



## Extraction of Acetaminophen from Aqueous Solution by Emulsion Liquid Membrane Using Taylor-Couette Column

N. D. Zaulkiflee<sup>a</sup>, M. M. H. Shah Buddin<sup>b</sup>, A. L. Ahmad<sup>\*a</sup>

<sup>a</sup> School of Chemical Engineering, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia

<sup>b</sup> Faculty of Chemical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

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### ABSTRACT

A study on the extraction of Acetaminophen (ACTP) which is also known as paracetamol, from aqueous solution by emulsion liquid membrane process using Taylor-Couette Column (TCC) was investigated. An ELM consists of three phase system which are the external, membrane and internal phases. The external phase containing the ACTP aqueous solution to be treated. Basically, the internal and membrane phase form the primary water-in-oil (W/O) emulsion using ultrasonic probe which is to be dispersed in the external phase. In this work, Triethylamine (TOA), Span 80 and kerosene were used as carrier, surfactant and diluent, respectively in membrane phase. Meanwhile ammonia solution was used as a stripping agent in the internal phase. The influence of several operating conditions such as surfactant and carrier concentration, ultrasonic power, emulsification time, treat ratio, stirring time and stirring speed were investigated. The present work proved that the ELM using TCC system was capable to effectively remove about 85 % ACTP from aqueous solutions under optimum conditions of 15 minutes of emulsification time, 6 wt.% of Triethylamine and Span 80, 20 W power of ultrasonic probe, 5 minutes of extraction time, frequency angular ratio of 1.0 and treat ratio of 3:1.

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## 1. INTRODUCTION

Diverse chemicals are being introduced by society in vast quantities for a range of purposes including agricultural, industrial, household as well as for human and animal healthcare. These chemicals are commonly known as 'contaminants of emerging concern' (CECs). The term does not necessarily correspond to newly discovered compound in the environment due to analytical developments, but also refers to compounds that are recently been categorised as contaminants [1]. CECs has emerged as an environmental problem and it may have adverse effects on aquatic ecosystem. There are several groups of compounds that emerged which are algal and cyanobacterial toxins, brominated flame retardants, disinfection by-products, gasoline additives, hormones and other endocrine disrupting compounds, organometallics, organophosphate flame retardants and plasticisers, perfluorinated compounds, pharmaceuticals and personal care products, polar pesticides and their

degradation/transformation products and surfactants and their metabolites [2].

The presence of pharmaceuticals is one of the most studied group of emerging contaminants has been widely reported in the aquatic environment at the low ng/L to mg/L range [3-6]. This includes more than 4000 molecules with different physico-chemical and biological properties and distinct modes of biochemical reaction. Most of medical substances are administered orally whereas some drugs are metabolised while others remain intact before excreted [7]. Therefore, a mixture of pharmaceuticals and their metabolites will enter municipal sewage and sewage treatment plants. Improper treatment of these chemicals will eventually cause major environmental pollution [8]. Besides that, it may also enter the environment through disposal of unused and emissions from manufacturing process of the products [9]. Though residues of the chemicals were detected in natural waters, however outputs of Waste Water Treatment Plants (WWTP) were identified as the main

\*Corresponding Author Email: chlatif@usm.my (A. L. Ahmad)

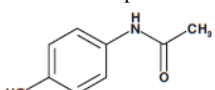
source of pharmaceuticals introduction into the ecosystem. Recently, 44 pharmaceuticals have been developed in a common priority list which are relevant for the water cycle based on consumption, physicochemical properties, toxicity, occurrence, persistence and resistance to treatment [10]. In fact, number of sources of water at high risk of contamination is expected to escalate as human population density increases. This is due to the fact that, these chemicals disrupt the endocrine balance in various ecological species and can adversely affect fish and other aquatic species living in the contaminated water [11]. Easily detected compounds in the contaminated water include acetaminophen, carbamazepine, diclofenac, ibuprofen and salicylic acid [12].

Among widely used pharmaceuticals is Acetaminophen (ACTP), also known as Paracetamol, which primarily used as analgesics and antipyretics. It is a drug used to relieve pain and to suppress inflammation in a way similar to steroids without side effects. Table 1 shows the chemical structures and properties for acetaminophen. Although the anti-inflammatory effect is weak, the impact on the environment is not different from others. As reported by Kim, Choi, Jung, Park [12], ACTP is one of the most frequently detected pharmaceuticals in sewage treatment plant effluents, drinking water or surface water. According to Stackelberg, Gibs, Furlong, Meyer [9], the maximum amount of acetaminophen compounds detected in source water is 0.12 µg/L. Even though this compound existed in trace amount, and at insignificant degree, but finding an effective method to prevent further pollution of our water sources is a major concern.

Major portion of the pharmaceuticals products were removed by conventional wastewater treatment processes. Reports on the inability of the conventional treatment processes applied in wastewater treatment plants to remove pharmaceutical compounds in water completely have been well documented [13]. To some extent, the accumulated chemicals were simply discharged into the groundwater while some were not treated properly in the WWTPs [11].

Among the hydrometallurgical methods available, solvent extraction provides an effective and simple separation method [14].

**TABLE 1.** Chemical structures and properties for acetaminophen [7]

Compound Chemical Structure	Log K <sub>ow</sub>	pK <sub>a</sub>	Water solubility (mg/L)
Acetaminophen 	0.4 6	9.3 8	1.40 x 10 <sup>4</sup>

To enhance this process, liquid membrane separation was looked at in this study to be utilized in separating ACTP from contaminated water.

To date, there are three configurations of liquid membrane, bulk, supported and emulsion liquid membrane. Out of these three types, emulsion liquid membrane (ELM) can achieve much higher mass transfer compared than the other two [15]. ELM 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 [16]. The combination of extraction and stripping processes in a single unit leads to solute purification and concentration simultaneously. The basic process of ELM system is the use of three phase dispersion system where primary emulsion which consists of organic and stripping phase is dispersed in the phase to be treated. 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.

Attempts to reduce emulsion instability have been made including the usage of Taylor-Couette column (TCC) to disperse the system. The unit was design to minimize emulsion instability while maintaining high extraction performance [17]. This column improves the stability of the emulsion in such a way that it provides relatively low and uniform fluid shear.

Previous study by Chaouchi and Hamdaoui [13] revealed that under optimum operating parameters, it was possible to extract ACTP molecules by ELM. To the best of our knowledge, no work has been reported on extraction of ACTP from aqueous solution by ELM using TCC. This study focuses on the development of an ELM system, which to be disperse in TCC to extract the targeted solute from aqueous solution. Optimum operating conditions are determined. Therefore, influence of operating conditions such as surfactant concentration, extractant concentration, ultrasonic power, emulsification time, treat ratio, stirring time and stirring speed were investigated.

## 2. EXPERIMENTAL

**2. 1. Materials** Chemicals and reagents used in this study composed of four main components which are carrier, diluent, stripping agent and surfactant. In present work, the Acetaminophen (ACTP) act as a feed solution in the external phase, Trioctylamine as extractant, Sorbitan Monooleate as surfactant, Kerosene as diluent and Ammonia as stripping phase. All chemicals used for ELM and its properties are listed in Table 2.

**2. 2. ELM Preparation** Emulsion Liquid Membrane (ELM) system was performed by dispersing primary emulsion consist of membrane and internal phase in the feed solution. 10 mg/L of ACTP feed solution, was prepared by dissolving the desired amount of solute ACTP in distilled water. 10 mg/L of ACTP feed solution, was prepared by dissolving the desired amount of solute ACTP in distilled water with the addition of hydrochloric acid solution, HCl for ACTP complexes protonation [13].

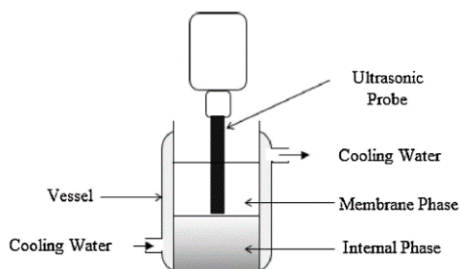
Meanwhile, the membrane phase was prepared by mixing Trioctylamine (TOA) and Span 80 in kerosene at speed of 500 rpm. The internal aqueous phase of ammonia solution was then added to the membrane organic phase where the volume ratio is internal aqueous phase to membrane phase is 1:3. These phases were homogenized with the assistance of the ultrasonic probe (USG-150) which is equipped with titanium horn as shown in Figure 1, at power and frequency of W, kHz, respectively.

In this work, ultrasound was employed to obtain the primary water-in-oil (W/O) emulsion as it is a meta-stable mixture that can be obtained by applying certain level of shearing energy. High monodisperse emulsion was achieved via this process.

**2. 3. ELM For ACTP Extraction** The prepared W/O emulsion and external aqueous solution (ACTP) were put in the feeding container and allowed to pass through silicon tube into the TCC. The content were mixed at predetermined shear stress defined by the stirring speed.

**TABLE 2.** List of chemicals and their properties

Chemicals	Molar Mass (g/mol)	Brand
Acetaminophen	151.163	Sigma Aldrich
Kerosene	198.39	Sigma Aldrich
Trioctylamine	323.68	Merck
Sorbitan Monooleate	428.60	Merck
Hydrochloric acid fuming 37 %	36.46	Merck
Ammonia Solution 25 %	35.04	Merck



**Figure 1.** Experimental setup of ultrasonic probe for the preparation of W/O emulsion [18]

In this system, the inner and outer concentric cylinders are independently rotatable.

At the end of the extraction process, the solution was then flowed into separating funnel and left for settling for 5 minute. The external phase sample was then taken out using a syringe for ACTP ions concentration measurement and the extraction efficiency, E(%) was calculated using Equation (1):

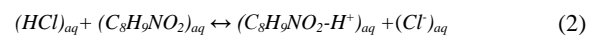
$$\text{Extraction Efficiency, } E(\%) = \frac{C_0 - C_f}{C_0} \times 100 \quad (1)$$

where  $C_0$  is the initial concentration of ACTP in the external phase (mg/L) while  $C_f$  is the final concentration of ACTP at the end of extraction process. The concentration of ACTP in the solution was determined by UV-Visible Spectrophotometer at the maximum absorbance of ACTP.

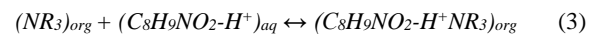
### 3. RESULTS AND DISCUSSION

**3. 1. Transport Mechanism of ACTP** The mechanism of carrier-facilitated transport of extraction and stripping of ACTP is schematically presented in Figure 2.

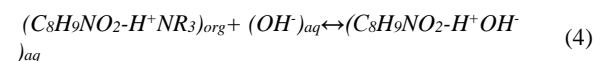
a) In the external feed phase, the ACTP was protonated by HCl as shown in Equation (2):



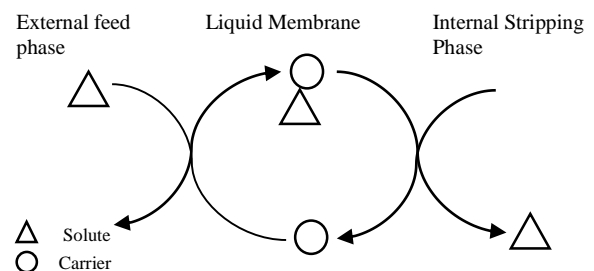
b) At the external-membrane interface, ACTP chemically react with TOA as expressed in Equation (3).



c) At the membrane-stripping interface, the complex ACTP-TOA then diffuses to the internal interface through the membrane phase by reacting with ammonia as shown in Equation (4):



where  $NR_3$  represents TOA,  $C_8H_9NO_2$  represent ACTP;  $C_8H_9NO_2-H^+NR_3$  represent ACTP-TOA complex;  $OH^-$  represent ammonia.

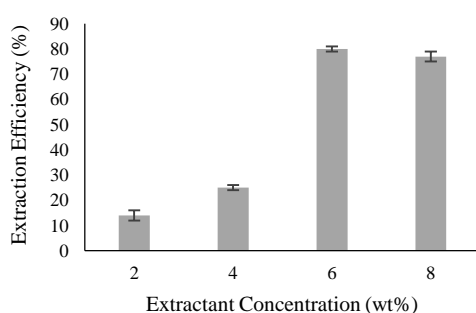


**Figure 2.** The mechanism of coupled transport in ELM

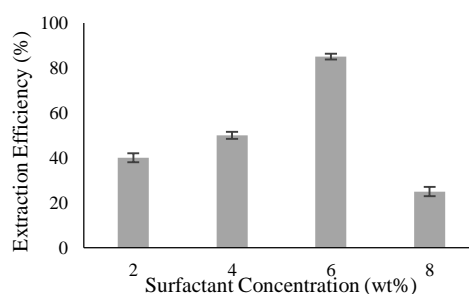
### 3. 2. Effect of Extractant Concentration

Extractant concentration is important in determining the extraction performance of ACTP. The effect of the TOA concentration on the ACTP extraction efficiency is shown in Figure 3. It can be seen that the ACTP extraction efficiency was highly affected by the concentration of the extractant. There was only 14 to 20 % of ACTP was extracted using TOA concentration of 2 to 4 wt.%, respectively. At such low extractant concentration, ACTP was not completely be entrapped and transported into the internal phase thus resulting in low extraction efficiency Chaouchi and Hamdaoui [13]. The extraction progressed as the extraction concentration increased to 6 wt.%. This can be attributed to the higher the concentration of extractant in the membrane phase which lead to the higher transportation of solute at the membrane-external interface. However, further increment of the extractant concentration beyond 6 wt.% has reduced the efficiency to 70%. This indicates that it is more than enough to remove ACTP from feed phase by utilizing 6 wt.% of extractant concentration. High amount of carrier in the membrane does not result much benefit due to increase in viscosity which leads to larger globules [19, 20]. Moreover, it is important to minimize the amount of carrier since it is the most expensive material for emulsion composition to make it economically feasible. Thus, the best condition of extractant concentration was identified as 6 wt.%.

**3. 3. Effect of Surfactant Concentration** Various surfactant concentration (2 to 8 wt.%) were used in the membrane phase to investigate its effect on ACTP extraction efficiency. The data obtained is shown in Figure 4. It was found that the extraction efficiency increased from 40 to 85 % together with the increment of Span 80 concentration from 2 to 6 wt.%, respectively. However, the usage of 8 wt.% of Span 80 turns out to be counterproductive as the efficiency was reduced to 25 %. Basically, increasing surfactant concentration resulted in more stable emulsion which provide better extraction efficiency.



**Figure 3.** Effect of extractant concentration on extraction efficiency. (Experimental condition: Organic to Internal Ratio = 3:1, Diluent = Kerosene)

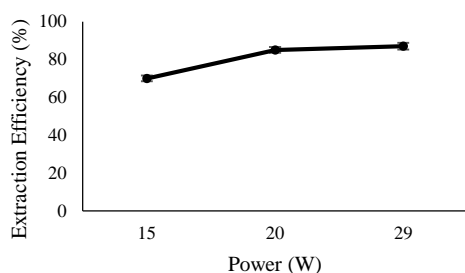


**Figure 4.** Effect of surfactant concentration on extraction efficiency. (Experimental condition: [Ammonia] = 0.1 M ; [TOA] = 6 wt.%; Organic to Internal Ratio = 3:1; Ultrasonic Power = 15W; Emulsification Time = 5 min ; Treat Ratio = 3:1 ;Speed =1;Diluent = Kerosene)

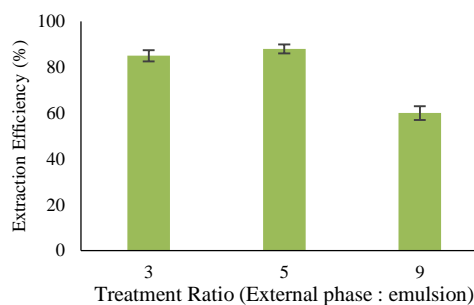
This is due to the reduction in interfacial tension which provides smaller internal droplets [17]. However, excessive amount of surfactants may inversely decrease the extraction efficiency due to the amount of surfactants or micelles was adsorbed to emulsion surface which triggered droplets coalescence [21]. On the other hand, lack of surfactant led to re-dispersion and coalescence during extraction process [22]. Therefore, 6 wt.% of Span 80 is sufficient to be used to develop the emulsion.

**3. 4. Effect of Ultrasonic Power** Figure 5 shows the effect of ultrasonic power used in preparing the primary emulsion towards ACTP extraction efficiency. As illustrated in the figure, the emulsion prepared using 15 W ultrasonic power provided the lowest extraction efficiency due to the incapability to effectively extract ACTP from the aqueous solution. The sound field provided is insufficient to yield enough energy to form a homogenized emulsion which causes large emulsion globules to be produced [18]. Meanwhile, the increment of power from 20 to 29 W increased the extraction efficiency improved from 85 to 87 %, respectively which almost plateau. With an increase in the ultrasonic power, the energy dissipation in the system increase [23]. At this stage, the production of the smaller emulsion globules resulting in the increment of solute transportation area. Further increment of power usage yields no significant increment in the extraction efficiency. Besides, higher ultrasonic power at 29 W does not benefit the emulsion as it may cause coalescence. Moreover, consumption of intensive energy to produce the emulsion will only cause a hike in the cost of production [24]. Therefore, 20 W of ultrasonication is taken as the best condition to produce the primary emulsion.

**3. 5. Effect of Emulsification Time** The effect of emulsification time on the extraction of ACTP was experimentally studied.



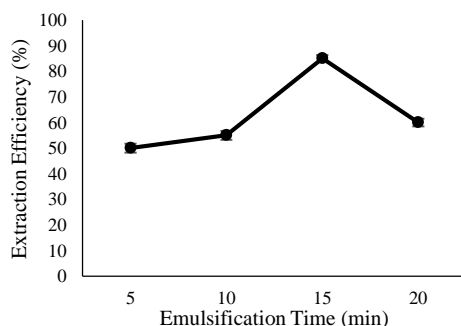
**Figure 5.** Effect of ultrasonic power on extraction efficiency (Experimental condition: [Ammonia] = 0.1 M ; [TOA] = 6 wt.% ; [Span 80] = 6 wt.% ; Organic to Internal Ratio = 3:1; Emulsification Time = 5 min ; Treat Ratio = 3:1 ; Speed = 1 ;Diluent = Kerosene)



**Figure 7.** Effect of treatment ratio on extraction efficiency (Experimental condition: [Ammonia] = 0.1 M ; [TOA] = 6 wt.% ; [Span 80] = 6 wt.% ; Ultrasonic Power = 20W ; Emulsification Time = 15 min; Organic to Internal ratio = 3:1; Diluent = Kerosene)

Experiments were carried out using the best conditions obtained, 6 wt.% TOA, 6 wt.% Span 80 and 20 W of ultrasonication, while varying the emulsification time from 5 to 20 minutes as shown in Figure 6. According to the result obtained, at short emulsification time of 5 minute, it was not enough to homogenize the emulsion thus membrane solution unable to cover the internal phase completely. Subsequently, the percentage was then increased from 55 to 85 % with the increasing emulsification time from 5 to 10 minutes, respectively. This indicates that longer emulsification time yields smaller emulsion diameter thus providing greater amounts of emulsion globules to perform better extraction efficiency [25]. However, extraction efficiency decreased if the emulsion was produced longer than 15 minutes. This is due to excessive amount of fine droplets which lead to the coalescence and resulted in bigger emulsion which in turn decreasing the extraction efficiency [24]. Optimum emulsification time was found to be at 15 minutes.

**3. 6. Effect of Treat Ratio** The effect of treat ratio (external phase: emulsion) on ACTP extraction efficiency was investigated as shown in Figure 7.



**Figure 6.** Effect of emulsification time on extraction efficiency (Experimental condition: [Ammonia] = 0.1 M; [TOA] = 6 wt.% ; [Span 80] = 6 wt.% ; Ultrasonic Power = 20W ; Organic to Internal Ratio = 3:1 ; Treat Ratio = 3:1 ; Speed = 1 ;Diluent = Kerosene)

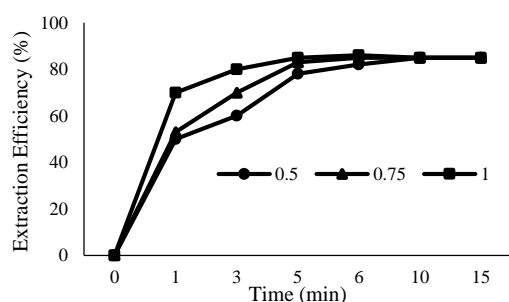
Observation was done at treat ratio of 3:1, 5:1 and 9:1. Diffusion process and extraction rate are related to the treatment ratio. Based on the figure, usage of neither high nor low emulsion volume has benefited the system. As can be observed, ACTP removal increased with the increasing of treatment ratio of 3:1 to 5:1, respectively. This is due to the fact that, increasing treatment ratio increased the amount of emulsion globules available for ACTP per unit volume of reaction mixture. This phenomenon has resulted in the significant increment of the interfacial surface area for mass transfer and evidently followed by the increment of the rate of mass transfer. This in turn increases the extraction efficiency of ACTP. The insignificant change in the extraction efficiency was observed by increasing the treatment ratio to 9:1. This can be explained by the fact that raising the amount of external phase cause the emulsion phase could not disperse very well in order to treat the external phase, hence decreasing the contact area between both external and emulsion phases [20]. This is strongly supported by the other studies where the higher treatment ratio is not preferable as the surface area for extraction is limited due to large emulsion globule size [26]. In addition, the possibility of membrane instability was also taken into account as high amount of emulsion enhances the globules interactions, leading to coalescence and re-dispersion of globules. Eventually, this phenomenon will cause membrane to rupture [27]. Therefore, in order to ensure good dispersion of emulsion, volume ratio of external phase to emulsion of 3:1 was selected as the best treatment ratio.

**3. 7. Effect of Stirring Speed** Optimal stirring speed and time was investigated by maintaining the rotational speed of the inner cylinder while the outer cylinder was varied. Ratio of angular frequency of outer and inner cylinder were varied at 0.5, 0.75 and 1.0. The obtained results are as shown in Figure 8. The extraction efficiency increase with the increase of angular frequency ratio.

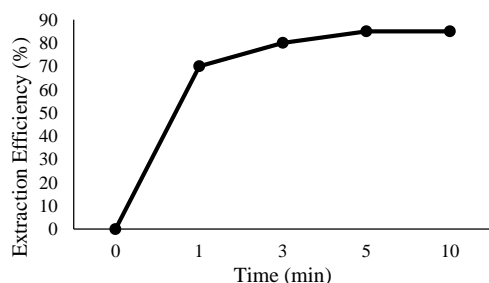
These results is in agreement with Ahmad, Kusumastuti, Buddin, Derek and Ooi [28] in which the extraction efficiency at the end of the process was almost similar except for the extraction time. Basically, the lower angular frequency ratio needed longer extraction time and vice versa. Therefore, increasing angular frequency ratio from 0.5 to 1.0 only shortened the extraction time.

On the other hand, the rotation speed of two counter rotating cylinder provide turbulent-based flow pattern which normally produce high fluid mixing along the cylinder. This condition provide a better extraction efficiency. Meanwhile, the increase in angular frequency of the outer cylinder while fixing the inner cylinder results in a featureless turbulent regimes [17, 28, 29]. High mixing activity which lead to the increment of ACTP extraction rate. Therefore, an angular frequency ratio of 1.0 was chosen for the stirring speed parameter.

**3. 8. Effect of Extraction Time** Figure 9 shows the effect of extraction time towards the extraction performance of ACTP was investigated by varying the extraction time from 1 to 10 minutes using the optimal conditions of the previous parameters studied.



**Figure 8.** Effect of stirring speed on extraction efficiency (Experimental condition: [Ammonia] = 0.1 M; [TOA] = 6 wt.%; [Span 80] = 6 wt.%; Ultrasonic Power = 20W; Emulsification Time = 15 min ; Organic to Internal Ratio = 3:1; Treat Ratio = 3:1; Speed = 1; Diluent = Kerosene)



**Figure 9.** Effect of extraction time to extraction efficiency (Experimental condition: [Ammonia] = 0.1 M ; [TOA] = 6 wt.% ; [Span 80] = 6 wt.% ; Ultrasonic Power = 20W ; Emulsification Time = 15 min ; Angular Frequency Ratio = 1 ; Organic to Internal Ratio = 3:1 ; Treat Ratio = 3:1 ; Speed = 1 ;Diluent = Kerosene)

The results showed that the extraction process occurred rapidly as almost 80 % of ACTP was extracted from the external phase solution in 3 minutes. After 5 minutes, extraction efficiency started to increase insignificantly. Data obtained is in agreement with Raja Norimie Raja Sulaimana, Othman, Amin and Noah [20] who indicated that the extraction time in ELM system was very fast due to the reaction kinetic occurs in a short time. Besides, as the remaining number of moles of ACTP-TOA complex reduces as a function of extraction time, the concentration gradient established was found to be insufficient to drive the complex into the internal phase. Thus, 5 minutes is enough to extract ACTP using TCC.

#### 4. CONCLUSION

The present work proved that the ELM using TCC system was capable to effectively remove about 85 % ACTP from aqueous solutions under optimum conditions of 15 minutes of emulsification time, 6 wt.% of Trioctylamine and Span 80, 20 W power of ultrasonic probe, 5 minutes of extraction time, frequency angular ratio of 1.0 and treat ratio of 3:1.

#### 5. ACKNOWLEDGEMENT

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## Extraction of Acetaminophen from Aqueous Solution by Emulsion Liquid Membrane Using Taylor-Couette Column

N. D. Zaulkiflee<sup>a</sup>, M. M. H. Shah Buddin<sup>b</sup>, A. L. Ahmad<sup>a</sup>

<sup>a</sup> School of Chemical Engineering, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia

<sup>b</sup> Faculty of Chemical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

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یک مطالعه بر روی استخراج استامینوفن (ACTP) که همچنین به عنوان پاراستامول شناخته شده است، از محلول آبی با روش غشای مایع امولسیون (ELM) با استفاده از ستون تیلور کویت (TCC) مورد بررسی قرار گرفت. یک ELM متشکل از سیستم سه فازی است که از فاز خارجی، غشا و فاز داخلی تشکیل شده است. فاز خارجی حاوی محلول آبی ACTP است که می بایست مورد درمان و جداسازی قرار گیرد. اساساً فاز داخلی و غشایی، امولسیون اولیه آب در روغن (W/O) است که با استفاده از پروب اولتراسونیک که در فاز خارجی پراکنده می شود، تشکیل می شود. در این کار، تری اکتیل آمین (TOA)، Span 80 و نفت سفید به عنوان حامل، سورفاکتانت و رقیق کننده به ترتیب در فاز غشاء مورد استفاده قرار گرفتند. در عین حال محلول آمونیاک به عنوان یک عامل پاک کننده و تمیزکننده در فاز داخلی استفاده شد. تأثیر چندین شرایط عملیاتی مانند سورفاکتانت و غلظت حامل، قدرت اولتراسونیک، زمان امولسیون، نسبت درمان و جداسازی، زمان جداسازی و سرعت همزدن جهت جداسازی مورد بررسی قرار گرفت. نتایج نشان می دهد که سیستم ELM با بهره گیری از سیستم TCC توانایی حذف حدود 85% ACTP از محلول های آبی را در شرایط بهینه 15 دقیقه زمان امولسیون، 6% وزنی تری اکتیل آمین و Span 80، قدرت پروب اولتراسونیک 20 W، و 5 دقیقه زمان استخراج با نسبت زاویه ای فرکانس 1.0 و نسبت ریکاوری 3:1 بهترین شرایط برای جداسازی می باشد.

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