

**EXTRACTION OF ACETAMINOPHEN FROM  
AQUEOUS SOLUTION BY EMULSION LIQUID  
MEMBRANE**

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MEMBRANE**

**by**

**NUR DINA BINTI ZAULKIFLEE**

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 Speed = 1 ; Diluent = Kerosene) -

## LIST OF SYMBOLS

M	Molar	-
W	Watt	-
%	Percent	-
ng/L	nanogram per litre	-
µg/L	microgram per litre	-
mg/L	miligram per litre	-
kg/m <sup>3</sup>	kilogram per meter cube	-
cP	centiPoise	-
d <sub>32</sub>	Sauter mean diameter	cm
ε	Membrane breakage	%
V <sub>i</sub>	Initial volume of the internal phase	L
V <sub>s</sub>	volume of the internal phase leaked into external phase	L
μ	Viscosity	mPa.s
E	extraction efficiency	%
C <sub>o</sub>	initial concentration of ACTP	mg/L
C <sub>f</sub>	Final concentration of ACTP	mg/L
Hz	Hertz	-
rad/s	Radian per second	-
K <sub>D</sub>	distribution coefficient	-
C <sub>i,II</sub>	ACTP-complex concentration in organic solution	mg/L
C <sub>i,III</sub>	ACTP concentration in feed phase	mg/L
wt%	weight percentage	-

### Subscripts

<i>i</i>	Responding variable
<i>n</i>	Order of membrane flux registered
1,2	Component/species to be evaluated

## LIST OF ABBREVIATIONS

ACTP	Acetaminophen
Adogen 283	Di-tridecylamine
Adogen 381	Tri-isooctylamin
Adogen 383	Tri-isodecylamine
Adogen 464	Tri (C8-C10) methylammonium chloride
Alamine 336, TOA	Tri-octylamine
alkyl poly	Ethylene oxide
Amberlite LA-1	N-5,5,7,7-tetramethylocten-2-yl-1,1,3,3,5,5,- hexamethylhexylamine
BLM	Bulk liquid membrane
$C_{24}H_{44}O_6$	Span 80
CEC	contaminant of emerging concern
$CH_3COOH$	acetic acid
CTAB	cetyltrimethylammonium bromide
D2EHPA	di(2-ethylhexyl)phosphoric acid
DoD	Department of Defense
ELM	Emulsion Liquid Membrane
EPA	Environmental Protection Agency
HCl	Hydrochloric acid
HLB	hydrophile-lipophile balance
$HNO_3$	nitric acid
HOE F 2562	Di-isotridecylamine
KCl	potassium chloride
$Na_2CO_3$	Sodium Carbonate
$NaHCO_3$	Sodium Bicarbonate
NaOH	Sodium Hydroxide
$NH_3$	ammonia
OWO	organic-water-organic
Peimene JMT	1,1,3,3,5,5,7,7,9,9- decamethyldecyl amine
poly	propylene oxide

PP	propylprarabane
PPCP	pharmaceuticals and personal care products
rpm	radian per minute
SDS	sodium dodecylsulfate
SLM / ILM	supported or immobilized liquid membrane
Span 80	Sorbitan Monooleate
TBP	Tributyl-phosphate
TCC	Taylor-Couette Column
TDA	Tri-dodecylamine
TDA	tridodecylanmine
TOA	Trioctylamine
TOPO	trioctylphosphine oxide
USGS	United States Geological Survey
UV-Vis	Ultra Violet Visible
W/O	water-in-oil
WOW	water-organic-water
WWTP	waste water treatment plants

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## **PENGEKSTRAKAN ASETAMINOFEN DARI LARUTAN BERAIR MENGUNAKAN MEMBRAN CECAIR EMULSI (ELM)**

### **ABSTRAK**

Kesedaran mengenai pencemaran alam sekitar menerusi bahan cemar farmaseutikal telah meningkat sejak beberapa tahun lalu. Pencemaran ini adalah sangat meluas, dengan beratus-ratus jenis dadah perubatan boleh didapati pada kadar kepekatan yang rendah di dalam sungai. Salah satu bahan farmaseutikal yang paling banyak digunakan ialah asetaminofen (ASTP). Proses rawatan konvensional yang digunakan oleh loji rawatan air sisa gagal untuk menyingkirkan sebatian farmaseutikal secara sepenuhnya. Di antara kaedah yang sedia ada, salah satu kaedah yang berpotensi untuk menyingkirkan ASTP adalah membran cecair emulsi (MCE). MCE terdiri daripada fasa dalaman dan fasa membran yang membentuk emulsi A / M (air dalam minyak) utama, yang kemudiannya akan diserakkan di dalam fasa luaran. Kajian telah dijalankan bagi mencari rumusan MCE dengan agen pembawa, agen pencair, dan agen perlucutan yang sesuai. Kesan parameter perumusan ini juga telah dikaji untuk mendapatkan rumusan MCE yang terbaik bagi penyingkiran ASTP. Pemilihan komponen MCE yang sesuai, penggunaan pengemulsian ultrabunyi, dan Turus Taylor-Couette (TTC) dijangka dapat meningkatkan kecekapan pengekstrakan ASTP. Kesan bagi beberapa keadaan pengendalian seperti kepekatan agen surfaktan, agen pembawa dan agen perlucutan, kekuatan ultrabunyi, nisbah isipadu, tempoh pengemulsian, nisbah rawatan, kepekatan awal dan kepekatan asid, tempoh adunan, dan kelajuan adunan telah dikaji. Keputusan eksperimentasi menunjukkan bahawa rumusan yang paling sesuai bagi pembentukan membran cecair bagi penyingkiran ASTP adalah dengan menggunakan kerosin sebagai agen pencair, trioktilamina (TOA) sebagai agen pembawa, dan ammonia (NH<sub>3</sub>) sebagai agen perlucutan. Keadaan optimum bagi

proses emulsifikasi telah didapati pada peratus berat TOA dan Span 80 sebanyak 6%, kepekatan agen perlucutan pada kadar 0.1M, tempoh pengemulsian selama 15 minit, nisbah isipadu pada kadar 3: 1, kuasa prob ultrabunyi pada nilai 20W, kepekatan awal pada kadar 10 bahagian per juta (ppm), kepekatan HCl pada kadar 0.1M, tempoh pengekstrakan dengan menggunakan TTC selama 5 minit dengan nisbah sudut frekuensi pada kadar 1.0, dan nisbah rawatan pada kadar 3:1. MCE yang dihasilkan didapati berkesan untuk menyingkirkan 85% ion ASTP daripada larutan akua. Oleh itu, proses MCE merupakan teknologi yang berpotensi untuk mengekstrak ASTP daripada air sisa pengeluaran farmaseutikal.

## **EXTRACTION OF ACETAMINOPHEN FROM AQUEOUS SOLUTION BY EMULSION LIQUID MEMBRANE**

### **ABSTRACT**

In recent years there is an increasing awareness of pharmaceutical contaminants in the environment. Pharmaceutical contamination in rivers is widespread with hundreds of drugs found at low concentrations. One of the main abundantly used pharmaceuticals are acetaminophen (ACTP). The application of conventional treatment process in wastewater treatment plants is unable to completely remove the residues. Thus, among the existing methods, one of the promising methods for ACTP removal is by emulsion liquid membrane (ELM). ELM comprises internal and membrane phase which form primary W/O (water-in-oil) emulsion. The formulation of ELM was investigated to find suitable carrier, diluent and stripping agent. The effect of emulsion formulation parameters of ELM was investigated in order to obtain its best formulation for removal of ACTP. Selection of suitable ELM components, use of ultrasound emulsification and Taylor-Couette Column (TCC) are expected to increase the extraction efficiency. The influence of several parameters such as carrier, surfactant and stripping agent concentration, ultrasonic power, volume ratio, emulsification time, treat ratio, initial and acid concentration, stirring time and stirring speed were investigated. The results show that kerosene as a diluent, Trioctylamine (TOA) as carrier or extractant and ammonia (NH<sub>3</sub>) as stripping agent were the most suitable for the liquid membrane formulation of ACTP removal. The optimum condition for the emulsification study was found at 6 wt.% of TOA and Span 80, 0.1 M concentration of stripping agent, 15 minutes of emulsification time, volume ratio of 3:1, 20 W power of ultrasonic probe, 10 ppm of initial concentration, 0.1M of HCl concentration, 5 minutes of extraction time using TCC with a frequency angular ratio of 1.0 and treat ratio of 3:1. The prepared ELM was found to effectively remove

85% of ACTP ions from aqueous solution. Thus, ELM process is a promising technology to extract ACTP from pharmaceutical production wastewater.

# CHAPTER 1

## INTRODUCTION

### 1.1 Research Background

For centuries environmental pollution has existed but started to be notable in consequence of the industrial revolution in the 19th century. This in turn has raised many critical issues on a vast and unprecedented scale around the globe. Pollution occurs when there is an introduction of contaminants into the natural environment where it harms humans and other living species as well as causes damage to the environment. It is one of the major challenges that the globe is presently facing and increases day by day causing irreversible damage to Mother Earth. Thus, as our environment changes, so does the need to become increasingly aware of the problems that surrounds it.

While present generation resumes to exert themselves in order to minimize traditional contaminants in environment, diverse “emerging” environmental contaminants are warranting attention and is labelled as ‘contaminants of emerging concerns’ or CECs (Richardson and Kimura, 2017). These contaminants are widespread in the aquatic and terrestrial environments, including anthropogenic and naturally occurring chemicals, pharmaceuticals and personal care products (PPCPs), illicit drugs, engineered nanomaterials, and antibiotic resistance genes. Even though it is not yet circulated in drinking water supplies and not monitored in the environment, these contaminants have the potential to cause harmful ecological and human health effects (Noguera-Oviedo and Aga, 2016).

PPCPs is one of the most common emerging pollutants present in wastewater and drinking water. This is because it is initiated not only by humans, but through

veterinary usage too resulting in their endless release to environment. The presence of pharmaceutical CECs in environmental waters are due to incomplete removal in wastewater treatment or diffuse-source contamination, which are threats to drinking waters which leads to estrogenic possibility and harmful effects to both wildlife and human. The major concern of pharmaceuticals CECs is that it is usually specifically designed to target certain metabolic, enzymatic or cell-signalling mechanisms as well as maximise their biological activity at low doses. Several research studies stated that over 30 mg/L of pharmaceutical waste was discharged daily (Fawell and Ong, 2012). Some of the most abundantly used pharmaceuticals are cimetidine, diltiazem, carbamazepine, acetaminophen, and six sulfonamide related antibiotics. According to Al-Odaini et al. (2013), acetaminophen has the highest concentration detected in Langat River, Malaysia with value as high as 350.3 ng/L. The same phenomenon was also noted to occur internationally such as in Spain with concentration of 250 ng/L and 1µg/L in UK.

The removal of PPCPs from wastewater and drinking water is really challenging since there are no comprehensive method in removing it. Removals of these pollutants in the wastewater treatment processes are generally good. Nevertheless, reports on the inability of the application of conventional treatment processes in wastewater treatments plants (WWTPs) to remove pharmaceutical contaminants in water completely have been well documented (Chaouchi and Hamdaoui, 2014). To some extent, the accumulated chemicals were simply discharged into the groundwater while some were not treated properly in the WWTPs (Jarrett, 2017). This is due to the notable concentrations remaining in the final effluents owing to the relatively high influent concentrations encountered. Some specific treatments have been implemented to eliminate PPCPs such as biodegradation, photocatalysis, ozonation and Fenton process

(Kyzas et al., 2015). However, some disadvantages have arisen from these methods including high investment and maintenance cost, formation of secondary pollutants and complex operation procedures (Grassi et al., 2012).

Therefore, integration of advance separation technology with conventional wastewater treatments such as liquid membranes has become a great interest for many. To enhance this process, liquid membrane separation was looked at in this study to be utilized in separating ACTP from contaminated water. Emulsion Liquid Membrane (ELM) which was invented by Li (1968) has shown to have a promising potential for the application of extraction of ACTP. Currently, ELM is introduced as an alternative technique to the separation process where it consists of three main stages which are emulsification, extraction and demulsification. ELM fulfils the promise of providing several attractive characteristics such as high interfacial area to volume ratio for mass transfer, economical, low energy consumption, simultaneous extraction and stripping process, efficient for low solute concentration and requirement of small quantity of solvent. Besides, it is also estimated that ELM is about 40% cheaper than the conventional extraction processes (Kislik, 2010). With these advantages, ELM has been widely studied for industrial applications such as for the separation of various types of metal ions (Zhao et al., 2010, Alaguraj et al., 2009, Ahmad et al., 2013), organic compound (Ng et al., 2010, Lee, 2011) and inorganic compound (Lichang et al., 2016).

## **1.2 Problem Statements**

These chemical compounds, such as acetaminophen, carbamazepine, diclofenac, ibuprofen, and salicylic acid can be easily detected in water (Kim et al., 2007). While the levels of individual pollutants are low, little is known about the long-

term health implications. To make matter worse, there is some concern regarding potential ‘cocktail’ effects of different species of pharmaceutical contaminants mixed together. Thus, even though these compounds existed in trace amount, and at insignificant degree, finding an effective method to prevent further pollution of our water sources are of a major and emerging concern.

Emulsion liquid membrane was given more attention due to having high interfacial area which able the system to selectively recover solute. Thus, it is a suitable method to implement in order to remove acetaminophen CECs in water. Unfortunately, emulsion stability remains as a great challenge that would hinder its wide applications. Emulsion instability occurs through various physical mechanisms such as swelling, breakage and coalescence. It is usually governed by membrane breakage in ELM systems which involves the rupture of the emulsion and leakage of internal phase and extracted solute to the external phase causes the decrease in volume of the stripping phase (Ho and Kamalesh, 1992). This causes the driving force for mass transfer, concentration gradient reduced and increases the external feed concentration, thereby lowering the extraction efficiency. The instability may cause by the emulsion formulation and condition of emulsification (Djenouhat et al., 2008). Therefore, the effects of several factors on liquid membrane formulation together with its effectiveness in removing ACTP were investigated in this study.

In order to obtain high performance of acetaminophen extraction, the selectivity of ELM formulation is very important. According to Chiha et al. (2010), carrier and surfactant concentration, emulsification time and W/O volume ratio have greatly influenced the efficiency and the stability of ELM. Besides that, investigation on process parameters during extraction are important to understand the process of acetaminophen extraction using ELM process. Attempts to reduce emulsion instability

have been made including the usage of Taylor-Couette column (TCC) to disperse the system. The unit was designed to minimize emulsion instability while maintaining high extraction performance (Park et al., 2004). This column improves the stability of the emulsion in such a way that it provides relatively low and uniform fluid shear.

### **1.3 Research Objectives**

The focus of this study is to develop an emulsion liquid membrane system for acetaminophen extraction from aqueous solution using Taylor-Couette Column. Listed below are the measurable objectives:

- i. To formulate emulsion liquid membrane for acetaminophen extraction.
- ii. To investigate the affecting parameters of acetaminophen extraction using emulsion liquid membrane.
- iii. To evaluate the effectiveness of emulsion liquid membrane formulation on acetaminophen removal.

### **1.4 Scope of Research**

The aim of this study is to develop Emulsion Liquid Membrane system for the extraction of acetaminophen from an aqueous solution. Firstly, an ELM was formulated where suitable components in the systems are required for selectively extract acetaminophen from the aqueous solution. Thus, the formulation was initiated by the screening of liquid membrane components where the compatibility of diluent with the other membrane phase components (carrier, surfactant and stripping solution) will be looked at before selection is made for the optimal ELM formulation.

The chosen carrier, diluent and stripping solution must comply with the reaction in the interface of the membrane to support the simultaneous processes of extraction and stripping. Also, the stoichiometry of the extraction reaction and the optimum operating conditions will be determined. Therefore, influence of operating conditions such as carrier concentration, surfactant concentration, stripping agent concentration, volume ratio, ultrasound power, emulsification time, treat ratio, angular frequency ratio and time using TCC will be investigated. These parameters were investigated to obtain the best emulsion formulation hence, a stable emulsion and maximum acetaminophen removal efficiency could be achieved.

### **1.5 Significance of Current Work**

Due to increasing demand of acetaminophen in many applications, it is essential to extract acetaminophen from biological production waste. ELM was implemented as promising alternative separation technology to the existing conventional technique such as electrochemical, ozonation and solar photoelectro-Fenton oxidation. It provides tremendous advantages where the extraction offers high interfacial area to volume ratio for mass transfer, economical, low energy consumption, simultaneous extraction and stripping process, efficient for low solute concentration and requirement of small quantity of solvent. This study will be significant in wastewater treatment due to its high efficiency in removing desired contaminants. Besides that, it is also beneficial to industry and treatment plants as it is an alternative economical way in dealing with emerging contaminants. In addition, it will also serve as reference to new researchers to achieve higher and better extraction efficiency.