On the Effect of the Modified Graphene/Graphite Composite Electrode in the Electricity Generation Efficiency of the Microbial Fuel Cell

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Abstract

As an alternative to traditional carbonaceous material, an electrode constructed by modified graphene film/modified graphite felt composite with enhanced biocompatibility, was employed in a traditional dual chamber Microbial Fuel Cell(MFC). The electrode was fabricated with surface coated graphite oxide felt, SNF composite graphene oxide film, and copper foil tape. In the controlled environment (40℃) with controlled conditions (concentration of electrolyte, surface area of electrode, and type/concentration of chemical catholyte), the MFC fabricated with this type of electrode shows a maximum output power density of 522.2 mW/m^3, which is 3 times higher than that of a carbon felt (175.117 mW/m^3). Also, such a structure successfully avoid most of the procedures that require nanotechnology, thus simplifying the process of fabrication and decreasing the cost.

Keywords: Dual Chamber MFC, Electrode, Graphene, Electricity generation Efficiency

Introduction

Microbial Fuel Cell (MFC) that is capable of converting chemical energy to electrical energy by the bio catalyzed reaction takes place in the anaerobic anode chamber[1,2] has been intensively investigated as a substitute of traditional energy sources (e.g. fossil fuel, solar energy, tidal energy). Different from traditional energy sources like fossil fuel, which emit a large amount of hazardous and greenhouse gases, MFC not only generates electricity without damaging the environment[3], but also helps to decompose the hazardous chemicals, such as Chlorophenol[4] and polluted air with hazardous chemicals (e.g. SO_x, NO_x, CO_2)[5]. Besides, compared to other alternatives which have relatively high cost and unstable energy conversion rate; MFC, however, is able to provide energy up to around 425-850 families (if 500L of wastewater which possess a capacity of diffusion of oxygen of 300mg/L was produced)[6]. The improvement during this century shows that it has great potential to be one of the best alternatives for fossil fuel due to its multiple advantages. Furthermore, the evolutional development of material science in this decade, especially the exploration of graphene; a type of 2D material with extremely outstanding electrical properties; creates a vast amount of opportunities to upgrade the function of an electrochemical energy storage system. MFC, which is also in this category, is worthwhile to explore.

Due to the rapid industrialization and urbanization, the amount of fossil fuel available has been drastically decreasing, imposing a problem of energy deficiency. In addition, as a result of the emission of the greenhouse gases and hazardous gases, the global temperature and the frequency of acid rain are constantly rising. Therefore, it is necessary to develop more effective solutions to both solve energy deficiency and balance the side effect brought by the development of the industry. For example, the 2,4-Dichlorophenol($C_6H_4Cl_2O$), a type of pollutant used as a

bleaching agent to decolorized the raw paper[7] and a component of herbicide[8], is highly corrosive and hazardous to the environment. Its insolubility and vaporization ability impose great danger on species, including humans ourselves[8]. Specifically, its exposure to humans is likely to cause anemia and irreversible nerve damage[8]. Used as the substrate of the MFC; however, this chemical not only allows bacteria to generate electricity but also can be decomposed into a new harmless substance[4].

Various types of model of MFC have been explored (e.g. single-chamber MFC, dual-chamber MFC, ACMFC)[9,10,11]. The configuration of the MFC constructed in this experiment is a traditional dual chamber MFC. While various parameters influence the performance of MFC, according to *Rahimnejad et al*[5], there are 4 predominant influencing factors that is critical for this device: 1). Amount of oxygen and the rate of reaction, 2). Oxidation status of substrate in the anode chamber, 3). electron shuttle from anode chamber to anode surface, and 4). permeability of the PEM. To design a device with higher efficiency, not only the factors listed above should be considered, the material and configuration of the components should also be investigated.

As reported previously, the electrode[5], catholyte[13], type & concentration of organic substrate[4], configuration[9], and type of bacteria[5] are all influential factors that will affect the power output of the MFC. In this experiment, the electrode is selected as a variable due to its important role in both cost and power output played in the device and lack of sufficient research[14].

The criteria of a best-fitting electrode include relatively low cost, outstanding physical and chemical properties (e.g. Coulomb Efficiency, conductivity, chemical and mechanical stability, surface area, specific surface area, and substance decomposed), and biocompatibility.

Previously, scientists categorized materials for electrodes in different groups, including carbonaceous electrodes and metallic electrodes[14].

For the anode, the three most popular structure employed is felt, plane, and packed structure. According to the data collected, Carbon & Graphite (carbon-based) is the group that possesses the highest power density (386W/m^3) with a relatively small volume (156 ml). Another group of materials is carbon with granular activated. With a relatively high working volume (450ml); however, its power density is relatively low, which is only 5W/m^3[14]. For the cathode, there were 4 types of structures have been employed, including plane, packed, tubular, and brush. Among the data collected[14], graphite felt bio-electrode with aerobic and anaerobic sludge possess the highest output power density (83±11W/m^3) with an ultra-small volume (only 40 ml). The next step moving forward is to focus on all aspects of the electrodes. To guarantee the biocompatibility, resistance and surface area, the carbon-based material with different microscopic and macroscopic structures will be employed at the same time in the experiment introduced in this article.

The polluted industrial wastewater purification efficiency and electricity production rate are also closely related to the taxonomy of bacteria used in the MFC. The incorporation of chemical or electronic shuttles in MFCs enables bacteria or even yeast to transfer electrons through biological oxidation-reduction reaction, giving rise to the enhancement of the efficiency of the device. According to *Babanova et al*[15], the utilization of yeast not only gives rise to the formation of current, but also improves the efficiency of the device. Since then, the utilization of shuttles, including neutral red[16] and methyl viologen[17], enabled the popularization of this technique in the experiment. It is plausible to conclude that with all other respect held still, pure bacteria groups are not capable of producing higher power density than the mixed counterpart[5].

Moreover, numerous experiments involved pure bacteria reveal that it has a relatively high resistance, obscuring the path of energy transfer, thus reducing efficiency and power.

The experiment reported here was designed to assess the effect of the use of a modified graphene/graphite composite electrode on the electricity generation efficiency of the MFC and explore the relationship between the resistance and the output power density. In this paper, the methodology for the fabrication of the electrodes, their properties, and their relationship with the electricity generation efficiency will be evaluated and compared with others, fundamentally assessing the role that the change of the electrodes plays in improving the power output of the MFC.

Materials and Methods

2.1 Experimental Set-up and Microorganisms

A dual-chamber MFC separated by the Nafion Proton Exchange Membrane(PEM. Dupon®) was assembled with Acrylic and latex, as shown in **Figure 2.1.** The volume of each device was 512ml with a working volume of 358ml. A removable lid with a pore, which allows the insertion of electrode and connecting wires, was fabricated. For the anode reaction chamber, such a designation ensured the anaerobic environment throughout the experiment.

The PEM was treated to ensure the full elimination of all impurities that are possibly attached on the membrane during the transportation. 1000ml if 5%(w/v) Hydrogen Peroxide solution prepared via the dilution of 30%(w/v) Hydrogen Peroxide solution heated to 80 ℃ with a Thermostat Water Bath(DZKW-4) was prepared for the first 1 hour of immersion. Afterward, the PEM in 1000 ml deionized water for 30 min. During this process, 1000 ml of 5% (w/v) Sulfuric Acid via the dilution of 98%(w/v) Sulfuric Acid solution was prepared and heated to 80℃. The PEM was immersed in the acid and deionized water, respectively for 1h and 30 min.

In this experiment, the Microorganisms cultured was mixed bacteria community, which was collected and segregated from the sludge near the discharging outlet of a paper mill. As a necessary prerequisite, the bacteria were cultivated and stored in an anaerobic environment to ensure the growth and survival of the community. To achieve this, the pump was employed. The medium prepared by the Magnetic heating stirrer(HJ-1) for the cultivation includes glucose, NaCl, KCl, Na_2HPO_4, and KH_2PO_4 with 14, 8, 0.2, 3.63, 0.24 g/l, respectively. The pH of the solution consisting of the chemicals listed above was adjusted to 7.4 with HCl and a pH meter(PHS-25) and will be stored in the ambient temperature, waiting to be used for the biofilm culturing process.

2.2 Electrode Preparation

2.2.1. Modified Graphite felt

The Graphite felt was treated with Collagen I(Y-J Biological®) solution for 24 hours to further improve its biocompatibility[18].

2.2.2. Modified Graphene/Graphite Composite

This type of electrode was assembled from 3 components: pristine graphite felts treated with H_2SO_4, SNF based biocompatible graphene film, and copper foil tape.

To extract the sericin protein, the cocoon was boiled for 30 minutes in 1000ml of Sodium Bicarbonate solution(0.02M). Afterward, the cocoon was rinsed for 2 minutes and soak in water for 1 minute to ensure the full elimination of the sericin protein. After drying, 10g of extracted silk fibroin was dissolved in a 50ml of LiBr(Cool Chemistry®) solution(9.3M) at 60℃ for 4 hours, yielding a 20%(w/v) solution. The solution was dialyzed against the distilled water for 72 hours using a Slide-A-Lyzer Dialysis Cassettes(Pierce®, 3.5K MWCO 30ml) with a 50ml syringe to achieve the complete removal of the salt. The dialyzed solution was centrifuged using an Ultracentrifuge(Beckman Coulter®, Optima XPN-80 Ultra) at 12,000 rpm for 45 minutes to extract the silk aggregates.[19] After 24 hours, the solution was slowly concentrated to over 20%(w/v) to form metastable nanoparticles. After the formation of the nanoparticle, the solution was diluted to 0.5% with double distilled water and incubated for over 24 hours in sealed beakers at 37℃ to ensure the formation of the nanofiber. Graphite and SNF were combined in a kitchen blender(SUPOR®, JS39D-250) with a blade speed of 15 krpm to perform the exfoliation for 20 minutes. The temperature of the solution was controlled below 40℃ during the exfoliation process. The samples were centrifuged at 1500 rpm for 45 minutes at 10 ℃ to segregate the unexfoliated graphite particles with an ultracentrifuge(Beckman Coulter®, Optima XPN-80

Ultra). The supernatant graphene dispersion was collected for further treatment. Fresh silk solution and graphene dispersions were blended at different ratios and cast on polystyrene Petri dishes(diameter 55mm) to prepare transparent composite films. The solution was dried at room temperature in a fume hood in the ambient temperature. The dried films cut into 55mm×5mm squares were treated with methanol for 30 minutes to introduce water insolubility.[20]

After the preparation of all electrodes, each graphite felt electrode was combined with 2 pieces of graphite felts and copper foil tape. Each of the composite electrodes was combined with 2 pieces of graphite felt and 2 pieces of SNF-based-graphene film with copper foil tape cut into the proper size. For convenience purposes, a copper wire was connected to the copper foil tape with solder for each electrode, as shown in **Figure 2.2.**

2.3. Biofilm Cultivation and Device Starting-up

Before the test with inserted electrodes, bacteria have to be attached on the surface of the electrode, especially near the copper foil tape, which is physically attached to the wire connected to the outer circuit, for transportation of electrons released. After the 4 days of cultivation for each electrode, the output voltage and physical appearance were observed. When both the biofilm could be observed with eyes, and the output voltage was stabilized, the electrode will be extracted for further test. The electricigens was tested with a Scanning Electron Microscope (HITACHI®, S-4800, SEM) at 22℃ and 67% of relative humidity with samples made using a dried sterilized paper or silicon plate by smearing the electrode.

After the test of SEM, the cleared anode reaction chamber was fully refilled with 2,4-Dichlorophenol solution (368μM), which was three times higher than the maximum concentration of the industrial sewage in the Baikalsk ecosystem[21]. With the fully dissolved solution, electrodes were inserted to initiate the acclimation and decomposition.

2.4. Physical, Stoichiometric, and Electrochemical Measurements

The resistance of the electrodes was determined by a multimeter(DY-2101) adjusted to 200Ω level across a 7 cm distance in the ambient environment. The samples from anode reaction chamber at time equal to 0 and 24h were placed in an adjusted UV-Visible Spectrophotometer(SOPTOP®, Model 752) with wavelength of 285 nm and deionized distilled water to test the absorbance. After the measurement, calculate the percentage of substrate decomposed with **Equation 1.**, as shown below:

$$\mathbf{^0/_0}\,\boldsymbol{Decomposed} = \mathbf{1} - \left(\frac{\boldsymbol{A_f}}{\boldsymbol{A_i}}\right)$$

where A_f is the final absorbance and A_i is the initial absorbance. The specific surface area of the electrode with **Equation 2.**, as shown below:

$$A_{SS} = \frac{A_S}{m}$$

where A_s is the surface area of the electrode and m is the mass of the electrode.

The time and voltage of the cell collected by Multifunction I/O device(National Instrument®, NI USB-6008) at 40℃ were summarized. Current density and power density were calculated. Surface Area, Specific Surface Area, and Resistance of device incorporated with each electrode were summarized and drawn in **Table 1.** The diagram of Time-Voltage, Current density-Voltage/Power density, Resistance-Power density, Surface Area-Power Voltage, Specific Surface Area-Power density, and Time-Concentration were drawn, as shown in the following sections.

Result and Discussion

3.1. Identification of Microbe and Biofilm Cultivation

Two necessary steps are required to confirm the successful cultivation of targeted bacteria and their distribution on the surface of electrode(e.g. density and survival): the existence of a visible membrane on the surface of electrode and the photographs taken by the Field Emission Scanning Electron Microscope(JSM-7401F, JEOL®) , as shown in the **Figure 3.1 a)-e).** Compare the other images with the **Figure 3.1 a).**, it can be identified that the bacteria cultivated on all electrodes are consistent with that of the sample taken from the bacteria community we collected and segregated(mixed anaerobic bacillus). However, the distribution of them, including the density and the survival rate, is drastically different from electrode to electrode due to their difference in biocompatibility. To ensure the validity of the result, all electrodes were cultivated for exactly 72 hours.

As shown in **Figure 3.1 b).**, the density of bacillus on the surface of carbon felt electrode is moderately dense. Comparatively, it is denser than that of the graphite felt electrode. Since it shows a lower surface area than that of the carbon brush electrode, it is sparser than the density of the latter. Most of the bacteria shown on this electrode survived, which means there is essentially no bio-hazardous material component in this electrode.

As shown in **Figure 3.1 c).**, the density of bacillus of the carbon brush electrode is denser than those of the carbon felt and graphite felt electrodes and slightly sparser than that of the composite electrode. Its large surface area allows it to surpass the other electrode made of the same material. However, without a special surface treatment, it is still more humble than that of the composite electrode. Furthermore, more than 90% of bacteria survived, which means this electrode shows good biocompatibility.

As shown in **Figure 3.1 d).**, the graphite felt electrode, the only electrode that failed to show a visible membrane on its surface, is the sparsest among all other electrodes. Besides, only 75% of bacteria survived, which means the electrode is not biocompatible and possesses bio-hazardous chemicals. Without other changes in material, this should be attributed to the utilization of Collagen I.

As shown in **Figure 3.1 e).** the composite electrode shows the greatest density among all electrode. As the carbon-based material as others, it shows better biocompatibility due to its special modification. In addition, more than 95% of the bacteria survived, which means this electrode shows ultrahigh biocompatibility, contradicting the traditional view of graphene material. The use of Silk Nano Fiber contributed to this.

In summary, the distribution of the bacteria mainly relies on biocompatibility, and surface area. To quantitively analyze the scale of effect of each of the electrode, further researches are required.

3.2. Analysis of the Electricity Generation Efficiency of the Device Incorporated with Each Electrode

As is shown in **Figure 3.2.**, the Voltage, Current density, and Power density are analyzed for each device in the controlled condition. Since the MFC is a device for long-term utilization, the trend of voltage over time and its stability is also crucial. According to **Figure 3.2. a).,** the voltage of the device over an 80 hours shows an overall tendency of increasing for all electrodes.

The modified graphene/graphite electrode shows the greatest voltage among all four electrodes, presenting the maximum voltage of 437.6 mV at 70 hour and fluctuation between 204.4 mV and 414.1mV. The carbon brush electrode shows the second-greatest result, a

maximum voltage of 336.9 mV at 70 hours and a fluctuation between 153.4 mV and 327.4mV. It is relatively stable than that of the composite electrode since, as shown by its relatively small range. However, the former provides more potential. The graphite electrode and carbon felt electrode show much unpromising result, maximum voltage of 254.3 mV and 260.1 mV, respectively. The lower data of the graphite electrode is caused by the failure in cultivating bacteria on the surface of electrode. Since both the amount of the bacteria and the rate of survival of them are much lower than that of the other electrodes, the reaction rate on the graphite electrode is insufficient to generate a higher voltage. As for the carbon felt electrode, this should be attributed to its physical properties since the cultivation status is much better than that of the graphite electrode.

All electrodes show fluctuation in output voltage. Among the four electrodes, the fluctuation of the carbon brush electrode is less intense, showing a rising tendency. However, all the other electrodes show greater fluctuation. The greatest range of the data is as large as 100 mV, showing that the other three electrodes is not as stable as the carbon brush electrode does. Since the temperature is controlled, it should be attributed to their relative humble chemical stability.

3.3. Physical Properties and Power Output

The power output is measured in the power density, as is shown in **Figure 3.2. b).-e).** To evaluate the relationship between the power output and the physical properties and the scale of effect caused by each physical property, the data of power output will be analyzed first.

Same as the voltage graph, the modified graphene/graphite composite electrode shows the maximum power density 522.2 mW/m^3, higher than all of other electrode. Comparatively, the carbon felt, carbon brush and graphite felt electrode shows their maximum power density at

175.1 mV/m^3, 301.1 mV/m^3, and 184.4 mV/m^3, respectively. As is shown in **Table 1.**, the graphite electrode shows the lowest resistance and the carbon brush shows the greatest specific surface area. However, the power density of the former is relatively lower, proving the analysis that the distribution of bacteria that affects it most. On the other hand, the carbon brush does show a promising result. However, since its resistance is much higher than that of other electrodes, it is hard for it to obtain a higher power output. Though the specific surface area of the modified electrode is relative low, its outstanding biocompatibility[20] compensate for this drawback, giving it a comparable result of the cultivation status. With lower resistance, it shows even higher power output than the carbon brush electrode. Showing the second-highest resistance and lowest specific surface area, the carbon felt electrode presents the lowest power output of 175.1 mV/m^3 as predicted.

Since each property affects the result uniquely, it is important to take account of all of the properties mentioned above, instead of focusing on one of them.

3.4. Decomposition Efficiency

During the course of electricity generation, the organic substrate, 2,4-Dicholorophenol is decomposed into water and other harmless organic substance. Using a UV-Visible Spectrophotometer and **Equation 1.**, the percentage of decomposed in the first 24 hours of each experiment is collected and calculated.

The carbon felt electrode shows that 14% of the substrate is decomposed in the first 24 hours. The graphite felt shows that 19% of the substrate is decomposed. The carbon brush shows that 35% of the substrate is decomposed. The modified electrode shows that 42% of the substrate is decomposed. The relation in scale between the decomposition efficiency shares the same trend

relation with the result of output power. Since the biochemical reaction is triggered by the activity of the bacteria, this correalation is understandable.

Conclusion

A new type of composite electrode is designed, fabricated, and operated in a traditional dual chamber MFC. In the data for electrochemical performance from the test, the maximum output power density is 1.7 times higher than that of the carbon brush electrode and 2.8 times higher than that of the graphite felt electrode. In addition, with the greatest rate of decomposition in the first 24 hours, it should be recognized as a proper device for the sustainable development. For now, although the current and power generated is not sufficient for the real-life application, it can be concluded that the modified electrode developed shows promising results for future application.

The data presented in this experiment is from a single MFC device, which means they can be improved with the incorporation of more devices. Acting as the source of energy for small electrical devices, which require higher current instead of voltage, multiple devices incorporated with this electrode can be connected in series. Furthermore, more work should be done on developing proper material and structure for electrode, catholyte. Proper temperature should also be investigated to improve the performance of MFC. With the improvement in every aspect, MFC can be a superior substitution for traditional energy sources.

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Tables

Table 1

Physical Properties of the Electrodes

Type of Electrode	Surface Area (cm^2)	Specific Surface Area (m^2/g)	Resistance (Ω)
Carbon Felt	60.5	1.01×10^{-3}	15.8
Carbon Brush	70.68	9.64×10^{-3}	25.3
Graphite Felt	60.5	1.06×10^{-3}	11.4
Modified	60.5	1.03×10^{-3}	12.2

Note: This table shows the critical physical properties investigated in this research.

Figures

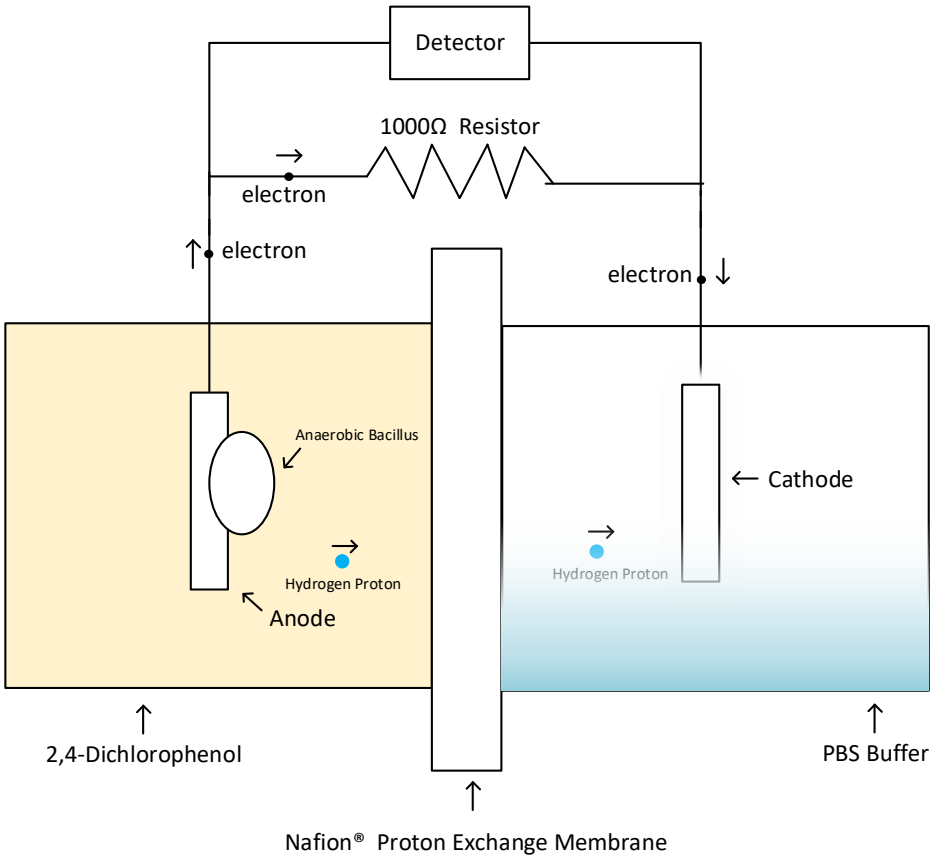


Figure 2.1 The traditional model of a dual-chamber Microbial Fuel Cell

a). b). c). d).

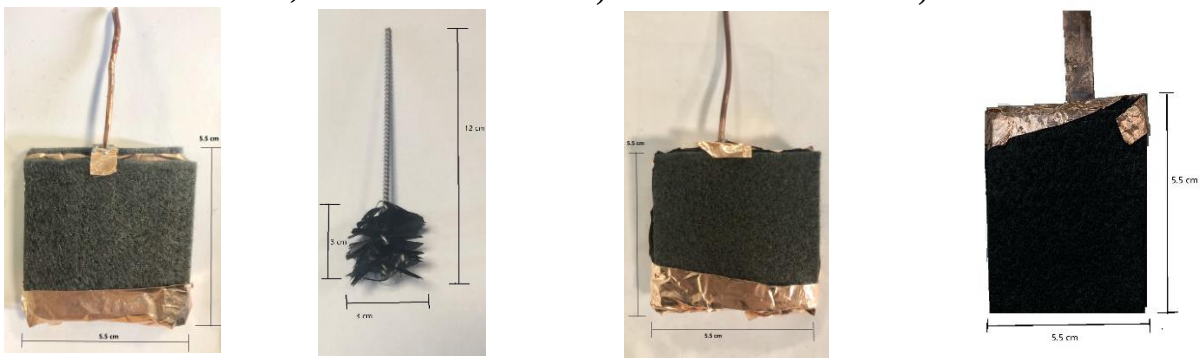
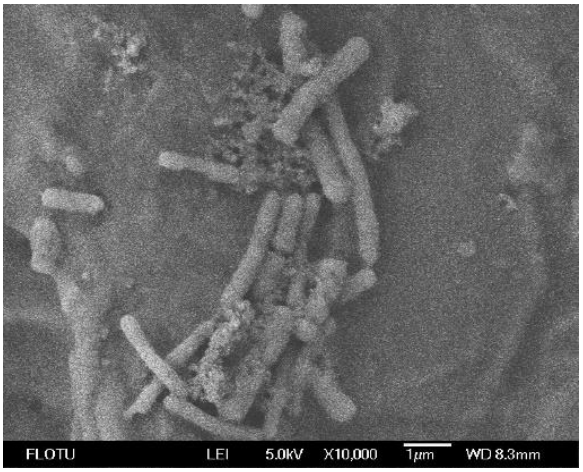


Figure 2.2 The model and structure of electrodes employed in the experiment a). the carbon felt electrode; b). the carbon brush electrode; c). the graphite felt electrode; d). the modified graphene/graphite composite electrode

a).



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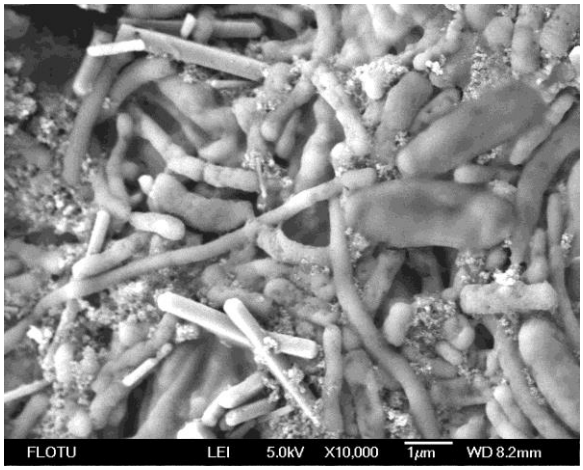

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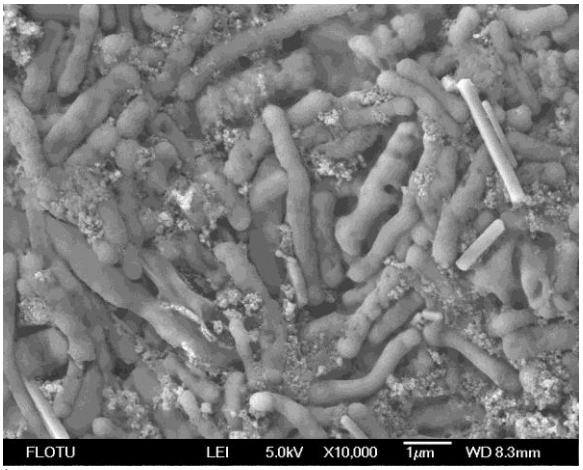

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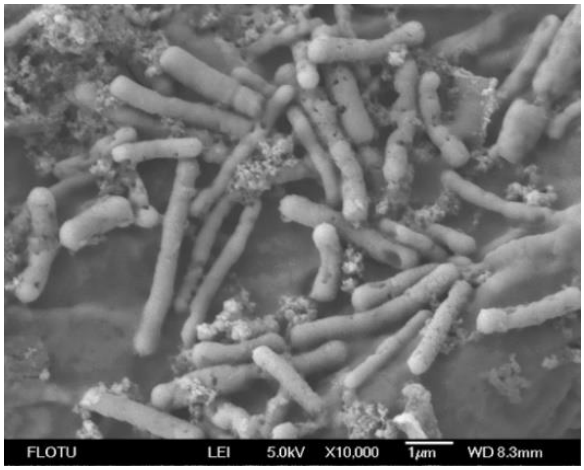
d).



e)

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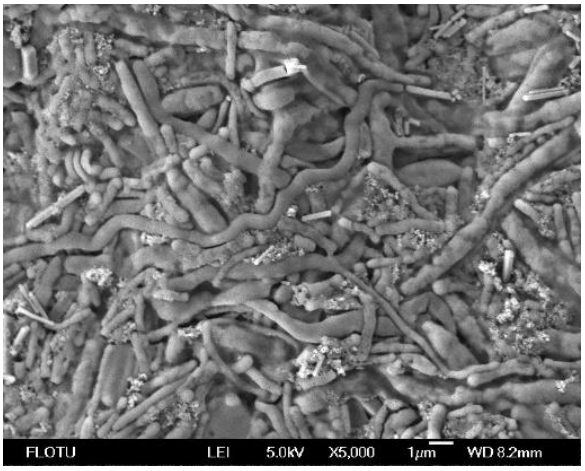

Figure 3.1. photograph of the sample of bacteria taken from the surface of electrode. a). the

bacteria employed; b). carbon felt; c). carbon brush; d). graphite felt; e). modified graphene/graphite composite electrode.

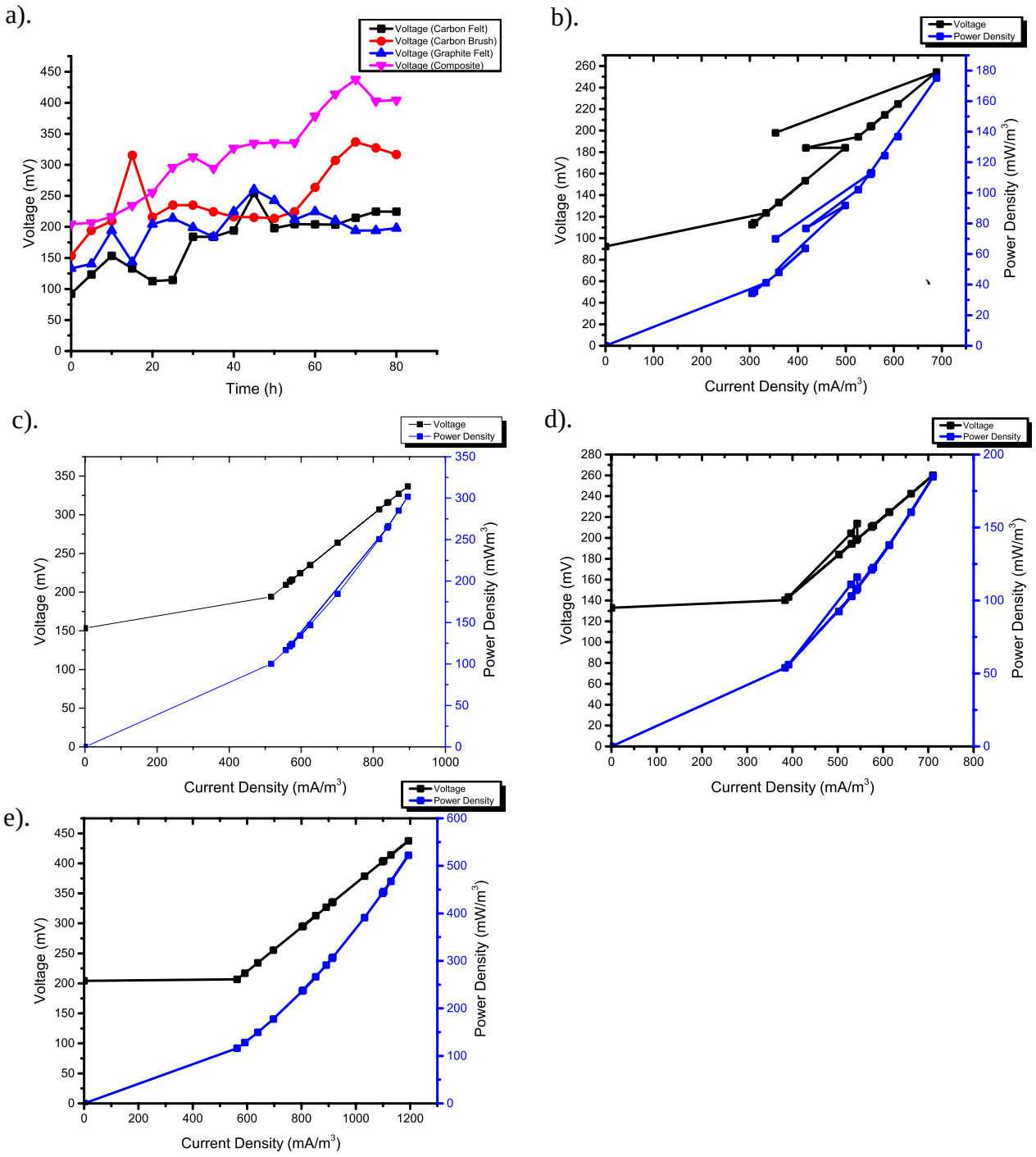

Figure 3.2. The Electrochemical performance of the MFC with different electrodes. a). Time-Voltage graph; b). Current Density-Voltage/Power Density graph of carbon felt; c). Current Density-Voltage/Power Density graph of carbon brush; d). Current Density-Voltage/Power

Density graph of graphite felt; e). Current Density-Voltage/Power Density graph of modified electrode

Contribution

Among the group, each of the three members is responsible for some of the procedures in the group, as introduced:

Zhou Yilin proposed the variable of the project. He was responsible for the designation of the experiment, distribution of the work, analysis of data, and the primary writer of the paper. In addition, he also went to the associated facilities for further tests that cannot be proceeded in Beijing Royal School.

Yuan Jianing acted as the co-designer of the experiment. He was responsible for the designing the procedures with Zhou Yilin, data collection and the graphics depiction. In addition, he also took credit for his prudent and contributing questions raised in the process of experiment, assisting Zhao Erzhuo to better complete his job.

Zhao Erzhuo was the primary experimenter and assistant designer(who looks for the problem of such designation after the primary version is completed) in this project. He was responsible for the purchase of material, operation of the experimental procedures, contact with the associated facilities and the tutor to help us, and check for the problem inside the original designation.