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Recovery of Nitrophenols from Aqueous Solutions by a Liquid Emulsion Membrane System

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Abstract

The application of liquid emulsion membrane technology to the extraction of nitrophenols from aqueous solutions is discussed. The effects of various parameters such as hydrophilic lipophilic balance of the surfactants, speed of agitation, water volume fraction in the water-in-oil emulsion, treatment ratio, and the concentration of surfactant on the extraction of *p*-nitrophenol are studied, and a suitable formulation for a liquid emulsion membrane is developed. The same formulation was used to extract *o*-nitrophenol and 2,4-dinitrophenol, and the results are compared with those of *p*-nitrophenol. The swelling of the emulsion during extraction is minimized by the addition of cyclohexanone to the membrane phase. The demulsification of the emulsion phase after extraction is also carried out by a chemical demulsification technique. Acetone is used for demulsification.

INTRODUCTION

The removal of phenols from aqueous solutions is a major waste treatment problem. Phenols are highly toxic and resistant to biological treatment. Most methods for the removal of phenols involve their destruction.

The liquid emulsion membrane (LEM) extraction process, which efficiently removes and recovers phenol, was developed by Li (*1*) in 1968. Since that time, LEM has been extensively investigated by many scientists. It is a highly selective method of separating organics, inorganics, and metal ions, and it has relatively low energy costs.

Some prominent applications of LEM are:

(a) Removal of phenol from wastewater (*2–4*).
(b) Removal of toxic heavy metals from wastewater (*5–9*).

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(c) Extraction of ammonia from municipal wastewater (*10*).
(d) Extraction of amines and amino acids (*11–13*).
(e) *In-situ* extraction of carboxylic acids from fermentation broth (*14*).
(f) Separation of hydrocarbons (*15–18*).

LEM have been successfully tested on a pilot-plant scale by Marr et al. (*19*) for the extraction of zinc, copper, cadmium, lead, tungsten, and chromium. On a plant scale, LEM has been used to recover zinc from the effluent of viscose fiber industries.

This work presents the application of LEM technology for the extraction of nitrophenols from dilute aqueous solutions. The present investigation attempts to carry out a systematic and detailed study of the effect of various operating parameters on extraction. The major problems posed by this extraction process are breakage of emulsion, swelling of emulsion, and demulsification. The present work has been carried out to study these problems. A suitable membrane phase was developed to extract *p*-nitrophenol. The same formulation was used to extract other nitrophenols, e.g., *o*-nitrophenol and 2,4-dinitrophenol.

MATERIALS

Span-80 and Tween-80 (Fluka grade) were used. Kerosene was obtained from a petroleum refinery, Hindustan Petroleum Corporation Limited. Other chemicals were of analytical reagent grade with a minimum purity of 99.0%.

EXPERIMENTAL

The extraction of *p*-nitrophenol was carried out by using a water-in-oil-in-water (w/o/w) multiple emulsion. It consisted of an external aqueous phase containing *p*-nitrophenol (Phase III), internal droplets of an aqueous NaOH solution (Phase I), and an oil membrane phase (Phase II), as shown in Fig. 1.

The water-in-oil (w/o) emulsion was prepared in the emulsification step (Fig. 1a) by initially dissolving the surfactant (Span-80) in the oil phase (kerosene) and then adding 5 mL NaOH solution to make volume of 50 mL. The emulsification was carried out at 2 ± 0.5°C for 15 min at 3000 rpm. The emulsion globule is shown in Fig. 1(b). Extraction experiments were performed in a vessel of 0.09 m inner diameter equipped with a six-bladed turbine impeller. A *p*-nitrophenol solution (300 mL) was first placed in the vessel. Then the 50 mL of emulsion prepared in the emulsification

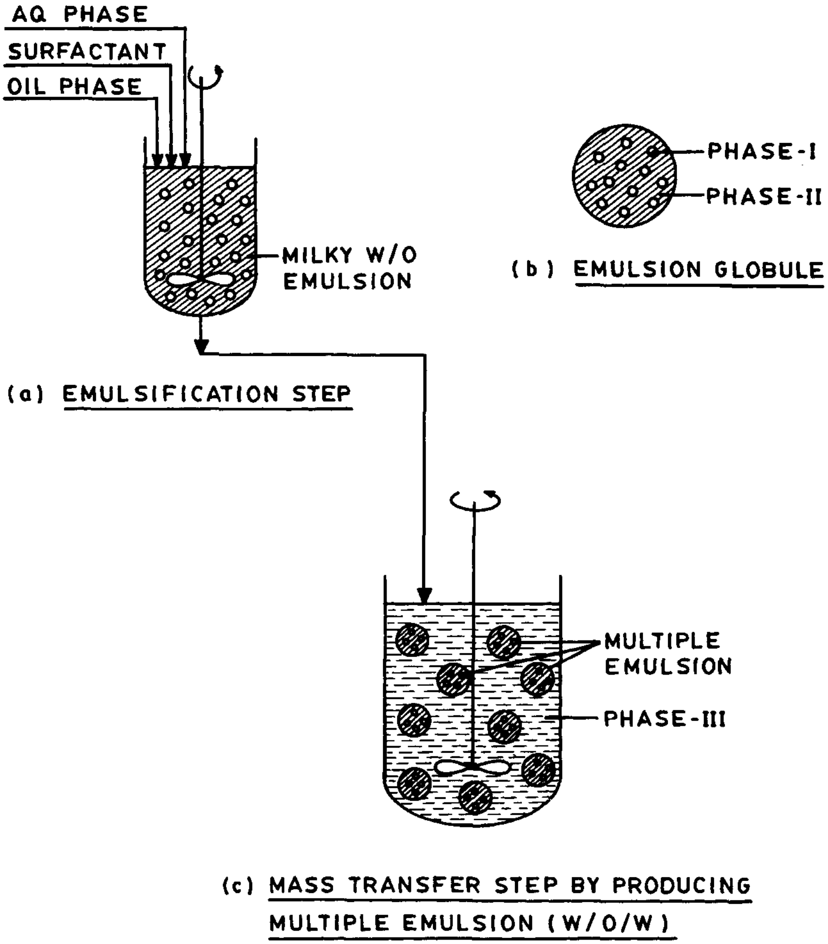


FIG. 1. Systematic representation of liquid emulsion membrane system.

step was dispersed in the solution under stirring at 350 rpm. Figure 1(c) shows the w/o/w multiple emulsion.

Analytical

Samples for analyzing the *p*-nitrophenol concentration in the external phase were periodically removed from the vessel. The samples were made alkaline with NaOH solution to increase their absorbance. The concentration of *p*-nitrophenol in each case was measured as the sodium salt of *p*-

nitrophenol with the help of a UV/VIS Spectrophotometer (Perkin-Elmer Lambda 3B).

Swelling Measurement

The initial and final volumes of the emulsion phase were compared to obtain the swelling percentage.

RESULTS AND DISCUSSION

1. Effect of Hydrophilic Lipophilic Balance (HLB) on Extraction

The HLB of an emulsifier is the balance of the sizes and strengths of the hydrophilic and the lipophilic groups of the emulsifier. This HLB system is generally used in the selection of an emulsifier. The HLB of an emulsifier is related to its solubility and is also used for the preparation of either a water-in-oil (w/o) or an oil-in-water (o/w) emulsion. Thus, an emulsifier having a low HLB will tend to be oil soluble and one having a high HLB will tend to be water soluble. Low HLB (range 4–6) is used for the preparation of a w/o emulsion and high HLB (range 8–18) is used to prepare an o/w emulsion. Experiments were carried out by varying the HLB factor (4.3 to 5.9), and its effect on extraction was studied. Table 1 shows the recipes for the preparation of emulsifiers having four different HLB values. The following emulsion composition was used for the extraction of *p*-nitrophenol (3000 ppm): surfactant (4% v/v) + kerosene (76% v/v) + aqueous phase [20% v/v (0.7 mol/L of NaOH)].

It was observed that as the HLB value of the surfactant increases, the stability of the emulsion decreases. This is because the properties of the blend of surfactants are different from those expected from averaging the properties of the individual emulsifiers involved due to molecular association at the interface, especially when a Span–Tween blend is used (*20*).

Figure 2 shows that the initial rate of extraction increases with an increase in the HLB value of surfactant, but the extent of extraction decreases with an increase in the HLB value of surfactant during an initial period of 10

TABLE 1
Recipe for the Preparation of Emulsifiers with Different HLB Values

Span-80 % × HLB value = A	Tween-80 % × HLB value = B	$A + B$ = HLB
100 × 4.3 = 4.3	0 × 15 = 0	4.3 + 0 = 4.3
95 × 4.3 = 4.085	5 × 15 = 0.75	4.085 + 0.75 = 4.885
90 × 4.3 = 3.87	10 × 15 = 1.5	3.87 + 1.5 = 5.37
85 × 4.3 = 3.655	15 × 15 = 2.25	3.665 + 2.25 = 5.905

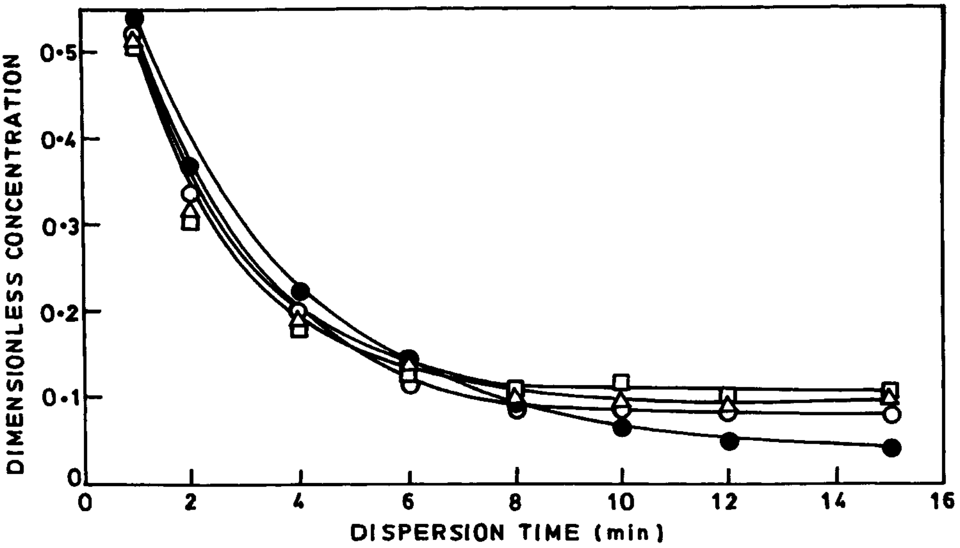


FIG. 2. Effect of HLB on extraction.

HLB	Symbol
4.3	●
4.835	○
5.37	△
5.9	□

min. As the HLB value increases, the water transport also increases because of the hydrophilic nature of the Tween-80 added. Solute is transported along with the water, causing a little higher depletion of the solute in the outer phase. This causes a little higher rate of extraction in the initial period as the HLB value increases. However, as the HLB value of the surfactant increases, the strongly hydrophilic surfactant predominantly dissolves in the water phase and the strongly lipophilic surfactant predominantly dissolves in the oil phase (*21*, *22*), as well as adsorbing at the interface. Hence, there is insufficient interaction between these surfactants to form a strong interfacial film (*23*). Because there is a weak film at high HLB values, spillage of the entrapped solute takes place. Thus, the extent of extraction decreases with an increase in the HLB value of the surfactant.

2. Effect of Speed of Agitation on Extraction

In order to obtain high mass transfer rates, a large membrane surface area is desirable. This may be obtained by increasing the speed of agitation.

At 240 rpm, it was observed that the emulsion globules were not uniformly distributed in the continuous phase, and the majority of globules were found to be in the upper part of the vessel.

During the initial stages of extraction, the following changes were observed. As a result of a steady increase in the mixing speed from 240 to 500 rpm, the emulsion globules are gradually distributed uniformly in the continuous phase. An increase in the speed also leads to a reduction in the size of the globules, thereby increasing the membrane surface area and hence the mass transfer rates (Fig. 3). However, liquid membranes are not strong enough to withstand high shear during intense mixing. This leads to a reduction in liquid membrane efficiency because of emulsion breakage and constant spillage of the internal reagent along with the extracted solute. A very clear indication of this is seen at 415 and 500 rpm when the extent of extraction decreases after dispersion times of 10 and 7 min, respectively.

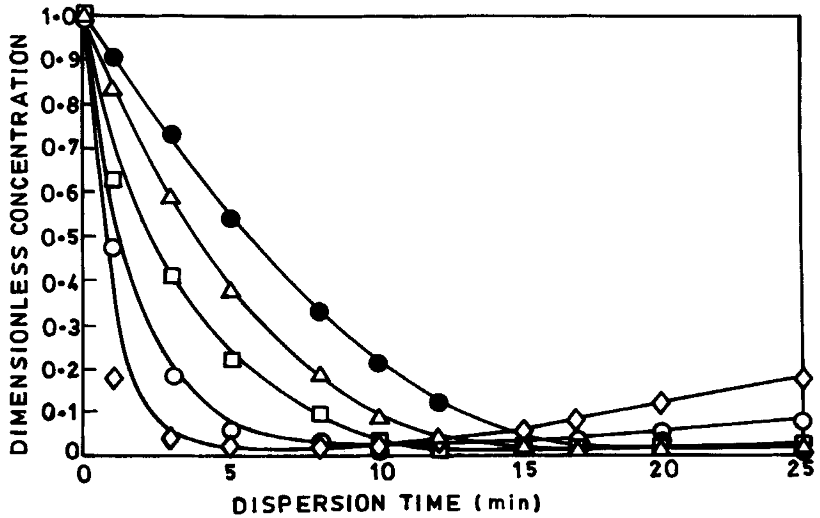


FIG. 3. Effect of speed of agitation on extraction.

rpm	Symbol
240	●
300	△
350	□
415	○
500	◇

Another disadvantage reported by Ohtake et al. (*24*) is that the extent of water entrainment increases at high agitation speeds. This is due to the increased drop surface area and the enhanced contact between the drop and external water. After considering all these factors, an agitation speed of 350 rpm was selected for further studies.

3. Effect of Water Volume Fraction in w/o Emulsion on Extraction

This effect was studied by using the following emulsion composition: surfactant 2% v/v + kerosene (98 − *A*)% v/v and an aqueous NaOH phase *A*% v/v (*A* = 10 to 30% v/v).

The stoichiometric ratio in each experiment was maintained between the number of moles of NaOH in the internal aqueous phase and that of *p*-nitrophenol in the feed solution.

Figure 4 shows that the initial rate of extraction, as well as the overall

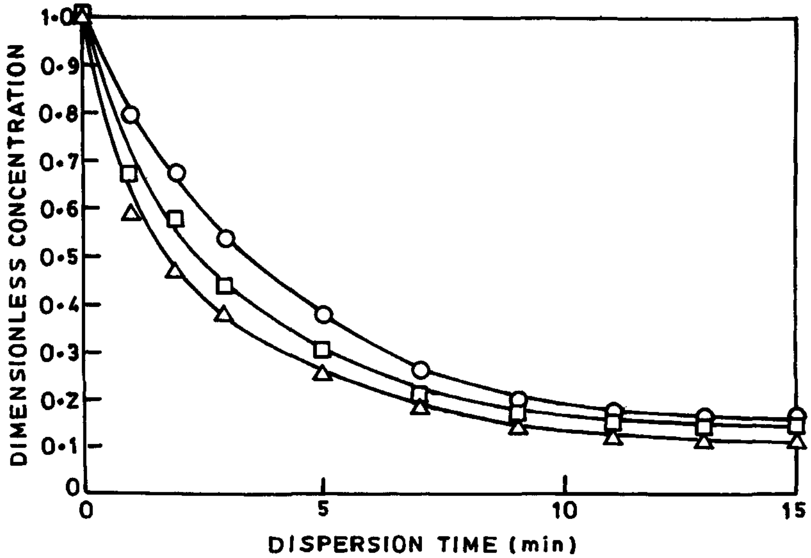


FIG. 4. Effect of water volume fraction in w/o emulsion on extraction.

Aqueous NaOH, % v/v	Symbol
10	○
20	□
30	△

mass transfer rate, increases with an increase in the volume percent of the aqueous phase at a constant surfactant concentration. This is due to the decrease in membrane thickness and the increase in surface area which are responsible for higher diffusion rates.

4. Effect of Treatment Ratio on Extraction

Figure 5 shows the effect of the treatment ratio (the ratio of the feed solution to emulsion) on the extraction of *p*-nitrophenol. A change in the treatment ratio was brought about by changing the volume of the feed while maintaining the volume of the emulsion phase constant. Although the concentration of *p*-nitrophenol in the continuous phase is the same, an increase in the treatment ratio increases the number of moles of *p*-nitrophenol in the continuous phase. Therefore, the depletion of *p*-nitrophenol from the feed is higher in the case of a low treatment ratio, although the flux may be a little higher because of the larger driving force. The

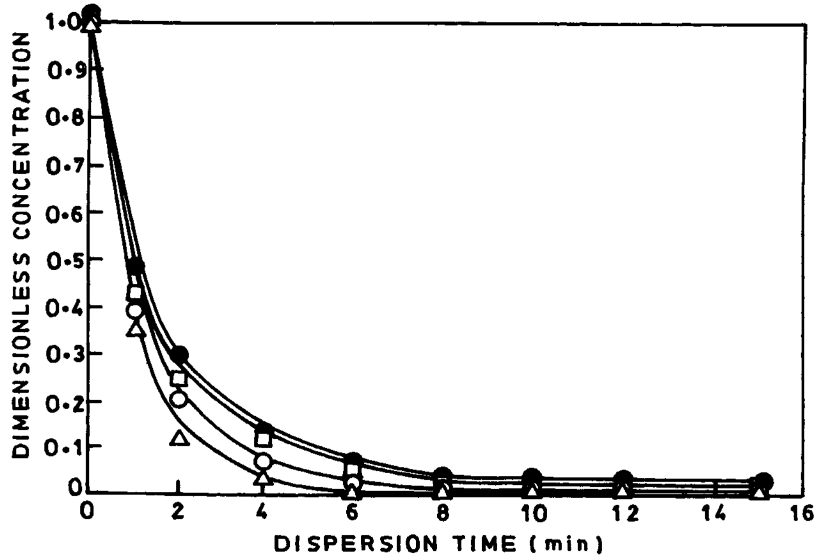


FIG. 5. Effect of treatment ratio on extraction.

Treatment ratio	Symbol
4:1	△
6:1	○
8:1	□
10:1	●

overall result is a lower mass transfer rate in the case of a higher treatment ratio. Similar results were observed by Terry et al. (*25*) and Goswami and Rawat (*17*).

5. Effect of Surfactant Concentration

The concentration of the surfactant determines the stability of the emulsion and also has a marked effect on the rate of extraction. Figure 6 shows the effect of surfactant concentration on the extraction of *p*-nitrophenol. It is observed that as the concentration of surfactant in the membrane phase increases, the initial flux increases. The initial higher rate of extraction at a greater surfactant concentration may be attributed to a decrease in the droplet size of the inner phase, resulting in a larger interfacial area for the mass transfer (*24*). However, the extent of extraction was found to slightly decrease with an increase in surfactant concentration (Fig. 6). This can be explained as the result of emulsion swell and rupture, causing spillage of the entrapped solute from the inner phase.

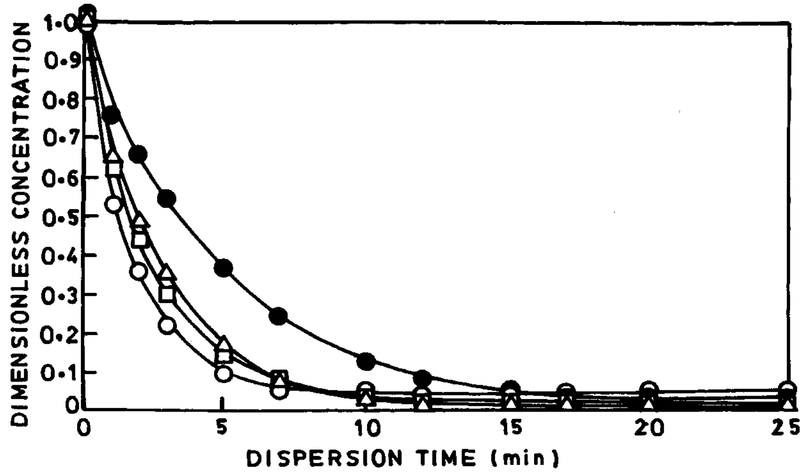


FIG. 6. Effect of surfactant concentration on extraction.

Span-80, % v/v	Symbol
1	●
2	△
4	□
6	○

6. Swelling

The major disadvantages of swelling of emulsion (transfer of water from the bulk phase into the emulsion phase) are dilution of the receiving phase and changes in the properties of the emulsion phase. Figure 7 shows the swelling of the emulsion with the same membrane formulation. It is observed that swelling of the emulsion is a function of surfactant concentration. An increase in the surfactant concentration was found to increase the swelling of the emulsion. Figure 8 shows the effect of the dispersion time on the percentage swell of the emulsion. It indicates that percent swell is directly proportional to mixing time. The following three possible factors are responsible for water transport (*26*, *27*):

(a) Shear on the system.
(b) Osmotic pressure difference between internal reagent and feed solution.
(c) Occlusion.

This swelling problem can be tackled by formulating a membrane phase which is not permeable to water. A drawback associated with the emulsifier Span-80 is that it is a good carrier for water molecules (*24*, *27*, *28*). The transport of water is mainly caused by hydration of the surfactant and inverse micelle formation. Draxler and Marr (*29*) reported that emulsion containing an anion exchanger (like Aliquat-336) can prevent water transport by Span-80 molecules without affecting the properties of the carrier.

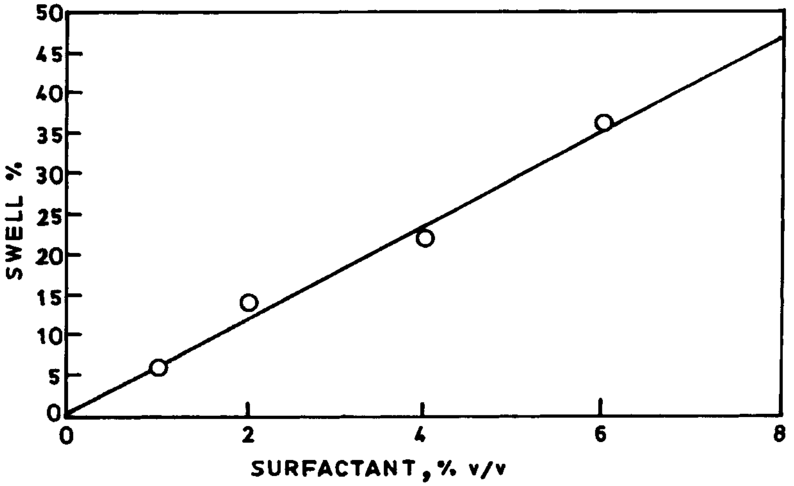


FIG. 7. Effect of surfactant concentration on swelling.

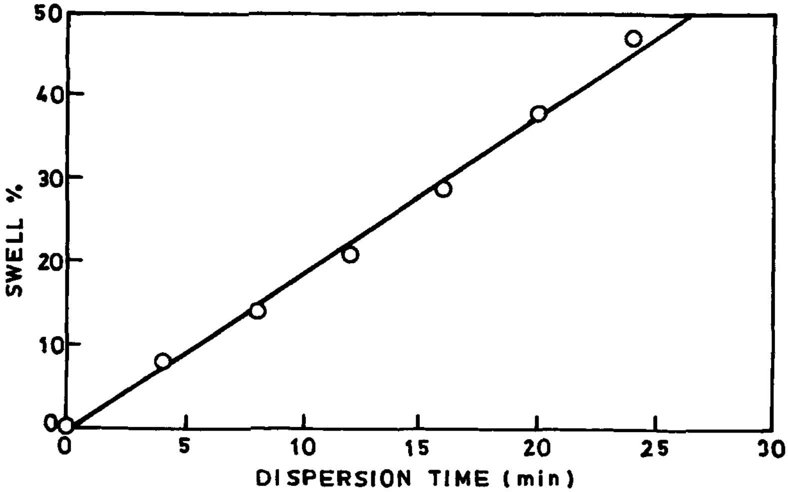


FIG. 8. Effect of the dispersion time on the percentage swelling of the emulsion.

Therefore, an additive was added to the membrane phase to reduce these effects. It was found that the addition of cyclohexanone decreases swelling in LEM extraction (*30*). It was observed that swelling is minimized by the addition of 2.5 and 5% v/v cyclohexanone with 2 and 5% v/v Span-80, respectively (Table 2).

This swelling can be further reduced to a minimum value by increasing the viscosity of the membrane phase. Figure 9 shows the effect of paraffin addition on the percent swelling at a dispersion time of 25 min. Minimum swelling of 2% is obtained at 20% v/v paraffin in the membrane phase. Thus, water transport can be greatly decreased by increasing the viscosity of the membrane phase, but the mass transfer rate decreases at a higher concentration of paraffin in the membrane phase due to the increased viscosity of the membrane (Fig. 10).

TABLE 2
Effect of Cyclohexanone Addition to Span-80 for Minimizing Swelling

Span-80, % v/v	Cyclohexanone, % v/v	% Swelling
2	0	24
2	2.5	10
5	0	44
5	5	14

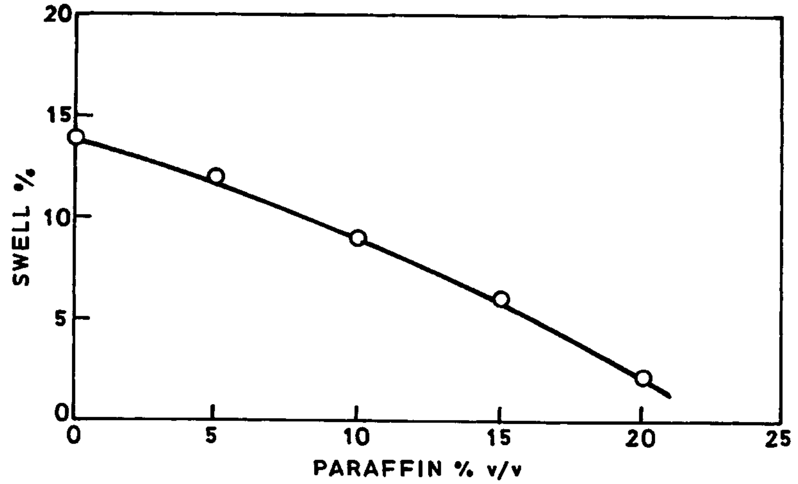


FIG. 9. Effect of paraffin concentration on the percentage swelling of the emulsion.

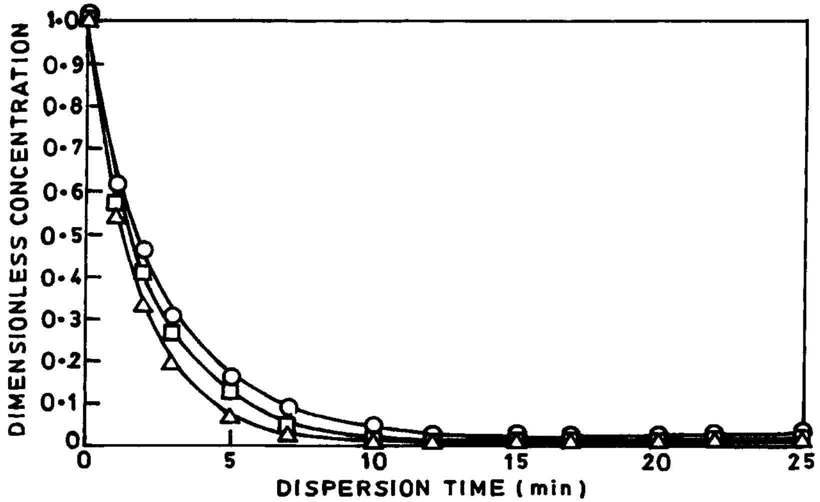


FIG. 10. Effect of paraffin concentration on extraction.

Paraffin, % v/v	Symbol
0	△
10	□
20	○

7. Effect of Breakage

Initially, some runs were carried out to study the permeation of metal ions through the liquid membrane. It was observed that Cu permeates through the membrane. Therefore, only Cu cannot be used as a tracer, but it was also observed that in presence of Cu, Ni does not permeate through the membrane. Therefore, a mixture of Cu and Ni was taken in the internal phase and the concentration of Ni in the outer phase was observed to quantify emulsion breakage. Therefore, Ni was used as the tracer in the presence of Cu. The validity of this tracer technique was studied in detail by Matsumoto et al. (*31*).

Figure 11 shows that the percentage of breakage increases with dispersion time because of shearing. However, the breakage was found to be negligible in the initial stage of the experiments. This is because the emulsion at 2 ± 0.5°C was introduced into the bulk phase at 27°C. Here, during the initial period, i.e., until the emulsion reached the temperature of the bulk phase, no breakage was observed. This was confirmed by carrying out the run with the bulk phase at 2 ± 0.5°C. In this case, breakage was found to be negligible up to 30 min extraction time. This could be explained by the fact that the viscosity of emulsion is high at low temperatures. Because of the high viscosity or low diffusivity, the rate of extraction also decreases. This is seen from the data presented in Fig. 12.

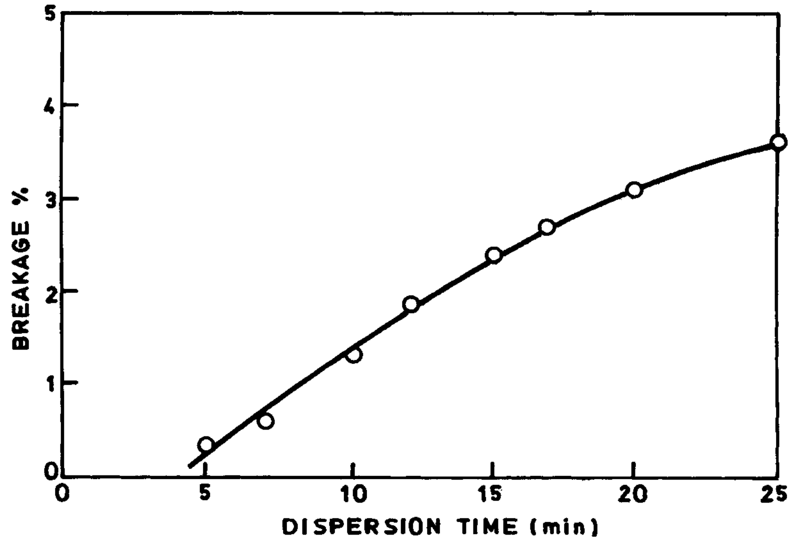


FIG. 11. Variation of emulsion breakage with dispersion time.

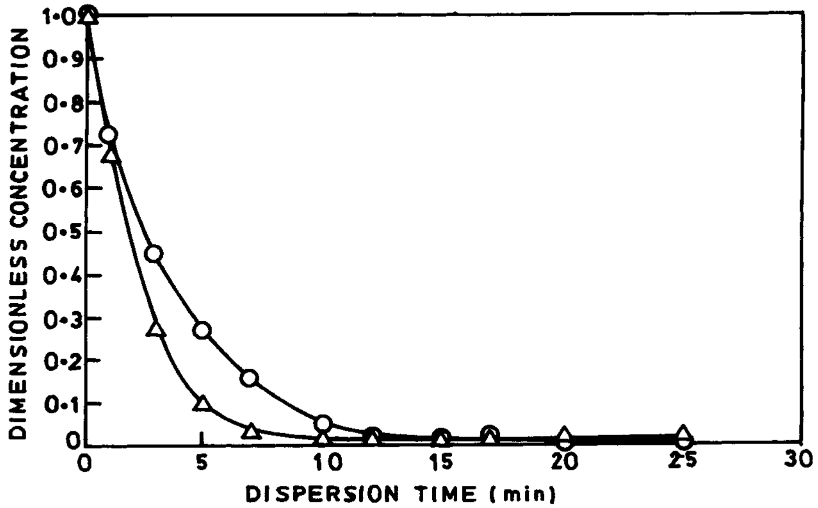


FIG. 12. Effect of temperature on extraction.

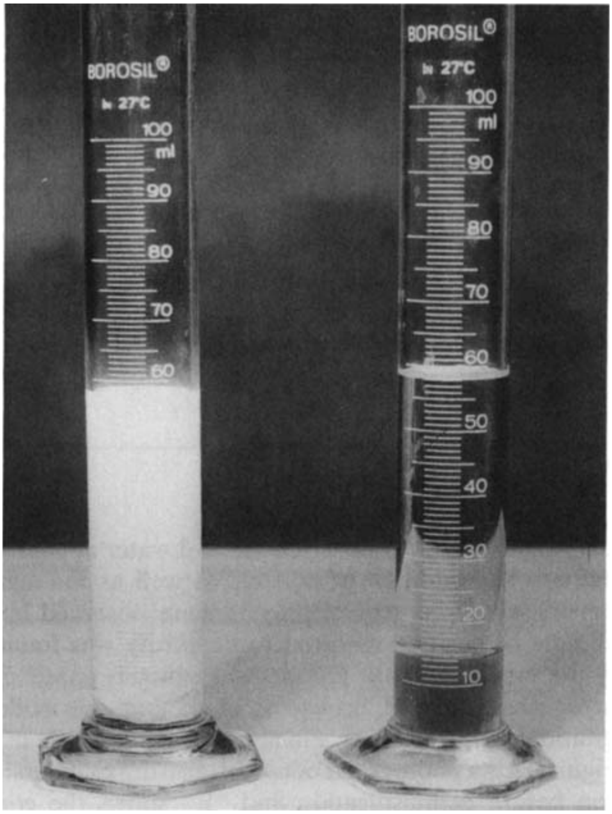
Temperature (°C)	Symbol
27	△
2	○

8. Demulsification

An important step in the LEM separation process is the breakdown of the emulsion after extraction and the reusability of the demulsified oil phase. Demulsification of a w/o emulsion can be carried out by using different methods such as settling, heating, mechanical, electrical, chemical, or a combination of these methods. Demulsification by a chemical method was used in the present investigation.

Li (*32*) has patented a chemical process for demulsification of w/o and o/w emulsions. According to this technique, the emulsion is mixed with a mixture of more than two comiscible, volatile solvents, at least one of which is miscible with water and one is miscible with oil, thereby causing the demulsification. However, in this study a process was developed for demulsification after extraction of *p*-nitrophenol by using a chemical which is miscible in both phases. Acetone was used for demulsification in the present study.

The following procedure was adopted for demulsification. After completing the extraction of *p*-nitrophenol, the complete solution was taken



(A) (B)

FIG. 13. A photograph of two measuring cylinders. (A) The condition before demulsification. (B) The contents after demulsification.

in a separating funnel. A settling time of 30 min was allowed to obtain separate layers of the feed solution and the emulsion phase. The volume of the emulsion was measured and the volume of the internal aqueous phase was calculated by comparing the volume of emulsion before and after extraction and considering the original aqueous phase in the w/o emulsion.

The above emulsion was taken in a glass vessel of 0.03 m inner diameter equipped with a turbine impeller. Then it was stirred at a constant agitation speed of 300 rpm. The addition of acetone was immediately started by using a burette.

It was observed that at a particular stage the color of the emulsion

TABLE 3
Typical Composition of LEM Separation System

Parameter	Value
Emulsification rpm	3000
Emulsification time (min)	15
Permeation rpm	350
Concentration of surfactant Span-80 (% v/v)	2
Concentration of cyclohexanone (% v/v)	2.5
Concentration of paraffin (% v/v)	10
Kerosene (% v/v)	75.5
Membrane oil phase to internal aqueous phase ratio (in w/o emulsion) (% v/v)	90:10
Treatment ratio	6:1
Emulsification temperature (°C)	2 ± 0.5°C
Extraction temperature (°C)	27

changes from bright yellow to faint yellow, and water appears on the side of the container. The addition of acetone as well as the agitation were stopped immediately. Two separate layers were observed in the vessel. Here the volume of acetone required to demulsify was found to be approximately the same as that of the internal aqueous phase. Because the upper oil layer was not clear, an excess of acetone was added at a low speed of agitation (150–180 rpm). Finally, two different clear layers were obtained. Figure 13 is a photograph of two measuring cylinders: "A" shows the condition before demulsification and "B" shows the contents after demulsification.

It was found that all the added acetone was present in the water phase only. The efficiency of the membrane phase was checked by using this oil phase in the preparation of the w/o emulsion with the same aqueous phase of NaOH solution. This emulsion was used for the extraction of *p*-nitrophenol by keeping all the parameters the same. It was observed that there was no difference in the mass transfer rate. Therefore, this method of demulsification was used in subsequent work.

Considering all the parameters discussed above, the conditions mentioned in Table 3 were used for the extraction of other phenolic compounds, viz. *o*-nitrophenol and 2,4-dinitrophenol.

9. Comparison of the Rate of Extraction of *p*-Nitrophenol, o-Nitrophenol, and 2,4-Dinitrophenol

Figure 14 shows the rate of extraction of three solutes: *p*-nitrophenol, *o*-nitrophenol, and 2,4-dinitrophenol. It is seen that the rate of extraction

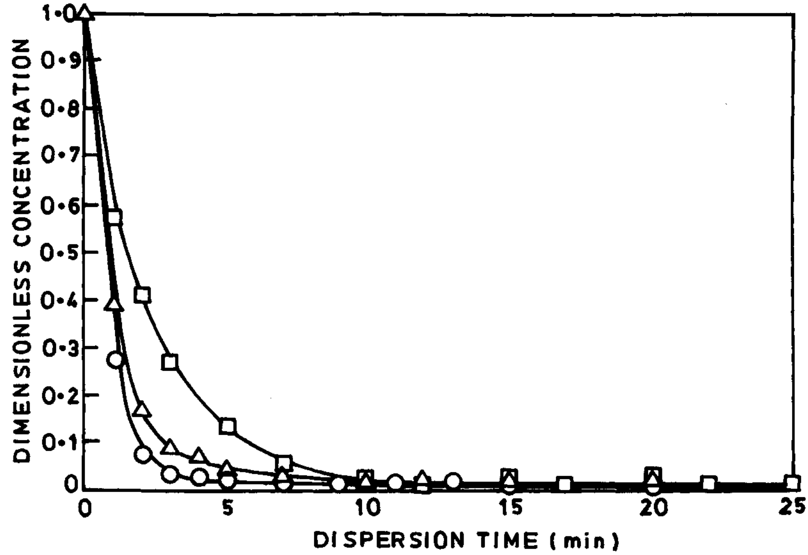


FIG. 14. Comparison of the rate of extraction of *p*-nitrophenol, *o*-nitrophenol, and 2,4-dinitrophenol.

Nitrophenols	Symbol
p-Nitrophenol	□
o-Nitrophenol	○
2,4-Dinitrophenol	△

varies in the following order: *o*-nitrophenol > 2,4-dinitrophenol > *p*-nitrophenol. The initial concentration was 1000 ppm for *p*-nitrophenol and *o*-nitrophenol and 500 ppm for 2,4-dinitrophenol.

The partition coefficient values of *p*-nitrophenol, *o*-nitrophenol, and 2,4-dinitrophenol are listed in Table 4. The partition coefficient of *o*-nitrophenol is higher than that of 2,4-dinitrophenol, and the partition coefficient

TABLE 4
Partition Coefficient of Nitrophenols [organic phase (membrane phase of emulsion) to aqueous phase ratio 1:1]

Nitrophenol	Initial concentration (ppm)	Partition coefficient
p-Nitrophenol	1000	0.19
o-Nitrophenol	1000	15.58
2,4-Dinitrophenol	500	6.86

of 2,4-dinitrophenol is higher than that of *p*-nitrophenol. Therefore the trend in the rate of extraction of the three solutes shown in Fig. 14 is explained on the basis of the solubility of the solute in the membrane phase. The rate of extraction of *o*-nitrophenol is higher than those of *p*-nitrophenol and 2,4-dinitrophenol due to higher solubility of *o*-nitrophenol in the oil phase.

CONCLUSIONS

The effects of the HLB value, speed of agitation, water volume fraction in w/o emulsion, treatment ratio, and concentration of surfactant on the extraction of *p*-nitrophenol using LEM were studied. The swelling phenomenon was studied by considering the effect of surfactant concentration, dispersion time, and addition of paraffin on emulsion swelling. A recipe for minimizing the swelling has been proposed. The demulsification was successfully carried out by the addition of acetone. Extraction of *o*-nitrophenol and 2,4-dinitrophenol was also studied.

REFERENCES

1. N. N. Li, U.S. Patent 3,410,794 (1968).
2. R. P. Cahn and N. N. Li, *Sep. Sci. Technol., 9,* 505 (1974).
3. Y. C. Chang and S. P. Li, *Desalination, 47,* 351 (1983).
4. L. Boyadzhiev, E. Benenshek, and Z. Lazarova, *J. Membr. Sci., 21,* 137 (1984).
5. T. Kitagawa, Y. Nishikawa, J. W. Frankenfeld, and N. N. Li, *Environ. Sci. Technol., 11,* 602 (1977).
6. J. W. Frankenfeld, R. P. Cahn, and N. N. Li, *Sep. Sci. Technol., 16,* 385 (1981).
7. S. Weiss, V. Grigoriev, and P. Muhl, *J. Membr. Sci., 12,* 119 (1982).
8. M. Protsch and R. Marr, *Proceedings of the International Solvent Extraction Conference, Denver, Colorado, August 26–September 2, 1983,* p. 66.
9. J. Draxler, W. Furst, and R. Marr, *J. Membr. Sci., 38,* 281 (1988).
10. H. H. Downs and N. N. Li, *J. Sep. Proc. Technol., 2,* 19 (1981).
11. M. Teramoto, H. Takihana, M. Shibutani, T. Yuasa, Y. Miyake, and H. Teranishi, *J. Chem. Eng. Jpn., 14,* 122 (1981).
12. R. S. Baird, A. L. Bunge, and R. D. Noble, *AIChE J., 33,* 43 (1987).
13. M. P. Thien, T. A. Hatton, and D. I. C. Wang, *Biotechnol. Bioeng., 32,* 605 (1988).
14. S. C. Boey, M. C. G. Del Cerro, and D. L. Pyle, *Chem. Eng. Res. Des., 65,* 218 (1987).
15. N. N. Li, *Ind. Eng. Chem., Process Des. Dev., 10,* 215 (1971).
16. E. R. Elzinga, U.S. Patent 3,389,078 (1968).
17. A. N. Goswami and B. S. Rawat, *J. Membr. Sci., 24,* 145 (1985).
18. S. K. Ihm, Y. H. Jeong, and Y. S. Won, *Ibid., 32,* 31 (1987).
19. M. Ruppert, J. Draxler, and R. Marr, *Sep. Sci. Technol., 23,* 1659 (1988).
20. M. J. Schick (ed.), *Nonionic Surfactants Physical Chemistry* (Surfactant Science Series, Vol. 23), Dekker, New York, 1987, p. 535.
21. H. L. Greenwald, E. B. Kice, M. Kenly, and J. Kelly, *J. Anal. Chem., 33,* 465 (1961).
22. E. H. Crook, D. B. Fordyce, and G. F. Trebbi, *J. Colloid Interface Sci., 20,* 191 (1965).
23. K. Shinoda, T. Yoneyama, and H. Tsutsumi, *J. Dispersion Sci. Technol., 1,* 1 (1980).
24. T. Ohtake, T. Hano, K. Takagi, and F. Nakashio, *J. Chem. Eng. Jpn., 21,* 273 (1988).

25. R. E. Terry, N. N. Li, and W. S. Ho, *J. Membr. Sci., 10,* 305 (1982).

26. C. Del Cerro and D. Boey, *Chem. Ind.,* p. 681 (1988).

27. P. Colinart, S. Delepine, G. Trouve, and H. Renon, *J. Membr. Sci., 20,* 167 (1984).

28. F. Nakashio, M. Goto, M. Matsumoto, J. Irie, and K. Kondo, *Ibid., 38,* 249 (1988).

29. J. Draxler and R. Marr, *Ber. Bunsenges. Phys. Chem., 86,* 64 (1982).

30. A. V. Mukkolath, P. T. Gadekar, and K. K. Tiwari, "A New Method for the Reduction of Swelling in Liquid Emulsion Membrane Systems," *Chem. Ind.,* In Press.

31. M. Matsumoto, M. Goto, K. Kondo, and F. Nakashio, *J. Chem. Eng. Jpn., 21,* 318 (1988).

32. N. N. Li, H. Taras, and R. P. Cahn, U.S. Patent 3,525,204 (1976).

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