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PALM OIL MILL EFFLUENT TREATMENT AND BIO-ENERGY GENERATION USING HYBRID MICROBIAL FUEL CELL – ADSORPTION SYSTEM

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BACHELOR OF ENGINEERING WITH HONOURS
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PALM OIL MILL EFFLUENT TREATMENT AND BIO-ENERGY GENERATION USING HYBRID MICROBIAL FUEL CELL – ADSORPTION SYSTEM

JUSTINA ROSE A/P SELVANATHAN

A Dissertation submitted in partially fulfillment of the
requirement for the degree of
Bachelor of Engineering with Honours
(Chemical Engineering and Energy Sustainability)

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Dedicated to my late father Selvanathan and beloved mother Anniammah, my lovely siblings and my friends who are always there for me for unconditional love, supports and encouragements

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ABTRACT

Palm oil mill effluent (POME) discharged without effective treatment into watercourses can pollute the water source due to its high acidity, biochemical oxygen demand (BOD), and chemical oxygen demand (COD). As an approach to overcome the limitation of the existing POME treatment methods, a hybrid microbial fuel cell-adsorption (MFC-Adsorption) system is introduced as an innovative and progressive technology that is able to treat wastewater effectively and at the same time generate electricity. However, the bio-energy generated by the system is relatively low which is not practical for industrial application. This study aims to investigate the effects of different types of mediator and pH on the performance of the hybrid MFC-Adsorption system by using POME as the substrate. The viability of the hybrid MFC-Adsorption system in generating bio-energy in the form of voltage, power density and current was investigated. The air-cathode single chamber hybrid MFC-Adsorption system with different types of mediator such as congo red, crystal violet and methylene blue was fabricated and the characterization of POME was conducted using COD, BOD, total suspended solids (TSS), ammoniacal-nitrogen (AN) and turbidity tests to evaluate the efficiency of the hybrid MFC-Adsorption system to treat POME. From this study, 120.58 mV, 168.63 mV and 189.25 mV of voltage was generated in MFC-Adsorption system with congo red, crystal violet and methylene blue as mediator, respectively when using 50 Ω external resistances. The current generation of 2.41 mA,3.37 mA and 3.79 mA as well as power generation of 290.79mW/m^3, 568.72 mW/m^3 and 716.31mW/m^3 were produced respectively by the MFC-Adsorption system with these three mediators. Besides, these systems achieved BOD removal of 73.2%, 74.2% and 75.3% while the COD removal up to 84.1%, 84.3% and 84.8% was achieved for the MFC-Adsorption system with congo red, crystal violet and methylene blue as mediator, respectively. The TSS removal of 90.8%, 90.77% and 91.5% as well as turbidity removal of 83.3%, 83.3% and 86.1% was obtained, respectively with these three mediators. The maximum AN removal of 21.5% was achieved by MFC-Adsorption system with congo red as the mediator while 22.0% and 23.31% was obtained respectively for system with crystal violet and methylene blue as the mediator. Overall, the MFC-Adsorption system fabricated in this study was feasible to be applied for POME treatment as the effluent concentration was able to comply with the discharge standards imposed by Department of Environment, Malaysia.

ABSTRAK

POME dilepaskan tanpa rawatan yang efektif ke dalam saluran air, boleh mencemarkan sumber air disebabkan oleh keasidan yang tinggi, kandungan oksigen biokimia (BOD), dan kandungan oksigen kimia (COD). Sebagai pendekatan untuk mengatasi batasan kaedah rawatan POME, sistem hibrid *MFC-Adsorption* diperkenalkan sebagai teknologi inovatif dan progresif yang membawa banyak kelebihan dalam merawat air sisa secara berkesan, ekonomi dan mesra alam telah ditekankan. Sistem *MFC-Adsorption* telah terbukti dapat menyingkirkan bahan pencemar dalam POME termasuk COD, BOD, jumlah pepejal terampai (TSS), ammoniacal-nitrogen (AN) dan kekeruhan, namun tenaga bio yang dihasilkan agak rendah. Kajian ini bertujuan untuk mengkaji kesan-kesan dari pelbagai jenis *mediators* dan pH terhadap prestasi sistem hibrid *MFC-Adsorption*. Daya maju sistem hibrid *MFC-Adsorption* dalam menghasilkan tenaga bio dalam bentuk voltan, ketumpatan kuasa dan arus juga dikaji. Sistem hibrid *MFC-Adsorption* dengan jenis-jenis *mediators* seperti kongo merah, kristal violet dan biru metilena telah direka dan pencirian POME mentah dan POME yang dirawat telah dijalankan dengan menggunakan ujian-ujian COD, BOD, jumlah pepejal terampai (TSS), ammoniacal-nitrogen (AN) dan kekeruhan untuk menilai kecekapan system hibrid *MFC-Adsorption* untuk merawat POME. Dari kajian ini, 120.58 mV, 168.63 mV dan 189.25 mV voltan telah dihasilkan dalam system hibrid *MFC-Adsorption* dengan kongo merah, kristal violet dan metilena biru sebagai *mediator* masing-masing apabila menggunakan 50 Ω rintangan luar. Penjanaan arus 2.41 mA, 3.37 mA dan 3.79 mA manakala penjanaan kuasa 290.79 mW/m³, 568.72 mW/m³ dan 716.31 mW/m³ dihasilkan masing-masing oleh system hibrid *MFC-Adsorption* dengan tiga jenis *mediator* yang dikaji. Di samping itu, sistem ini berjaya menyingkirkan BOD sebanyak 73.2% (78 mg/L), 74.2% (75 mg/L) dan 75.3% (72 mg/L) manakala penyingkiran COD adalah 84.1% (68.21 mg/L) , 84.3% (67.38 mg/L) dan 84.8% (65.21 mg/L). Peratusan penyingkiran TSS sebanyak 90.8% (24 mg/L), 90.77% (24 mg / L), dan 91.5% (20 mg / L) dan penyingkiran kekeruhan sebanyak 83.3% (12 NTU), 83.3% 86.1% (10 NTU) diperoleh masing-masing dengan tiga jenis *mediator*. Penyingkiran AN maksimum sebanyak 21.5% dicapai untuk sistem MFC-Adsorption dengan kongo merah sebagai pengantara manakala 22.0% dan 23.31% masing-masing untuk sistem kristal ungu dan metilena biru sebagai mediator. Secara keseluruhannya, system hibrid *MFC-Adsorption* yang dihasilkan dalam kajian ini boleh digunakan untuk rawatan POME kerana kualiti efluen yang dihasilkan dapat mematuhi *regulation standard* Jabatan Alam Sekitar, Malaysia.

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ABBREVIATION

ACFF	Activated carbon fiber felt
AN	Ammoniacal nitrogen
BOD	Biochemical oxygen demand
COD	Chemical oxygen demand
FFB	Fresh fruit brunch
GAC	Granular activated carbon
GFB	Granular fibre brush
GG	Graphite granular
HRT	Hydraulic retention time
MFC	Microbial fuel cell
PAC	Powdered activated carbon
PEM	Proton exchange membrane
POME	Palm oil mill effluent
TOC	Total organic carbon
TS	Total solid
TSS	Total suspended solid
TVS	Total volatile solid

NOMENCLATURE

°C	Degree Celsius
μl/L	Microliter per liter
A	Ampere
cm	Centimeters
cm^3	Cubic centimeters
g	Grams
h	Hours
I	Electric current
kg	Kilograms
L	Liters
L/min	Liters per minute
mA	Milliampere
mg/L	Milligram per liter
ml	Milliliters
mW/m^2	Milliwatt per square meter
mW/m^3	Milliwatt per cubic meter
mV	Millivoltage
NTU	Nephelometric turbidity unit
V	Volts
W/m^2	Watt per square meter
W/m^3	Watt per cubic meter
μA	Microampere
μM	Micromolar
Ω	Ohms

CHAPTER 1

INTRODUCTION

1.1 Palm Oil Mill Effluent (POME)

The palm oil mill industry is the continuous and successive developmental industry in Malaysia over current era where according to the search it is stated that Malaysia produced approximately 80 million dry tonnes of solid biomass in 2010 and is expected to increase to about 100 million dry tonnes by 2020 (Islam et al., 2016). The positive progress of palm oil industry gives serious environmental impact. Palm oil mill industries are large industrial consumer of water as well as producer of wastewater known as palm oil mill effluent (POME). POME is a yellowish acidic wastewater with fairly high polluting properties with complex substrates comprising amino acids and some inorganic nutrients such as sodium, potassium, calcium, magnesium, short fibers; organelles, nitrogenous constituents, free organic acids, and a mixture of carbohydrates ranging from hemicelluloses to simple sugars (Aremumuyibi et al., 2014). Besides that, there are also some microorganisms associated in POME (Soleimaninanagegani et al., 2014). POME discharged without effective treatment into watercourses, can pollute the water source due to its high acidity, temperature, biochemical oxygen demand (BOD), and chemical oxygen demand (COD). Therefore, a sustainable approach to handle this problem will be to convert it to a useful recyclable product at site by an eco-friendly and economical method (Hayawin et al., 2013).

1.2 POME Treatment Methods

Over the past decades, several technological solutions have been introduced for the treatment of POME. Table 1.1 shows the various POME treatment systems with their respective advantages and disadvantages.

Table 1.1: Comparison of POME treatment systems (Bala et al., 2014)

POME Treatment Systems	Advantages	Disadvantages
Ponding System	Low cost	Requires high land space
Anaerobic Digestion	Does not require additional energy source and easy to conduct and operate	Requires long retention time, produces undesirable odors, produce sludge deposit and able to reduce small amount of ammonia
Aerobic Bacteria	Shorter retention time, more effective in handling toxic wastes	High energy requirement (aeration), rate of pathogen inactivation is lower in aerobic sludge compared to anaerobic sludge, thus unsuitable for land applications
Membrane	Produces consistent and good water quality after treatment, smaller space required for membrane treatment plants, can disinfect treated water	Short membrane life, membrane fouling, expensive compared to conventional treatment
Evaporation	Solid concentrate from process can be utilized as feed material for fertilizer manufacturing	High energy consumption

1.3 Energy Source

Today we are witnessing a global energy crisis due to huge energy demands and limited resources. The usage of fossil fuels, which have supported industrialization and economic growth in all world economies as the main energy source, has led to the danger of extinction and increasing environmental concerns (Ozturk & Onat, 2017). It is due to the growth of world population, an escalating in demand and continued dependence on fossil-based fuels for generation. Moreover, the non-renewable energy sources are depleting and renewable energy sources are not properly utilized. There is an immediate need for search of alternative routes for energy generation. Therefore, many alternatives of energy around the globe and energy from fuel cell has been identified as one of the potential renewable energy sources. In a fuel cell, chemical energy of a reaction is converted to electricity with by-products of water and heat (Tee et al., 2016b).

1.4 Microbial Fuel Cell (MFC)

Considering the negative effects of the existing techniques to treat POME, it is necessary to come up with an effective and practical approach to protect the environment as well as to balance the economy. Microbial fuel cell (MFC) technology, which uses microorganisms to transform chemical energy of organic compounds into electricity, is considered a promising alternative method for solving this problem (Chaturvedi et al., 2016). MFC is gaining more and more interest as an alternative eco-friendly energy source which is simultaneously used for the treatment of POME and generation of electricity (Khan et al., 2016). MFC has operational and functional advantages over the technologies currently used for generating energy from organic matter found in wastewater. MFC technology offers novel alternative and cost effective approach of energy generation directly from the oxidation of waste organic matter and renewable biomass in the form of electricity with less sludge production as compared to aerobic processes (Baranitharan et al., 2013).

MFC technology produce harnesses energy from metabolism of microorganisms, seems to be attractive to warrant energy generation. The use of MFC as an alternative source for power generation is considered as a reliable, clean, efficient process, which utilizes renewable method and does not generate any toxic by-product (Chaturvedi et al., 2016). Therefore, MFC is feasible to be used for POME treatment.

1.4.1 Working Principle of Single Chamber MFC

MFC consists of anode and cathode chambers separated by a proton (ion) exchange membrane (PEM) (Agarry et al., 2016). An anode respiring bacterium digests the organic waste to carbon dioxide and transfers the electrons released to the anode. This are several possible mechanisms for electron transfer from the microorganism to the anode, which involve direct electron transfer via outer membrane cytochromes, mediators and nanowires (Hisham et al., 2013). The electrons thus produced are transferred to the anode with the help of mediators which are produced by the bacteria; exogenous mediators (ones external to the cell) such as methylene blue, thionine, neutral red or by direct transfer of electrons from electrochemically active bacteria cells (cytochromes) to the electrode. Therefore, these mediators are considered electrophores and support current generation. Application of mediators in MFC is necessary due to the non-conducting nature of most of the microbial cell surfaces which implies low electron transfer efficiency (Agarry et al., 2016). Figure 1.1 shows the schematic diagram of single MFC.

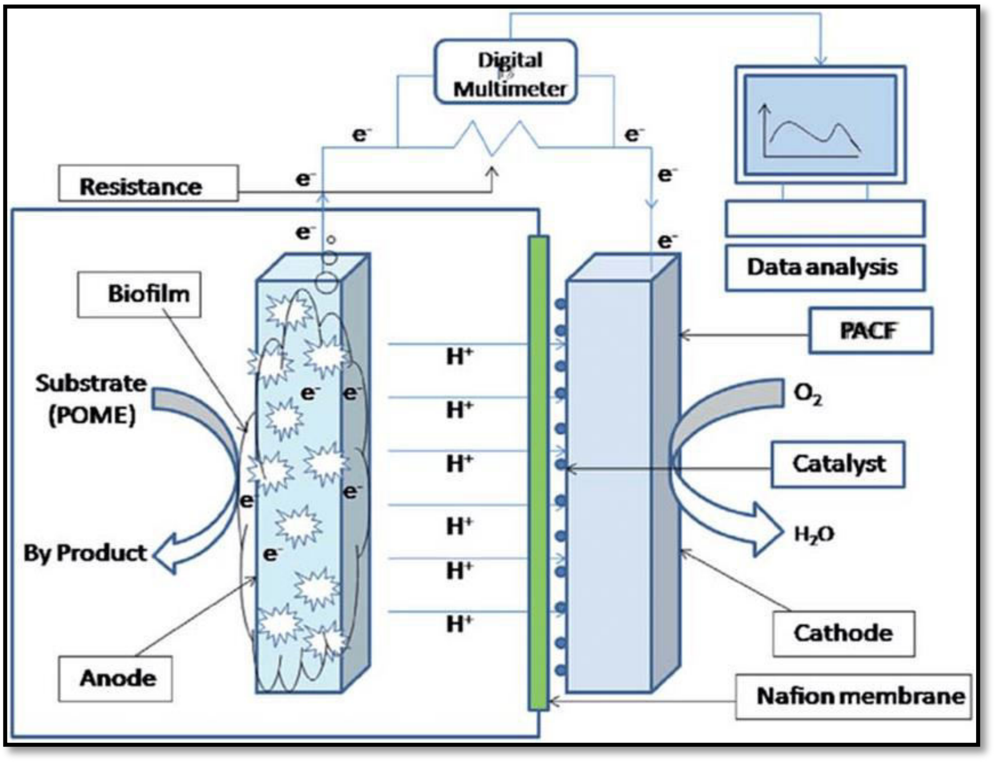

Figure 1.1: Schematic diagram of single microbial fuel cell (Islam et al., 2016)

1.4.2 Hybrid MFC-Adsorption System

According to Tee et al. (2016b), up to 70% of the COD could be removed from various wastewaters by utilizing MFC. Furthermore, according to Tee et al. (2016a), single MFC could be categorized under chemical-biological hybrid system while the hybrid MFC could be categorized under physical-chemical-biological hybrid system with the addition of granular or powdered activated carbon as an adsorbent. This system made of earthen pot was designed and tested simultaneously for wastewater treatment and energy recovery. Biological, electrochemical as well as physicochemical processes from the activated carbon used in the system would improve the wastewater quality. Usage of granular activated carbon (GAC) with MFC was able to increase the electricity generation at low COD concentration due to adsorptive capacity of GAC. Moreover, combined a graphite fiber brush (GFB) with graphite granules (GG) as composite electrodes was designed by Li et al. (2013) as an anode material for a tubular MFC which demonstrated higher power density. Besides that, usage of mediator such as methylene blue, natural red, tetramethyl-p-phenylenediamine, thionine and ferricyanide in anode chamber of hybrid MFC-Adsorption system was examined by Agarry et al. (2016) and showed that mediator improved the electricity generated in MFC and helped the electrochemically active bacteria from POME to transfer the electrons to the electrode as well as to improve the performance of MFC by increasing the power output of the cell.

Activated carbon plays as the adsorbent for contaminant removal in water treatment plant in order to remove COD, TSS, toxicity, color, odor, and turbidity. Integration of MFC with adsorption system and mediator is considered a more efficient method because integration of GAC and mediator with MFC are able to deliver higher electricity generation at low COD concentration due to the adsorptive capacity of GAC that helps in bacteria adhesion on the anode and mediator as electron promoter (Tee et al., 2016b). Thus, integration of MFC system with adsorption system can be a great hybrid system to improve the wastewater quality besides achieving bio-energy from the system.

1.4.3 pH as Factor Affecting MFC Performance

The value of pH in MFC plays a significant role on the activity of bacteria in terms of COD removal rate and electrical energy production. The optimal range of pH best for the methane-producing bacteria was observed to be in the range of 6.3 – 7.8 by Lim (2012). Apparently, any pH value lower than 5.5 is suitable for acidogenic bacteria to survive. Within this condition, the organic removal rate is expected to decrease as compared to neutral and alkaline conditions and only hydrogen production would be the dominant mechanism. Due to low removal rate, less electron will be produced and leads to lower electricity generation (Marashi & Kariminia, 2015). In the study conducted by Marashi and Karimina (2012), the highest power density was observed under alkaline condition with pH 8.5 due to inactivation of acidogenic and methanogenic bacteria in favor of more activity for electrogenic bacteria. Therefore, by stabilizing the pH value to an optimum point to achieve the required high power density is one of the efficient methods.

1.5 Problem Statement

POME containing high COD and BOD can cause environmental pollution when discharged without proper treatment (Chin et al., 2013). The POME bioremediation technology using ponding system requires large amount of land, long hydraulic retention time, produces bad odour and difficult in maintaining the liquor distribution and biogas collection. Besides that, aerobic treatment system requires high energy and is not suitable for land application due to its lower rate of pathogen inactivation. Due to the deficiency of these technologies, numerous searches for an eco-friendly method that can treat POME effectively and economically have been given priority. Microbial fuel cell (MFC) is considered as a new source of energy where it uses bacterial metabolism to generate electrical current and at the same time serving as solution for wastewater treatment while removing the organic compounds. Moreover, activated carbon adsorption process has been proven effective in removal of contaminants in POME.

Hybrid microbial fuel cell-adsorption system is an innovative and progressive technology. It carries the advantages in treating wastewater effectively, economically and environmental friendly as well as being able to generate bio-energy simultaneously. Besides that, reduction of cost due to integration of MFC with adsorption system using activated carbon will eliminate the aeration unit that consumes 50% of the electricity used at a treatment plant (Tee et al., 2016b). Hybrid MFC-Adsorption system has been proven effective in removing contaminants from POME and is capable to deliver higher electricity generation at low COD concentrations (Tee et al., 2016b).

Addition of methylene blue as mediator in the MFC-Adsorption system has been shown to be effective in reducing the pollutants in POME, however, the system still demonstrated relatively low bio-energy generation (Salim, 2017) which not practical for industrial application. Besides that, pH is a significant parameter affecting the performance of MFC-Adsorption system where the optimal range of pH for the methane-producing bacteria was reported to be in the range of 6.3 – 7.8 by Lim (2016). Therefore, this study aims to investigate the effects of different types of mediator and pH on the performance of the hybrid MFC-Adsorption system in term of pollutant removal efficiency and bio-energy generation.

1.6 Research Questions

The research questions to be addressed in this study are:

1. Which type of mediator will give the best treatment efficiency and bio-energy generation using the hybrid MFC-Adsorption system?
2. What is the effect of pH on performance of the hybrid MFC-Adsorption system?