

Liquid membrane extraction of 2-chlorophenol from aqueous solution

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Abstract

Experiments on liquid membrane extraction of 2-chlorophenol were conducted to examine the extraction efficiencies under various operating conditions. Emphasis was placed on identifying the optimal conditions of operating variables in the extraction process and in the demulsification of exhausted water-in-oil (W/O) emulsion. Also considered was multi-stage process in lieu of single-stage operation for efficiency enhancement. An operating model was established for the former by coupling the material balance equations with extraction equilibrium correlation. Experiments were performed to verify the formulated model and to prove the advantages of the multi-stage process. © 1999 Published by Elsevier Science B.V. All rights reserved.

Keywords: Liquid membrane; 2-Chlorophenol; W/O emulsion; Multi-stage extraction; Demulsification

1. Introduction

Phenol and its derivatives are among the most important raw materials used in the manufacturing of various commercial products. Hence, those organic compounds appear very frequently in wastewaters from many heavy chemical, petrochemical and oil refining industries. Because of their toxicity to human and marine life, increasingly stringent restrictions have been imposed on the phenol concentration in the wastewater for safe discharge [1]. In Taiwan, for instance, the phenol concentration in the industrial wastewater for safe discharge has been reduced from 2 to 1 mg/l due primarily to health consideration. Therefore, treatment of industrial wastewater containing phenolic

Abbreviations: *a*, *b* Isotherm parameters of the Langmuir model; *C_e* Equilibrium 2-CP concentration in the wastewater; *L_i* Amount of emulsion added to the *i*-th extraction stage; *Q_e* Equilibrium extraction capacity of the W/O emulsion; *V_i* Amount of wastewater entering the (*i* + 1)-th extraction stage; *X_i* 2-CP concentration in the solution entering the (*i* + 1)-th extraction stage; *Y_i* 2-CP concentration in the W/O emulsion added to *i*-th extraction stage

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compounds has become an integral part in the wastewater treatment system of various chemical industries.

Activated sludge process has been the most widely used traditional method to deal with phenolic wastewater because of its simplicity and relatively low cost [2]. However, the microorganisms in an activated sludge system, even well acclimated, can only deal with chemical wastewater containing relatively low phenol concentration, usually less than 100 mg/l due to low biodegradability and inhibitory effects of those compounds [3]. Unfortunately, chemical wastewaters from many heavy chemical, petrochemical and oil refining industries contain phenolic compounds far exceeding this concentration level. Hence, treatment of high concentration phenolic wastewaters by efficient chemical or physical alternatives is needed.

Many chemical or physical methods have been employed for handling the industrial wastewaters containing high concentration phenolic compounds [4–11]. They included extraction by liquid membrane [4], adsorption by activated carbon [5], macroporous resin [6] and organoclays [7], chemical decomposition by Fenton's reagent [8], wet air oxidation [9,10] and ozonation [11]. Among these methods, extraction by emulsion liquid membrane has been generally regarded as a good attractive alternative because of its high efficiency and recovery of phenolic compounds for reuse as raw materials [12].

Since its discovery by Li [13], liquid membrane has been receiving considerable attention as effective tools for an increasing variety of separation [4,14,15]. Research has been done on various practical and theoretical facets of liquid membrane extraction of organic compounds and heavy metals from aqueous solution [4,14,15]. In a liquid membrane extraction process, many operating variables are involved. Proper selection of those operating variables to achieve optimal system performance is not an easy task. Furthermore, liquid membrane extraction, like other extraction processes, seldom achieve its maximum extraction efficiency in a single stage [16]. Very often, multi-stage operation is necessary. These two aspects have received very little attention in the previous investigations. The objective of this study is first to identify the optimal operating conditions of 2-chlorophenol (2-CP) extraction and the demulsification of exhausted water-in-oil (W/O) emulsion. The model pollutant, 2-chlorophenol, is an important chemical widely used as a solvent or in the syntheses of pharmaceutical, dye and other organic products. Second, a multi-stage liquid membrane extraction process is examined in the present study instead of single-stage operation. A theoretical model was formulated by combining stage-wise material balances with empirical extraction equilibrium correlation for both single- and multi-stage extraction processes. Experiments were then performed to prove that significant improvement in extraction efficiency can be achieved by the multi-stage process in comparison with that of single-stage operation.

2. Experimental studies

The main constituents for the preparation of water-in-oil (W/O) emulsion consisted of: (1) surfactant Span-80 (sorbitan monooleate), obtained from Aldrich, WI, USA; (2) kerosene, obtained from China Petroleum, Taiwan; and (3) sodium hydroxide (NaOH).

The model pollutant, 2-chlorophenol (2-CP), was obtained from Fu Chang Chemicals, Taiwan. According to the manufacturer, Span-80 had a relatively low HLB (hydrophile–lipophile balance) value of 4.3 which renders it appropriate for hydrophobic W/O emulsion preparation. The W/O emulsion was prepared by putting appropriate amounts (feed mixture) of Span-80, kerosene and NaOH in a 1-l beaker, as shown on the top of Fig. 1. A high-speed homogenizer was turned on and maintained at a constant speed of 4000 rpm for 20 min. After homogenizing mixing was completed, the W/O emulsion was left still for 30 min to insure emulsion stability.

The batch extraction tests were conducted in an extraction cell (the bottom graph of Fig. 1). The pyrex extraction cell had a dimension of 6.5 cm ID and 13.3 cm high. It was equipped with a cooling jacket for temperature control. The extraction cell had four pieces of baffles attached to the cell wall, 90° apart from each other. A low-speed stirrer was provided so that the W/O emulsion and the 2-CP wastewater could be kept well mixed. The W/O emulsion was expected to have a wide range of globule sizes [4,14,15]. The average globule size was estimated graphically from enlarged photos to be approximately 2 mm in this study, confirming those reported by the previous investigators [4]. An extraction run was started by putting a desired amount of 2-CP wastewater (with 1000 mg/l initial 2-CP concentration) in the extraction cell and the system was kept at a constant temperature (25°C). The stirrer was turned on and maintained at a constant speed of 100 rpm. A proper amount of prepared W/O emulsion was added to the 2-CP wastewater. Small samples were then taken periodically and the 2-CP concentration was determined using a HP gas chromatograph (Model HP 5890 series II, Hewlett Packard Instrument, CO, USA) equipped with a FID detector and a GP80/100 Carboxpack capillary column.

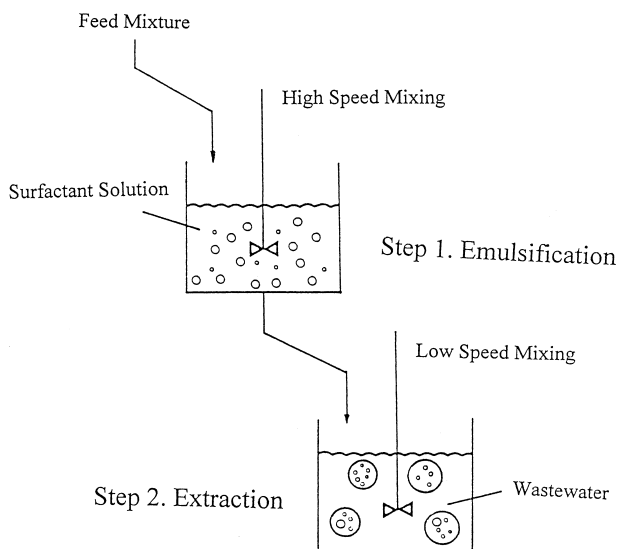


Fig. 1. Experimental schematic.

As would be elaborated in detail later, an liquid membrane extraction usually lasted no more than 20 min because of high extraction efficiency. At the end of an extraction step, mixing was stopped and the mixture of W/O emulsion and wastewater rapidly separated into two phases with the W/O emulsion on the top and the wastewater in the bottom. The treated wastewater contained only very little amount of 2-CP at this stage because of the large majority of 2-CP being removed by the W/O emulsion. The W/O emulsion was easily decanted due to immiscibility between the two phases. In order to recover sodium 2-chlorophenolate, the W/O emulsion was put in a demulsification device, as shown in Fig. 2. The device consisted of a high-voltage AC power supply (designed and manufactured by Chang Yao Electric, Taipei, Taiwan) with adjustable output voltage between 0 and 30 kV and current between 20 and 200 mA. The demulsification container was a glass cylinder of 6 cm ID and 9.5 cm high. The inner electrode was a pyrex cylinder of 5.7 OD and 9.5 cm high. Located at the bottom of the pyrex cylinder was a circular SS-316 electrode disc which was covered with 200 ml silicone oil for isolation. A similar circular SS-316 electrode disc of 6 cm in diameter was attached to the bottom of the outside glass cylinder. A gap of about 2 cm was maintained between the bottom of the inner electrode and the bottom of the outer demulsification container and this gap permitted approximately 50 ml of exhausted W/O emulsion to be demulsified. Hence, for a demulsification run, 50 ml of exhausted W/O emulsion was placed in the demulsification container first. Turn on the power supply and adjust the operating voltage to the desired level to start the demulsification process.

The progress of a demulsification operation could be measured or monitored by the heights of the oil phase (Span-80 and kerosene) on the top and water phase (containing sodium 2-chlorophenolate) in the bottom, as depicted in Fig. 3. At the beginning of a

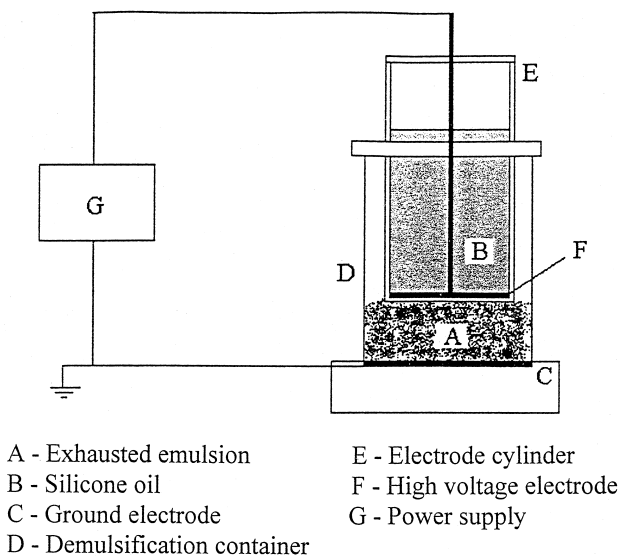


Fig. 2. High voltage demulsification device.

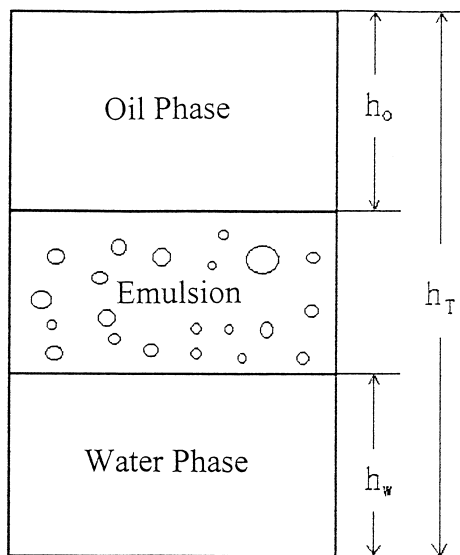


Fig. 3. Schematic of demulsification process.

demulsification run, the gap was filled with opaque exhausted emulsion. After the demulsification started, a clear bottom water phase and a top oil phase became visible. As the demulsification progressed, the thickness of the top oil phase (h_o) and the bottom water phase (h_w) increased while that of the middle opaque phase decreased. The values of h_o and h_w were read with reasonable accuracy through a magnifying glass window outside the demulsification container. Note that the value of h_o was measured from the top while that of h_w measured from the bottom. The thickness of the original opaque exhausted W/O emulsion was represented by h_T which was the gap between the two electrodes. The recovered oil, containing only Span-80 and kerosene, was added with a desired amount of new NaOH solution for preparing a new batch of W/O emulsion for the next experimental run.

3. Results and discussion

3.1. Effects of operating variables on the extraction efficiency

In the extraction test runs, both baffled and non-baffled cells were employed. The effect of baffle on the 2-CP extraction is demonstrated in Fig. 4. Significant difference in the liquid phase 2-CP reduction (C/C_0) for baffled and non-baffled extraction cells is obvious in this figure for the first 20 min of the extraction operation. The improvement in the extraction efficiency for baffled cell is apparently due to better mixing and suspension of W/O emulsion in the wastewater in a baffled extraction cell over that in the non-baffled one. However, after 20 min of extraction, a small decrease in the 2-CP

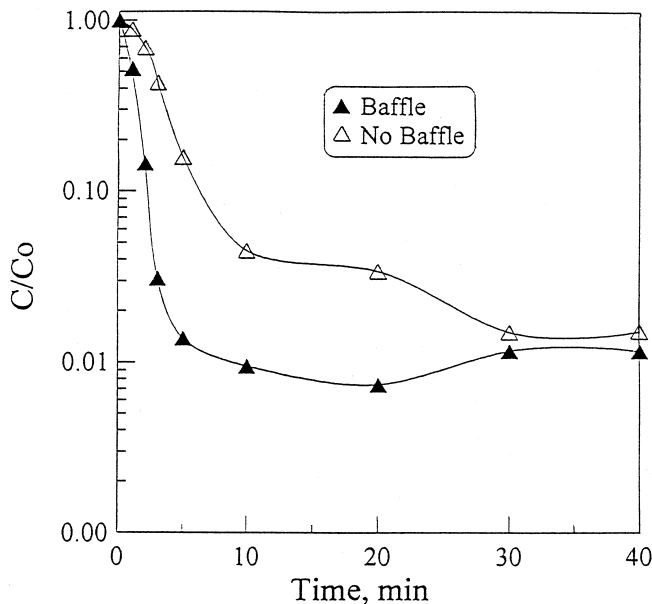


Fig. 4. Effect of baffles on the 2-CP reduction with 1000 mg/l initial 2-CP concentration, 5% Span-80, 1% NaOH, 0.1 V_c/V_w ratio.

reduction was apparent in the baffled case, attributable presumably to breakup of some W/O emulsion globules, and the two extraction efficiencies become essentially the same. This time frame serves as the upper limit of the 2-CP extraction by liquid membrane. Such a short extraction time was possible because of very large surface of the W/O emulsion per unit weight. Hence, for efficient extraction operation, baffled extraction cell is definitely a good choice and was adopted for the rest of all experimental investigations.

The effect of the surfactant (Span-80) on the liquid membrane extraction is twofold. More surfactant in the W/O emulsion tends to reduce the surface tension of the emulsion, leading to smaller emulsion globules and much larger surface area for the same amount of W/O emulsion. This in turn enhances the 2-CP extraction efficiency. However, as more surfactant is added, the thickness of the emulsion globules increases also, resulting in an increase in the 2-CP mass transfer resistance which adversely influences the 2-CP extraction. The combined effect of these two conflicting factors is shown in Fig. 5. As the initial surfactant concentration of W/O emulsion increases from 1% to 5%, the efficiency of 2-CP extraction was steadily improves. The improvement in the extraction efficiency, however, was drastically reversed as the surfactant concentration is further elevated to 11%. In the latter case, the increased mass transfer of 2-CP due to thicker emulsion layer apparently more than offset the beneficial effect of reduced surface tension. Hence, 5% surfactant concentration offers the best choice for the present liquid membrane system. It is further noticed in Fig. 5 that 10 to 15 min

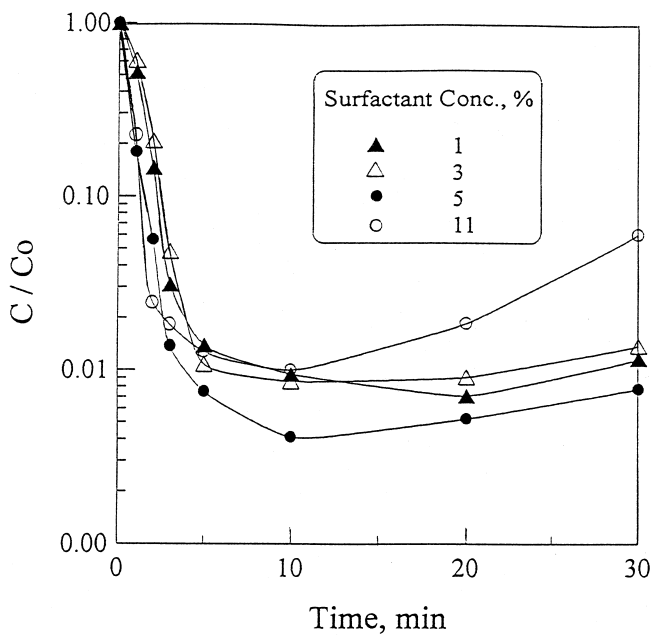


Fig. 5. Effect of surfactant (Span-80) concentration on the 2-CP reduction with 1000 mg/l initial 2-CP concentration, 1% NaOH, 0.1 V_c/V_w ratio.

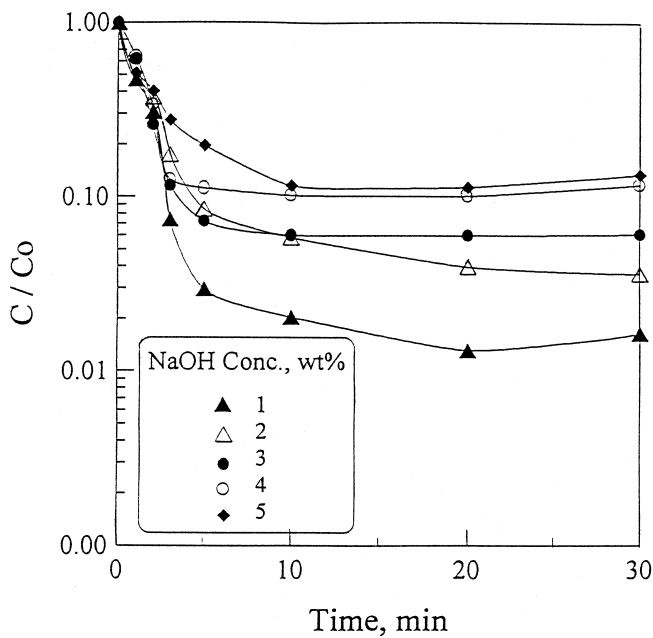


Fig. 6. Effect of NaOH concentration on the 2-CP reduction with 1000 mg/l initial 2-CP concentration, 5% Span-80 and 0.1 V_c/V_w ratio.

appear to be a better time duration for the 2-CP extraction which is in line with that observed in Fig. 4.

NaOH inside the emulsion droplets serves to convert 2-CP, that had diffused through the liquid membrane, into sodium 2-chlorophenolate. The latter compound could not diffuse back to the outside wastewater phase and thus was retained within the emulsion droplets [17,18]. Hence, the NaOH concentration is expected to exert some effect on the 2-CP extraction efficiency. The NaOH effect is revealed in Fig. 6. It is apparent that 1% NaOH concentration yields the best results among all the cases tested. This is due primarily to the fact that 1% NaOH solution offers sufficient amount of NaOH to react with 2-CP to form sodium chlorophenolate. Additional amount of NaOH negatively influences the formation of W/O emulsion and is not conducive to the extraction process.

Another factor that deserves some attention is the relative amounts of W/O emulsion and wastewater to realize the best 2-CP extraction. Fig. 7 shows the 2-CP removal as a function of the volume ratio of W/O emulsion and wastewater containing 1000 mg/l initial 2-CP concentration. As the volume ratio increases, the 2-CP removal increases rapidly until it reaches 0.1 where no further extraction improvement could be realized. It is further noted that by maintaining at this optimum volume ratio, the present liquid membrane extraction can be employed to deal with wastewater of high phenolic concentration. This clearly shows the unique advantages of the liquid membrane extraction method.

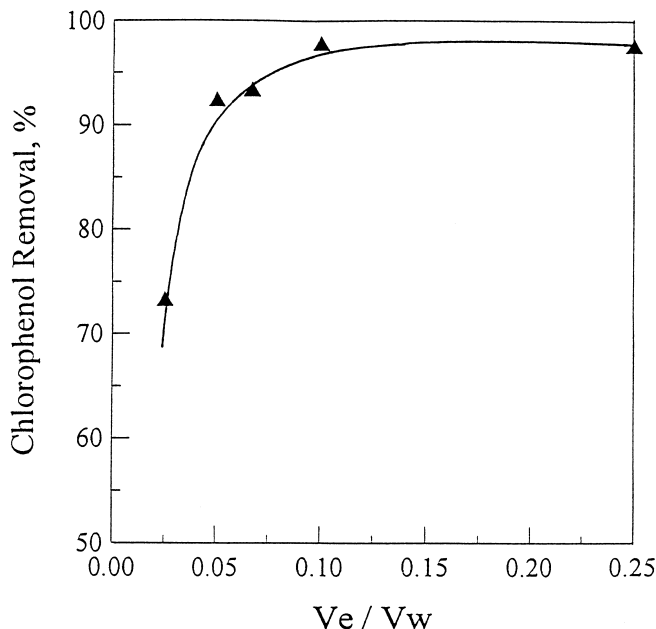


Fig. 7. 2-CP removal as a function of the volume ratio of W/O emulsion and wastewater with 1000 mg/l initial 2-CP concentration, 5% Span-80 and 1% NaOH.

3.2. Multi-stage liquid membrane extraction

Dispersion of W/O emulsion in an extraction cell under well mixed conditions results in suspension of emulsion globules in the phenolic wastewater. This suspension phenomenon is in fact not much different from that of solid adsorbent (such as granular activated carbons, adsorption resins, etc.) in an aqueous wastewater. By visualizing the W/O emulsion globules as adsorbent particles, 2-CP extraction by the present liquid membrane from the wastewater is functionally equivalent to an adsorption process. Hence, an adsorption equilibrium isotherm could be adopted for describing the 2-CP equilibrium established between the external aqueous wastewater and the internal W/O emulsion. The simple monolayer Langmuir isotherm is first adopted for representing the extraction equilibrium. This isotherm is given by [19]

$$Q_e = \frac{abC_e}{1 + bC_e} \quad (1)$$

in which Q_e is the equilibrium extraction capacity of W/O emulsion (mg 2-CP removed per ml emulsion), C_e the equilibrium 2-CP concentration in the external wastewater (mg/l), and a and b the constant model parameters. To determine the two parameters, Eq. (1) is rewritten as

$$\frac{1}{Q_e} = \frac{1}{a} + \frac{1}{abC_e} \quad (2)$$

The equilibrium 2-CP concentration (C_e) in the external phase was measured in a test run and the equilibrium extraction capacity (Q_e) was determined using the measured data and the known initial conditions. According to Eq. (2), a plot of $1/Q_e$ vs. $1/C_e$ would yield a straight line and the model parameters (a and b) were obtained from the slope and intercept. Fig. 8 reveals that the equilibrium Langmuir extraction model, represented by the solid lines, describes reasonably well the equilibrium 2-CP relation in the liquid membrane extraction system. The constant parameters for this extraction system obtained from the model fit are listed in Table 1. This table reveals that the parameter a is strongly influenced by the surfactant concentration while the second parameter b remains constant. The above extraction equilibrium correlation can be conveniently utilized in establishing the following multi-stage material balances of the extraction process.

Table 1
Equilibrium model parameters of 2-CP extraction by liquid membrane

Surfactant concentration (%)	Langmuir parameter	
	a	b
1	30.46	0.06
3	35.90	0.06
5	37.26	0.07
11	29.63	0.06

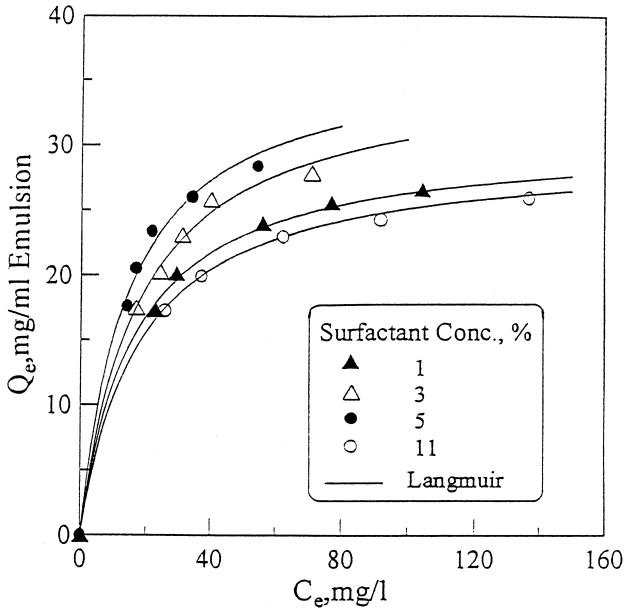


Fig. 8. Effect of surfactant concentration on the equilibrium 2-CP relation with 1000 mg/l initial 2-CP concentration, 1% NaOH and 0.1% V_e/V_w ratio.

A cocurrent k -stage liquid membrane process is shown in Fig. 9 in which V_i and Y_i denote, respectively, the volume and 2-CP concentration of wastewater entering the $(i + 1)$ -th extraction stage and L_i and X_i the corresponding quantities of emulsion entering the same stage. Note that X_i and Y_i are equal to the equilibrium extraction capacity (Q_e) and 2-CP concentration (C_e) in Eq. (1), respectively. Since the emulsion used in each stage of the present cocurrent extraction process was fresh, hence,

$$X_1 = X_2 = \dots = X_0 = 0 \tag{3}$$

It was also assumed that the amount of emulsion added in each stage was equal and

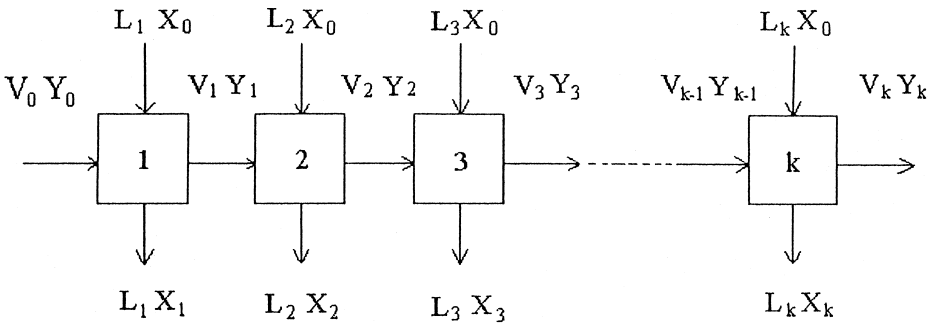


Fig. 9. Multi-stage cocurrent 2-CP extraction by liquid membrane.

there was no loss or gain in the volumes of wastewater and emulsion entering and leaving an extraction stage because of their high immiscibility. Hence,

$$V_1 = V_2 = \dots = V_0 \quad (4)$$

and

$$L_1 = L_2 = \dots = L_0 \quad (5)$$

A material balance of 2-CP for an arbitrary i -th stage yields

$$V_0(Y_{i-1} - Y_i) = L_0(X_i - X_0) \quad (6)$$

which is rearranged to

$$Y_i = (-L_0/V_0)X_i + Y_{i-1} \quad (7)$$

In the above equation, $(-L_0/V_0)$ represents the slope of the operating line for the i -th extraction stage in Fig. 9. X_i and Y_i in the equation are related by Eq. (1)

$$X_i = \frac{abY_i}{1 + bY_i} \quad (8)$$

which is the equilibrium line in Fig. 9. Combining Eqs. (6) and (7) gives

$$Y_i = -\left(\frac{L_0}{V_0}\right)\frac{abY_i}{1 + bY_i} + Y_{i-1} \quad (9)$$

The above equation represents the material balance of liquid membrane extraction for the i -th stage. For a single-stage process, Eq. (8) reduces

$$Y_1 = -\left(\frac{L_0}{V_0}\right)\frac{abY_1}{1 + bY_1} + Y_0 \quad (10)$$

For known L_0 , V_0 , Y_0 and a given set of parameters a and b , Eq. (9) can be solved for Y_1 and the 2-CP removal determined. For the two-stage operation, the total amount of emulsion (L_0) is equally divided for each stage. Hence, the 2-CP concentrations of the wastewaters exiting the first and second stages become

$$Y_1 = -\left(\frac{L_0}{2V_0}\right)\frac{abY_1}{1 + bY_1} + Y_0 \quad (11)$$

and

$$Y_2 = -\left(\frac{L_0}{2V_0}\right)\frac{abY_2}{1 + bY_2} + Y_1 \quad (12)$$

Again, for known L_0 , V_0 , Y_0 and a given set of a and b , Eqs. (10) and (11) can be solved iteratively for Y_1 and Y_2 . The 2-CP removal for each stage and the overall 2-CP removal can then be determined. Material balances can be similarly established for extraction of any number of stage.

To verify the theoretical model of multi-stage liquid membrane extraction formulated above and ascertain its advantage in comparison with that of single-stage process, test

Table 2

Comparison of the extraction efficiencies of single- and two-stage processes

Item	Single-stage	Two-stage	
		Stage 1	Stage 2
Amount of emulsion (ml)	20	10	10
Inlet 2-CP concentration (mg/l)	1000.0	1000.0	526.6
Theoretical 2-CP concentration at the end of that stage (mg/l)	142.1	521.3	76.1
Observed 2-CP concentration at the end of that stage (mg/l)	157.7	526.6	78.5
Observed 2-CP removal (%)	84.2	47.4	44.8
Accumulated 2-CP removal (%)			92.2

Note: 2% NaOH, $V_e/V_w = 1/15$; $V_{oil}/V_{NaOH} = 1/1$.

runs were conducted for both single- and two-stage processes. Table 2 compares the test results for a typical test run with a 1000 mg/l initial 2-CP concentration and 20 ml emulsion solution for single-stage process. The table reveals that the theoretical model predicts well the 2-CP concentration in the wastewater exiting each stage when compared with the measured ones. Also observed in this table is that the 2-CP removal was elevated from 84.2% for the single-stage process to 92.2% for the two-stage process, an 8% improvement over the single-stage removal efficiency. The 2-CP removal improvement for two-stage or even three-stage process was considerably more pronounced as the 2-CP removal for single-stage process is below 80%. In addition to

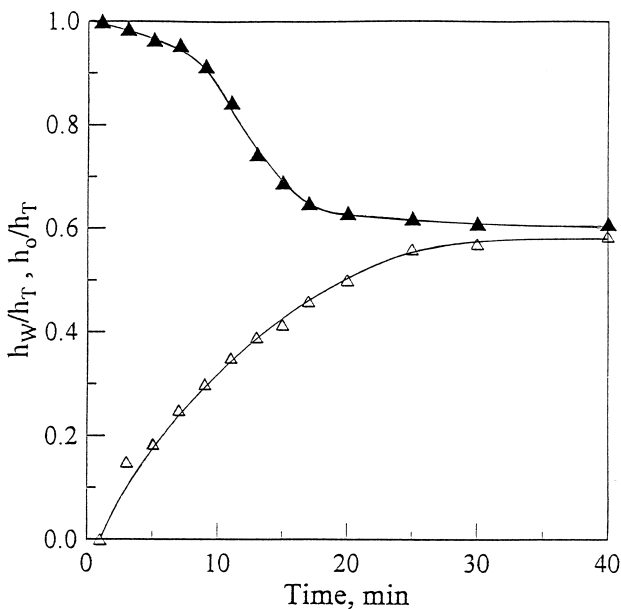


Fig. 10. Progress of demulsification process with 1000 mg/l initial 2-CP concentration, 5% Span-80, 2% NaOH and 0.1 V_e/V_w ratio.

the improvement in extraction efficiency, multi-stage process has another advantage in that the time to reach equilibrium in each stage is significantly shorter than that in single-stage process due to smaller amount of emulsion used in the former. In general, a two-stage process is usually sufficient and can be recommended for practical purposes.

3.3. Demulsification of exhausted W/O emulsion

Demulsification of the exhausted W/O emulsion was performed in an demulsification device, as described in detail earlier in Section 2. The progresses of h_0/h_T and

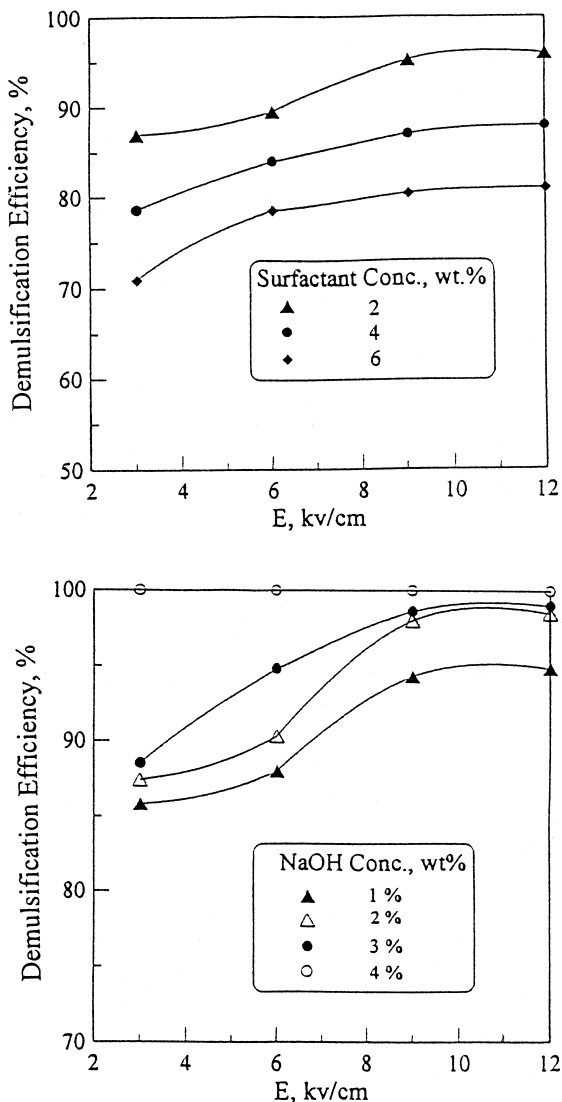


Fig. 11. Effect of voltage on the demulsification performances with 1000 mg/l initial 2-CP concentration, 5% Span-80, 2% NaOH and 0.1 V_c/V_w ratio.

h_w/h_T as a function of time for a typical demulsification case is demonstrated in Fig. 10. The thickness of the middle opaque exhausted emulsion layer (distance between the solid and open triangles) apparently shrank rapidly. The demulsification was seen to approach a steady state after about 30 min with the h_0/h_T and h_w/h_T curves being rather close to each other.

The operating voltage is an important factor affecting the demulsification efficiency of the exhausted emulsion. Such an effect is demonstrated in Fig. 11. The demulsification efficiency (%) at any time was defined as the ratio of the volume of wastewater measured to that originally used for preparing the W/O emulsion. Both the top and bottom graphs in this figure reveals that at least 9 kV/cm voltage was a good choice. It is also seen in this figure that an increase in surfactant concentration in the original W/O emulsion is not conducive to demulsification. However, as the surfactant concentration becomes too low (less than 5%), the liquid membrane extraction efficiency was compromised. Hence, proper balance between the extraction efficiency and the demulsification efficiency is necessary in order to come up with an optimum surfactant concentration. The bottom graph demonstrated that a NaOH concentration above 2% yields very good demulsification efficiency.

A key advantage of the liquid membrane extraction process is the reuse of recovered oil (Span-80 and kerosene) and, hence, experiments on recycling the recovered oil for preparing a new batch of emulsion were performed. The experiments were conducted to test whether there would be any degradation of the recovered oil for reuse. Fig. 12

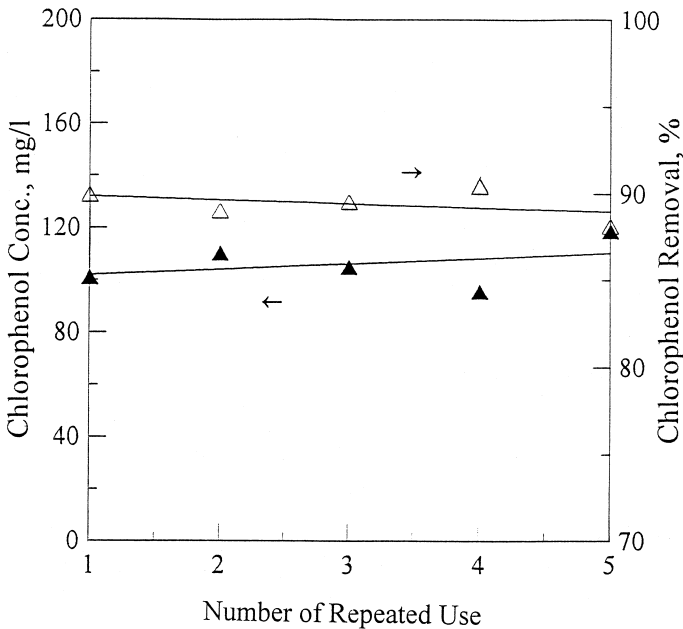


Fig. 12. 2-CP removal vs. the number of repeated use of recovered oil (Span-80 and kerosene) with 1000 mg/l initial 2-CP concentration, 5% Span-80, 2% NaOH and 0.1 V_c/V_w ratio.

displays the 2-CP removal vs. the number of reuse of oil. This figure clearly demonstrates that after 5 reuses of oil, there was little change in the 2-CP removal efficiency, implying that there was essentially no quality degradation of the recovered oil and reuse of those oil is possible.

4. Conclusions

Extraction of 2-chlorophenol (2-CP) from phenolic wastewater by liquid membrane was investigated in this work. Test runs were performed under various operating conditions in order to identify the optimal conditions of those variables. Multi-stage process in lieu of single-stage operation was considered in this study. Demulsification of exhausted emulsion at the end of the extraction process was also experimentally examined. From the results of experimental investigations, the following conclusions can be drawn.

(1) Extraction cell with baffles was found to perform significantly better than that without. Hence, baffled extraction cell is recommended for practical purposes.

(2) Four to five percent surfactant and 1–2% NaOH concentrations in the preparation of W/O emulsion were observed to yield rather good extraction and demulsification results for all the cases tested here. Also an optimal 0.1 volume ratio of W/O emulsion to wastewater was found in the present liquid membrane system.

(3) A theoretical model was established for describing multi-stage process by combining the material balances and the Langmuir-type equilibrium correlation proposed here. The model was experimentally verified and the multi-stage process was proved to be significantly more advantageous than the single-stage operation in terms of extraction efficiency improvement and shorter time to reach extraction equilibrium in each stage. Such advantages are even more pronounced as the single-stage extraction efficiency is lower than 80%.

(4) An optimum operating voltage of 9 kV/cm was observed for the demulsification process of exhausted W/O emulsion. The recovered oil (Span-80 and kerosene) was found to suffer essentially no quality degradation and can be reused.

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