## ELECTROCOAGULATION OF PALM OIL MILL EFFLUENT TREATMENT

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#### DEDICATION

This thesis is dedicated to my beloved husband who loved me unconditionally. To my late father, who taught me that there is always a way to achieve a dream. To my mother, who taught me to become stronger every time. To my late child, who motivated me to finish this study

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#### ABSTRACT

Palm oil mill effluent (POME) categorized as highly polluted wastewater. The conventional methods have limitations. Electrochemical process offers several advantages however the previous study on the electrode material such as aluminum and iron show removal rate lower than 50%. Thus, this study investigates the electrochemical process of POME treatment using platinum and graphite as anode materials after the selection process. Response surface methodology was used to study the effect between different operating conditions such as voltage, electrolysis time and electrolyte support and responses of the treatment including chemical oxygen demand (COD) removal, colour reduction and electrolyte support. A quadratic mathematical model was developed for all responses using Box-Behnken Design (BBD) with  $R^2$ 0.9853 for COD removal, R<sup>2</sup> 0.9478 for colour reduction and R<sup>2</sup> 0.9185 for oil and grease removal on platinum as anode material. Meanwhile R<sup>2</sup> 0.9844 for COD removal, R<sup>2</sup> 0.9412 for colour reduction and R<sup>2</sup> 0.9724 for oil and grease removal. According Derringer's function desirability, under the optimum condition such as voltage 15 V, electrolysis time 2 hours and 19.95 mg/L NaCl of POME treatment using platinum could achieve  $84\pm2\%$  COD removal,  $98\pm2\%$  colour reduction and  $99\pm1\%$  oil and grease removal. For graphite material under optimized conditions such as voltage 14 V, electrolysis time of 3 hours and electrolyte amount of 13.41 g/L show that electrochemical process achieve 56±4% COD removal, 65±1% colour reduction and 94±2% oil and grease removal. The total operating cost both for platinum and graphite under optimum conditions are US\$ 18.346/L and US\$16.6641/L of treated POME. On the other hand, the operating cost for conventional method US\$ 6.63/L of POME with drawback 50% removal of COD and requires more than 24-hour retention time. These results indicate the platinum and graphite as anode material is applicable and effective for POME treatment.

#### ABSTRAK

Sisa pembuangan kilang minya kelapa sawit (POME) di kategorikan sebagai sisa pembuangan yang sangat tercemar. Kaidah konvensional memiliki banyak kekurangan. Sistem elektrokimia menawarkan beberapa kelebihan, tetapi kajian terdahulu mengenai penggunaan bahan elektrod menunjukkan hasil rawatan kurang dari 50%. Oleh karena itu, kajian ini di lakukan untuk mengkaji POME menggunakan proses elektrokimia dengan menjadikan platinum dan grafit sebagai bahan anod. Metodologi permukaan tindak balas digunakan untuk mengkaji kesan antara keadaan pengoperasian seperti voltan, masa pengoperasian elektrolisis dan bahan elektrolit penyokong serta hasil dari rawatan tersebut yang termasuk pengurangan kadar organik komponen (COD), pengurangan kadar warna serta pengurangan kadar kandungan minyak. Sebuah kuadratik model matematik telah di kembangkan untuk semua kesan hasil dari pengoperasian menggunakan reka bentuk Box-Behnken dengan nilai R<sup>2</sup> 0.9853 untuk pengurangan kadar COD, R<sup>2</sup>0.9478 untuk pengurangan kadar warna dan  $R^2$  0.9185 untuk pengurangan kadar kandungan minyak yang menggunakan platinum sebagai bahan anod nya. Sedangkan nilai R<sup>2</sup> 0.9844 untuk pengurangan kadar COD,  $R^2$  0.9412 untuk pengurangan kadar warna and  $R^2$  untuk pengurangan kadar kandungan minyak dengan grafit sebagai bahan anod nya. Berdasarkan fungsi Derringer, dalam keadaan optima seperti 15 voltan dalam masa 2 jam dan 19.95 mg/L NaCl dengan menggunakan platinum dapat mencapai 84(±2)% pengurangan kadar COD, 98±2% pengurangan kadar warna dan 99±1% pengurangan kadar kandungan minyak. Sedangkan untuk grafit pula dalam keadaan optima seperti 14 voltan dalam masa 3 jam dan 13.41 g/L NaCl menunjukkan pengurangan kadar COD sebanyak 56±4 %, pengurangan kadar warna 65±1% dan pengurangan kadar kandungan minyak 94±2%. Jumlah kos pengoperasian masing-masing bahan anod ialah US\$ 18.346/L and US\$16.6641/L dalam jumlah POME yang di rawat. Dalam masa yang berlainan, jumlah kos pengoperasian rawatan menggunakan kaidah konvensional sebanyak US\$ 6.63/L dengan kekurangan sebanyak 50% pengurangan kadar COD dan memerlukan masa pengoperasian melebihi 24 jam. Hasil ini bermaksud bahwa platinum dan grafit sesuai di guna pakai dan efektif untuk rawatan POME.

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## LIST OF ABBREVIATIONS

ANOVA	-	Analysis of Variance
AP	-	Adequate Precision
BBD	-	Box-Behnken Design
BOD	-	Biological Oxygen Demand
COD	-	Chemical Oxygen Demand
CV	-	Coefficient Variance
DC	-	Direct Current
HRT	-	Hydraulic Retention Time
MFC	-	Microbial Fuel Cell
OOMW	-	Olive Oil Mill Wastewater
POME	-	Palm Oil Mill Effluent
RSM	-	Response Surface Methodology

## LIST OF SYMBOLS

°C	-	Celcius
\$	-	Dollar
w/v	-	Weight per volume
μm	-	Mikrometer

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#### **CHAPTER 1**

#### **INTRODUCTION**

#### **1.1 Background of Study**

The supplies of water usage in the world contribute to 8% local, 22% industry and 70% in agriculture (UNESCO, 2003). Agriculture is the biggest sector which consume water supply. A substantial part of this water is discharged into environment as wastewater (Zeraatkar et al., 2016). One of major agriculture sector is vegetable oil, which comes from a variety of sources including coconut, cottonseed, olive, palm, peanut, grapeseed, soybean and sunflower seed. The wastewater characteristic varies in both quantity and the organic content depending on the types of industry and the types of milling process (Chipasa, 2001).

One of the well-known oil industries in several ASEAN countries is palm oil. The contribution of palm oil on economical is significant. However, palm oil industries produce huge amount of palm oil mill effluent (POME) (Tan et al., 2018). In fact, Malaysia as second largest production palm oil in Southeast Asian produces 19.6 MT on 2020 (USDA, 2020). As the production of palm oil industries is high therefore the demand for POME treatment is high. POME is generated from sterilization of fresh oil palm fruit bunches, clarification of palm oil and effluent from hydro-cyclone operation when it is undergoes on palm oil milling (Borja et al., 1996). A study reported by Prasertsan and Prasertsan (1996) shows that on the process of palm oil milling, more than 70% (w/v) of the processed fresh fruit bunch (FFB) was left over as oil palm waste.

Untreated POME is a major source of inland water pollution when released in the local irrigation system (Abdurrahman et al., 2018). The conventional method for POME treatment includes physical, chemical and biological treatment. They are adsorption, coagulation, flocculation (Keeley et al., 2014), composting (Adam et al., 2016) and open ponding system. The most common method has been used in Malaysia is open ponding system. The final discharge from this system is released to the river (Chin et al., 2013). However, the discharged of this system from several palm oil industries do not meet standard regulatory discharge limit (Zahrim et al., 2014) in term of parameters including colour (Tamrin and Yaser, 2017), chemical oxygen demand (COD), biological oxygen demand (BOD) and ammonia content (Hosseini and Wahid, 2013). This system also requires long hydraulic retention time (HRT), extensively large land space and greenhouse gasses emission (Ahmed and Theydan, 2014). In addition, there are no studies reported on oil and grease removal using open ponding system.

Current research for POME treatment as an alternative varies from biological, physical and chemical treatment processes. They are including MFC (Microbial Fuel Cell) (Islam et al., 2016), microalgae cultivation (Saidu et al., 2017), photocatalysis (Aris et al., 2017), catalytic steam reforming (Cheng et al., 2018), etc. However, these methods are expensive and require more effort to improve performance (Gormez et al., 2020). Another alternative is electrochemical treatment that possible for POME treatment. Compared to the conventional open ponding system, electrochemical treatment has several advantages such as easy to control, inexpensive, less sludge formation (Mussa et al., 2015), less external chemicals, lower secondary pollution and lower retention time compare to conventional method (Prajapati et al., 2014)

This study aims to using electrochemical process for POME treatment. Electrode materials have significant effect for electrolysis performance. The effective combinations of electrode materials will be selected from different type of electrode materials. The electrolysis performance will be optimized using RSM (Response Surface Methodology) followed by operating cost analysis from optimum condition.

#### **1.2 Problem Statement**

Fresh POME typically contains a high amount of organic matter with complex characteristics (Healy and O'flynn, 2011). POME has been classified as highly

pollutant-containing wastewater due to its' high amount of nutrient (i.e., ammoniacal nitrogen = 220 mg/L) and organic matter (i.e., COD value = ~ 50,000 mg/L, BOD value = ~ 20,000 mg/L) (Saeed et al., 2016; Razali et al., 2020). POME has been well-known as great source of water and groundwater pollution. The characteristic of POME which is acidic, viscous and contaminating nature cause serious water pollution and aquatic ecosystem (Abram et al., 2015). Conventional method for POME treatment such as open ponding system has been widely used by palm oil industries. Several limitations of this system are the performance is not sufficient to meet regulations for the final discharge of POME, especially in term of colour and COD removal. POME also contains high concentration of oil and grease reaches 4,000 mg/L (Kamyab et al., 2017). Previous study reported adsorption method of POME treatment resulting only 57.7% removal of oil and grease (Wahi et al., 2016). There is also no study reported on oil and grease removal using electrochemical treatment. Thus, the demand of another alternative for POME treatment is in increase (Mohan et al., 2007).

Electrochemical process is one of the alternatives for POME treatment. Despite it comes with several advantage, some limitation remained. For instance, electrode materials play major role on the performance of electrolysis, whereby previous study hase demonstrated the use of aluminium (Al) (Agustin et al., 2008), iron (Fe) (Phalakornkule et al., 2010) and steel wool (Nasrullah et al., 2018) during electrochemical process of POME treatment. Those electrode materials have several limitations such as treatment performance of COD and colour removal below than 50% and the electrolysis time more than 4 hours means more energy to consume. Thus, in this study new proposed potential electrode materials will be selected and optimized to achieve better performance in term of COD removal, colour reduction and oil and grease removal.

In addition, other factors that may affect treatment efficiencies, including voltage, electrolysis time and electrolyte support need to be addressed to achieve optimal treatment condition. The reason of optimization due to performance result of POME treatment should be achieve the Department of Environment (DOE) final discharge limit. In addition, the optimization process could reduce the operating cost for this system and make this system more applicable to the industries. Response surface methodology (RSM) is an analytical statistic tool for studying the interaction

between operating condition with the response from treatment performance. Limited report on optimization of electrochemical treatment of POME using RSM therefore in this study the selected electrode materials will be optimized with RSM using box-behnken design (BBD) as design experiment. Moreover, to ensure the applicability of this system for POME treatment, cost analysis need to be conducted once optimum condition achieved. Thus, the operating cost will be evaluated based on three main components including electrical energy consumption, electrode materials consumption and chemical consumption.

#### **1.3** Objectives of Study

The objectives of the study are outlined as follow:

- To select potential anode materials for the electrochemical treatment of POME from different type of electrode material (Aluminium, copper, graphite and platinum).
- To optimize the electrochemical treatment of POME operating conditions (electrolysis time, electrolyte amount and voltage) using graphite and platinum as anode materials
- iii. To investigate the cost analysis of the electrochemical treatment of POME using potential electrodes at the optimum condition

#### **1.4** Scope of the Research

In this study, fresh POME samples were collected in POMTEC, Negeri Sembilan Malaysia. The raw sample was analysed based on pH, chemical oxygen demand (COD), colour intensity and oil and grease. In term of COD removal performance, effective combination electrode materials (anode and cathode) were selected from various type of combination of electrode materials. The combinations are Copper-Graphite, Platinum-Graphite, Aluminium-Graphite and Graphite-Graphite. The selected combinations of electrode materials were optimized using

RSM. The parameter including voltage, electrolysis time and electrolyte support while the responses are COD removal and colour removal. In addition, the operating cost from optimum condition was evaluated.

#### 1.5 Significance of Study

This study will be useful for contribution to POME treatment in Malaysia. It is expected that POME treatment at local palm oil industries will be improved following the findings of this study. The COD, colour intensity and oil and grease removal in POME by electrochemical process are expected to improve the efficiency of final discharge. The effect of different operating parameters was optimized to fulfil the discharge limit by Department of Environment (DOE). This work would contribute to sustainable development goals (SDGs) agenda specifically goal number three on clean water and sanitation. The contribution target from this work includes improve water quality released to environment by reducing pollution, minimizing release of hazardous chemical and reduce portion untreated wastewater.

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