REDUCTION OF CHROMIUM (VI) TO CHROMIUM (III) USING CONTINUOUS EMULSION LIQUID MEMBRANE

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ABSTRACT

Recently, the application of emulsion liquid membrane (ELM) process as an alternative technology for solute separation is highlighted due to the simple operation of simultaneous extraction and stripping process. The most important aspects for a successful ELM process are liquid membrane formulation and emulsion stability. This study was carried out to investigate the liquid membrane formulation for the reduction of chromium (VI) to chromium (III) from electroplating wastewater using continuous ELM process (CELM). Liquid membrane system comprises of three liquid phases which are external (electroplating wastewater), organic liquid membrane and internal phase. Liquid membrane and internal phase were emulsified and dispersed into the external phase to be treated. The experimental work consisted of four major parts which were ELM component formulation, stability study of ELM in batch process, screening of parameters and optimization of chromium removal efficiency by response surface methodology (RSM) in continuous operation process and recovery of the chromium at optimum process conditions. The results show that the favourable conditions for liquid membrane formulation are 0.04 M TOMAC as a carrier, palm oil as a diluent and 0.1 M thiourea in 0.1 M sulfuric acid as a stripping agent. The best condition of stable water-in-oil (W/O) emulsion was obtained at 7000 rpm of homogenizer speed, 5% (w/v) Span 80 as surfactant and 1 minute of emulsifying time. Meanwhile, the most stable water-in-oil-in-water (W/O/W) emulsion obtained during the continuous process operation was at 350 rpm agitation speed, pH<5 of external phase and 1 to 5 of treat ratio. The optimization results by RSM show that 99% of chromium was extracted at 2.83 minutes of retention time, 342 rpm rotational speed and 1 to 5 of treat ratio. As a conclusion, about 81% of less-toxic chromium (III) has been recovered into the internal phase using 2.0 M thiourea in 2.0 M sulfuric acid as the stripping agent. The favourable process condition of the formulated membrane study was satisfactory and is suitable to treat wastewater as low as 20 ppm up to 200 ppm of chromium concentrations. This study reveals that CELM is a simple process and practical technology to remove chromium (VI) from industrial wastewater while solving the environmental problem simultaneously.

ABSTRAK

Pada masa kini, penggunaan proses emulsi membran cecair (ELM) sebagai teknologi pemisahan alternatif bahan larut telah diberi penekanan disebabkan proses pengoperasiannya yang mudah bagi pengekstrakan dan pelucutan secara serentak. Perkara yang paling penting bagi menjayakan proses ELM adalah formulasi membran cecair dan kestabilan emulsi. Kajian ini dijalankan untuk mengkaji formulasi membran cecair untuk menurunkan kromium (VI) kepada kromium (III) daripada air sisa buangan penyaduran menggunakan proses ELM berterusan (CELM). Sistem membran cecair terdiri daripada tiga fasa cecair iaitu luaran (air sisa buangan penyaduran), membran cecair organik dan fasa dalaman. Membran cecair dan fasa dalaman telah diemulsi dan diserakkan ke dalam fasa luaran yang akan dirawat. Eksperimen ini terdiri daripada empat bahagian utama iaitu formulasi ELM, kajian kestabilan ELM dalam proses berkelompok, penyaringan pembolehubah dan pengoptimuman kecekapan penyingkiran kromium dengan menggunakan kaedah sambutan permukaan (RSM) dalam operasi proses berterusan dan perolehan semula ion kromium pada keadaan optimum. Hasil kajian menunjukkan bahawa keadaan yang bersesuaian bagi formulasi membran cecair adalah 0.04 M TOMAC sebagai pembawa, minyak sawit sebagai bahan pencair dan 0.1 M thiourea dalam 0.1 M asid sulfurik sebagai agen pelucutan. Keadaan terbaik bagi kestabilan emulsi air-dalam-minyak (W/O) diperolehi pada 7000 rpm kelajuan penghomogen, 5% (w/v) Span 80 sebagai surfaktan dan 1 minit masa pengemulsian. Sementara itu, emulsi air-dalam-minyak-dalam-air (W/O/W) yang paling stabil semasa operasi proses berterusan adalah pada 350 rpm kelajuan pengadukan, pH<5 bagi fasa luaran dan nisbah rawatan 1 kepada 5. Keputusan pengoptimuman oleh RSM menunjukkan bahawa 99% kromium telah diekstrak pada 2.83 minit tempoh penahanan, 342 rpm kelajuan pengadukan dan nisbah rawatan 1 kepada 5. Kesimpulannya, sebanyak 81% kromium (III) yang kurang toksik telah berjaya diperoleh ke dalam fasa dalaman pada 2.0 M thiourea dalam 2.0 M asid sulfurik sebagai agen pelucutan. Keadaan proses yang sesuai untuk membran yang telah diformulasikan adalah memuaskan dan sesuai untuk merawat air sisa pada kepekatan kromium serendah 20 ppm hingga 200 ppm. Kajian ini menunjukkan bahawa CELM adalah satu proses mudah dan merupakan teknologi yang praktikal untuk menyingkirkan kromium (VI) daripada air sisa buangan industri sekaligus menyelesaikan masalah persekitaran.

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LIST OF ABBREVIATION

AAS	–	Atomic Absorption Spectrometer
Au	–	Gold
BLM	–	Bulk Liquid Membranes
CELM	–	Continuous Emulsion Liquid Membrane
Cr	–	Chromium
Cu	–	Copper
CYANEX 272	–	bis(2,4,4-trimethylpentyl)phosphinic acid
CYANEX 302	–	Diisooctylthiophosphinic acid
D2EHPA	–	Bis(2-ethylhexyl)phosphate
DAF	–	Dissolved air flotation
DOE	–	Department of Environment
ELM	–	Emulsion Liquid Membrane
EPA	–	Enviroment Protection Agency
H_2S	–	Hydrogen Sulphide
H_2SO_4	–	Sulphuric Acid
HLB	–	Hydrophilic-Lipophilic Balance
IC	–	Ion Chromatography
ILM	–	Immobilized Liquid Membranes
LM	–	Liquid Membrane
O/W	–	Oil In Water Emulsion
PAC	–	Poly Aluminum Chloride
ppb	–	Parts Per Billion
ppm	–	Parts Per Million
RDC	–	Rotating Disk Contactor
SLM	–	Supported Liquid Membranes
Span 80	–	Sorbitan Monooleate

TBP	–	Tributyl Phosphate
TDA	–	Tridecyl-amine
TOA	–	Tryoctyl-amine
TOPO	–	Tri-n-octylphosphineoxide
USEPA	–	United State Environment Protection Agency
W/O	–	Water In Oil Emulsion
W/O/W	–	Water-In-Oil-In-Water
WHO	–	World Health Organization
Zn	–	Zinc

LIST OF SYMBOLS

T – Temperature

g/mol – gram per mol

m^3 – Meter cubic

M – Molar concentration

ppm – part per million

ppb – part per billion

rpm – rotation per minute

t – Time (s)

wt – Weight

w/v – weight per volume

% – Percentage

g/cm^3 – gram per centimeters cubic

V – Volume (m^3)

pH - – Logarithmic measure of hydrogen ion concentration

D – Dissociation constant

d_{32} - – Sauter mean diameter

H/D – Height-to-diameter ratio

X_n – Independent variables

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Heavy metal ions can be found naturally in the environment, however nowadays, their concentration is getting higher due to the increase of industrial wastewater. Thus, discharging wastewater containing heavy metals into the water bodies directly without any treatment can pose severe effects to the environment as well as public health. Meanwhile, rapid industrialization and urbanization in Malaysia has alarmingly increased the amount of toxic heavy metals entering the environment. According to the World Health Organization (WHO), metals that are deemed among the most toxic existed in the industrial wastewater are chromium (Cr), zinc, lead, nickel, iron, aluminium, copper, cobalt, mercury, and cadmium [1].

There are several sources of wastewater that contribute to Cr pollution such as wood preservatives, plants producing industrial inorganic chemicals and pigments, textile dyeing, leather tanning, aluminum conversion coating operations, electroplating, and mining [2]. Above all, electroplating processes create significant amounts of wastewater containing heavy metals (Cr) from a numerous of applications. These include milling and etching, anodizing-cleaning, conversion-coating, electroless depositions, and electroplating [3]. Consequently, it is essential to treat heavy metals-contaminated wastewater prior to its release to the environment. Furthermore, instead of removing the heavy metals from electroplating wastewater, a study on metal recovery is significantly important. Therefore, it requires more efficient techniques for the recovery process. In principal, the recovery process has several incentives such as reduction in the volume and toxicity of the waste effluents, recovery of valuable/monetary metal and saving of disposal costs. Moreover, these wastes will cause a lot of environmental problems if they are directly discharged into the natural water system.

Several conventional treatment processes have been used in metal ions extraction from industrial wastewater such as precipitation, solvent extraction, ion exchange, adsorption, and electrochemical recovery. These techniques, however, are not effective due to unsuccessful fulfilment to the regulation levels for technical, economic, and environmental reasons [4-8]. Conventionally, precipitation is the most used method to extract heavy metals. Among the existed precipitation techniques, sulphide, and hydroxide precipitations are the two preferred techniques that are presently been utilized with, and by far the most commonly used technique is hydroxide precipitation. But, as not all metal hydroxide completely precipitated at a single pH, this technique does not guarantee a total compliance for a variety of metals existing in the waste stream [9]. Many researchers found that emulsion liquid membrane (ELM) extraction has a great potential to overcome the problem. ELM is also reported as an advanced technique for separating and concentrating metals. This process gives less chemical consumption, energy saving, fast, and simple operation [10].

Conventionally, the membrane phase of liquid membrane is formed by organic diluents derived from petroleum resources, and thus is toxic, non-renewable, and could be extremely expensive due to the limited resources. Environmental aspects are frequently connected to the concept of sustainable development, which has become a common goal and from time to time a demand in the industrial sector. Recently, liquid membrane was improved to “green liquid membrane” through the usage of environmental friendly diluents. Plant oils (for instance coconut or palm oil) can be used as non-toxic and biodegradable diluents as an alternative of common organic diluents such as kerosene, toluene and benzene. It has the capability to reduce the amount of common toxic and hazardous chemicals used in liquid membrane formulation. Yet, among the nine types of commercially available vegetable oils in India, Venkateswaran and Palanivelu [11] found that palm oil is the best green oil based on LM. In addition, palm oil has been found to work well for the extraction of Cr (VI) using ELM [12] and the extraction of phenol in supported liquid membranes (SLM) [11]. Therefore, competitive vegetable oils are used as alternative and renewable organic diluent as they are non-toxic and readily available.

The implementation of small laboratory batch process is unpractical at the industrial scale as hundreds or even thousands of process cycles would be necessary for commercial purposes. The solution to this problem is to use a continuous mode. Consequently, ELM can be operated in both batch and continuous modes [13]. Currently, ELM has difficulties for commercialize processes and still operate in batch process and laboratory scale due to the membrane instability encountered as reported by Kislik [13]. On the other hand, upgrading all or parts of a process from batch to continuous yields many benefits such as 24 hour production, less retention time, more cost-effective owing to constant extraction and recovery of targeted solutes, having less total operating cost for large scale as well as higher recovery rate compared to the batch process [14].

1.2 Problem Statement

The strong release of Cr ions into the environment by several manufacturing industries will not simply contaminate the wastewater but the nature as well. Meanwhile, wastewater treatment is crucial in ensuring safer and healthier environment. Hexavalent Chromium, Cr (VI) is broadly found in electroplating wastewater. It is mostly presents in the form of oxyanions such as bichromate ($HCrO_4^{-}$), dichromate ($Cr_2O_7^{2-}$) or chromate (CrO_4^{2-}) which are reliant on the pH [15]. According to the provisional guideline by WHO, the permitted concentration value for Cr in drinking water is 0.05 ppm [1]. In addition, based on Environmental Quality (Industrial Effluent) Regulations, 2009 (Malaysia), plating industries need to pre-treat the wastewater prior to discharge in accordance with discharge limits for industrial effluent [16]. Therefore, a cost-effective recovery process for Cr (VI) is a great concern due to its growing importance in the environmental protection problems. On the other hand, studies on Cr reduction are significantly important in order to find suitable alternative of Cr removal from industrial wastewater [17]. As Cr (VI) is known as human carcinogen, it is vital to evaluate the oxidation-reduction characteristics of Cr (VI) species [18]. Although Cr (VI) can be reduced to trivalent state, detailed information on this in workplace environments is limited. The most common conventional method for Cr (VI) removal is Cr (VI) reduction to Trivalent

Chromium, Cr (III) followed by precipitation of Cr $(OH)_3$ with lime at pH 9-10 [19]. In principal, reduction offers several incentives for example disposal costs decrement, recovery of valuable/monetary Cr, and low toxicity of waste effluents. However, precipitation possesses solid waste disposal disadvantage [19]. Therefore, it requires more efficient techniques for removal and reduction of Cr (VI) to Cr (III). Meanwhile, the study on Cr (VI) reduction to Cr (III) has been studied by Maxcy *et al.* [20]. Excellent performance for Cr (VI) reduction to Cr (III) was attempted using thiourea in strongly acidic medium. Besides that, thiourea also has been applied as reducing agent for gold and silver [21]. Therefore, in order to focus on metal reduction toxicity and recovery, it is requires more efficient techniques for removal and reduction of Cr (VI) to Cr (III).

In order to solve the problem, ELM which is one of the configurations in liquid membrane technology was chosen in this present work due to several advantages such as less energy requirement, both extraction and stripping occurred simultaneously in one single-step operation, less chemical consumption, ease of functioning, large mass transfer interfacial area, and low cost factor. ELM allows a highly selective transport and efficient enrichment of solute ions through a very thin liquid membrane with suitable tailor made liquid membrane (LM) formulation. In this study, palm oil as a green based diluent is formulated with suitable carrier and stripping agent to selectively extract the Cr (VI) from real electroplating wastewater.

On the other hand, the industries also generate huge volume of hazardous wastewater and require proper disposal and treatment. Instead of batch treatment, the continuous operation is more suitable for treatment of large volume of wastewater. Several studies for continuous ELM has been done and demonstrated as an effective alternative technology for separation and purification processes for metal extraction [22-23]. However, there is drawback such as big possibility of re-emulsification for Oldshue-Rushton type extraction column, poor mass transfer efficiency for spray column and deficiency of mixing due to the disc limitation for rotating disc contactor (RDC). Although few methods had been established for the continuous ELM processes, improvements on the processes as well as their design are still required especially in studying the purpose of scale-up and practical applications in the industries.

The key obstacle in employing this method for industrial separations is the stability of emulsion. Thus, the result of the emulsion droplets and globule size distribution was investigated in the ELM stability study. Also, the investigation on emulsion stability using CELM was done by manipulating the total volume level based on height to diameter ratio (H/D). Based on the literature review, there were no researches reported for the stability study using CELM process.

To the best of our knowledge, this thesis reports, for the first time, a detailed investigation on the Cr (VI) reduction to Cr (III) in CELM process from real rinse electroplating wastewater. The investigation was carried out to study the formulation and stability of the emulsion liquid membrane and also to establish optimum condition for Cr extraction and recovery in CELM process. Hence, this technology is expected to be suitable and relevant in treating Cr ions present in the real rinse electroplating wastewater.

1.3 Objectives of the Research

The main purpose of this research is to study the feasibility of using continuous emulsion liquid membrane (CELM) process to extract and recover Cr from real rinse electroplating wastewater with selected LM formulation. The following are the objectives of this research.

i. To formulate suitable liquid membrane for Cr (VI) reduction to Cr (III) from real rinse electroplating wastewater in ELM process.
ii. To investigate the ELM stability for water-in-oil (W/O) and water-in-oil-in-water (W/O/W) emulsion in a batch process.
iii. To set-up the bench scale of CELM and to study the effect of parameters on emulsion stability and extraction performance in the continuous extractor using response surface methodology (RSM) method.

iv. To establish optimum process condition for Cr extraction and recovery in CELM process.

1.4 Research Scopes

In liquid membrane formulation, the study focused on the selection of liquid membrane components for Cr extraction from real rinse electroplating wastewater. Electroplating wastewater was characterized in terms of anionic and ionic content, pH, density, and viscosity. Then, a screening process was carried out using liquid-liquid extraction to determine the suitable types of carriers, diluents and stripping agents for Cr ions extraction. During the experiments, different types of carriers (acidic, basic and solvating) were used and the amounts of Cr extracted were recorded, while the other parameters were fixed. After finding the most suitable carrier for Cr, the carrier concentrations were varied in order to find the best concentration of carrier to extract the Cr. At the same time, several stripping agents (basic, acidic and chelating) were screened out to extract the loaded carrier-Cr complexes. Span 80 was used as surfactant while corn oil, chloroform, toluene, kerosene, and palm oil were used as diluents. Then, the liquid membrane formulation was developed for Cr extraction and recovery and the mass transfer mechanism of Cr extraction was determined in the second objective.

The third objective was achieved by conducting the batch ELM system. There are three main components which are liquid membrane phase (consists of diluent, carrier and surfactant), external phase (feed phase), and internal phase (stripping solution). Several affecting parameters for ELM stability, swelling and breakage were identified in this objective. Investigation on the stability of primary water-in-oil (W/O) emulsion was carried out by manipulating the emulsifying times (1 to 10 minutes), homogenizer speeds (5000 to 13500 rpm) and the concentrations of surfactant (1 to 7% (w/v)) during the emulsification stage. 1-Octanol was used as the phase modifier in this study. Besides that, the stability of water-in-oil-in-water (W/O/W) was also studied by varying agitation speed (200 to 500 rpm), contact time (1 to 7 minutes), treats ratio of emulsion to external phase (1:3 to1:10) and pH of the external phase solution. The influence of these parameters on the emulsion droplets and globules size distribution was determined under the microscope.

Next, the possibility of continuous ELM as promising technique for Cr extraction and recovery was investigated. The optimum conditions of liquid membrane formulation and process conditions of batch system were used as a guide to set up a continuous process. Then, the CELM rig was set-up and configures as well as the investigation of its stability was done by manipulating the total volume level in continuous extraction vessel based on the height to diameter ratio (H/D).

After obtaining the stable CELM process condition, the extraction of Cr in the continuous emulsion liquid membrane process was tested. Several factors affecting the extraction and recovery of Cr were investigated in the fifth objective. In order to screen the factors affecting the extraction efficiency, the design matrix was used in the 2^{6-3} fractional factorial design. Six process parameters which are retention time (1 to 10 minutes), rotational speed (150 to 450 rpm), modifier concentration (1 to 5% (w/v)), treat ratio (1:3 to 1:10), carrier concentration (0.04 to 0.5 M) and stripping agent concentration (0.1 to 1.0 M) were studied in this research to screen the most significant parameters. Then, optimization of the selected parameters from the screening process was proceeded with 3 parameters considered such as treat ratio, rotational speed, and retention times. The optimum conditions were obtained using RSM. Finally, the Cr recovery was investigated. A few parameters have been studied on their effect of recovery process such as acidic thiourea and H_2SO_4 concentrations in the internal phase and external feed phase concentration.

1.5 Significance of Study

Liquid membrane (LM) separation provides a promising method in the extraction of various solutes from aqueous solution. The main advantage of this process compared to conventional processes is the extraction and recovery/enrichment of the solute ion which occured simultaneously in one single stage operation. Furthermore, it has some attractive features such as high efficiency, simple operations, larger interfacial area, reduced operation costs due to less chemicals consumption and selectively extract the solute. In this research, CELM was used to extract Cr from real rinse electroplating wastewater. Cr is used extensively in electroplating and numerous industries due to its stability which helps to protect materials from degradation by the environment. However, as its form can vary, it can exist in its toxic form; thus pose hazard to the environment. Therefore, removal and recovery of Cr from wastewater has become a great concern and significance. Optimized condition of Cr recovery in CELM process may benefit the manufacturing industries due to its simple and cost-effective technology.

1.6 Thesis Outline

This thesis contains 5 chapters, presenting the research in sequential order. Chapter One introduces the brief research background, problem statement, research objectives, and research scopes. Chapter Two provides the detailed reviews on researches related to the Cr process in electroplating and their alternatives in extracting and recovering ELM components and future development of ELM process. Chapter Three described the methodology that was involved in this study. All results and discussions about the findings are presented in Chapter Four. Chapter Five stated the general conclusion and suggestions for future work.

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Appendix A List of Publications

1. **Noah, N. F. M.,** Jusoh, N., Othman, N., Sulaiman, R. N. R., & Parker, N. A. M. K. (2018). Development of stable green emulsion liquid membrane process via liquid–liquid extraction to treat real chromium from rinse electroplating wastewater. Journal of Industrial and Engineering Chemistry, 66, 231-241.

2. **Noah, N. F. M.,** Sulaiman, R. N. R., Othman, N., Jusoh N., Rosly M. B. (2019). Extractive Continuous Extractor for Chromium Recovery: Chromium (VI) reduction to Chromium (III) in Sustainable Emulsion Liquid Membrane Process Journal of Cleaner Production, Accepted.

3. Othman, N., **Noah, N. F. M.,** Poh, K. W., & Yi, O. Z. (2016). High performance of chromium recovery from aqueous waste solution using mixture of palm-oil in emulsion liquid membrane. Procedia Engineering, 148,765-773.

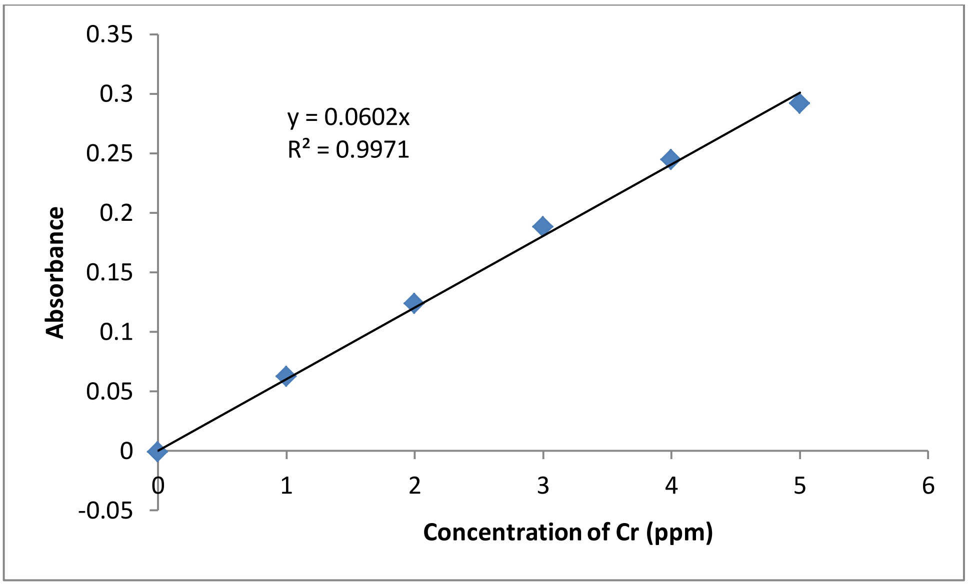
4. **Noah, N. F. M.,** Othman, N., and Jusoh, N. (2018). Emulsion breakage behaviour on chromium (VI) removal using emulsion liquid membrane containing quaternary ammonium compounds. (Malaysian Journal of Fundamental and Applied Sciences) 14(2), 298–302.

5. **Noah, N. F. M.,** Othman, N., and Jusoh, N. (2018). The Use of Factorial Design in Screening of Factors Influencing Hexavalent Chromium Extraction by Continuous Green Emulsion Liquid Membrane (International Conference on Process Engineering and Advanced Materials (ICPEAM2018). 458: 1-13.

6. **Noah, N. F. M.,** Othman, N., 2017. Precious Metals Enrichment from Liquid Waste Solution Using Emulsion Liquid Membrane Process. Journal of Applied Environmental and Biological Sciences. 7(3S): 42-48.

7. Emulsion Liquid Membrane (ELM) And Method of Removing Metal From A Liquid Body/Aqueous Streams Using Said Emulsion Liquid Membrane. PI 2016 001612.

Appendix B Quantitative Analysis Report: Standard of AAS



Wavelength of chromium: 540 nm

Appendix C Result of Experiment for the Liquid Membrane Component Selection

The general equation of extraction and stripping as state in Equations (C.1) and (C.2):

$$\text{Extraction (\%)} = \frac{[Cr]_{i(aq)} - [Cr]_{f(aq)}}{[Cr]_{i(aq)}} x100 \qquad \text{(C.1)}$$

$$\text{Stripping (\%)} = \frac{[Cr]_{fs(aq)}}{[Cr]i_{(org)}} x100, \qquad \text{(C.2)}$$

Where,

$[Cr]_{i(aq)}$ is the initial chromium concentration in aqueous phase (ppm)

$[Cr]_{f(aq)}$ is the chromium concentration in aqueous phase after extraction (ppm)

$[Cr]_{fs(aq)}$ is the chromium concentration in aqueous phase after stripping (ppm) and
$[Cr]_{i(org)}$ is the chromium concentration in the organic phase after extraction (ppm)

Table C1 Extraction of chromium using different types of carrier from rinse electroplating wastewater (Experimental conditions: [Carrier] = 0.1 M, [Cr] = 38.35 ppm, Aqueous : Organic = 10 mL : 10 mL, Agitation speed = 320 rpm, Extraction time = 18 hrs, T = 25±1 °C, Diluent = Palm oil)

Types	**Carrier**	**$[Cr]_{initial}$ (ppm)**	**$[Cr]_{final}$ (ppm)**	**% Extraction**
Acidic	D2EHPA	38.35	26.59	31
Acidic	Cyanex 302	38.35	20.34	47
Acidic	Cyanex 272	38.35	28.86	25
Basic	TOMAC	38.35	0.058	100
Basic	TOA	38.35	37.59	2
Basic	TDA	38.35	33.57	12
Solvating	TOPO	38.35	27.87	27
Solvating	TBP	38.35	37.67	2

Table C2 Effect of carrier concentration in chromium extraction (Experimental conditions: [Cr] = 38.35 ppm, Aqueous : Organic = 10 mL : 10 mL, Agitation speed = 320 rpm, Extraction time = 18 hrs, T = 25±1 ºC, Diluent = Palm oil)

Concentration TOMAC (M)	**[Cr]initial (ppm)**	**[Cr]final (ppm)**	**% Extraction**
0.1	38.35	0.3733	99
0.05	38.35	0.3453	99
0.04	38.35	0.2146	99
0.03	38.35	1.873	95
0.02	38.35	18.81	51
0.01	38.35	34.1	11
0.005	38.35	37.95	1
0	38.35	37.08	0

Table C3 Loq D and Log [TOMAC] for TOMAC concentration

Concentration TOMAC (M)	**$[Cr]_{initial}$ (ppm)**	**$[Cr]_{final}$ (ppm)**	**D =** $\frac{[Cr]initial - [Cr]final}{[Cr]final}$	**Log D**	**Log [TOMAC]**
0.05	38.35	0.35	99	1.99	-1.30
0.04	38.35	0.21	99	1.99	-1.40
0.03	38.35	1.87	19	1.28	-1.52
0.01	38.35	34.10	0.12	-0.91	-2
0.005	38.35	37.95	0.01	-1.99	-2.30

Table C4 Screening process using different types of stripping agent for extraction of chromium from aqueous solution (Experimental conditions: [Stripping agent] = 0.1 M, [Cr] = 38.35 ppm, Aqueous : Organic = 10 mL : 10 mL, Agitation speed = 320 rpm, Extraction time = 18 hrs, T = 25±1 °C, Diluent = Palm oil)

Types	**Stripping agent**	**[Cr]mi**	**[Cr]mf**	**[Cr]s**	**% Stripping**
Basic	NaOH	38.04	22.17	27.14	71
Basic	Na_2CO_3	38.04	26.71	11.33	30
Basic	NaCL	38.04	36.23	1.809	5
Basic	$(NH_4)_2CO_3$	38.04	37.07	0.9746	3
Acidic	HCl	38.04	37.42	0.6232	2
Acidic	H_2SO_4	38.04	28.04	10	26
Acidic	Thiourea in H_2SO_4	38.04	4.54	33.5	88
Chelating	Thiourea	38.04	38.04	0	0

Table C5 Screening process using different concentration of acidic thiourea for extraction of chromium from aqueous solution (Experimental conditions: [Cr] = 38 ppm, Aqueous : Organic = 10 mL : 10 mL, Aagitation speed = 320 rpm, Extraction time = 18 hrs, T = 25±1 °C, Diluent = Palm oil, [H_2SO_4] = 0.1 M)

Thiourea (M)	**[Cr]mi (ppm)**	**[Cr]mf (ppm)**	**[Cr]s**	**% Extraction**
0.01	38.04	36.88	1.16	100
0.03	38.04	22.50	15.54	99
0.05	38.04	21.56	16.48	99
0.1	38.04	4.54	33.5	99
0.15	38.04	13.26	24.78	95
0.2	38.04	21.56	16.48	51

Table C6 Loq D and Log [Thiourea] for thiourea concentration

Thiourea concentration (M)	**[Cr]mi (ppm)**	**[Cr]mf (ppm)**	**[Cr]s (ppm)**	**D =** $\frac{[Cr]initial - [Cr]final}{[\mathrm{Cr}]\mathrm{final}}$	**Log D**	**Log [Thiourea]**
0.01	38.04	36.88	1.16	0.03	-1.50	-2
0.03	38.04	22.50	15.54	0.69	-0.16	-1.52
0.1	38.04	4.54	33.5	7.38	0.87	-1

Appendix D Result of Experiment for the Stability Study

Table D1 Volume of internal phase broken at different observation period using different homogenizer speed (Experimental result: [TOMAC] = 0.004 M, [Acidic Thiourea] = 0.1 M, [1-Octanol] = 5% (w/v), [Span 80] = 3% (w/v), Emulsifying time = 3 minutes)

	Times to broke, minutes		
Homogenizer speed (x1000 rpm)	**1**	**10**	**60**
5	42	50	50
6	10	20	20
6.5	10	10	40
7	0	0	20

Table D2 Effect of homogenizer speed on W/O emulsion viscosity (Experimental conditions: [TOMAC] = 0.004 M, [Acidic Thiourea] = 0.1 M, [1-Octanol] = 5% (w/v), Aqueous : Organic = 5 mL : 5 mL, Span 80 concentration = 3% (w/v), Emulsifying time = 3 minutes and T = 25±1 ºC)

Homogenizer Speed	**Average Droplet Size (µm)**	**Viscosity (cP)**
5000	25.88	138
6000	15.05	139
6500	7.38	142
7000	4.64	158

Table D3 Volume of internal phase broken at different observation period using different surfactant concentration (Experimental result: [TOMAC] = 0.004 M, [Acidic Thiourea] = 0.1 M, Emulsifying time = 3 minutes, [1-Octanol] = 5% (w/v), and T = 25±1 ºC).

Span 80 (M)	**Times to broke, minutes**		
	10	**60**	**120**
1	50	50	50
2	50	50	50
3	0	20	45
4	0	10	30
5	0	0	3

Table D4 Effect of Span 80 concentration on liquid membrane and W/O emulsion viscosity (Experimental conditions: [TOMAC] = 0.004 M, [Acidic Thiourea] = 0.1 M, [1-Octanol] = 5% (w/v), Aqueous : Organic = 5 mL : 5 mL, Emulsifying time = 3 minutes and T = 25±1 ºC).

Span 80 concentration (% (w/v))	**LM viscosity (cP)**	**W/O emulsion viscosity (cP)**
1	84.0	134.5
2	85.5	140.7
3	87.2	154.1
4	87.2	156.0
5	87.6	162.1
6	88.2	781.4

Table D5 Volume of internal phase broken at different observation period using different emulsifying time ([TOMAC] = 0.004 M, [Acidic Thiourea] = 0.1 M, [Span 80] = 5% (w/v), Homogenizer Speed = 7000 rpm, [1-Octanol] = 5% (w/v) and T = 25±1 ºC).

Emulsifying time (min)	**Times to broke, minutes**		
	10	**60**	**120**
1	0	0	0
2	0	2	10
3	0	1	12
4	0	20	50

Table D6 Effect of emulsifying time on W/O emulsion viscosity (Experimental conditions: [TOMAC] = 0.04 M, [Acidic Thiourea] = 0.1 M, [1-Octanol] = 5% (w/v), Aqueous : Organic = 5 mL : 5 mL, Span 80 concentration = 5% (w/v)).

Emulsifying time (min)	**Average Droplet Size (µm)**	**Emulsion viscosity (cP)**
1	3.09	100.4
2	3.16	124.3
3	3.22	154.5
4	4.39	162.8

Table D7 Favorable condition from primary emulsion stability study

Parameter	**Best condition**
Homogenizer speed	7000 rpm
Emulsifying time	3 min
Surfactant concentration	5% (w/v) Span 80

Table D8 Effect of agitation speed on W/O/W and emulsion stability in Cr extraction (Experimental conditions: [TOMAC] = 0.004 M, [Acidic thiourea] = 0.1 M, Aqueous : Organic = 5 mL : 5 mL, Treat ratio = 1:3, Span 80 concentration = 5% (w/v), Emulsifying time = 1 minute, Homogenizer speed = 7000 rpm, Contact time = 3 minutes, and Initial pH of waste = 3).

Agitation Speed (rpm)	**Breakage (%)**
150	40
250	20
350	10
450	20

Table D9 Effect of contact time on W/O/W and emulsion stability in chromium extraction (Experimental conditions: [TOMAC] = 0.004 M, [Acidic thiourea] = 0.1 M, Aqueous : Organic = 5 mL : 5 mL, Treat ratio = 1:3, Span 80 concentration = 5% (w/v), Emulsifying time = 1 minute, Homogenizer speed = 7000 rpm, Agitator speed : 350 rpm, and Initial pH of waste = 3).

Contact Time (min)	**Breakage (%)**
1	20
3	7
5	18
7	30

Table D10 Effect of treat ratio on W/O/W emulsion stability in chromium extraction (Experimental conditions: [TOMAC] = 0.004 M, [Acidic thiourea] = 0.1 M, Aqueous : Organic = 5 mL : 5 mL, Agitation speed = 350 rpm, Span 80 concentration = 5% (w/v), Emulsifying time = 1 minute, Homogenizer speed = 7000 rpm, Contact time = 3 minutes, and Initial pH of waste = 3).

Treat ratio (Emulsion : External phase)	**Breakage (%)**
1:2	40
1:3	8
1:5	8
1:7	18

Table D11 Effect of pH external phase on W/O/W emulsion stability in chromium extraction (Experimental conditions: [TOMAC] = 0.004 M, [Acidic thiourea] = 0.1 M, Aqueous : Organic = 5 mL : 5 mL, Agitation speed = 3, Span 80 concentration = 5% (w/v), Emulsifying time = 1 minute, Homogenizer speed = 7000 rpm, Contact time = 3 minutes, Treat ratio = 1:4, and Initial pH of waste = 3).

pH Electroplating wastewater	**Breakage (%)**
1	8
3	8
5	9
7	15
9	20

Table D12 Favourable conditions for W/O/W emulsion stability toward chromium extraction

Parameter	Condition
Agitation speed (rpm)	350
Contact time (minutes)	3
Treat ratio (emulsion : external phase)	1:5
pH external phase	3

Appendix E Prospect of Continuous Emulsion Liquid Membrane (CELM) Process

Table E1 The liquid height-to-diameter ratio (H/D ratio) of the stirred tank reactor at different volumes tank

Tank volume (mL)	H	H/D ratio
750	6.63	0.55
1000	8.84	0.74
1250	11.05	0.92
1500	13.26	1.11

*Diameter = 12 cm

Table E2 Effect of total extractor volume on emulsion stability (Experimental conditions: [TOMAC] = 0.022 M, [Acidic thiourea] = 0.1 M, Aqueous : Organic= 1 : 1, Agitation speed= 350 rpm, Span 80 concentration = 5% (w/v), Emulsifying time = 1 minute, Homogenizer speed= 7000 rpm, Retention time = 3 minutes, and Initial pH of waste = 3).

Time	Sample Total volume (mL)	Initial Emulsion (mL)	Final Emulsion (mL)	ph after	% breakage
5	750	125	134	2.02	-7.2
10			167	1.98	-33.6
15			166	1.98	-32.8
20			165	2.98	-32
5	1000	167	172	2.37	-3.20
10			169	2.4	-1.40
15			175	2.27	-5.00
20			174	3.27	-4.40
5	1250	208	242	2.37	-20.16
10			276	2.4	-36.48
15			280	2.27	-38.4
20			277	3.27	-36.96
5	1500	250	300	2.2	-20
10			340	2.14	-36
15			341	1.98	-36.4
20			345	2.98	-38

Table E3 Design Matrix for 2^{6-3} fractional factorial design and chromium extraction performance

Std Order	Run Order	Blocks	Variables						% Extraction
			X_1	X_2	X_3	X_4	X_5	X_6	
4	1	1	5	0.040	0.1	450	0.10	1	97.84
2	2	1	5	0.004	0.1	150	0.10	5	91.05
7	3	1	1	0.040	1.0	150	0.10	5	81.16
1	4	1	1	0.004	0.1	450	0.25	5	100.00
3	5	1	1	0.040	0.1	150	0.25	1	94.05
5	6	1	1	0.004	1.0	450	0.10	1	94.63
8	7	1	5	0.040	1.0	450	0.25	5	100.00
6	8	1	5	0.004	1.0	150	0.25	1	100.00

X_1 : t(min), X_2 : [TOMAC] (M), X_3 : [Tu Acidic], X_4 : Rotational speed (rpm), X_5 : treat ratio (Emulsion : Feed), and X_6 : [1-Octanol] (% (w/v)). All variables are in uncoded units.

Table E4 Experimental validation (Experimental conditions: Emulsifying time = 1 minute, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, [Acidic thiourea] = 0.55 M, Aqueous : Organic = 1 : 1, Rotational speed = 300 rpm, Homogenizer speed = 7000 rpm, Retention time = 3 minutes).

Extraction time (s)	[Cr]i	[Cr]f	% Extraction
0	41.81	41.8	0
1	41.81	14.565	65.17
2	41.81	8.378	79.96
3	41.81	0.975	97.67
5	41.81	0.308	99.26
10	41.81	0.271	99.35

Table E5 Best Stability Conditions for Chromium Extraction by ELM

Factors	Symbols	Screened value	Chromium extraction (%)		Error (%)
			Observed value	Predicted value	
Retention time	X_1	3	99	95	4
TOMAC concentration	X_2	0.022			
Acidic Thiourea	X_3	0.55			
Rotational speed	X_4	300			
Treat ratio	X_5	0.175			
1-Octanol	X_6	3			

Table E6 Box-Behnken design (BBD) matrix together with experimental and predicted results of chromium removal percentage

Run	**X_5 Treat ratio**		**X_4 Rotational speed (RPM)**		**X_1 Retention Time, min**		**Removal (%)**	
							Experimental	**Predicted**
1	-1	0.142	-1	300	0	3	99.47	99.82
2	+1	0.142	-1	300	0	3	82.11	82.11
3	-1	0.142	+1	450	0	5	100.00	100.00
4	+1	0.142	+1	450	0	1	96.58	96.58
5	-1	0.100	0	300	+1	5	91.05	91.05
6	+1	0.250	0	300	+1	5	100.00	100.00
7	-1	0.250	0	450	-1	3	100.00	100.00
8	+1	0.100	0	300	-1	1	94.47	94.47
9	0	0.142	-1	300	+1	3	91.58	91.58
10	0	0.142	+1	150	+1	5	100.00	100.00
11	0	0.100	-1	450	-1	3	100.00	99.82
12	0	0.142	+1	150	-1	1	100.00	100.00
13	0	0.250	0	300	0	1	100.00	100.00
14	0	0.250	0	150	0	3	92.11	92.11
15	0	0.100	0	150	0	3	100.00	99.82

Table E7 Verification of optimized data for chromium extraction study

Optimum condition		**Chromium extraction (%)**		**Error (%)**
		Observed value	**Predicted value**	
Rotational speed	342 rpm	99	100	1
Retention Time	170 s			
Treat Ratio	1:5			

Table E8 Experimental validation (Experimental conditions: Emulsifying time = 1 minute, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, [Acidic thiourea] = 0.55 M, Aqueous : Organic = 1 : 1, Rotational speed = 342 rpm, Homogenizer speed = 7000 rpm, Retention time = 170 s).

Extraction time (s)	**[Cr]i**	**[Cr]f**	**% Extraction**
0	41.81	41.8	0
1	41.81	10.44	75.04
2	41.81	6.498	84.46
3	41.81	0.922	97.79
5	41.81	0.429	98.97
10	41.81	0.421	98.99

Appendix F Chromium Recovery Performance

Table F1 Effect of H_2SO_4 acid concentration on chromium extraction (Experimental conditions: Emulsifying time = 1 minute, Rotational speed = 342 rpm, Aqueous : Organic = 1 : 1, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, [Thiourea] = 0.55 M, Homogenizer speed = 7000 rpm, Treat ratio = 1:5, Retention time = 170 s)

H_2SO_4 concentration (M)	**Extraction times (min)**						
	0	**1**	**2**	**3**	**5**	**10**	**15**
0.1	0	39.71	50.57	51.90	52.50	51.84	52.02
0.55	0	72.81	90.72	98.11	99.08	99.06	99.08
1	0	73.39	98.11	98.23	99.11	99.08	99.08
2	0	86.56	98.69	98.75	99.37	99.37	99.37
3	0	98.11	98.69	98.63	99.34	99.37	99.37

Table F2 Effect of H_2SO_4 acid concentration on chromium recovery (Experimental conditions: Emulsifying time = 1 minute, Rotational speed = 342 rpm, Aqueous : Organic = 1 : 1, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, Homogenizer speed = 7000 rpm, [Thiourea] = 0.55 M, Treat ratio = 1:5, Retention time = 170 s)

H_2SO_4 concentration (M)	**Extraction times (min)**						
	0	**1**	**2**	**3**	**5**	**10**	**15**
0.1	0	23.38	23.60	26.38	27.77	27.56	27.81
0.55	0	28.99	34.12	34.23	33.77	33.77	34.00
1	0	33.18	36.53	45.85	43.98	44.90	43.98
2	0	38.52	41.41	45.55	49.38	49.38	49.38
3	0	35.31	35.23	37.83	29.54	29.16	29.16

Table F3 Effect of thiourea concentration on chromium extraction (Experimental conditions: Emulsifying time = 1 minute, Rotational speed = 342 rpm, Aqueous : Organic = 1:1, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, Homogenizer speed = 7000 rpm, Treat ratio = 1:5, Retention time = 170 s, [H_2SO_4] = 2.0 M)

Thiourea concentration (M)	**Extraction times (min)**						
	0	**1**	**2**	**3**	**5**	**10**	**15**
0.1	0	39.71	91.64	98.63	98.63	98.63	98.75
0.55	0	72.81	90.72	98.11	99.08	99.06	99.08
1	0	98.69	98.23	98.11	99.08	99.14	99.14
2	0	73.16	98.11	99.08	99.07	99.08	99.08
3	0	79.57	87.60	90.07	91.46	92.90	90.88

Table F4 Effect of thiourea concentration on chromium recovery (Experimental conditions: Emulsifying time = 1 minute, Rotational speed = 342 rpm, Aqueous : Organic = 1 : 1, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, Homogenizer speed = 7000 rpm, Treat ratio = 1:5, Retention time = 170 s, [H_2SO_4] = 2.0 M)

Thiourea concentration (M)	**Extraction times (min)**						
	0	**1**	**2**	**3**	**5**	**10**	**15**
0.1	0	1.03	8.73	13.07	23.33	27.56	21.27
0.55	0	39.13	38.14	37.39	43.80	44.29	43.92
1	0	32.91	61.15	66.12	82.35	71.61	83.80
2	0	72.47	85.30	82.06	84.43	81.43	86.74
3	0	6.39	10.09	28.81	13.44	13.44	4.26

Table F5 Effect of initial feed concentration on chromium extraction (Experimental conditions: Emulsifying time = 1 minute, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, [Acidic thiourea] = 2.0 M thiourea in 2.0 M H_2SO_4, Aqueous : Organic = 1 : 1, Rotational speed = 342 rpm, Homogenizer speed = 7000 rpm, Treat ratio = 1:5, Retention time = 170 s)

Chromium concentration (M)	**Extraction times (min)**						
	0	**1**	**2**	**3**	**5**	**10**	**15**
20	0	80.02	96.28	99.74	99.74	99.74	99.74
40	0	71.51	90.68	98.62	98.62	98.66	98.62
60	0	63.18	80.40	98.60	98.76	98.76	98.76
80	0	61.95	75.11	98.71	98.71	98.95	98.95
100	0	60.66	74.70	84.65	84.46	85.18	86.81
200	0	28.40	36.84	47.08	49.41	49.99	48.29

Table F6 Effect of initial feed concentration on chromium recovery (Experimental conditions: Emulsifying time = 1 minute, Rotational speed = 342 rpm, Aqueous : Organic = 1 : 1, Span 80 concentration = 5% (w/v), [TOMAC] = 0.022 M, Homogenizer speed = 7000 rpm, [Acidic thiourea] = 2.0 M thiourea in 2.0 M H_2SO_4, Treat ratio = 1:5, Retention time = 170 s)

Chromium concentration (M)	**Extraction times (min)**						
	0	**1**	**2**	**3**	**5**	**10**	**15**
20	0	67.01	97.72	95.54	96.27	95.68	95.54
40	0	69.12	78.71	89.17	76.98	79.00	78.60
60	0	49.76	49.84	51.06	51.54	58.41	60.96
80	0	36.29	41.32	39.63	39.69	44.42	45.87
100	0	28.74	34.51	41.02	37.21	37.28	36.94
200	0	14.26	16.49	18.73	12.53	10.82	6.92

Table F7 Recovery of chromium in internal phase

External/Feed phase (ppm)	**Internal/recovery phase (ppm)**	**Enrichment ratio**
20	191.08	9.55
40	330.11	7.86
60	365.77	6.10
80	366.98	4.59
100	369.41	3.69
200	138.45	0.69

Table F8 CELM performance summary

Parameters	**Properties**
Treat ratio	1:5
Extractor volume	1000 mL
Emulsion Flow rate (mL/min)	~167
External phase Flow rate (mL/min)	~833
Total treated wastewater in 10 minutes (mL)	~8330

Appendix G Sauter Mean Diameter

The Sauter mean diameter of the dispersed phase droplets/globules is defined as follows:

$$\boldsymbol{D_{32}} = \left(\mathbf{3}\sum \boldsymbol{D_p^3}\right)/\left(\mathbf{3}\sum \boldsymbol{D_p^2}\right) \qquad \textbf{(D1)}$$

Where, Dp is the diameter of each droplet. When the emulsion liquid membrane is in the extractor, the size of the emulsion globules in the extractor can be obtained by photography. Then, Sauter mean diameter is calculated by its definition. For determining the size of emulsion droplet of W/O emulsion, the procedures are followed.

Reagents:
Membrane phase: Palm oil as diluents with 0.004 M TOMAC as carrier and 5% (w/v) Span 80 as surfactant
Stripping phase: 0.1 M thiourea in 0.1 M H_2SO_4

The emulsion was made similarly to the previous methods. A Stereomicroscope with colour camera was used to snap emulsion picture with is direct connected to computer. A few repeating step should be done until a clearly emulsion picture observed in the computer monitor. A small scale was also recorded in the microscope focus area. For obtaining more exact result, the caption picture was analyzed with image analyzer. Around 40 of clear droplets were calculated according to Equation D1.

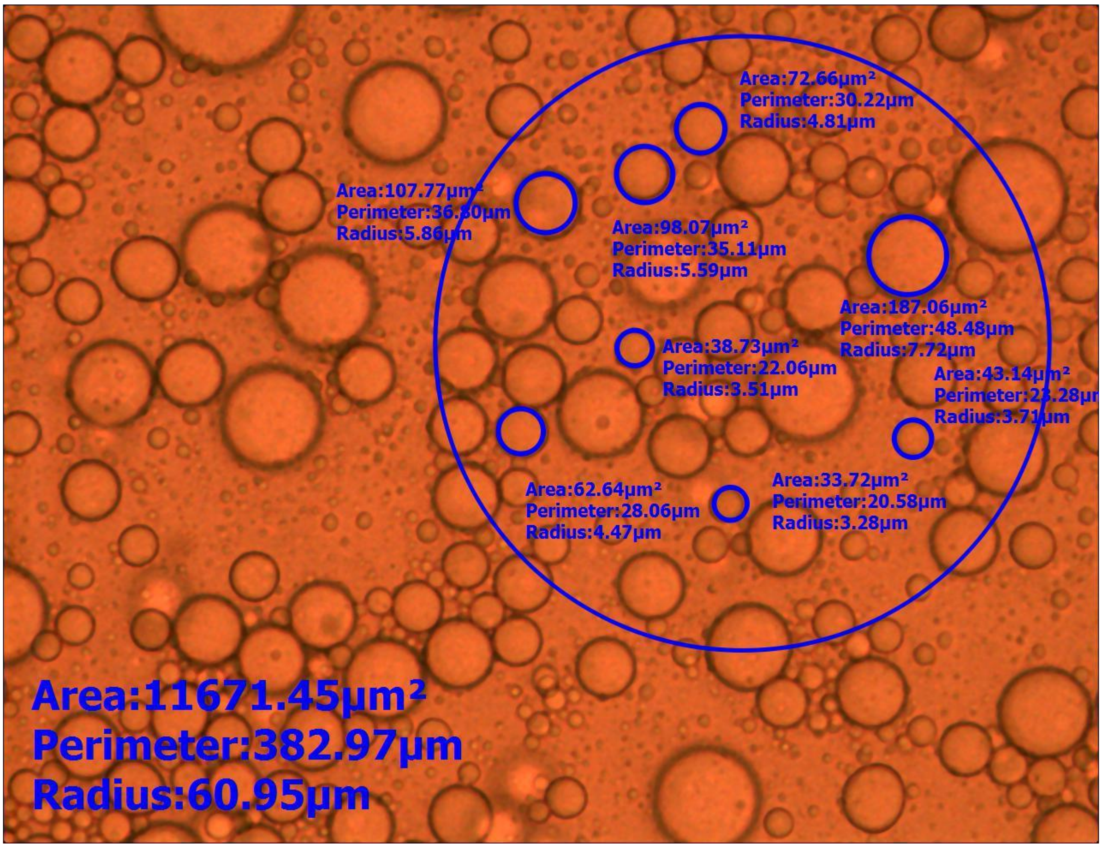


Figure G1 Example of microscope image of the primary emulsion for determination of droplet size.

Table G1 Size of emulsion droplets at different emulsifying time (Experimental conditions: [Cyanex 302] = 0.1 M, [Stripping phase] = 1.0 M thiourea in 1.0 M H_2SO_4, [Span 80] = 5% (w/v), Aqueous : Organic = 5 mL : 5 mL, Homogenizer speed = 12000 rpm, Agitation speed = 250 rpm, Mixing time = 5 minutes, and T = 26 ºC)

Emulsifying time											
1 min			**2 min**			**3 min**			**4 min**		
Diameter	**Dp^3**	**Dp^2**	**Diameter**	**Dp^3**	**Dp^2**	**Diameter**	**Dp^3**	**Dp^2**	**Diameter**	**Dp^3**	**Dp^2**
2.27	11.77	5.17	1.60	4.07	2.55	1.80	5.86	3.25	1.60	4.12	2.57
2.40	13.78	5.75	1.88	6.61	3.52	1.90	6.86	3.61	1.90	6.83	3.60
2.43	14.30	5.89	1.93	7.22	3.74	2.74	20.51	7.49	1.98	7.78	3.93
2.47	15.09	6.11	1.95	7.43	3.81	3.01	27.29	9.06	2.22	10.88	4.91
2.68	19.26	7.19	2.04	8.51	4.17	3.02	27.67	9.15	2.36	13.21	5.59
2.68	19.26	7.19	2.07	8.90	4.29	3.20	32.70	10.23	2.82	22.40	7.95
2.68	19.26	7.19	2.67	19.13	7.15	3.54	44.41	12.54	2.84	22.93	8.07
2.73	20.43	7.47	2.96	25.95	8.77	3.59	46.40	12.91	2.85	23.11	8.11
2.85	23.14	8.12	2.98	26.58	8.91	3.63	47.75	13.16	2.91	24.53	8.44
2.85	23.14	8.12	3.08	29.17	9.48	3.63	47.75	13.16	3.17	31.89	10.06
2.90	24.37	8.41	3.22	33.28	10.35	3.76	53.30	14.16	3.52	43.75	12.42

Table G1 Continued

Emulsifying time											
1 min			**2 min**			**3 min**			**4 min**		
Diameter	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**
2.95	25.64	8.69	3.72	51.64	13.87	3.80	54.66	14.40	3.54	44.41	12.54
3.00	26.92	8.98	3.81	55.11	14.48	3.84	56.79	14.77	3.68	49.79	13.53
3.00	26.92	8.98	2.01	8.13	4.04	3.99	63.36	15.89	3.68	49.79	13.53
3.00	26.92	8.98	2.11	9.36	4.44	1.27	2.03	1.60	3.74	52.41	14.00
3.06	28.55	9.34	2.19	10.56	4.81	1.75	5.40	3.08	3.75	52.55	14.03
3.13	30.55	9.77	2.37	13.39	5.64	1.95	7.43	3.81	4.02	64.85	16.14
3.23	33.63	10.42	2.68	19.26	7.19	2.13	9.65	4.53	4.02	64.85	16.14
3.23	33.63	10.42	2.76	21.07	7.63	2.21	10.80	4.89	4.42	86.06	19.49
3.23	33.63	10.42	2.77	21.37	7.70	2.24	11.29	5.03	4.46	88.54	19.87
3.24	33.98	10.49	2.80	21.93	7.83	2.29	12.01	5.25	4.58	96.12	20.98
3.24	33.98	10.49	2.83	22.56	7.98	2.47	15.09	6.11	4.59	96.97	21.11
3.39	38.98	11.50	2.85	23.09	8.11	2.56	16.87	6.58	4.73	105.65	22.35

Table G1 Continued

Emulsifying time											
1 minute			**2 minutes**			**3 minutes**			**4 minutes**		
Diameter	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**	**Diameter**
3.39	38.98	11.50	3.22	33.28	10.35	2.66	18.79	7.07	4.79	110.09	22.97
3.41	39.71	11.64	3.72	51.64	13.87	2.76	21.02	7.62	4.98	123.74	24.83
3.49	42.69	12.21	3.81	55.11	14.48	3.20	32.70	10.23	5.64	179.20	31.79
3.58	45.74	12.79	4.01	64.55	16.09	3.54	44.41	12.54	5.68	183.42	32.28
3.60	46.51	12.93	4.35	82.50	18.95	3.59	46.40	12.91	5.99	214.94	35.88
Sauter mean diameter = 3.09			Sauter mean diameter = 3.17			Sauter mean diameter = 3.22			Sauter mean diameter = 4.39		