These two chambers which are separated by Nafion 117 membrane and Cathode chamber was maintained in aerobic process.

6. In cathode chamber platinum coated carbon cloth taken as cathode. Both chambers which are separated by cation exchange membrane (Nafion). The membrane forms a bridge between cathodic and anodic chamber facilitates the transfer of ions (protons).

7. Microbes at anode oxidize organic matter generates electrons and protons. Protons move to the cathode compartment through the membrane. Electrons transferred to the cathode compartment through external circuit to generate current.

8. An external resistance was connected from anode chamber and cathode chamber copper wires are externally connected to 100ohm resistance box to complete the circuit and the readings (voltage and current) were measured using a digital multimeter.

9. The experimental run was carried out for 7 days.

Calculation and Reporting Data

Voltages and currents generated were measured using a digital multimeter, and converted to power P (milliWatts) and power density pd (mW/m^2) and current density cd (mA/m^2).

Power: The overall performance of an MFC is evaluated in many ways, but principally through power output. Power is calculated as

$$P = IV$$

Power density: Power density is the amount of power (time rate of energy transfer) per unit volume. The power output is usually normalized to the projected anode surface area because the anode is where the biological reaction occurs. The power density (pd, W/m^2) is therefore calculated on the basis of the area of the anode (A) as

$$Pd = P/A$$

Current density: The current produced is normalized to the projected anode surface area. The current density (mA/m^2) is calculated as

$$Cd = I/A$$

Where I (mA) is the current, V (volts) is the voltage, and A (m^2) the surface area of the projected anode.

Characterization and morphology

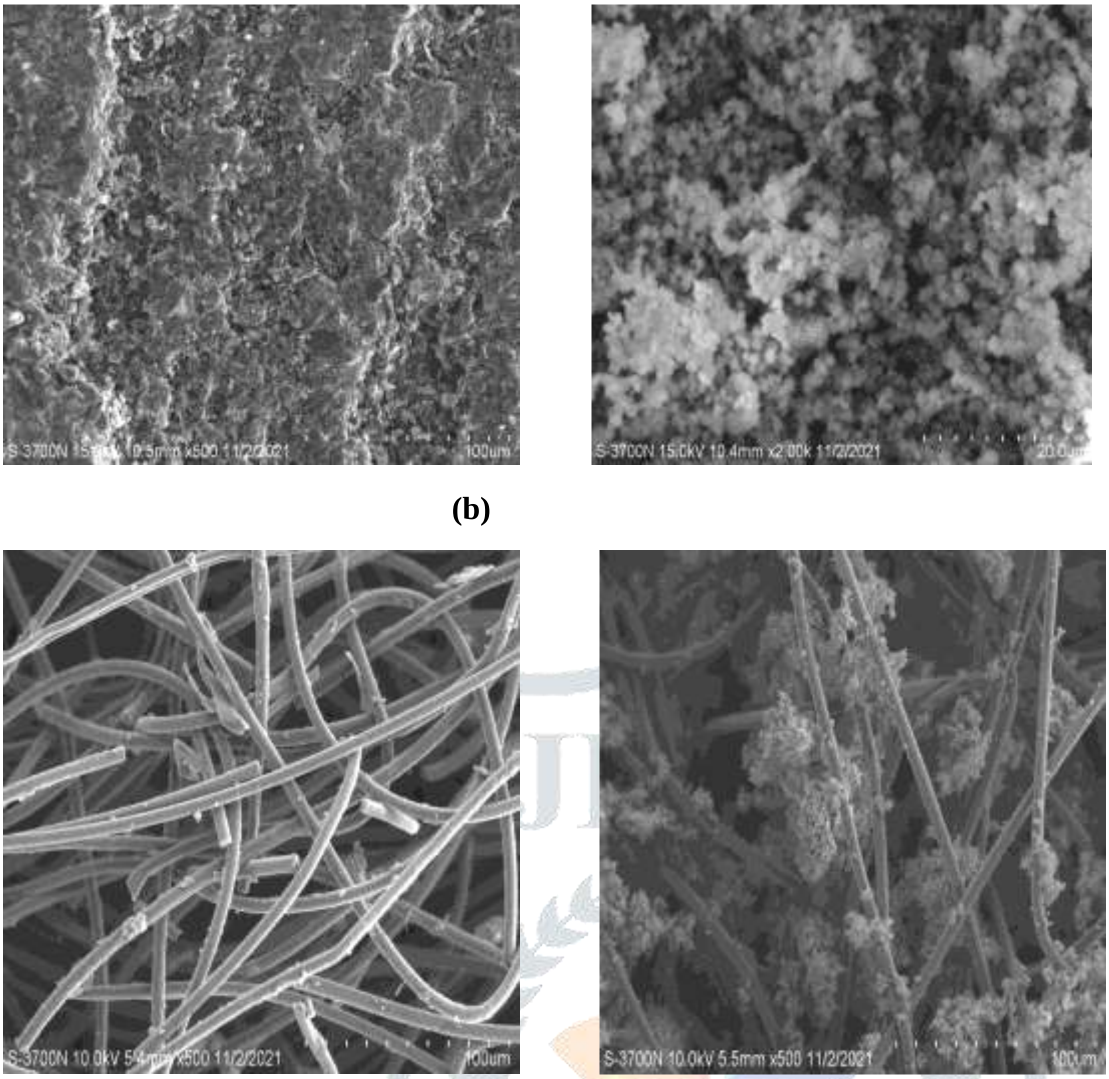
Various characterization tests are performed on graphite anodes and coated graphite anodes as SEM, XRD for analysis of chemical and surface changes underwent by chemical modification. The performance of the fuel cell is critically evaluated based on the power output.

Scanning electron microscopy was performed to analyze the bacterial morphology.

Analysis of Anode Electrode

SEM analysis of graphite anodes and coated graphite anodes

Scanning electron micrograph of graphite anodes. Coated with Na Y resulted in more rougher surface on graphite anodes owing to more hydrophilicity, therefore provided more space for bacterial attachment. A marked change of morphology on graphite anode was caused by microbe adhering to them and colony formation, likely because oxygen-containing functional group on the coated graphite anode surfaces significantly increased their biocompatibility.



(a) **(b)**

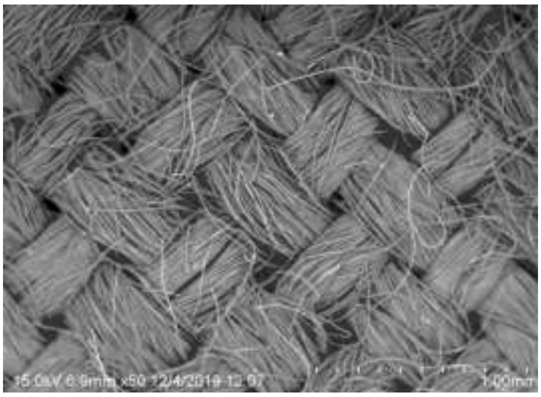
(c) **(d)**

Figure 6: SEM analysis of (a) Graphite rod (b) Zeolite Na Y graphite rod

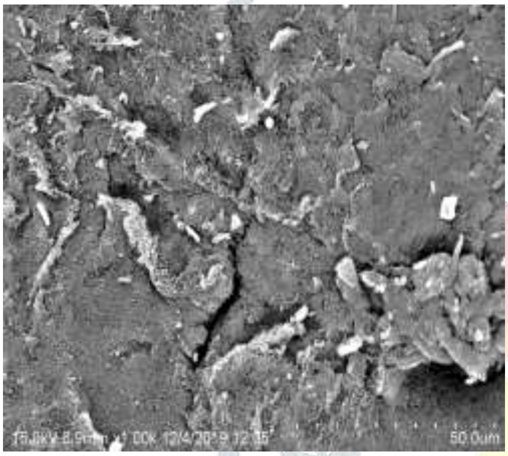
(c) Graphite plate (d) Zeolite Na Y graphite plate

Analysis of cathode electrode

SEM analysis of carbon cloth and Pt coated carbon cloth



(a)



(b)

Figure 7: SEM Analysis of (a) Carbon cloth (b) Pt coated carbon cloth

Scanning electron microscopy was performed to analyze the morphology. The cathode as gas diffusion electrode Pt/c loaded on carbon cloth and a normal carbon are analyzed for surface morphology in which we have observed some whitish Pt coating on the coated electrode while the normal carbon cloth has appeared to be fibrous surface with nano-fiber threading on it. The SEM analysis is done at 15.0kV to observe the porous structure of cathode electrode.

XRD Analysis of Graphite Anodes

To determine the chemical composition of the chemical scale, elemental analysis was performed with XRD. Quantitative analysis of XRD data could not be performed due to a limitation of number of XRD images. XRD analysis of both the electrodes showed the difference of roughness as can been seen in the figures the graphite anodes had more rough and blunt peaks and while the coated graphite anodes had slim and sharp peaks

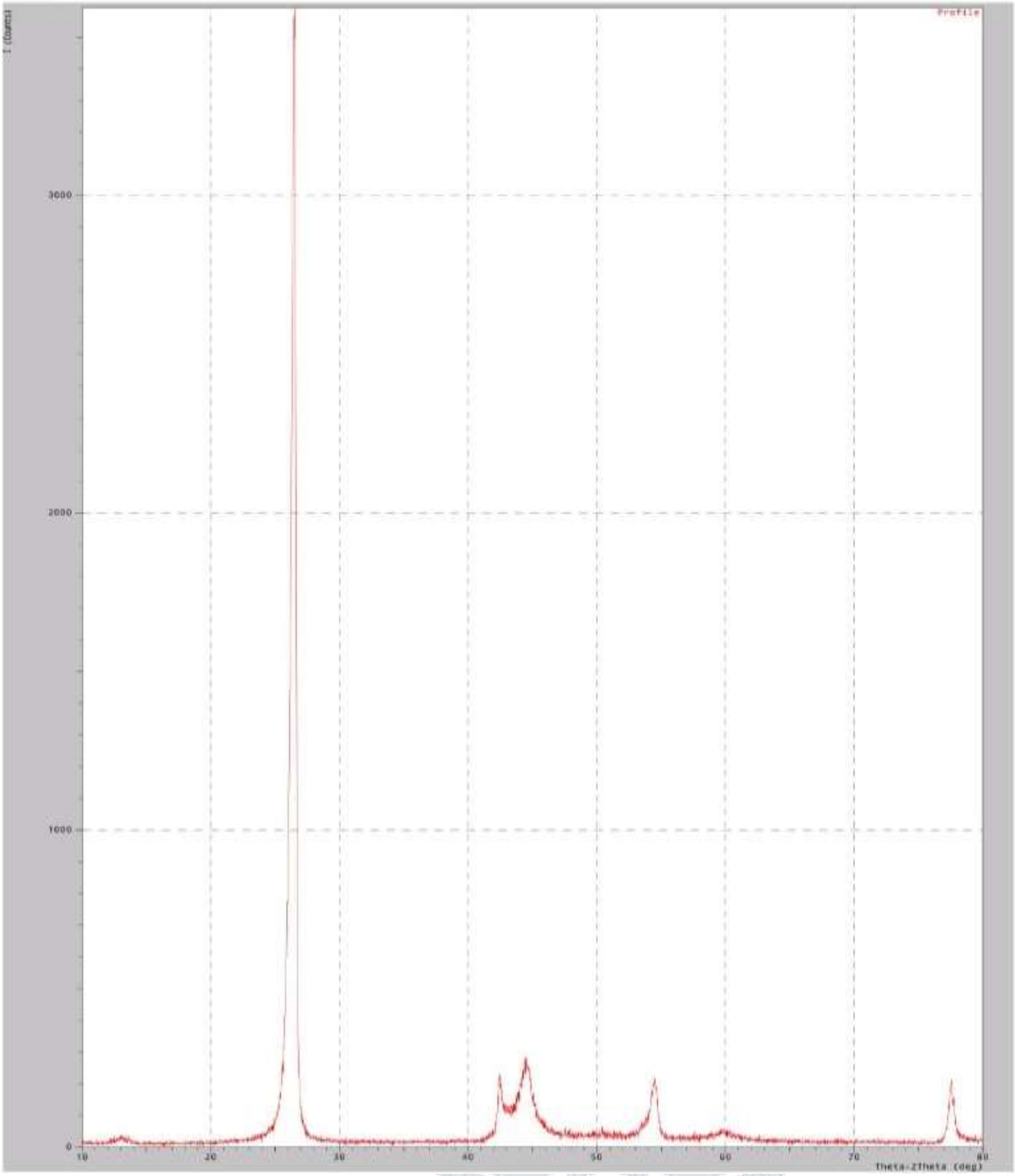


Figure 8: XRD analysis for graphite rod

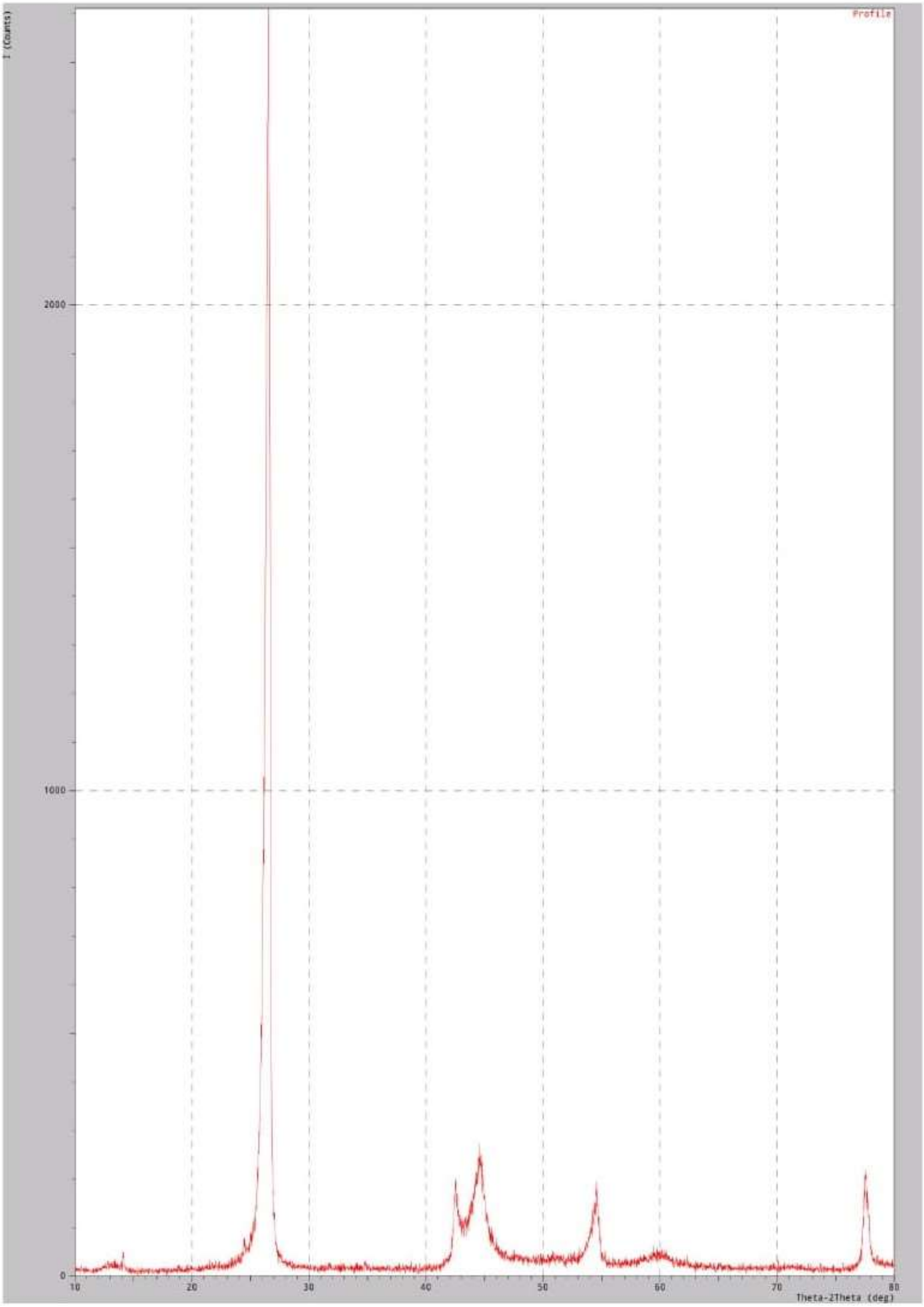


Figure 9: XRD graph for coated graphite rod

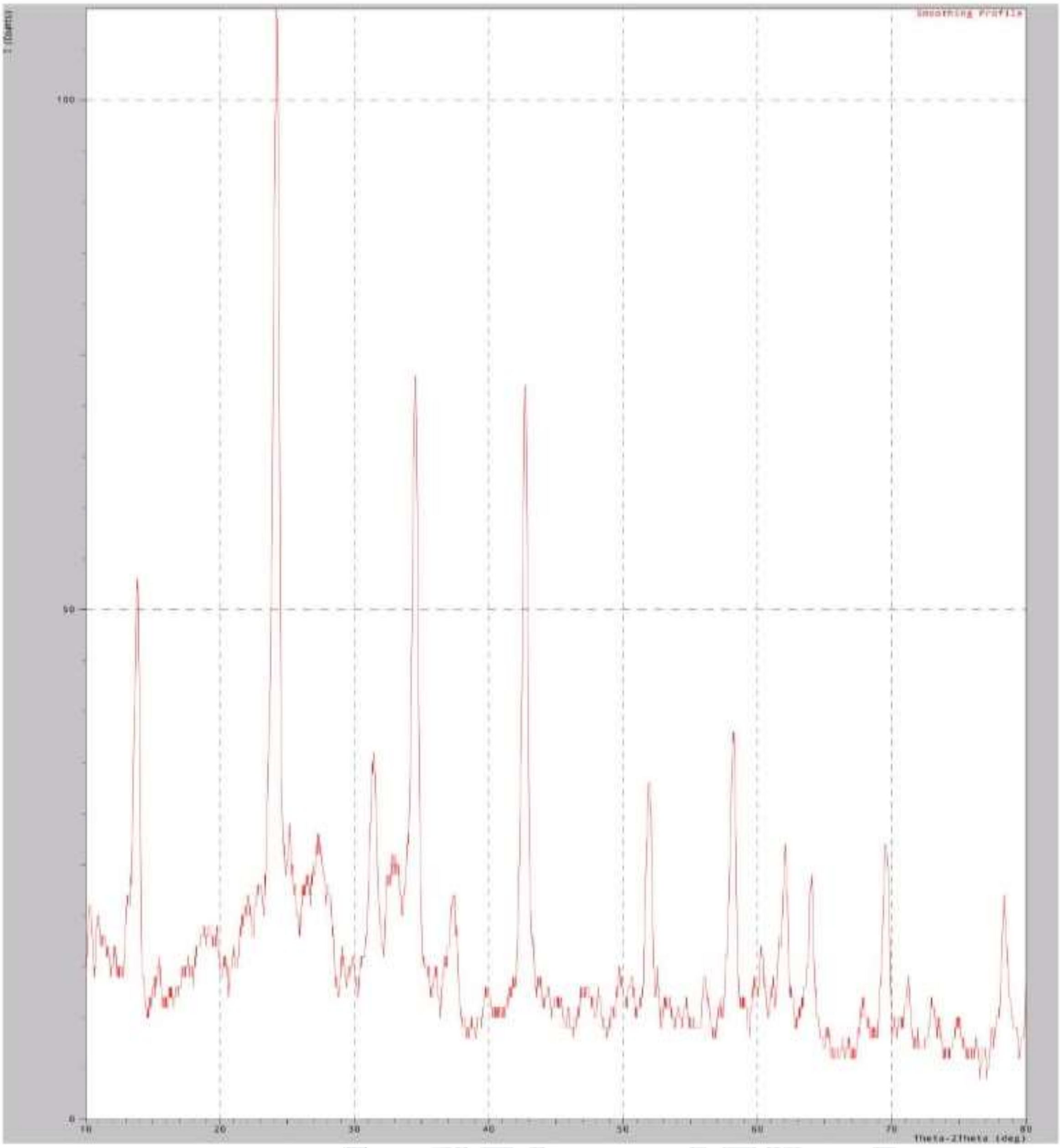


Figure 10: XRD analysis for coated graphite plate

In summary, we successfully prepared a novel Coated graphite anodes material by hydrothermal and chemical treatment methods for the application of MFC. The results of SEM, XRD indicated that the Coated graphite anodes had a good morphology and a higher wet ability with larger specific surface area and less surface impurities than graphite anodes, which is beneficial for the attachment of microorganism groups and electronic transmissions. Electrochemical test results showed that the coated graphite anodes held a lower charge transfer rate, which facilities the transfer of electrons from microorganisms to the outer circuit. Graphite anodes were coated using Zeolite Na Y to increase hydrophilicity, improve biocompatibility, enhance electron transfer rates, promote the electrocatalytic properties of bio-anode especially increase the power output of the resultant MFCs substantially.

RESULTS AND DISCUSSION

The maximum power generation (mW/m^2) under optimized operational conditions was obtained and used to compare the reactor performance of the dual chambered MFC.

Graphite rod as anode and Pt coated carbon cloth as cathode. Anolyte solution used as wastewater, sodium acetate, catholyte solution used as potassium ferricyanide, phosphate buffer solution of pH 7.0.

Table 1 Data from dual chamber MFC using Graphite rod

Days	Voltage (V)	Current (mA)	Current density (A/m^2)	Power density (mW/m^2)
1	0.15	0.12	0.084	12.64
2	0.32	0.21	0.147	47.23
3	0.39	0.25	0.175	68.56
4	0.48	0.42	0.295	141.74
5	0.30	0.38	0.267	80.16
6	0.25	0.34	0.239	59.75

Table 2: Data from dual chamber MFC using coated graphite rod

Days	Voltage (V)	Current (mA)	Current density (A/m^2)	Power density (mW/m^2)
s1	0.18	0.20	0.140	25.30
2	0.38	0.32	0.225	85.5
3	0.56	0.50	0.351	196.89
4	0.82	0.72	0.506	415.16
5	0.42	0.68	0.478	200.8
6	0.35	0.65	0.457	159.98

Table 3: Data from dual chamber MFC using Graphite plate

Time in days	Voltage (V)	Current (mA)	Current density (A/m^2)	Power density (mW/m^2)
1	0.16	0.14	0.444	71.04
2	0.34	0.22	0.698	237.3
3	0.40	0.28	0.888	355.2
4	0.50	0.48	1.523	761.5
5	0.42	0.35	1.111	466.6
6	0.32	0.30	0.952	304.7

Table 4: Data from dual chamber MFC using coated Graphite plate

Time in days	Voltage (V)	Current (mA)	Current density (A/m^2)	Power density (mW/m^2)
1	0.21	0.25	0.793	166.5
2	0.40	0.38	1.206	482.4
3	0.58	0.55	1.746	1012
4	0.90	0.82	2.603	2342
5	0.83	0.72	2.285	1896
6	0.62	0.68	2.158	1337

The current, voltage and power generation showed a gradual increase for few days, and then declined. This variation was due to the availability of less oxidizable substrates in the waste samples. The peak current, voltage and power generation were observed, in all cases, on the 4^{th} day of the preliminary experiments, as can be clearly observed from Tables (1 to 4) and figures (11 to 13). After 5^{th} day, the peak values started to decrease continuously. The current, voltage and power density from the waste substrate samples with coated graphite anodes has higher voltage generation than graphite anodes, in all cases, especially under anaerobic condition of the anode chambers of the MFCs.

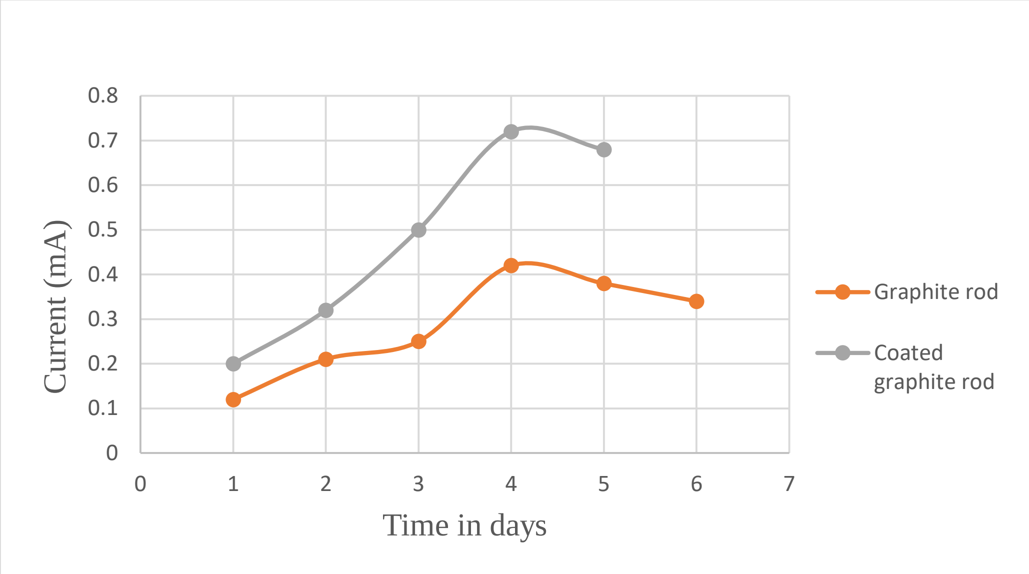


Fig 11: MFC Graphite rod, Coated graphite rod anode, Pt coated carbon cloth cathode

From the above graph we observed that MFC with coated graphite rod anode as electrode maximum current produced 0.72mA, which was higher than MFC graphite rod respectively. The current of the MFC with coated graphite anode increased compare with that with coated graphite rod.

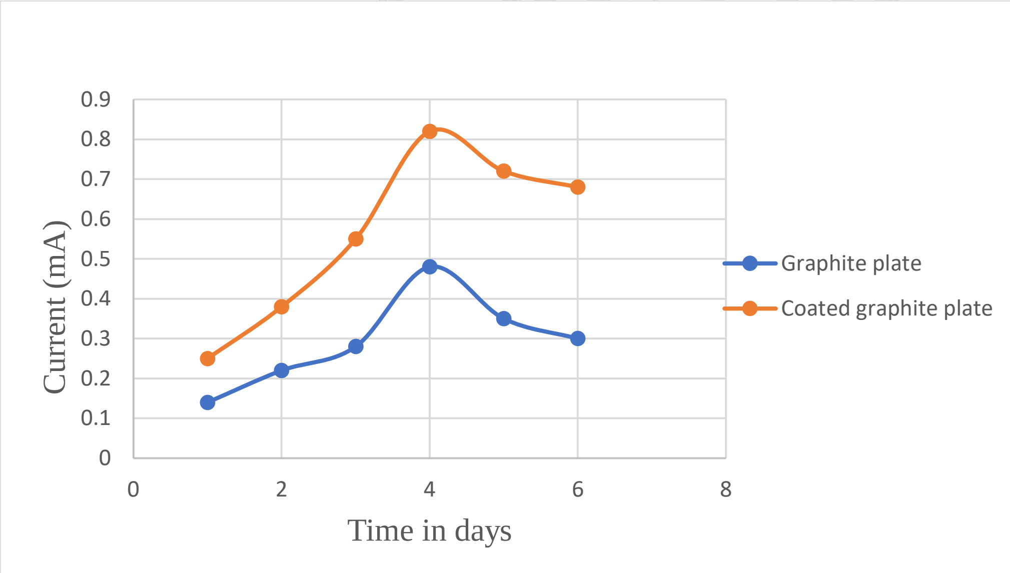


Fig 12: Graph for MFC Graphite plate, Coated graphite plate anode, Pt coated carbon cloth cathode

From the above graph we observed that MFC with coated graphite plate anode as electrode maximum current produced 0.82mA, which was higher than MFC graphite plate respectively. The current of the MFC with coated graphite plate increased compare with that with graphite plate.

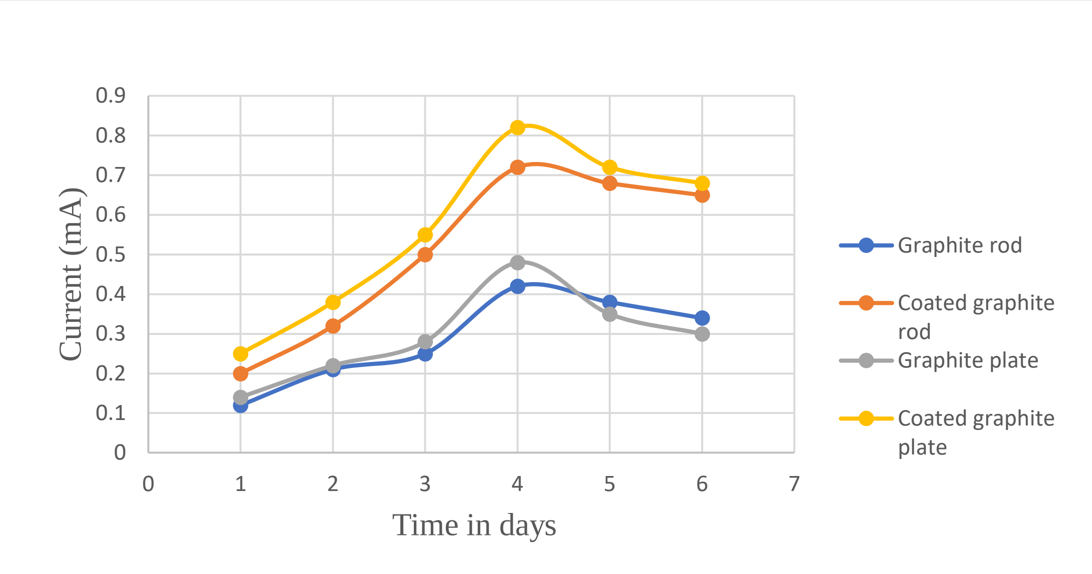


Fig 13: Graph for MFC Graphite anodes, Coated graphite anodes, Pt coated carbon cloth cathode

From the above graph we observed that current showed a gradual increase for few days and then decreased. This variation was due to the availability of less oxidizable substrate in the wastewater. MFC with coated graphite rod anode as electrode maximum current produced is 0.72mA, which was higher than MFC graphite rod respectively. MFC with coated graphite plate anode as electrode maximum current produced is 0.82mA, which was higher than MFC graphite plate respectively. And also we observed that MFC with coated graphite plate anode produce higher current 0.82mA compare to coated graphite rods.

The maximum power generation (mW/m^2) under optimized operational conditions was obtained and it is used to compare the reactor performance of the dual chambered MFC. The production of voltage and current with respect to time when MFC is operated with Nafion membrane is shown in the above table. After inoculating the mixed microbial flora into MFC, voltage production initiated and reached to a maximum value after 4 days. Later, the voltage decreased due to substrate exhaustion. The maximum produced voltage and current were reported to be 0.90V and 0.82mA for Coated graphite plate respectively. The time vs current density and power density curves were obtained through the experiments conducted with PEM membranes. The maximum current density and power density achieved by Nafion were 2.603mA/m^2 and 2342mW/m^2 respectively. By using the table and data obtained from MFC for 6 days the Voltage vs Time graph is drawn. The voltage production also increased with time and reached maximum, but after 3 days it was decreased as substrate is completely used and bacteria being dead after used for production of electricity.

CONCLUSIONS

In the present work graphite anodes were coated using zeolite Na Y and also the effects of parameters of both anodes used in MFC were investigated. Based on the present investigation and from the available scientific information derived from the review of relevant literature, following conclusions were drawn.

The Zeolite Na Y coating of graphite anodes was most efficiently done.

From the results it was concluded that the maximum current produced using Zeolite Na Y coated graphite plate 0.82mA, which was higher than the graphite rod & plate.

The surface characterization of different anodes used have shown different variations and was concluded that increase of oxygen containing functional groups on the coated graphite plate anode favored the adsorption and growth of bacteria and acceleration of electron transport.

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