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A Practicable Process for Phenol Removal with Liquid Surfactant Membrane Permeation Column

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ABSTRACT

A practicable liquid surfactant membrane process for phenol removal is proposed with a stirred countercurrent column used as the liquid membrane contact equipment. The constituents of liquid membranes, such as internal aqueous phase and surfactant, the type of column, and the operating conditions for efficient and continuous performance of the liquid surfactant membrane process, have been examined. When NaOH solution was used as the internal aqueous phase and ECA4360J was used as the surfactant, the W/O emulsion was stable for the duration of column operation. More than 97% phenol could be removed from the feed solution. Nearly complete demulsification was also achieved by gentle agitation with an electrostatic demulsifier.

Key Words. Liquid surfactant membrane process; Phenol; Permeation column; Demulsification; Continuous operation

INTRODUCTION

The separation method using liquid surfactant membranes is a highly selective and concentrating separation technique of potential value to the chemical industry. The method is an energy-saving separation technique

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and is regarded as a combined extraction/stripping process, where extraction and stripping in the conventional liquid–liquid extraction take place simultaneously in one apparatus. There have been many papers (1) suggesting a possibility of applications of liquid surfactant membranes to a variety of separations. However, only a commercial liquid membrane permeation plant for the recovery process of zinc from the wastewater of the viscose and rayon industry is in operation (2). This plant was built in Austria in 1986, and thus the separation process by liquid surfactant membranes is just beginning to be industrialized.

To advance the practical application of liquid surfactant membranes, it is necessary to investigate continuous operation of the separation process. The liquid surfactant membrane separation process has to be established through a set of tests from bench scale to pilot plant.

The liquid surfactant membrane process consists of four steps: 1) an emulsification step for producing W/O emulsion from an internal aqueous phase and a liquid membrane phase, 2) a permeation and concentration step transferring solutes from a continuous phase into emulsion drops, 3) a phase separation step for separating a continuous and dispersed phase, and 4) a demulsification step for splitting a dispersed emulsion phase into an enriched aqueous phase and an oil membrane phase. Many studies on these individual steps have been performed to date (3–19). However, in order to operate these four steps continuously, it is necessary to investigate the process as a whole.

The purpose of our work is to develop a continuous separation process with the liquid surfactant membrane technique when a stirred countercurrent column is used as the liquid membrane contact equipment. In this study we adopted phenol, which is one of the representative contaminants in the environment, as a separating solute, and examined the possibility of operating the above four steps continuously. W/O emulsions were prepared using an ultradisperser with a flow chamber. Continuous permeation experiments of phenol were carried out with different types of stirred countercurrent columns. In the demulsification of W/O emulsions after the permeation and concentration step, a cylindrical electrostatic demulsifier was used. For an efficient and continuous performance of such a liquid surfactant membrane process, we investigated the constitution of a liquid membrane, i.e., the internal aqueous phase and surfactant, the type of column, and the operating conditions.

EXPERIMENTAL

Apparatus

Figure 1 shows a schematic diagram of the experimental setup for the continuous liquid surfactant membrane process. As the liquid membrane

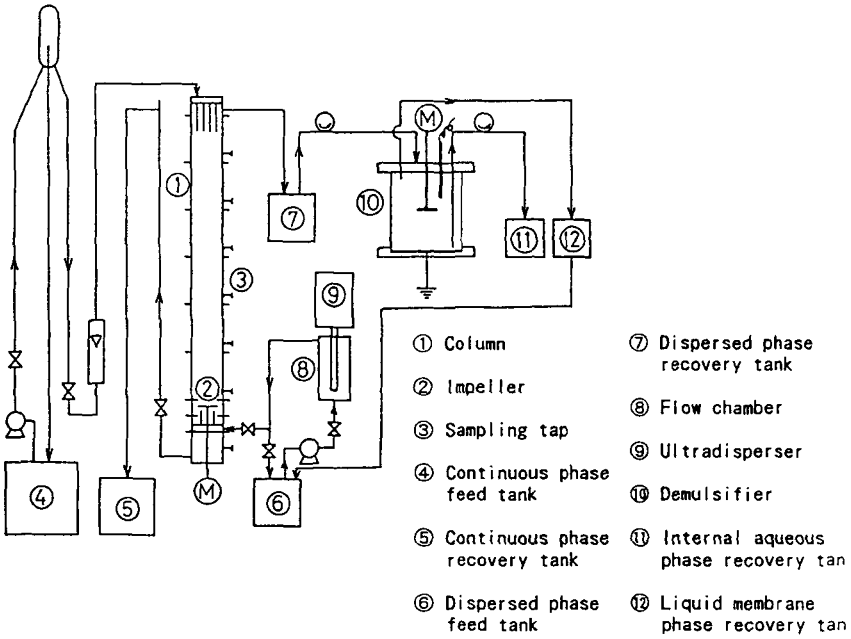

FIG. 1 Schematic diagram of the experimental setup for continuous liquid surfactant membrane process.

contact equipment we used three types of stirred countercurrent column, identified as A, B, and C, of different construction, whose details are described later. Column C, schematically described in Fig. 2, was the column mainly used. The column, which is made of acrylic resin, is 50 mm in inner diameter and 2 or 1 m high. In the upper part above the stator ring of 30 mm diameter, six units of 300 mm height each are joined in a column of 2 m height. This part is hollow except for the stator ring and baffle plate in order to prevent the adhesion of W/O emulsions. It is possible to attach impellers for agitation to a shaft inserted from the top of the column. Below the stator ring, one unit of 55 mm height with four baffle plates of 5 mm width is connected. In this section, one impeller is attached at 20 mm, just above the nozzles, to a shaft inserted through the bottom of the column. The W/O emulsion introduced through the nozzles is dispersed as fine drops by the impeller, a four-blade fan turbine of 30 mm diameter. Two nozzles of 0.51 mm inner diameter and six nozzles of 3 mm inner diameter are mounted for the feed of the dispersed phase and of the continuous phase, respectively. All nozzles are made of stainless steel.

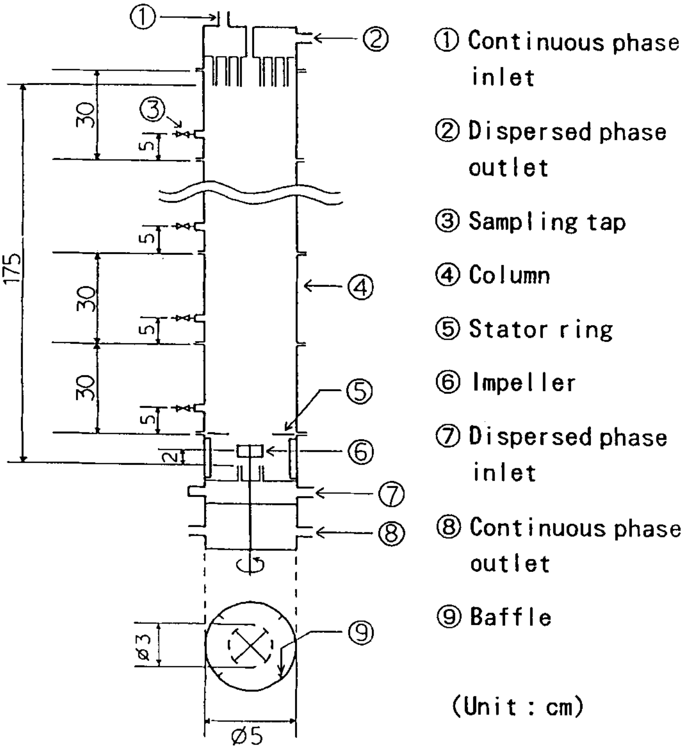

FIG. 2 Schematic diagram of Column C.

The W/O emulsions were prepared using an ultradisperser (LK-42, Yamato Scientific Co., Tokyo) with a flow chamber (F41).

The demulsification experiments were carried out with the electrostatic demulsifier (19) diagramed in Fig. 3. It is made of acrylic resin with 70 mm inner diameter and 90 mm height. One of the electrodes was prepared by filling an 8-mm-diameter glass tube with saturated aqueous sodium chloride solution into which a 2-mm-diameter copper wire was inserted; then the glass tube was sealed. A copper disk electrode of 41 mm diameter was set at the bottom of the demulsifier and grounded. A glass, two-blade fan turbine of 25 mm diameter was used to agitate the W/O emulsion in the demulsifier.

Procedure

The W/O emulsion was prepared by mixing and circulating equivolumes of the internal aqueous phase and the liquid membrane phase with the

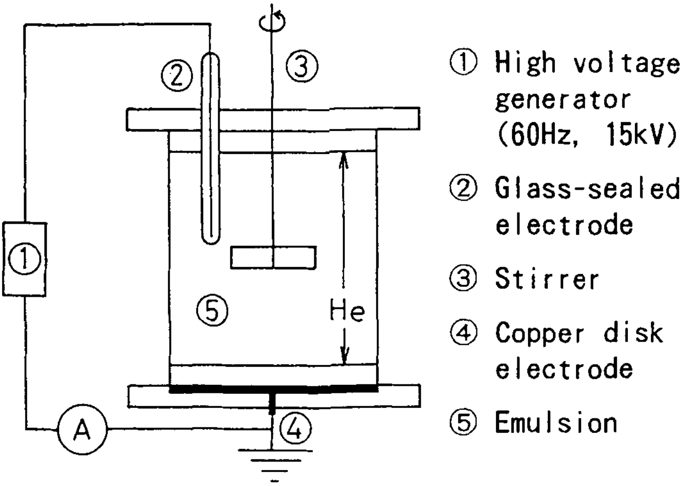

FIG. 3 Electrostatic demulsifier.

ultradisperser and the flow chamber at 6000 rpm for 10 minutes. The continuous phase, a feed solution containing phenol, was continuously introduced through six nozzles at the top of the column. The W/O emulsion was fed from two nozzles at the bottom of the column and dispersed as fine drops by agitation just above the nozzles. The dispersed emulsion drops rose to the top of the column and coalesced to form an emulsion layer which overflowed continuously. The interface between the continuous and dispersed phases in the top settling zone was kept constant in location by adjusting a constant level device. The flow rates of the continuous and dispersed phases were respectively measured with a graduated cylinder and a stopwatch. After the start of contact of the two phases, the continuous solution was sampled at specific time intervals from the continuous phase outlet. The time-dependent concentrations of phenol were also measured. The observed result indicated that the phenol concentration at the continuous phase outlet was nearly constant 30 minutes after the start of contact. Therefore, because steady state had been reached after 1 hour of operation the continuous solutions were sampled from sampling taps to obtain the concentration profile of phenol along the column. The location of the interface in the top settling zone was kept constant, and the dispersed phase holdup, ϕ', was measured by a shutdown method.

The phenol concentrations of the continuous phase were determined from absorption at 269.2 nm in a spectrophotometer. The breakage of liquid membranes, ϵ, was calculated from the following equation by measuring the concentration of sodium ion in the continuous phase by flame analysis:

$$\text{Breakage } (\epsilon) = \frac{\text{amount of Na}^+ \text{ at the continuous phase outlet}}{\begin{array}{c}\text{amount of Na}^+ \text{ in the internal aqueous phase}\\ \text{introduced into the column}\end{array}} \times 100$$

The water content, W, in the dispersed emulsion phase which overflowed from the top of the column was determined by Karl-Fischer titration. The extent of swelling of the emulsion was obtained as the ratio W/W_0, where W_0 is the water content in the feed emulsion.

The W/O emulsion in a fixed volume was supplied from the dispersed phase recovery tank into the demulsifier using a tubing pump. The batchwise demulsification experiment was carried out by applying 15 kV A.C. at 60 Hz between the two electrodes. The time for demulsification was obtained by calculating the extent of demulsification, B, defined as follows:

$$\text{Extent of demulsification } (B) = \frac{\begin{array}{c}\text{volume of the separated}\\ \text{aqueous phase}\end{array}}{\begin{array}{c}\text{volume of the aqueous phase}\\ \text{in the supplied emulsion}\end{array}} \times 100$$

RESULTS AND DISCUSSION

Constitution of Liquid Membrane

Comparison of Diluent

The continuous permeation experiments of phenol were carried out by using a 1-m high column to compare the diluents kerosene (which has commonly been used) and Shellsol 71 (Shell Chemicals Japan Co.) (which was newly used in this study). Figure 4 shows the concentration profiles of phenol along the column when the internal aqueous phase was a NaOH solution of 100 mol/m^3 and the concentration of Span 80 was 4 wt%. There was little difference in the concentration profiles by the various diluents, but breakage of the liquid membrane for Shellsol 71 was lower than that for kerosene. Thus, Shellsol 71 formed stable emulsions in comparison with kerosene, so we used Shellsol 71 as the diluent in the following experiments.

Comparison of Internal Aqueous Phase

When Span 80 was used as the surfactant and NaOH solution was used as the internal aqueous phase, phenol was removed efficiently but the

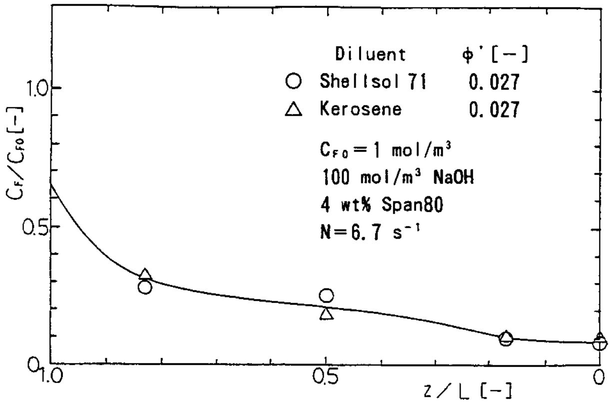

FIG. 4 Effect of diluent on the concentration profile of phenol along the column.

emulsion could not be demulsified. On the other hand, when Na_2CO_3 solution was used as the internal aqueous phase, demulsification became possible but the removal of phenol was reduced drastically. Therefore, we examined the use of a $NaOH/Na_2CO_3$ mixed solution. The experimental conditions and results are summarized in Table 1. In the case of a NaOH concentration of 50 mol/m³, demulsification did not occur in a Na_2CO_3 concentration of 50 mol/m³ but became possible above 125 mol/m³. However, an increase in the removal of phenol was not observed with an increase in Na_2CO_3 concentration. The result suggests that NaOH mainly contributes to the removal of phenol. In the case of a NaOH concentration of 200 mol/m³, the emulsion could not be demulsified even though the

TABLE 1
Comparison of Internal Aqueous Phase[a]

Internal aqueous phase	N (1/s)	ϕ' (—)	Removal (%)	B (%)
300 mol/m³ NaOH	5.8	0.0068	80.8	[b]
50 mol/m³ NaOH + 50 mol/m³ Na_2CO_3	6.7	0.029	94.9	[b]
50 mol/m³ NaOH + 125 mol/m³ Na_2CO_3	6.7	0.025	93.6	59.2
50 mol/m³ NaOH + 250 mol/m³ Na_2CO_3	5.8	0.013	77.2	51.4
100 mol/m³ NaOH + 100 mol/m³ Na_2CO_3	6.7	0.017	95.1	52.3
200 mol/m³ NaOH + 300 mol/m³ Na_2CO_3	6.7	0.017	94.1	[b]

[a] C_{F0} = 1 mol/m³, 2 wt% Span 80, V_F = 2.7 mL/s, V_E = 0.28 mL/s.
[b] No demulsification.

Na_2CO_3 concentration was increased up to 300 mol/m^3. Thus, in the NaOH/Na_2CO_3 mixed solution system, there were limited conditions under which demulsification of W/O emulsions was possible and more than 90% phenol could be removed.

Comparison of Surfactant

Table 2 shows the experimental conditions and results of comparing surfactants. When Span 80 was used as the surfactant, emulsification was possible at a 1 wt% concentration but stable operation in the column was not possible. Therefore, Shellsol 71 solution containing 2 wt% Span 80 was used as the liquid membrane phase. On the other hand, in the case of ECA4360J (polyamine), Shellsol 71 solution containing 5 wt% ECA4360J was used, because emulsification was difficult at the 2 wt% concentration. As shown in Table 2, the emulsion prepared using ECA4360J could be demulsified even though the internal aqueous phase was NaOH solution.

Comparison of the concentration profiles along the column is shown in Fig. 5 when a feed solution containing 10 mol/m^3 phenol was treated by using an internal aqueous phase of 300 mol/m^3 NaOH. Similar tendencies in the concentration profiles were observed, with the removal in Span 80 lower than that in ECA4360J owing to a difference in holdup, ϕ'. Although the reason for this is not clear, it is presumed that the emulsion became unstable due to a change of its physical properties due to a reaction between Span 80 and NaOH at high concentration (20). On the other hand, when the internal aqueous phase was a mixed solution of 100 mol/m^3 NaOH and 100 mol/m^3 Na_2CO_3 there were little differences in both ϕ'

TABLE 2
Comparison of Surfactants[a]

Surfactant	ϕ' (—)	Removal (%)	B (%)
2 wt% Span 80[b]	0.0094	89.0	[d]
5 wt% ECA4360J[b]	0.017	97.1	45.3
2 wt% Span 80[c]	0.017	95.1	52.3
5 wt% ECA4360J[c]	0.014	96.0	53.3

[a] V_F = 2.7 mL/s, V_E = 0.37 mL/s, N = 6.7 s^{-1}.

[b] C_{F0} = 10 mol/m^3, internal aqueous phase: 300 mol/m^3 NaOH.

[c] C_{F0} = 1 mol/m^3, internal aqueous phase: 100 mol/m^3 NaOH + 100 mol/m^3 Na_2CO_3.

[d] No demulsification.

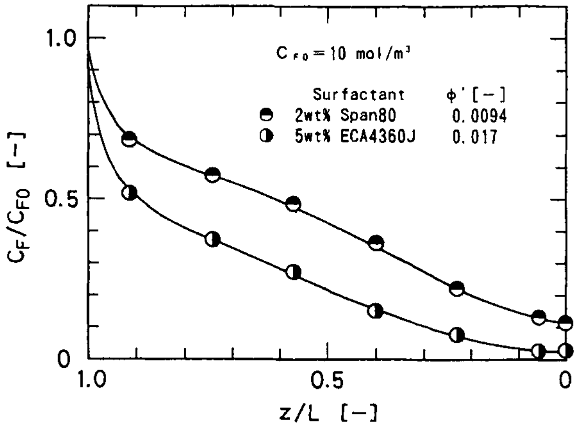

FIG. 5 Effect of surfactant on the concentration profile of phenol along the column.

and removal by the surfactants, as is evident in Table 2. In the case of ECA4360J, the use of NaOH/Na_2CO_3 mixed solution as the internal aqueous phase also led to a slight increase in the extent of demulsification, *B*.

When Span 80 was used as the surfactant, there was no problem for column operation, but Na_2CO_3 addition to NaOH solution was necessary to make demulsification possible, as noted previously. Up to the present, demulsification has been impossible above a NaOH concentration of 200 mol/m^3, so NaOH concentration has kept lower than 100 mol/m^3 in order to operate the process continuously. If a feed solution containing phenol in high concentration were to be treated with such an internal aqueous phase, the removal of phenol might become low. In the case of ECA4360J, demulsification was possible even when an internal aqueous phase containing only NaOH was used, and good removal could be established in the treatment of phenol solution in high concentration. However, foams sometimes occurred at the interface of the continuous and dispersed emulsion phases in the top settling zone. The foaming gradually grew in some types of column and made continuous column operation difficult. It is therefore important to select the proper type of column when ECA4360J is used.

Comparison of Column Type

The continuous permeation experiments were carried out using three 1-m high columns (illustrated in Fig. 6) to find a liquid membrane permea-

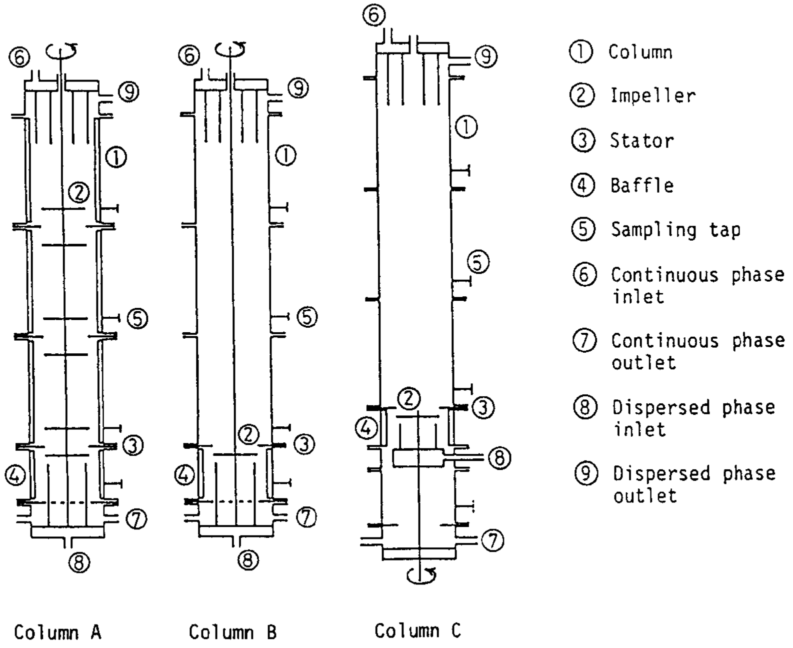

FIG. 6 Type of liquid surfactant membrane permeation column.

tion column suitable for the removal of phenol. Table 3 shows the representative experimental conditions and results. Column A is divided into four sections with stator rings. It has baffle plates and six impellers fitted above and below near the respective stator rings. Although the impellers are located so that the adhesion of emulsion drops to the attachments can be prevented, such adhesions occur in all three columns because of many attachments mounted in the columns. In the lowest section the emulsion phase can be dispersed into fine drops. Column A leads to the largest holdup among the columns at the same agitating speed, but at high speeds there is an entrainment of very fine droplets when the flow of the continuous phase increases due to repeated dispersions.

Column B consists of two parts, the lower one for dispersing emulsion drops and the upper one for contacting the continuous and dispersed phases countercurrently. The upper part has no impeller for suppressing backmixing in the column, and it is also without the stator ring and baffle plate in order to prevent the adhesion of emulsion. High-speed agitation is needed to obtain the same holdup as in Column A. It seems that there

TABLE 3
Comparison of Type of Permeation Column[a]

Column type	N (1/s)	ϕ' (—)	Removal (%)	ϵ (%)	W/W_0 (—)
A	5.0	0.037	92.1	2.19	1.32
B	5.0	0.018	85.9	1.59	1.03
	5.8	0.026	92.2	2.85	1.10
C	5.8	0.017	86.8	1.66	1.09
	6.7	0.027	91.5	2.34	1.05

[a] C_{F0} = 1 mol/m^3, internal aqueous phase: 100 mol/m^3 NaOH. Liquid membrane phase: 4 wt% Span 80–kerosene. V_F = 3.0 mL/s, V_E = 0.37 mL/s.

is a distribution of holdup with a tendency to decrease toward the top of the column.

Column C is basically the same as Column B except that a rotating shaft is inserted from the bottom of the column. Holdup becomes small because there is no influence due to shaft rotation in the upper part of the column. Therefore, high-speed agitation is needed for the extent of removal to be nearly the same as it is in Column B.

When Span 80 was used as the surfactant, continuous operation was possible in all three columns and good removal of phenol could also be achieved under appropriate operating conditions. In particular, Column A, with a large holdup at low agitating speed, and Column B, which suppresses backmixing, are suitable. On the other hand, in the case of ECA4360J, foaming tends to occur at the interface of the continuous and dispersed emulsion phases in the top settling zone, as noted previously. Therefore, the type of column in which the influence of agitation appears near the interface is unsuitable. In practice, operation using Column A was impossible, and it was undesirable to operate Column B under high rotating speeds of the shaft. Column C, without the influence of agitation near the interface, is suitable for the use of ECA4360J.

Demulsification

The above results indicated that W/O emulsions prepared with ECA4360J as the surfactant could be demulsified even though the internal aqueous phase was NaOH solution. However, the extent of demulsification was not enough for the continuous operation of the liquid surfactant membrane process to be performed smoothly. To investigate the possibility for further improvement in the extent of demulsification, we carried out batch demulsification experiments of W/O emulsions with ECA4360J under various conditions. In the experiments we used W/O emulsions, produced by agitating the internal aqueous and liquid membrane phases by a homogenizer with an emulsifying head (Silverson Machines Ltd., England) instead of the ultradisperser described above. The agitating speed and time were 4000 rpm and 3 minutes, respectively, based on earlier work (10).

The experiments were first carried out by heating W/O emulsions to examine the effect of temperature on demulsification. The extent of demulsification at room temperature was 12–15% at a demulsifying time of 30 minutes, but at 50°C it reached 80% after 10 minutes. Thus, both the rate and the extent of demulsification were greatly improved by operating at high temperature, as indicated in a previous paper (21). Next, we investigated demulsification with gentle agitation in the emulsion. The experimental results obtained by changing the location of the impeller and the agitating speed are summarized in Table 4. The impeller was set at $\frac{1}{4}$, $\frac{1}{2}$, and $\frac{3}{4}$He, respectively, of the height of the supplied emulsion phase, He, as illustrated in Fig. 3. It was found that the extent of demulsification, *B*, was greatly improved by an agitation of about 60 rpm in the middle of the emulsion sample. This result is similar to that reported by Hano et al. (19).

On the basis of the above results for demulsification of W/O emulsions, the effect of agitation ($\frac{1}{2}$He, 60 rpm) was examined for demulsification of the emulsion overflowing from the top of the column. A representative plot for the time course of the extent of demulsification is shown in Fig. 7. It is clear from the figure that demulsification was accelerated remarkably, and the extent of demulsification reached 90% after 30 minutes by adding gentle agitation.

TABLE 4

Effects of Impeller Location and Agitating Speed on the Extent of Demulsification[a]

Agitating speed: 1.0 s^{-1}		Impeller location: $\frac{1}{2}$He	
Impeller location	*B* (%)	Agitating speed (1/s)	*B* (%)
$\frac{1}{4}$ He	77.1	0.5	78.4
$\frac{1}{2}$ He	80.8	1.0	80.8
$\frac{3}{4}$ He	50.0	1.5	79.5

[a] W/O emulsion (6.7 s^{-1}, 180 s). Internal aqueous phase: 300 mol/m^3 NaOH + 100 mol/m^3 PhOH. Liquid membrane phase: 5 wt% ECA4360J–Shellsol 71.

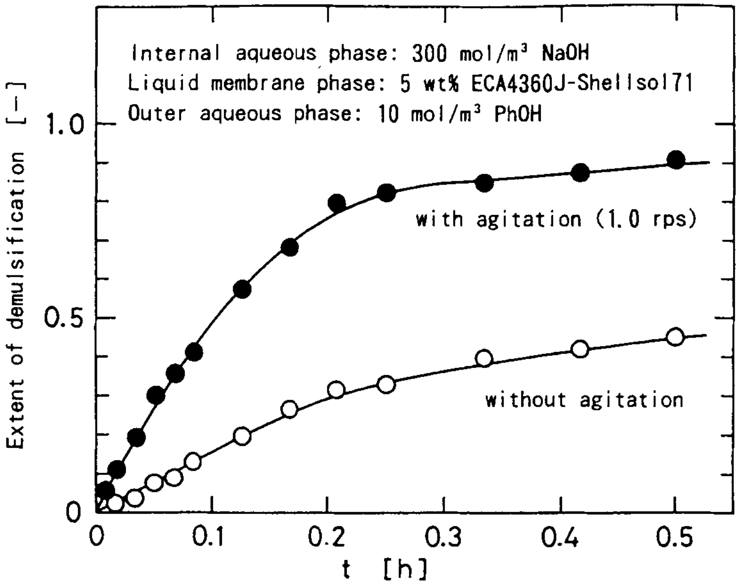

FIG. 7 Effect of agitation on the demulsification of emulsions overflowing from the column.

Setting up Process

Based on the above results, for the treatment of a feed solution containing 10 mol/m^3 of phenol, for example, the process shown in Fig. 8 is recommended.

Internal aqueous phase: 300 mol/m^3 NaOH solution. Liquid membrane phase: 5 wt% ECA4360J–Shellsol 71 solution when the feed solution is treated under the following conditions;

1. Preparation of emulsion. Volume ratio of internal aqueous phase to liquid membrane phase: 1/1. Mixing speed and time: 6000 rpm, 10 minutes.
2. Column operation. Feed rate of continuous phase: 160 mL/min. Feed rate of dispersed phase: 22 mL/min. Agitating speed: 400–450 rpm.
3. Demulsification. Applied voltage: A.C., 15 kV, 60 Hz. Agitating speed in the demulsifier: 60 rpm.

The W/O emulsion is stable for the duration of the column operation and can be demulsified above 80%; more than 97% phenol can be removed. The phenol concentration of the internal aqueous phase at the

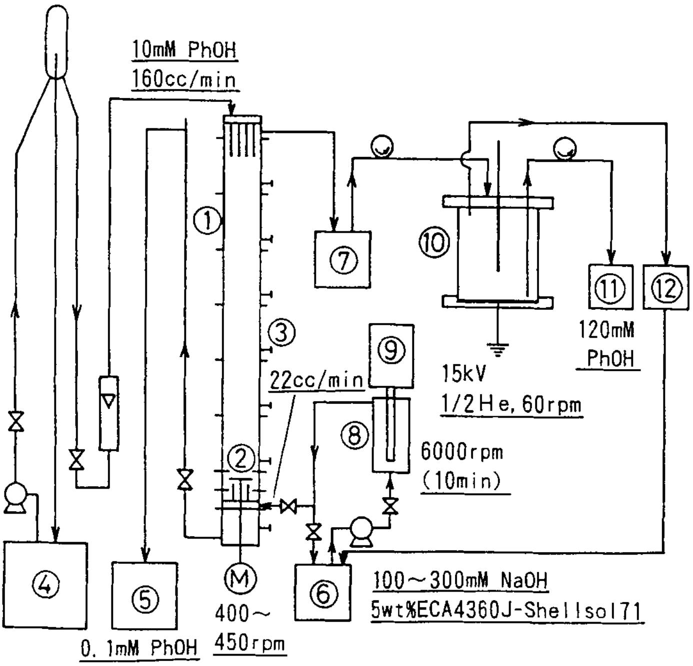

FIG. 8 Setting up a liquid surfactant membrane process for phenol removal. See the keys in Fig. 1.

dispersed phase outlet is about 120 mol/m^3, and phenol is enriched to twelve times its value in the feed concentration. In this case, 47% of the NaOH introduced into the column as the internal aqueous phase is utilized.

CONCLUSIONS

To develop a practical liquid surfactant membrane process when a stirred countercurrent column is used as the liquid membrane contact equipment, permeation experiments of phenol and the demulsification of W/O emulsions overflowing from the column were carried out under various conditions. When Span 80 was used as the surfactant, a stable continuous operation in the column was possible and a good removal of phenol could be achieved, but it was difficult to demulsify the W/O emulsion without Na_2CO_3 addition to NaOH solution of the internal aqueous phase. On the other hand, the W/O emulsion prepared by using ECA4360J as the surfactant could be demulsified even though the internal aqueous phase contained only NaOH. The extent of demulsification was greatly improved by demulsification with gentle agitation. A problem in column operation is foaming at the interface of the continuous and the dispersed emulsion phases in the top settling zone. A stable operation was possible and phenol could also be removed efficiently by using a column type without the influence of agitation near the interface.

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NOTATION

B	extent of demulsification (%)
C_F	concentration of phenol in the continuous phase (mol/m^3)
C_{F0}	concentration of phenol in the feed solution (mol/m^3)
He	height of W/O emulsion supplied into the demulsifier (m)
L	height of column (m)
N	agitating speed (1/s)
t	time (h)
V_E	flow rate of dispersed emulsion phase (cm^3/s)
V_F	flow rate of continuous phase (cm^3/s)
W	water content in W/O emulsion (kg/dm^3)
W_0	water content in W/O emulsion introduced into the column (kg/dm^3)
z	axial distance (m)
ϵ	breakage of liquid membrane (%)
ϕ'	dispersed phase holdup (—)

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