

BIOGRANULATION TECHNOLOGY FOR THE TREATMENT OF
ANAEROBICALLY DIGESTED PALM OIL MILL EFFLUENT USING
SEQUENCING BATCH REACTOR

MARIA BINTI NUID

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Faculty of Engineering
Universiti Teknologi Malaysia

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DEDICATION

This thesis is dedicated to:

Special dedication to my beloved father & mother, Nuid Soh & Rusmidi Linggam, who taught me how to read and how to write. The person dreamed to see me one day highly educated.

Special dedication to my beloved mother-in-law and late father-in-law, Minah Undas & Norbek Bisari, without them none of my success would be possible.

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ABSTRACT

Ponding treatment system has been widely used to treat palm oil mill effluent (POME) due to its low operating cost and ease of operation. However, it has some disadvantages such as requirement for a large area, long hydraulic retention time (HRT), and emittance of bad odour. Biogranulation system tested on different types of wastewater offers a solution for the problems. Although this system has been tested for different types of POME, its application in treating anaerobically digested POME has not been previously explored. Therefore, this study was conducted mainly to investigate the feasibility of biogranulation system for treatment of anaerobically digested POME. This study was carried out using a laboratory-scale sequencing batch reactor (SBR) under intermittent anaerobic and aerobic conditions with a working volume of 2 L. *Serratia marcescens* SA30 (SMSA30) strain was added to the system to treat high concentration of oil and grease (O&G) in wastewater. The system was operated with the variable organic loading rates (OLR) of 0.69 to 9.90 (kg/m³ d), HRT of 4 to 24 h, and superficial air velocity (SAV) of 2 cm/s. After 60 days addition of SMSA30 strain, the flocculent biomass was transformed into biogranules with excellent settleability with improvement in treatment efficiency of the system. Stable biogranules with an average diameter of about 2 mm, sludge volume index (SVI) of 43 mL/g and settling velocity of 81 m/h, were obtained in the biogranular system following 256 days of operation. Upon the biogranules formation, the average removal efficiencies of total chemical oxygen demand (COD_T), soluble chemical oxygen demand (COD_S), total nitrogen (TN), total phosphorous (TP) and O&G increased from 19 to 52%, 24 to 61%, 51 to 88% and 65 to 84% and 6 to 99.92%, respectively. The results also show that the HRT, OLR and food to microorganism (F/M) ratio influenced the removal efficiency. The removal efficiency of COD, TN, TP and O&G improved with increase in HRT; the percentage of COD and TP removal were slightly increased with the increase of OLR and F/M ratio. By contrast, the removal of TN and O&G was reduced with the increase of the OLR and F/M ratio. The modified mass transfer factor (MMTF) models were used to scrutinise the mechanisms of mass transfer for the biosorption of organic matter and nutrient i.e., COD_T and COD_S, TN and TP, accumulated onto biogranules. The application of MMTF models verified that resistance of mass transfer is dependent on the film mass transfer for the biosorption of COD_T, COD_S, TN, and TP. The performance of the biogranular system would increase with increase in global mass transfer factor ($[k_L a]_g$) value and thus, provides a new insight in the dynamic response of aerobic digestion to biogranules development. This study demonstrates that the biogranular system, with addition of SMSA30 is feasible for treatment of anaerobically digested POME.

ABSTRAK

Sistem rawatan kolam telah banyak digunakan untuk mengolah efluen kilang kelapa sawit (POME) kerana kos operasi yang murah dan kemudahan operasi. Walau bagaimanapun, ia mempunyai beberapa kelemahan seperti keperluan ruang yang luas, masa pengekal hidraulik yang panjang (HRT), dan pelepasan bau busuk. Sistem biogranulasi yang telah diuji pada pelbagai jenis air buangan menawarkan penyelesaian untuk masalah tersebut. Walaupun sistem ini telah diuji untuk pelbagai jenis POME, penerapannya dalam merawat POME yang dicerna secara anaerobik belum pernah diterokai sebelum ini. Oleh itu, kajian ini dilakukan terutamanya untuk mengkaji keupayaan sistem biogranulasi untuk rawatan POME yang telah dicerna secara anaerobik. Kajian ini dilakukan menggunakan reaktor berkelompok berjujukan (SBR) berskala-makmal di dalam keadaan anaerobik dan aerobik yang beroperasi secara berselang-seli dengan jumlah isipadu sebanyak 2L. Strain *Serratia marcescens* SA30 (SMSA30) ditambahkan ke dalam sistem untuk merawat minyak dan gris (O&G) yang berkepekatan tinggi dalam air sisa. Sistem ini dikendalikan dengan kadar muatan organik (OLR) yang berubah-ubah dari 0.69 hingga 9.90 ($\text{kg/m}^3 \text{ d}$), HRT dari 4 hingga 24 jam, dan halaju udara dangkal 2 m/s . Setelah 60 hari penambahan SMSA30, enapcemar berubah membentuk biogranul dengan keupayaan mengenang yang sangat baik dan juga meningkatkan kecekapan sistem rawatan. Biogranul yang stabil dengan diameter lebih kurang 2 mm, indeks isipadu enapcemar (SVI) 43 mL/g dan halaju penganapan 81 m/h , telah dicapai dalam sistem biogranular selepas 256 hari beroperasi. Setelah pembentukan biogranul, purata kecekapan penyingkiran purata permintaan oksigen kimia jumlah (COD_T), permintaan oksigen kimia terlarut (COD_S), jumlah nitrogen (TN), jumlah fosforus (TP) dan O&G telah meningkat daripada 19 kepada 52%, 24 kepada 61%, 51 kepada 88% dan 65 kepada 84% dan 6 kepada 99.92%. Hasil kajian juga menunjukkan bahawa HRT, OLR dan nisbah makanan kepada mikroorganisma (F/M) mempengaruhi kecekapan penyingkiran. Kecekapan penyingkiran COD, TN, TP dan O&G bertambah baik dengan peningkatan HRT; peratusan penyingkiran COD dan TP meningkat sedikit dengan peningkatan OLR dan nisbah F/M. Sebaliknya, penyingkiran TN dan O&G berkurangan dengan peningkatan nilai OLR and nisbah F/M. Model faktor pemindahan jisim diubah suai (MMTF) telah digunakan untuk meneliti mekanisme biopenjerapan bahan organik dan nutrien iaitu COD_T , COD_S , TN dan TP, yang terjepap dalam biogranul. Penggunaan model MMTF mengesahkan bahawa rintangan pemindahan jisim adalah bergantung kepada pemindahan jisim filem untuk biojerapan COD_T , COD_S , TN dan TP. Prestasi sistem biogranular akan meningkat dengan peningkatan nilai $[k_L a]_g$ dan seterusnya memberikan pemahaman baru kepada tindak balas dinamik pencernaan aerobik dalam pembentukan biogranul. Kajian ini menunjukkan bahawa sistem biogranular, dengan penambahan SMSA30, berkeupayaan untuk merawat air sisa POME yang dicerna secara anaerobik.

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LIST OF ABBREVIATIONS

2-FP	-	2-flouorophenol
2,4-DNT	-	2,4-Dinitrotoluene
4-CP	-	4-chlorophenol
ABR	-	Anaerobic Baffled Reactors
ACFR	-	Advanced Continuous Flow Reactor
AN	-	Ammoniacal nitrogen
ASBR	-	Anaerobic Sequencing Batch Reactor
ASP	-	Activated Sludge Process
AT-POME	-	Aerobically treated POME
BPAC	-	Banana Peel Activated Carbon
BOD	-	Biochemical Oxygen Demand
BOD3	-	Biochemical Oxygen Demand (sample is uncubated for 3 days at 30 °C)
BOD5	-	Biochemical Oxygen Demand (sample is uncubated for 5 days at 25 °C)
BWS	-	Brouers, Weron and Sotolongo
Ca ²⁺	-	Calcium Cation
CaCl ₂	-	Calcium Chloride
Cd ²⁺	-	Cadmium (II) Ions
CFRs	-	Continuous-Flow Reactors
CFR-TST	-	Continuous-Flow Reactor With Two-Zone Sedimentation Tank
CH ₄	-	Methane
CMC		Critical Micelle Concentration
CO ₂	-	Carbon Dioxide
COD	-	Chemical Oxygen Demand
CODS	-	Soluble Chemical Oxygen Demand
CODT	-	Total Chemical Oxygen Demand
CPO	-	Crude Palm Oil
Cr ³⁺	-	Chromium (III) Ion

CSTR	-	Continuous Stirred Tank Reactor
Cu ²⁺	-	Copper (II) Ions
DSM	-	Department of Statistics Malaysia
DO	-	Dissolved Oxygen
EBPR	-	Enhanced Biological Phosphorus Removal
EPS	-	Extracellular Polymeric Substances
EGSB	-	Expanded Granular Sludge Blanket
EM	-	Effective Microorganisms
EMT	-	External Mass Transfer
EQA	-	Environmental Quality Act
Fe (III)	-	Iron Ions
FeCl ₃	-	Ferric Chloride
F/M	-	Food and Microorganism Ratio
GDP	-	Growth Domestic Product
GMT	-	Global Mass Transfer
H ₂	-	Hydrogen
HCl	-	Hydrochloric Acid
H/D	-	Height to Diameter ratio
HRT	-	Hydraulic Retention Time
IC	-	Integrity Coefficient
IFAS	-	integrated fixed-film activated sludge
IMT	-	Internal Mass Transfer
IUMAS	-	Integrated Ultrasonic Membrane An Aerobic System
K ₂ HPO ₄		Dipotassium Phosphate
MB	-	Methylene Blue
MBR	-	Membrane Bioreactor
MgSO ₄	-	Magnesium Sulphate
MLSS	-	Mixed Liquor Suspended Solids
MLVSS	-	Mixed Liquor Volatile Suspended Solids
MMFM	-	Modified Mass Transfer Factor Model
Mn (II)	-	Manganese Ions
Na ₂ SO ₄	-	Sodium Sulphate Anhydrous
NH ₃ -N	-	Ammonia Nitrogen

NH ₄ Cl		Ammonium Chloride
NH ₄ ⁺ N	-	Ammonium Nitrogen
Ni ²⁺	-	Nickel cation
O&G	-	Oil and Grease
OLR	-	Organic Loading Rate
P	-	Phosphorus
PHA	-	Polyhydroxyalkanoate
PVDF	-	Polyvinylidene fluoride
PVDF-TNT	-	Polyvinylidene fluoride/titanate nanotube
PO ₄ ³⁻ -P	-	Phosphate Ion
PNP	-	p-nitrophenol
POME	-	Palm Oil Mill Effluent
RBC	-	Rotating Biological Contractor
RG	-	Residual granules
RO	-	Reverse Osmosis
R&D		Research and Development
SAV	-	Superficial Air Velocity
SBAR	-	Sequencing Batch Airlift Reactor
SBBGR	-	Sequencing Batch Biofilter Granular Reactor
SBR	-	Sequential Batch Reactor
SBR-CF	-	Sequencing Batch Reactor-Continuous-Flow System
SDBS	-	Sodium Dodecylbenzenesulfonate
SDS		Sodium Dodecyl Sulfate
SG	-	Settled granules
SLES	-	Sodium Lauryl Ether Sulfate
SLR	-	Sludge Loading Rate
SMSA30	-	Serratia Marcescens SA30
SMPR	-	Submerged Membrane Photo Reactor
SOUR	-	Specific Oxygen Uptake Rate
SS	-	Suspended Solids
SV	-	Settling Velocity
SVI	-	Sludge Volume Index
SVI5	-	Sludge Volume Index at 5 minute

SVI10	-	Sludge Volume Index at 10 minute
SVI30	-	Sludge Volume Index at 30 minute
TIN	-	Total Inorganic Nitrogen
TN	-	Total Nitrogen
TP	-	Total Phosphorus
TOC	-	Total organic carbon
TS	-	Total Solids
TKN	-	Total Kjeldahl Nitrogen
UAASB	-	Upflow Aerobic-Anoxic Flocculated Sludge Bioreactor
UASB	-	Up-flow Anaerobic Sludge Blanket Reactors
UASFF	-	Up-flow Anaerobic Sludge-Fixed Film Reactor
UASB-HCPB	-	Up-flow Anaerobic Sludge Blanket-Hollow Centered Packed Bed
UF	-	Ultrafiltration
USEPA	-	United States Environmental Protection Agency
VER	-	Volumetric Exchange Ratio
VSS	-	Volatile Suspended Solids
WHO	-	World Health Organization
Zn (II)	-	Zinc ions

LIST OF SYMBOLS

$[k_L]_f$	-	Film mass transfer coefficient (m/h)
$[k_L a]_f$	-	Film mass transfer factor or external mass transfer factor (1/h)
$[k_L a]_d$	-	Porous diffusion factor or internal mass transfer factor (1/h)
$[k_L a]_g$	-	Global mass transfer factor (1/h)
a, b	-	Elovich constants
b	-	Binding energy or affinity parameter of the adsorption system
β	-	Adsorbate-adsorbent affinity parameter
B	-	Potential mass transfer index relating to driving force of mass transfer
C	-	Indicates thickness of the boundary layer
C_e	-	The equilibrium solute concentration
COD_{in}	-	Influent concentration of COD
C_{eff}	-	Effluent concentration
C_{in}	-	Influent concentration
C_0	-	Initial concentration of the adsorbate
C_S	-	Concentration of the adsorbate to depart from SBR
C	-	Concentration of the adsorbate
$(C - C^*)$	-	Driving force
h	-	height of the cylinder
K_1, K_2	-	Adsorption rate constant
K_3, K_4, A	-	Diffusion rate constant
K_A, n_A	-	Avrami constants
k_{ad}	-	Constant related to adsorption energy
k_f	-	The Freundlich constants related to adsorption capacity
K_m	-	Adsorption rate constant
m	-	Kuo-Lotse constant
$m, \alpha, \beta,$	-	Bangham constants

N_A	-	Quantity of the solute A transferred per unit of time or molar flux of the solute A
n	-	Adsorption intensity and is always greater than unity
n, α, τ	-	BWS constants
$n, B, t,$	-	Boyd constants
Q	-	Flow rate
Q_m	-	The theoretical saturation capacity
q		Accumulative quantity of the solute adsorbed onto adsorbent
q_e	-	The amount of adsorbate adsorbed at equilibrium per unit weight of adsorbent
q_{max}	-	Langmuir constants which are related to maximum monolayer adsorption capacity
q_t	-	Capacity at t
R	-	The universal gas constant
S	-	Surface area
T	-	The absolute temperature
t	-	Adsorption time / time take
V	-	Volume of the treated water
ε	-	Polanyi potential

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Palm oil industry is one of the most important determinants of agriculture-based commerce and has been considered as one of the key economic drivers of the agricultural sector for Indonesia, Malaysia, Thailand, Nigeria and Colombia (Lee, Chin, Lim, Witoon, & Cheng, 2019; Gobi & Vadivelu, 2013). Malaysia produced 19.86 million tonnes of crude palm oil (CPO) in the year 2019, which generated between 49 and 75 million tonnes of palm oil mill effluent (POME) (MPOB, 2019; Ding et al., 2020). MPOB estimated that the oil palm planted area in 2019 to increase to 5.90 million hectares, an increase of 0.9% from 5.85 million hectares in the previous year (MPOB, 2019). However, the discharge of POME from the oil palm industry poses both economic and technical difficulties to wastewater managers and may cause the degradation of the environmental ecosystem. In terms of the water pollution, the discharge of POME into the natural water could lead to poor water quality available for consumption, water pollution for aquatic habitation and environmental degradation.

POME is a mixture of steriliser condensate, separator sludge and hydrocyclone wastewater at a ratio of 9:15:1, respectively (Abu Bakar et al., 2018). Raw POME has the values of biochemical oxygen demand (BOD), chemical oxygen demand (COD), O&G, total solids (TS), suspended solids (SS), ammoniacal nitrogen (NH₃-N), and TN in the ranges of 10,250-43,750 mg/L, 15,000-100,000 mg/L, 4000 mg/L, 11,500-79,000 mg/L, 5000-54,000 mg/L, 4-80 mg/L, 180-1400 mg/L, respectively (Iskandar, Baharum, Anuar, & Othaman, 2018). Typically, POME has been treated using the common conventional methods of anaerobic, facultative and

aerobic ponds (Ng et al., 2018; Ahmed, Yaakob, Akhtar, & Sopian, 2015; Lam & Lee, 2011).

The biological processes in the ponding system are capable to break down all biodegradable substances in the POME through anaerobic and facultative actions and thus, they release methane, carbon dioxide, hydrogen sulphide and water (Abu Bakar et al., 2018). The system is widely used mainly due to its low operational cost and ease of operation; however, the system also has several drawbacks which include long hydraulic retention time, large land requirement, sensitivity of microorganisms to variations in environmental conditions, foul smell and greenhouse gas emissions (Udaiyappan, Hasan, Takriff, Abdullah, Maeda, Mustapha, & Yasin et al., 2020; Nwuche, Aoyagi, & Ogbonna, 2014). The increase of environmental awareness and the toughening of government policies have made it necessary to develop new environmentally friendly, effective and low-cost treatment system to treat the POME.

The continuous increase in the demand for palm oil worldwide has led to an urgency in terms of the environmental monitoring of the released generated POME (Zolkefli et al., 2020). Conventionally 85% of the palm oil mills in Malaysia adopt biological methods to reduce the BOD into an acceptable limit (100 mg/L and 20 mg/L in Sabah and Sarawak) as it has been discharged into the environment while there are no specific discharge limits for COD as there is no legislation imposed upon it (Iskandar, Baharum, Anuar, & Othaman, 2018). The existing treatment process has deficiencies due to the ineffective residual oil removal from the waste stream and a large amount of greenhouse gas emission owing to the ponding system (Khadaroo, Grassia, Gouwanda, & Poh, 2020).

After the oil removal pit, otherwise known as the cooling ponds, POME is allowed to cool down to a suitable mesophilic temperature, after which it is sent directly to anaerobic digestion. However, often, the treated effluent quality is poor and does not conform to stringent environmental standards (Ahmad, Yaakob, Akhtar & Sopian, 2015). Therefore, releasing poorly treated POME directly to the surrounding will cause a serious pollution problem towards the environment. Instead,

there is an urgent need to find an efficient and practical approach to preserving the environment while maintaining the sustainability of the economy.

The increasing amounts of nitrogen and phosphorus compounds in water and wastewater cause toxicity to aquatic organisms, leading to the distortion of natural nutrient cycles and contributing to the occurrence of eutrophication in water bodies, resulting in oxygen depletion and organism death (Sarvajith, Reddy, & Nancharaiah, 2020; Sidiarto, Renggaman, & Choi, 2019; Yu et al., 2014). An outbreak of harmful cyanophytes, commonly known as cyanobacteria, will eventually result in the increased mortality of aquatic organisms (Bao et al., 2016). Hence, the discharge of treated wastewater must meet the effluent quality standards for nitrogen and phosphorus.

The biogranulation system is considered as an emerging treatment technology that provides several advantages over the activated sludge-based systems. These include reduced surface area requirement, lower energy demand, lower reactor volume, easier process control, faster settling sludge, higher toxic tolerance and excellent effluent quality (Najib, Ujang, Salim, & Ibrahim, 2017; Pronk et al., 2015; Li, Ding, Cai, Huang, & Horn, 2014). Biogranules, which are the key component in the biogranulation system, are aggregates of self-immobilised bacteria, induced by environmental stress conditions involving physical shear, physicochemical or biochemical interactions and microbial attraction mechanisms (Khan, Mondal, & Sabir, 2013).

The initial formation of aggregates is triggered by several stressors such as substrate composition, OLR, hydrodynamic shear force, settling time, HRT, aerobic starvation, presence of multivalent cations, intermittent feeding strategy, dissolved oxygen (DO), pH, temperature, seed sludge, reactor configuration, and inhibition to granulation (Franca, Pinheiro, Loosdrecht, & Lourenço, 2018; Khan et al., 2013; Adav, Lee, Show, & Tay, 2008).

1.2 Problem Statement

Research on biogranulation technology has increased rapidly due to its advantages over activated sludge technology. Biogranules has been studied in well-controlled lab-scale reactors with high or middle-strength synthetic wastewaters (Sengar, Basheer, Aziz, & Farooqi, 2018; Sarma, Tay, & Chu, 2016; Ab Halim et al., 2015; Muda et al., 2010) and real wastewaters from various industries, such as domestic wastewater (Sguanci, Lubello, Caffaz, & Lotti, 2019), rubber wastewater (Rosman, Nor Anuar, Chelliapan, Md Din, & Ujang, 2014), livestock wastewater (Othman et al., 2013), brewery wastewater (Corsino, Ryan, Munz, Torregrossa, & Oleszkiewicz, 2017), municipal wastewater (Derlon, Wagner, Helena, & Morgenroth, 2016), fermented soy sauce (Harun et al., 2014), seafood industry (Val del Río, Figueroa, Mosquera-Corral, Campos, & Méndez, 2013) and dairy industry (Schwarzenbeck, Borges, & Wilderer, 2005).

Although some studies have also been conducted with POME (Abdullah, Ujang & Yahya, 2011), the literature on the treatment of anaerobically digested POME with a high concentration of O&G by biogranules is apparently lacking. Although the wastewater can be treated using ponding system, it requires long HRT and area. Biogranulation system provides smaller footprint. Furthermore, the samples used in this research an anaerobically digested POME. The sample in this work could not develop biogranules because of high O&G concentration.

The issues involving biogranular system to treat real wastewaters such as long cultivation or start-up period, problems in structural and functional, the stability of the biogranules and possible negative impacts of high suspended solid concentrations on granulation have not been fully addressed (Cetin, Karakas, Dulekgurgen, Ovez, & Kolukurik, 2018). Long start-up period is usually required for the development of biogranules and the loss of biomass in this period may lead to poor nutrient removal performance (Verawaty, Pijuan, Yuan, & Bond, 2012). These shortcomings impede the application of this technology for real wastewater treatment.

Many researchers have shown great interest in the cultivation of biogranules by bioaugmentation. Ivanov and co-worker (2006) have applied an enrichment culture with an increased cell surface hydrophobicity for a faster formation of biogranules. The addition of fungal chlamydo spores to reactor accelerates biogranulation during the phenol wastewater treatment has been reported by Hailei et al. (2011). In addition, the cultivation of biogranules capable of degrading O&G compounds is much more difficult than the cultivation of biogranules degrading common organic pollutants as the cultivation generally requires several months or even longer time. To date, limited research has been published on treating diluted raw POME (Abdullah, Ujang & Yahya, 2011) by biogranules and no report has shown that biogranules could be cultivated by bioaugmentation in the treatment of anaerobically digested POME.

Different strategies have been proposed to enhance the formation of biogranules. High concentrations of O&G in the POME can lead to the loss of biogranules' strength and stability, growth of filamentous bacteria, the formation of scum and stable foam, and increased washout of slow settling biomass (Cammarota & Freire, 2006). Ahmad et al. (2003) proposed a solvent addition to extract oil from POME. Another comparative study was carried out for the residual oil and TSS removal in POME utilizing chitosan, bentonite and activated carbon (Ahmad, Sumathi, & Hameed, 2005).

More recently, Abdullah et al. (2015) revealed that the modified *Ceiba Pentandra* enhanced the removal of O&G from POME at the secondary level of treatment. As O&G is an important component in POME, there is a need to evaluate the feasibility of other types of biodegradation approach for the removal of O&G from POME. Reports on biogranules aiming at the bioremediation of O&G from POME are rare. In addition, the formation of O&G biogranules from pure cultures capable of degrading O&G has not previously been explored.

Mathematical modelling has been proven to be very useful in studying complex processes such as those found in biogranules (Beun, Heijnen, & Loosdrecht, 2001). To date, mechanism models that explain the mechanism of biogranules is still

lacking although several hypotheses and mathematical models have been proposed. Although biogranules technology has been studied for the treatment of a wide variety of wastewaters (Muda et al., 2010; Dahalan et al., 2015; Salmiati, Dahalan, Mohamed Najib, Salim, & Ujang, 2015), the kinetics and mechanism of mass transfer for the biosorption of organic matters on the surface of the biogranules need to be verified.

Although the wastewater can be treated using ponding system, it requires long HRT and area. Biogranulation system provides smaller footprint. Furthermore, the samples used in this research an anaerobically digested POME. The sample in this work could not develop biogranules because of high O&G concentration.

The present study can be regarded as the first study dedicated to the development of biogranules using anaerobically digested POME. The system utilised the concept of sequential anaerobic and aerobic biological reactions for the complete degradation of the wastewater. The factors influencing the efficiency of the biogranular system in treating anaerobically digested POME are addressed in this study. The addition of SMSA30 strain into the biogranular system aims to solve the problem with the high concentration of O&G presence in anaerobically digested POME. The addition of SMSA30 strain for the removal of O&G was tested for the potential enhancement of the system. Moreover, the application of the modified mass transfer factor models may help to determine the resistance of mass transfer for the biosorption of organic matter and nutrients onto the biogranules.

1.3 Objective of Study

The objectives of this study are as follows:

- (a) To develop the biogranules to treat anaerobically digested POME in intermittent anaerobic and aerobic biogranular systems with bioaugmentation of SMSA30 strain.
- (b) To evaluate the efficiency of the biogranulation system in treating anaerobically digested POME in terms of COD_T, COD_S, TN, TP and O&G.
- (c) To evaluate the effect of HRT, OLR and F/M ratios on the removal performance of biogranules in terms of COD, TN, TP and O&G.
- (d) To determine the kinetics and mechanism of mass transfer and to determine the resistance of mass transfer for the biosorption of COD_T, COD_S, TN and TP accumulated on biogranules by using the mass transfer kinetic models.

1.4 Scope of Study

This study explored the development of biogranules in a lab-scale reactor system to treat an anaerobically digested POME. The anaerobically digested POME was obtained from an anaerobic pond. The reactor has a working volume of 2 L and was operated according to SBR's sequence for 6 h cycle, which includes the sequential anaerobic and aerobic reaction phases. Wastewater from the palm oil mill was used as feed, while sewage was used as flocculant sludge seed. During the development of the biogranules, bioaugmentation was applied by add SMSA30 strain to enhance the removal of O&G in anaerobically digested POME. This is because the presence of O&G can lead to the loss of biomass, growth of filamentous bacteria, the formation of scum and stable foam in the biogranular system.

During the granulation process, samples of the biogranules were collected and examined in terms of the morphology and physical characteristics namely settling velocity (SV), sludge volume index (SVI), mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS) and biogranules strength.

The system performances were determined based on the COD_T, COD_S, TN, TP and O&G removal efficiencies during the development of the biogranules and post-development phase. The effects of HRT, OLR and F/M on the organic and nutrient removal efficiencies were also analysed. The application of the modified mass transfer factor (MMTF) model was used to predict the mass transfer kinetics of organic matters, TN and TP biosorption in the biogranular system. It was also used to describe the mechanisms of organic matters, TN, and TP removal before and after the addition of SMSA30 strain in the biogranular system.

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1.5 Significance of Study

The biological treatment of POME is considered the most environment-friendly and cost-effective way for a high-rate biological removal of organic and nutrients matter, such as COD, nitrogen and phosphorus, from wastewater, as opposed to physicochemical methods (Winkler et al., 2018; Franca et al., 2017; Gobi, Mashitah, & Vadivelu, 2011). Biogranulation is anticipated to be one of the main advanced technologies for POME treatment (Abdullah, Ujang, & Yahya, 2011; Gobi et al., 2011; Gobi & Vadivelu, 2014; Najib et al., 2017). However, the majority of the studies were conducted using diluted POME and the addition of light sources to

provide a condition conducive for the photosynthesis environment. Notably, biogranulation studies treating anaerobically digested POME wastewater in a biogranular system is apparently missing. Hence, the significance of this study in the field of biotechnology and environmental engineering are:

- (a) The characteristics of the anaerobically digested POME are suitable for the biological process without the additional cost of adding nutrients. However, the presence of O&G in POME can cause the reduction of the cell-aqueous phase transfer rates through the formation of a lipid coat around the biological flocs. Hence, many practical aspects of the treatment in developing biogranules need to be explored. This study presents the viability of the biogranulation system in treating anaerobically digested POME.
- (b) This study provides new insights on the application of SMSA30 strain to remove O&G in POME and to enhance the formation of biogranules. Although bioaugmentation with specified pure culture expedites the process of biogranulation, it is complex and expensive in practice. Therefore, applying a simple operational approach to enhance biogranulation is desirable and promising for real applications.
- (c) The study also provides the effects of HRT, OLR and F/M ratios using anaerobically digested POME in relation to the removal performance by the biogranules.
- (d) This study introduces the application of the modified mass transfer factor models to assess the behaviours of external and internal mass transfer and to determine the resistance of mass transfer for the biosorption of organic and nutrients matter onto biogranules (before and after the addition of SMSA30 strain applied to the biogranular system).

1.6 Organisation of the Thesis

The thesis is presented in five chapters. The first chapter explains the problems generated from the anaerobically digested POME and the importance of developing biogranules in the biogranular system using real wastewater. The second chapter presents the review of the literature on the characterisation of POME, selected treatment approaches used for POME treatment, developments of biogranules, and both the modelling and application of the kinetic models. Then, the third chapter describes the methodology used for this study and explains the experimental work involved throughout the research. Chapter four discusses the findings of the research focusing on each objective. Finally, chapter five presents the conclusions that are derived from this study and the recommendations for future research.

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LIST OF PUBLICATIONS

Journal with Impact Factor

1. Mohamad, A. F., **Nuid, M.**, Aris, A., Muhamad, F., Kasni, S., & Muda. K., (2019). Mass transfer kinetics of phosphorus biosorption by aerobic granule sludge in sequencing batch reactor. *Journal of Water Process Engineering*, 31(July), 100889. <https://doi.org/10.1016/j.jwpe.2019.100889> (Q1, IF: 3.465)
2. Mohamad, A. F., Nuid, M., Aris, A., & Muda, K., (2018). Mass transfer kinetics of biosorption of nitrogenous matter from palm oil mill effluent by aerobic granules in sequencing batch reactor. *Journal of Environmental Technology*, 39:17, 2151-2161. <https://doi.org/10.1080/09593330.2017.1351494> (Q3, IF: 2.213)
3. Mohamad, A. F., Nuid, M., Aris, A., & Muda, K., (2017). Kinetics and mass transfer studies on the biosorption of organic matter from palm oil mill effluent by aerobic granules before and after the addition of *Serratia Marcescens* SA30 in a sequencing batch reactor. *Journal of Process Safety and Environmental Protection*, 107, 259-268. <https://doi.org/10.1080/09593330.2017.1351494> (Q1, IF: 4.966)

Indexed Journal

1. Salim, N. A. A., Puteh, M. H., Yusoff, A. R. M., Abdullah, N. H., Fulazzaky, M. A., Rudie, A., Mohd. A. Z., Khamidun, M. H., Zaini, M. A. A., Syafiuddin, A., Ahmad, N., Lazim, Z. M., Nuid, M., Zainuddin, N. A., (2020). Adsorption isotherms and kinetics of phosphate on waste mussel shell. *Malaysian Journal of Fundamental and Applied Sciences*, 16, 393 – 399. <https://mjfas.utm.my/index.php/mjfas/article/view/1752> (Indexed by Web of Science)

2. Lazim, Z. M., Salmiati, Hadibrata, T., Yusop, Z., Nazifa, T. H., Abdullah, N. H., Nuid, M., Salim, N. A. A., Zainuddin, N. A., Ahmad, N., (2020). Bisphenol A Removal by Adsorption Using Waste Biomass: Isotherm and Kinetic Studies. *Biointerface Research in Applied Chemistry*, 11, 8467 – 8481. <https://biointerfaceresearch.com/wp-content/uploads/2020/08/20695837111.84678481.pdf> (Indexed by Scopus)
3. Salim, N. A. A., Puteh, M. H., Khamidun, M. H., Fulazzaky, M. A., Abdullah, N. H., Yusoff, A. R. M., Zaini, M. A. A., Ahmad, N., Lazim, Z. M., Nuid, M., (2021). Interpretation of isotherm models for adsorption of ammonium onto granular activated carbon. *Biointerface Research in Applied Chemistry*, 11, 9227-9241. <https://www.scopus.com/inward/record.uri?eid=2-s2.0-85090961519&doi=10.33263%2FBRIAC112.92279241&partnerID=40&md5=a8f9b07a673ecf5fdffb63336dd51901> (Indexed by Scopus)

Non-indexed Journal

1. Sulaiman, A. A. P., Jamil, S. Z., Jupri, M. A., Abdullah, N. H., Azmi, M. A. M., Azman, T. M. F. H. T., Hamid., N. B., Suwandi, A. K., Ahmad, N., Salim, N. A. A., Lazim, Z. M., Nuid, M., Zon, N. F., (2020). The Application of Waste Mussel Shell and Nitrogen - Phosphorus - Potassium (NPK) Fertilizer on The Growth Components of Jasmine (Jasminum). *Multidisciplinary Applied Research and Innovation Vol. 1, No.11-12*. DOI: <https://doi.org/10.30880/mari.2020.01.01.001> (UTHM)
2. Nuid, M., Aris, A., Mohamad, A. F., & Muda, K., (2020) Biogranules Technology: Application on Real Wastewater Treatment. *Issues and Technology in Water Contaminants*. ISBN No.978-983-52-1691-6, UTM, 2020. (Book Chapter)
3. Sabri, S., Azman, S., Low, W.P, **Nuid, M.** Metal Removal by Banana Peel as Bio-adsorbent. (Submitted to UTM Press for book chapter)