Parametric Optimization and performance assessment of polyaniline graphene-impregnated electrode-based low-cost Microbial Fuel Cell using mixed culture obtained from Canteen wastewater

Ponmani Padmanaban[1], Parthiban P.[2], Ramanujam Mahesh[2], Murugaboopathy Sivanantham[1], Balraju Nilavu[2], Tewari Chetna[3], Sahoo Nandagopal[3] and Das Ashutosh[2]*

1. Department of Physics, PRIST Deemed to be University, Thanjavur -613403, Tamil Nadu, INDIA
2. Centre for Environmental Engineering, PRIST Deemed to be University, Thanjavur -613403, Tamil Nadu, INDIA
3. Department of Chemistry, Kumaun University, Nainital-263001, Uttarakhand, INDIA
*scientists.crd@gmail.com

Abstract

The development of a high-efficiency anode is essential for the use of microbial fuel cells. The metallic conductivity, corrosion resilience, low cost of conducting polyaniline (PANI) and recycled graphene (RGn) pique our interest. In this study, PANI-RGn nanoparticles are polymerized in situ on cotton fabric wrapped with aluminium mesh as the anode for a microbial fuel cell. The MFC was designed as a dual-chambered system and the chambers are separated by a proton exchange membrane (PEM) salt bridge using canteen wastewater. The process parameters including cathodic chamber aeration, nutrient dosage, nitrogen fluxing, stirring and anode geometry, were optimized.

The potential difference (mV) and these systems' current (mA) performance have been analyzed. On the use of PANI-RGn carbon cloth as electrodes, the output voltage was found to be of the order of 180mV. The system developed herewith is highly cost-effective because of the use of graphene that has been generated from waste plastic through catalytic pyrolysis. Hence, the present study revealed that a potential green energy alternative is PANI-RGn in situ polymerized cotton fabric and circular aluminium-mesh electrode in anoxic MFC.

Keywords: Bio-electricity, Biofilms, Canteen wastewater, Microbial fuel cell, Recycled graphene, Wastewater treatment.

Introduction

Microbial fuel cells (MFCs) operate on the principle of electrochemical operation of bacteria towards steady excerption of energy from organic waste[2,8,16] and its conversion unto an electrical current, thereby making a persistent[20,26] and environmentally constructive technology[27,38,41].

Many wastewater treatment proceedings need a significant level of power usage. In a classic municipal wastewater treatment facility, the chemical energy in organic wastewater compounds is significantly more than the power required for wastewater treatment[19].

Hence, usage of the microbial fuel cell (MFC) with electrochemically active bacteria (EAB) at the anode has been reported to transform the chemical energy in organic molecules directly into electricity[3,6,25]. Wastewater from the food-processing and agro-based industries[1] by a high concentration of readily biodegradable organics[7] have been reported to yield electricity using MFCtechnology[29].

However, the implementation of MFC is severely constrained by their extremely low power density, mainly due to the low performance of extracellular electron transfer (EET) mechanisms. This mechanism includes redox-active outer membrane proteins, electron shuttles and conductive bacterial pili of electroactive bacteria and the electrode[31,35]. Regardless of the extracellular electron transfer mechanism, biofilm development is critical to continuous current production in most MFCs[17,28,44]. Pseudomonas, on the other hand, is a classic exoelectrogen strain that solely employs self-generated electron mediators for extracellular electron transmission for MFCs catalyzed by exoelectrogens reliant on their self-mediated electron transfer[11,33,39].

Besides, the studies using *E.coli* have shown higher charge collection using carbon-electrode devices with conjugated oligoelectrolytes[37]. The rates of electron transfer and electron losses, which are heavily influenced by the operating voltage generated by different variables interconnected with biological and chemical processes, are critical concerns that require considerable attention in influencing power generation in MFC[15,34,36]. Performance of a typical MFC is governed by bacterial growth on the electrode surface, interfacial electron generalizability from bacteria to the electrode, conductivity and electrode biodegradability[21]. As a result, an anode exhibiting excellent conductivity, high specific surface area, cheap cost, strong biocompatibility and durability for MFC is required[12].

Polyaniline (PANI) is a highly conductive polymer that has been widely researched in MFCs to improve anodic conductivity and extracellular electron transfer (EET) efficiency. This lengthy polymer chain may infiltrate the bacterial cell membrane and direct electrons out via the

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metabolic route. The bio-compatibility-modified anode and the behaviour of the bacterial growth benefit from PANI have been reported for greater surface area and layer thickness[42]. In fact, the embellishment of PANI nanocomposites containing carbon materials such as graphite, graphene, carbon nanotubes and reduced graphene oxide can strengthen the capability and durability of MFC.

Moreover, for its excellent conductivity and enormous specific surface area, which creates many active sites, graphene is being used to increase the electrode performance of MFCs[40,43]. Numerous research is giving on at the use of binary PANI RGn composites as anode catalysts in MFCs, thereby confirming the incorporation of conducting polymers with graphene in boosting MFC functionality by increasing electron transfer efficiency[14,24].

Although graphene usage in energy applications is well-documented, its outlay in the overseas market remains prohibitively high[46]. Given this context, researchers are exploring several precursor materials for graphene synthesis to achieve cheap cost with yield potential[9,32,37]. Because of its actual carbon content, plastic waste has recently got much attention as a starting material for graphene production[22]. Furthermore, because of its negative impact on the environment and living beings, plastic trash has become a hot topic among scientists. As a result, plastic trash is repurposed to produce carbon nanomaterials (including graphene) as an emerging solution to the plastic waste problem[13].

The purpose of the current work is to fabricate PANI-RGn using graphene obtained from catalytic pyrolysis of waste plastic with due optimization of process parameters to develop a low-cost and eco-friendly MFC.

Material and Methods

Procurement of wastewater for the study: The wastewater used in the study was obtained from the University campus centralized canteen (before secondary treatment).

Preparation of PANI nanoparticles and PANI-RGn nanocomposite: The reactants used to synthesize PANI-RGn composite fabric were aniline hydrochloride, sulphuric acid (H_2SO_4) and ammonium peroxydisulfate (APS) obtained from Sisco Research Laboratories Pvt. Ltd. Chennai, India. The graphene used in the study was synthesized from waste plastic through catalytic pyrolysis[30].

Synthesis of PANI-RGn nanocomposite-coated cotton fabrics: The PANI-RGn composite fabrics were prepared by *in situ* polymerization. The cotton fabric was dipped in the graphene-dispersed aniline monomer solution for 24 h. The polymerization was carried out by the dropwise addition of ammonium persulfate in the graphene aniline solution and the reaction mixture was kept stable at 0–5 °C for 24 h. As the fabric turns to green colour which indicates the polymerization of nanocomposite on the material, the fabric was removed from the solution and subsequently washed with water followed by ethanol until the supernatant became colourless in the case of each of these two reagents. The resultant fabric was oven-dried in a vacuum (60 °C, 24 h).

Assembly of microbial fuel cell: The MFC (Fig.1) consisted of two cylindrical chambers (the first chamber, or anode, with canteen wastewater and the second chamber, or cathode, with saturated salt water; 600 ml each) separated by a salt bridge housed in a PVC pipe and joined by adhesive to ensure a complete leak-proof system. The salt bridge was prepared using a thick cotton wick dipped in a super-saturated solution of NaCl and sun-dried (16 h), encaged by a hollow cylindrical PVC pipe (with an inner diameter matching the wick diameter to prevent leakage). Aluminium mesh (8x8 cm, four-fold) was used as electrodes in both chambers connected by a copper wire, which in turn are connected to both ends of a resistor (10 ohms). Handheld multimeters (Make: Meco, Model: 101B) were used for recording the voltage and current across the resistor.

Experimental setup: Several studies were carried out to study the effect of various process parameters.

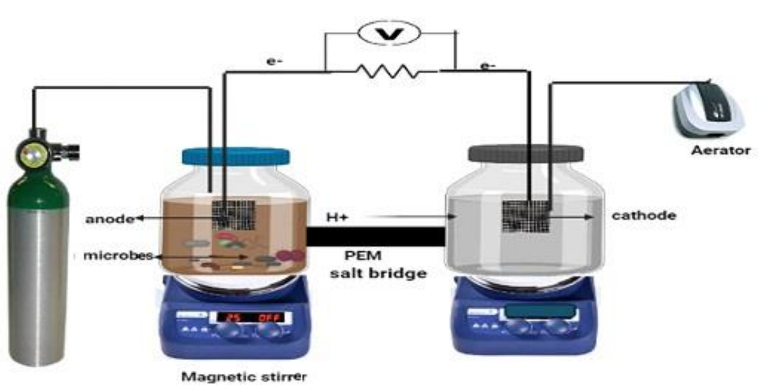

Fig. 1: Microbial Fuel Cell Setup

The cathode chamber was subjected to diffused aerations using an aerator (RS-628A) at two levels (0.5 and 2 l/min). Three levels of glucose dosage (viz. 0 gpl, 5 gpl and 7.5 gpl) were provided unto the anode chamber as a carbon source for the microbial community (at optimized aeration conditions) for enhancement of microbial activities. To evaluate the effect of anaerobic microbial activity, nitrogen fluxing was carried out in the closed anode chamber (6kg/cm^2) vis-à-vis the open anode chamber, using the optimized systems previously developed.

The stirring (@ 100 rpm) of the wastewater segment of the MFC was undertaken to assess its possible contribution to the release and transport of hydrogen in the anodic chamber. Various geometrical configurations of the anode (namely, cross-type, circular-type and cylindrical-type) were studied to evaluate the electron trapping potential. Finally, *in situ*polymerized PANI-RGn composite fabric studies were carried out as augmentation onto the existing aluminium mesh electrodes.

Microbial Community Analysis: The sample was aseptically collected and it was serially diluted up to 10^{-7}. Then it was inoculated in nutrient agar plates by spread plate method and the same incubated at 37^0 C for 24 hrs. After 24 hrs, colony morphology was observed. Besides, a gram-staining test was also carried out to evaluate the nature of microorganisms[23].

Morphological Analysis of the Composite Fibre: To evaluate the effect of the operation of MFC on the composite fibre electrode used for the study and to assess the characteristics and growth of the microbial community to augment the microbial study, Scanning electron microscopy (SEM) images were obtained for the PANI-RGn composite fabric electrode before and after the studies.

Results and Discussion

Effect of aeration in the cathode: The control (without aeration) shows a drastic decline in output voltage in less than four minutes. In contrast, aeration in the cathodic chamber was found to support the stability of voltage generation. But, the low and high levels in aerations do not seem to make many differences in voltage generation (Fig. 2).

Effect of nutrient dosage: Based on the study of the effect of varying glucose concentrations (at two levels viz. 5 g/l and 7.5 g/l), it is observed that glucose concentrations seemed to make no significant differences in the voltage output and in both cases, output voltage reduced to less than 10 mV within first four hours.

However, the stepwise addition of the glucose makes a stepwise increment in voltage immediately followed by a steep decline (Fig. 3).

Effect of nitrogen fluxing: For enhancing the activity of anaerobic degradation in the anodic chamber, nitrogen flux was provided to the anodic chamber. The results were fairly encouraging (Fig.4), showing a stable voltage of 60 ±10 mV after the first 2hrs in contrast to a drastic decline to less than 10 mV within one hour of operation as in the case of the control. Thus, maintenance of anaerobic conditions in the chamber seems to play a vital role in enhancing and stabilizing electricity generation.

Effect of stirring: A high rate of stirring (150 rpm) in the anode chamber showed a distinct higher initial voltage within the first ten minutes compared to the control whereas low stirring seems to have an adverse effect compared to the control.

Interestingly, after the first half an hour, the stirring did not seem to affect the voltage generation. In fact, rather, it showed a marginal lowering of electricity generation than even the control (Fig. 5). Perhaps, this is because of higher voltage generation at optimized turbulence, leading to maximum nutrient availability, as confirmed by studies elsewhere[5].

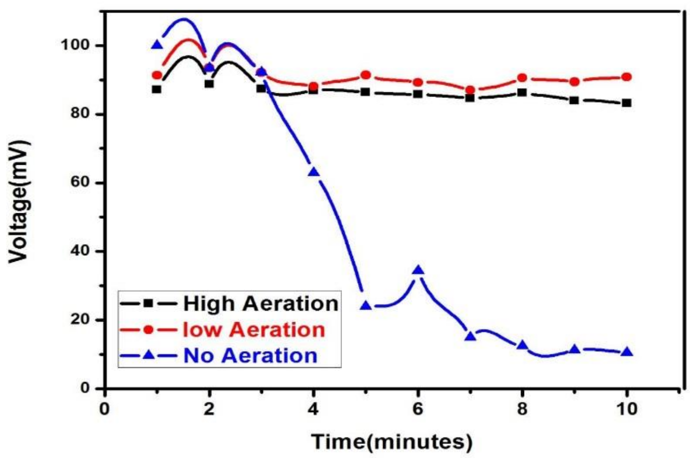

Fig. 2: Effect of Aeration

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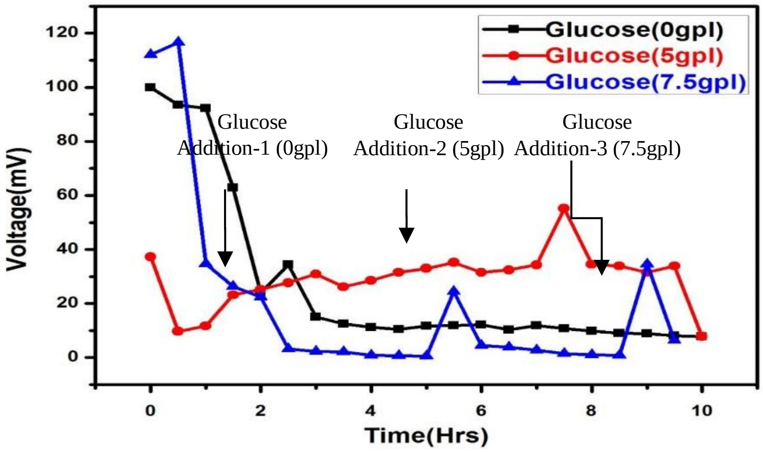

Fig. 3: Effect of Glucose Dosage in Anodic Chamber

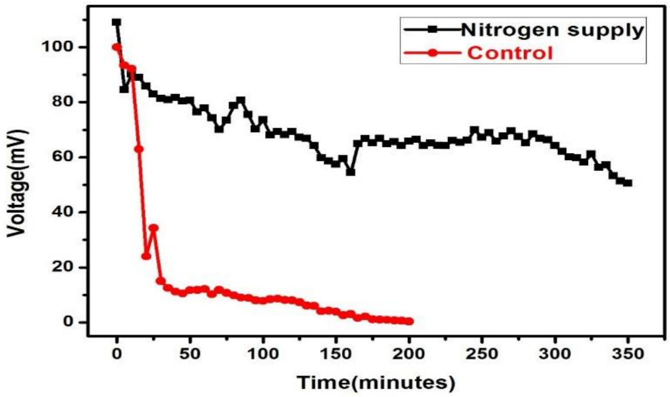

Fig. 4: Effect of Nitrogen Supply

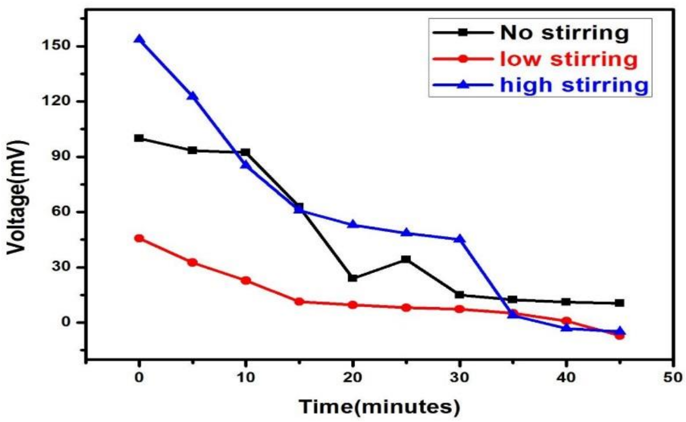

Fig. 5: Effect of stirring

Effect of Anode Geometry: The geometric variables of the anodes were used in the study (namely cross-type, cylindrical-type and circular-type, but with the same total surface area) to evaluate their electron trapping potential through the facilitation of anodic electrons generated in microbial complexes. As the study indicates (Fig. 6), the circular anode showed distinctively higher voltage generations followed by the cylindrical (hollow) and the cross-shaped anode. However, the voltage reduction observed over time in all the cases may be accountable to the conventional decline of voltage in normal MFC rather than specific to the electrode performance. The present work augments the studies by Choudary et al[4] claiming the scope of enhancement in power output in an MFC through the size and surface area of the electrode[4].

Effect of PANI-RGn Composite as Electrode: The study of the PANI-RGn wrapped Al-mesh electrodes showed distinct improvement in voltage output compared to regular Al-mesh electrodes used in the present study. Both the initial voltage and the temporal stabilization of the voltage seemed to show remarkable improvement (30-100%) along with the entire range of the period studied i.e. up to 30 mins (Fig. 7).

Assessment of Bacteriological Status during MFC operation: Micro-morphological state of nanocomposite fabric as viewed in SEM images (before MFC operation) reveal attachment of the PANI-RGn nanocomposite to the material (Fig. 8a). However, post MFC-operational image of spent PANI-RGn nanocomposite illustrates the thick biofilm covering the surface of the PANI-RGn fabric anode (Fig. 8b). The arrow mark displayed on the magnified image in fig. 8b shows the inhabitance of the microorganisms and the biofilm appeared to be completely encased and mixed with the nanoparticles. The microorganisms viewed in the SEM image appear rod-shaped. The gram staining of the surface deposits on the PANI-RGn fabric after the MFC operation indicated the microbe to be gram-negative rod-shaped. Colony morphology based on microbial community analysis (Fig. 9a) displays a yellowish pinpoint colony (construed as Micrococcus sp.), round-shaped smooth colonies (construed as Bacillus sp.) and a mucoidal spread colony (construed as Pseudomonas sp) which are predominant in the microbial community. This rod-shaped structure observed from SEM morphology corresponds to the above mention mucoidal spread colony, which seems to be Pseudomonas sp. as indicated by the arrow mark under microscope to be gram-negative rod-shaped (Fig. 9b).

In fact, Pseudomonas is a typical exoelectrogen strain that solely employs self-generated electron mediators for extracellular electron transfer in MFC's[18,45]. Since, conventionally, the life span of Pseudomonas sp. is 6 hours to 16 months[10] and each division of the present experimental setup (using PANI-RGn fabric) is less than 6 hours, there seems to be an increase in the microbial population during the investigation.

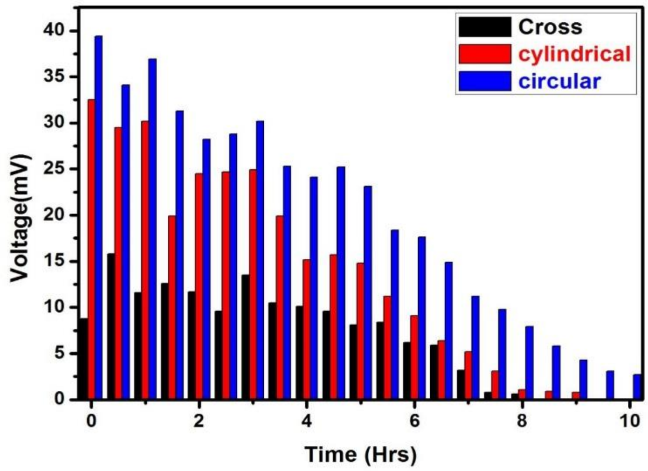

Fig. 6: Effect of Electrode Surface Area

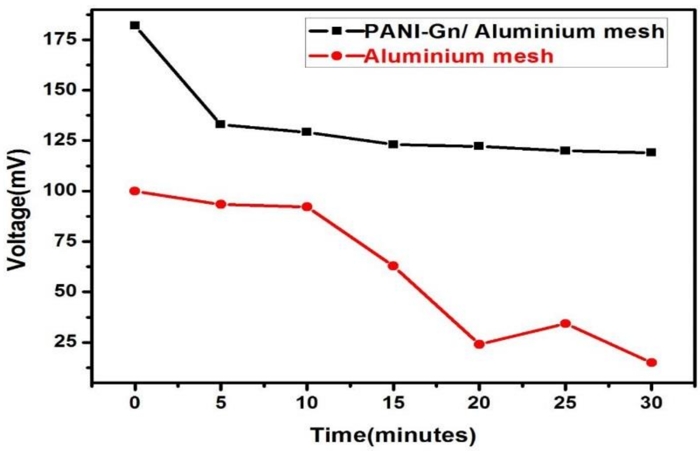

Fig. 7: Effect of PANI-RGn composite as Electrode on Voltage

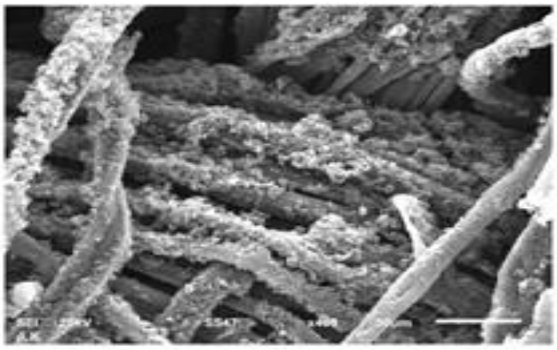

Fig. 8a: SEM image of PANI-RGn composite fabric before experiment

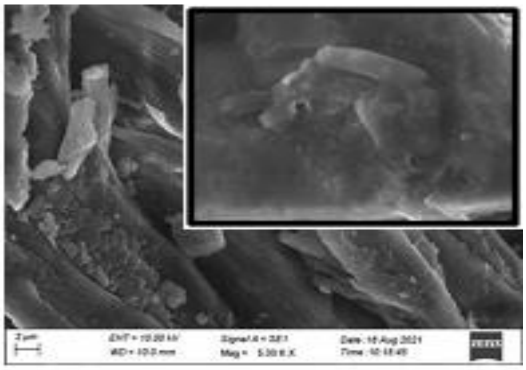

Fig. 8b: SEM image of PANIRGn composite fabric after experiment

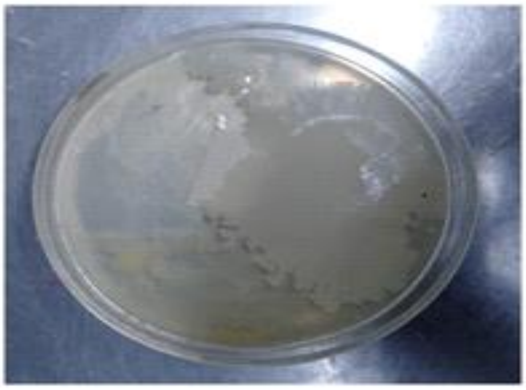

Fig. 9a: Microbial community analysis

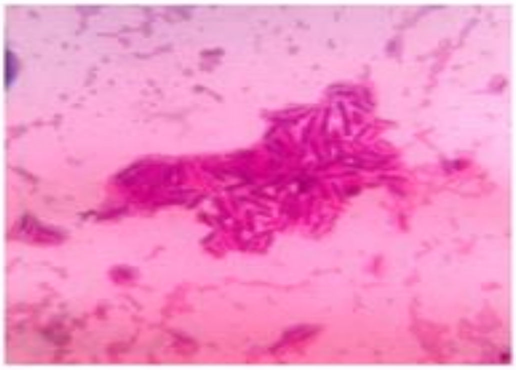

Fig. 9b: Micro copic image of wastewater sample after experiment (Gram-Staining)

Conclusion

The studies on various process parameters of a two-chambered Aluminum electrode and salt bridge PEM-based MFC reveal the following necessary inferences. Of the different parameters used in the present study, the most negligible and most transient effects have been witnessed concerning the cathodic chamber are aeration and anodic stirring, resulting in only a marginal improvement in power generation potential. Although optimal nutrient dosage (glucose in the present study) seems to play a catalytic role in enhancing the MFC's performance, the effect is transient and sporadic, even with repeated addition. The nitrogen fluxing seemed to enhance yield and stability of the power generation through the creation of better anaerobic conditions and thus release in hydrogen molecules.

However, this results in increased operational costs. The study also indicated improvement in output voltage through suitable anode geometry (preferably circular-shaped discs as anodes) supporting more efficient trapping of the electrons from the hydrogen molecules. The most prominent outcome of the current research activity is the distinct, significant and sustained improvement in the voltage output of the MFC through the usage of PANI-RGn composite wrapped onto an Al-mesh electrode, which could provide up to two-fold improvements in the voltage generation, even without the employment of other optimizing parameters necessitating high cost of manufacture and operation (such as nitrogen-fluxing, aeration, glucose-dosage etc.).

The present research using synthetic composite anodes is likely to contribute towards an integrated bioelectricity generation and waste treatment system and this research will also stimulate the construction of moderate, environmentally responsible biocompatible electrodes for bioelectricity generation and treating wastewater. Further research is recommended to explore low-cost alternatives to nitrogen fluxing (possibly via selected microbial culture) and electrode material type, size and shape to obtain more efficient, reliable and sustainable microbial fuel cell systems.

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