AEROBIC GRANULATION WITH INDUSTRIAL WASTEWATER IN SEQUENCING BATCH REACTORS

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requirements for the award of the degree of
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Dedicated to my beloved dien,

my family and nation

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ABSTRACT

The high operational cost of drinking water supply is mostly encountered following the upsurge of pollutant concentration in wastewater, surface water and ground water. Palm Oil Mill Effluent (POME) generated from palm oil industries and the micropollutant from pharmaceutical industries are amongst the main contributor to this problem. Two identical SBR bioreactors without granules (R1) and with granules (R2) were fed with 5 mg acetaminophen per 1 L of POME, organic loading rate (OLR) in a range 2.5 to 3.5 g COD L^{-1} were used. The reactors were operated at 27^{o}C (room temperature) with 12 hrs successive cycles involved 5 min of feeding period and 2 min for effluent withdrawal, resulting 50% of volumetric exchange ratio (VER). The reaction time was initially set as an anaerobic and aerobic period, 45 min and 10 hrs 35 min, respectively. On day 50 the biomass concentration and the sludge volume index (SVI) for both reactors were between 11,160 mg L^{-1} to 11,430 mg L^{-1} and 15 mL g^{-1} SS to 17 mL g^{-1} SS, respectively, indicating fair biomass accumulation in the reactor and good settling properties of granular sludge. Moreover, COD removal was 81% and 67% for R2 and R1, respectively. Phosphate was removed at 28% in R1 while R2 removed a higher phosphorous removal at 55%. The average color removal for both reactors ranged from 45% to 54%. In addition, the paracetamol concentration was depleted and the aerobic granules was developed 0.5 to 3.0 mm in size for both reactos. This study provided an insight of the feasibility of aerobic granular sludge formation in SBR to be an effective biodegradable process for high strength wastewater with emerging pollutants, such as pharmaceutical wastewater.

ABSTRAK

Kos operasi bekalan air minuman yang tinggi kebanyakannya mengikut peningkatan kepekatan bahan di dalam air sisa, air permukaan dan air bawah tanah. Air sisa kelapa sawit (POME) yang terhasil dari industri minyak sawit dan bahan cemar mikro dari industri farmaseutikal adalah antara penyumbang utama kepada masalah ini. Dua bioreaktor SBR yang sama tanpa butiran enapcemar (R1) dan dengan butiran enapcemar (R2) telah diberi makan dengan kombinasi POME dan parasetamol yang diketahui, dan kadar masukan organik adalah di antara 2.5 hingga 3.5 g COD L^{-1} telah digunakan. Reaktor telah beroperasi pada suhu $27^{o}C$ (suhu bilik) dengan 12 jam kitaran berturutan melibatkan 5 min tempoh makan dan 2 min pengeluaran efluen, menghasilkan 50% kadar nisbah pertukaran isipadu (VER). Masa tindak balas pada mulanya ditetapkan sebagai tempoh anaerobik dan aerobik, iaitu pada 45 min dan 10 jam 35 min. Pada hari ke 50, kepekatan biomas dan paras SVI untuk kedua-dua reaktor adalah di antara 11,160 mg L^{-1} hingga 11,430 mg L^{-1} dan mL g SS^{-1} hingga 17 mL g SS^{-1}, memperlihatkan pengumpulan biomas di dalam reactor dan kadar kemampuan enapcemar yang baik di dalam reaktor. Lebih-lebih lagi, penyingkiran COD adalah 81% dan 67% bagi R2 dan R1. Fosfat dapat disingkirkan pada kadar 28% di dalam R1 sedangkan R2 menyingkirkan kadar fosfat yang lebih tinggi pada 55%. Purata penyingkiran warna bagi kedua-dua reaktor adalah di antara 43% hingga 54%. Di samping itu, kepekatan parasetamol berkurang dan butiran enapcemar terbentuk pada saiz 0.5 – 3.0 mm untuk kedua-dua reaktor. Kajian ini menyediakan anggapan tentang kebolehan butiran enapcemar yang terbentuk di dalam SBR untuk menjalankan proses penguraian yang berkesan bagi air sisa berkekuatan tinggi dengan bahan cemar yang baru, seperti air sisa farmaseutikal.

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LIST OF ABREVIATIONS

ADI	-	Acceptable daily intake
APHA	-	American Public Health Association
BOD	-	Biochemical oxygen demand (mg L^{-1} or g L^{-1})
CPO	-	Crude palm oil
COD	-	Chemical oxygen demand
DOC	-	Dissolved organic carbon (mg L^{-1} or g L^{-1})
DNA	-	Deoxyribonucleic acid
DO	-	Dissolved oxygen (mg L^{-1} or g L^{-1})
EDCs	-	Endocrine Disrupting Chemicals
EPS	-	Extracellular polymeric susbstances
H/D	-	Colomn height to diameter ratio (mm)
HPLC	-	High-performance liquid chromatography
HRT	-	Hydraulic retention time
MIEX	-	Magnetic ion-exchange
MLSS	-	Mixed liquor suspended solid (mg L^{-1} or g L^{-1})
MLVSS	-	Mixed liquor volatile suspended solid (mg L^{-1} or g L^{-1})
MTD	-	Minimum therapeutic doses
OLR	-	Organic loading rate (mg COD L^{-1} g COD L^{-1})
PCT	-	Paracetamol
pH	-	Power hydrogen

POME	-	Palm oil mill effluent
PPCPs	-	Pharmaceuticals and Personal Products
R	-	Reactor
RO	-	Reverse osmosis
SBAR	-	Sequanceing batch airlift reactor
SBR	-	Sequanceing batch reactor
SRT	-	Sludge retention time
SS	-	Suspended solid (mg L^{-1} or g L^{-1})
SVI	-	Sludge volume index
TN	-	Total nitrogen (mg L^{-1} or g L^{-1})
TSS	-	Total suspended solid (mg L^{-1} or g L^{-1})
UASB	-	Up-flow anaerobic sludge blanket
USEPA	-	US Environmental Protection Agency
UTM	-	Universiti Teknologi Malaysia
UV	-	Ultraviolet
VER	-	Volumetric exchange rate
VSS	-	Volatile suspended solid (mg L^{-1} or g L^{-1})

LIST OF SYMBOLS

CO_2	-	Carbon Dioxide (mg L-1 or g L-1)
Fe_3O_4	-	Iron Oxide
Fe_2O_3	-	Ferric Oxides
H_2O	-	Water
H_2O_2	-	Hydrogen Peroxide
HOCI	-	Hypochiorous acid
OCI^-	-	Hypochiorite ion
OH^-	-	Hydroxyl
PHAs	-	Poly-hydroxyalkanoates
PO_4^{3-}	-	Total Phosphorous
NH_2	-	Amidogen
NH_2Cl	-	Cloramine
$NHCl_2$	-	Dicloramine
THMs	-	Trihalomethanes

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CHAPTER 1

INTRODUCTION

1.1 Introduction

Waste Water Treatment Plants (WWTPs) are designed only to handle conventional pollutants with normal parameter. However the increasing number and the soaring parameter of wastewater pollutants have reduced the effluent quality of WWTPs, affecting ground water with diverse organic contaminants such as high-strength organic substances and persistent micropollutant from pharmaceutical industries for example Palm Oil Mill Effluent (POME) and acetaminophen.

Palm oil plantation has dominated Malaysian agro-base crop. It has increased from 4.85 million hectares in 2010 to 5.1 million hectares in 2012 (Oil World, 2013) and has continue to increase. Alongside its oil production a large amount of solid and liquid waste in the form of empty fruit bunch (EFB) and POME are produced during the process. At almost 80 million dry tones of solid biomass per annum were produced by Malaysian palm industry (Agensi Inovasi Malaysia, 2011).

On the other hand, acetaminophen or universally known as paracetamol (N-acetyl-4-aminophenol) is an emerging persistent organic pollutant which has come to the fore front of environmental issues due to its frequent occurrence in aquatic environments and drinking water. Some of the unfavourable health effects raise concerns about their potential impacts on the environment and human health including aquatic toxicity, resistance development in pathogenic bacteria, genotoxicity, and endocrine disruption. Synthetic and natural chemical of acetaminophen can be found in prescription medicines, over-the-counter therapeutic drugs and veterinary drugs. The contamination can enter the environment through various forms such as human and livestock excretion, as a consequence of drugs consumption or additive animal feeds, or even directly as a disposal from hospital effluent and pharmaceuticals industry.

The composition of POME and acetaminophen in wastewater have increased for the last 10-years (Ratola *et al.,* 2012). There is a big potential for both pollutants to combine together in water stream and produces numerous hazardous chemicals for living organism on earth.

1.2 Problem Statement

Conventional WWTPs where acetaminophen will inevitably end up, uses chlorination process to treat the pharmaceutical compound. However, the procedure generates more toxic products than the reactant itself. Reactions with hypochlorite produce N-acetyl-p-benzoquinone imine and 1,4-benzoquinone as their product, both are known as toxic metabolites, which have shown to result in hepatic necrosis (Bedner & MacCrehan, 2006). The alternative of aqueous paracetamol removal through electrochemical (Waterston *et al.,* 2006), ozonation, H_2O_2/UV oxidation (Vogna *et al.,* 2004) and semiconductor photocatalysis (Yang *et al.,* 2008) methods has been reported. Another common treatment is biological and to date it is still seen to have prospective to be the most economical, energy efficient, and environmental friendly approach to treat waste (Zein *et al.,* 2004)

Aerobic granulation is one of the most recent innovations in biological treatment. A number of factors affecting the granulation process (Jiang *et al.,* 2006). Furthermore the application of this technology to treat industrial wastewaters (Hu *et* *al.*, 2012) indicates the possibility to grow aerobic granules in SBRs and has able to decrease pharmaceuticals compound such as paracetamol efficiently as the sole carbon and energy resources in the system.

1.3 Objective

The objectives of this study are as follows:

i. To investigate possibility of developing aerobic granules using pharmaceuticals wastewater.

ii. To evaluate new emerging pollutant in wastewater treatment using SBR

iii. To investigate the potential of aerobic system for high-strength wastewater treatment.

1.4 Scope and Limitation

The scopes of this study are divided into following phase; (a) the investigation of possible aerobic granular sludge development from pharmaceutical industrial wastewater; (b) Observation on removal rate of acetaminophen concentration in wastewater system; (c) Observation on ability of the stabilize granules to degrade acetaminophen compounds.

1.5 Significance of The Study

Hopefully the outcome of this study can show the feasibility of SBR to develop aerobic granules from pharmaceutical industrial wastewaters regarding to their poor settling properties development with the flocculent sludge characteristic

In addition, aerobic granules could be an effective biodegradable for acetaminophen compounds that are detected in the environment as a subject to both waste and drinking water treatment processes and gives imminent into the microbial involvement in that process.

1.6 Thesis Organization

This thesis is organized into five chapters. The first chapter is the introduction of the whole research. This chapter will discuss a general overview and the background of study of the project. The first chapter also includes the problem statement in pharmaceuticals wastewater treatment. Meanwhile, this chapter describes the objective of the project and the aims the project achieves.

Chapter 2 describes the literature review and basic concept of the project. This chapter will discuss palm oil mill effluent (POME) and paracetamol as micropollutant from pharmaceutical industries, with some alternate technologies that can be used in the wastewater treatment. On the other hand, this chapter shows the potential ability of aerobic granules in sequencing batch reactors (SBR) as an advance biotreatment. It also will discuss the influence of feast and famine, aggregate selection, hydrodynamic shear force, organic loading rate (OLR), reactor configuration, and a number of factors that effecting granulation process in SBR.

Chapter 3 introduces the feature of aerobic granulation in sequencing batch reactor from the previous research. This chapter begins with the characterization of POME and seed sludge for main substrate in the experiment. Then, this chapter also covers the experimental setup of SBR such as Organic Loading Rate (OLR), Hydraulic Retention Time (HRT), the ration (H/D) in cylindrical bioreactors and volumetric exchange rate that suitable for granules development. Finally this chapter will discuss the instrument data analysis for sample data collection and evaluation.

Chapter 4 will describes the result and discussion on the aerobic granulation process with the emerging of industrial pollutants in SBR. This chapter also will discuss on the ability of granulation process to treat high-strength industrial wastewater. Thus, the chapter include the evaluation of the organic, solids and nutrient removal efficiency in the effluent withdrawal. The result also include the observation of others physical parameter and the development of granular sludge itself.

Chapter 5 presents the conclusion of the reasarch and some suggestions for future development of the system. This chapter include the problem faced throughout the research. Some solution to improve the system also being suggested. Lastly, this chapter will describes the possible improvement and enhancement of the research in the future.

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