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Effect of Preparation Parameters on Leakage in Liquid Surfactant Membrane Systems

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Abstract

The stability of double emulsions or liquid surfactant membranes, which is an important topic in liquid membrane extraction processes, was investigated. The percentage of liquid membrane leakage which reflects the stability of the liquid surfactant membranes was measured as a function of time using sodium hydroxide as a tracer. Water-in-oil emulsions were prepared with SOLTROL 220, an isoparaffinic solvent, and solvent-extracted neutral oils, S100N and S500N. The surfactant studied was Span 80 (sorbitan mono-oleate). The influence of microdroplet volume fraction, surfactant weight percent, agitation speed, and emulsifying device on leakage was studied using a half fraction of 2^4 experimental design. Microdroplet volume fraction and percent surfactant showed significant effect on the extent of leakage in the case of SOLTROL 220 runs at the 99% level or better. For runs with S100N, the effect of emulsifying device and that of percent surfactant on the rate of leakage was found to be significant at the 99 and 95% level or better, respectively. Several interactions between variables were also significant. Emulsions prepared with solvent extracted neutral oils, S100N and S500N, were quite viscous, which limited the influence of factors being considered on the extent of leakage.

INTRODUCTION

Liquid membranes are typically made by first dispersing the internal phase in an immiscible liquid and then dispersing this emulsion in a third phase (commonly referred to as the external phase). Normally, the

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internal and the external phases are miscible, but are immiscible with the membrane phase separating them. Depending on the internal phase, the double emulsion is either of the water/oil/water or oil/water/oil type. The membrane phase essentially contains surfactants, membrane strengthening additives, and a base material which is a solvent for the other ingredients. Liquid membranes can be adapted for specific applications by using proper additives to obtain desired stability, permeability, and selectivity.

When the emulsion containing the microdroplets is dispersed in an external phase, it forms macrodroplets which are maintained in suspension by agitation, see Fig. 1. The size of these globules is determined by the balance of inertial forces and the surface tension. Hence a size estimate may be obtained by equating the Weber number, which is the ratio of shear stresses to surface forces, to unity. This holds provided the conditions of isotropic turbulence exist. Typical macrodroplet sizes observed in our experiments were 1-3 mm and those of the microdroplets were 1 to 10 μm. A large number of macrodroplets can be easily obtained, providing a large surface area for rapid mass transfer from the external phase to the internal phase or vice versa. After a desired degree of separation has been achieved, the agitation is stopped, allowing the macrodroplets to coalesce and form a layer of emulsion. The heavier or lighter emulsion phase can be easily separated from the external phase. The contacting operation can be either batchwise or continuous, cocurrent, or countercurrent.

Since their discovery by Li (*1*), liquid surfactant membranes have been exploited for a wide variety of separations (*2*). Typical applications are selective extraction of hydrocarbons (*3–9*); extraction of organic contaminants like phenol, acetic acid, or cresol from wastewater (*6*, *10–14*); recovery and purification of metal ions (*15–21*); controlled release of drugs or solutes; and extraction of ammonia and amines.

The main advantages of the double emulsion system are the high surface area per unit volume and short diffusion distances. In spite of these advantages, double emulsions have not been fully exploited on an industrial scale due in part to the problem of leakage. Leakage of the internal reagent is mainly due to the process of macrodroplet breakup and hence is inevitable in agitated equipment.

Experimental studies on the stability of liquid membrane systems have been performed by Hochhauser and Cussler (*16*, *22*), Martin and Davies (*18*), Takahasi et al. (*23*), Kita et al. (*24*), Tanaka et al. (*25*), and Kondo et al. (*17*). It is difficult to compare these findings since the liquid membrane systems used were different. Often the experiments conducted were over a relatively narrow range of conditions and did not lead to a

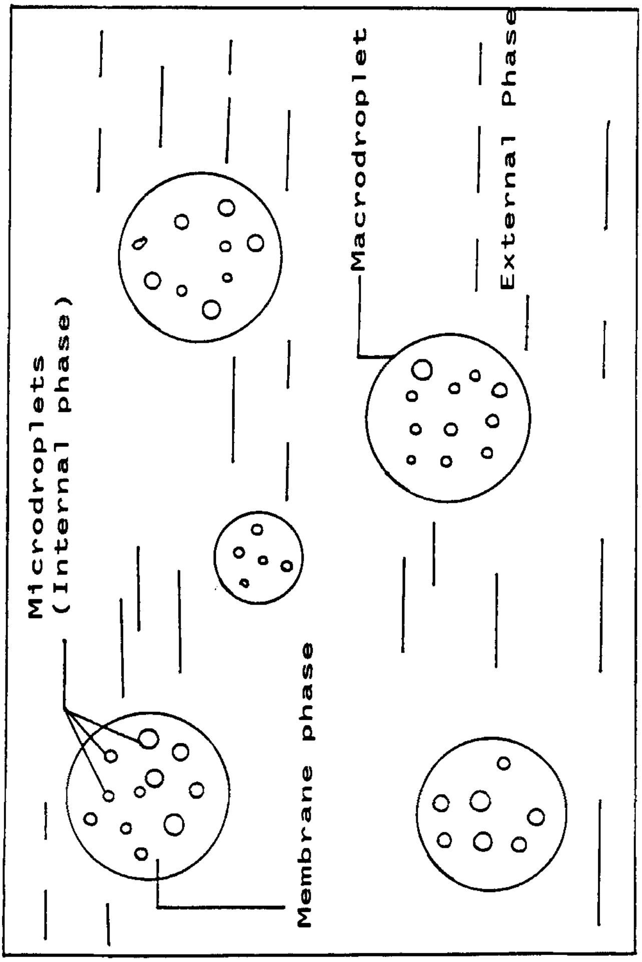

FIG. 1. Liquid membrane system.

full appreciation of the total process. A model for leakage developed by us and published elsewhere (*26*) is in qualitative agreement with all of the observations of these investigators.

Experimental data for leakage of sodium hydroxide into deionized water were measured. The experimental system and procedure were designed to provide a reasonable test of the stochastic model developed for leakage in our aforementioned work (*26*). The purpose of this paper is to present the results of a half fraction of 2^4 factorial design used to study the effects of four experimental factors on the extent and rate of leakage. The factors studied were percent surfactant in the membrane phase, speed of agitation at the extraction stage, emulsifying device (ultrasonic emulsifier or high-speed blender), and volume fraction of microdroplets in the emulsion.

EXPERIMENTAL

Experimental Design

Factorial design of experiments (*27*, *28*) provides a systematic procedure to study the effect of several factors in an objective function. This design not only requires the least number of experiments to evaluate the response due to a particular factor, but also provides insight into the interaction effects of the factors under study.

Leakage of the internal reagent of the liquid membrane into the external phase was assumed to be due to breakage of liquid membranes only. That is, the transfer of internal reagent (NaOH) was attributed to mechanical rupture of microdroplets, neglecting the diffusion of NaOH through the liquid membrane. The percent leakage is then given by

$$L = \frac{(C_t - C_0)(1 - \varphi_m)100}{C_i \varphi_M \varphi_m} \tag{1}$$

where C_i is the concentration of NaOH used in preparing liquid membranes. Here C_t and C_0 are NaOH concentrations in the external phase at time t and $t = 0$, respectively, and φ_m and φ_M are the volume fractions of microdroplets in the liquid membrane and macrodroplets in the overall system, respectively.

The following factors were chosen for study:

(1) Percent surfactant in the membrane phase (w/w)

(2) Speed of agitator at the extraction stage
(3) Mixing device: High-speed blender and Ultrasonicator
(4) Volume fraction of microdroplets in the emulsion

A fractional factorial design provides essentially the same information as a simple factorial design, in fewer experiments. Hence it is usually used in cases with more than three factors to prevent the number of experiments from becoming unmanageable. The only assumption required is that the ternary and higher order interactions are negligible. Hence a four-variable, two level, half fraction of 2^4 factorial design resulting in 8 experiments was used to evaluate the effect of the four factors mentioned above on the extent of leakage. The design matrix and high (+) and low (−) levels for each factor are shown in Tables 1 and 2. Experiments (1), a(d), b(d), ab, c(d), ac, bc, and abc(d) were performed. Symbol c(d) indicates Factors C and D are to be kept at the upper level and all other factors at the lower level. The last column of Table 1 shows the effects. ABC + D indicates that D is aliased with the interaction ABC. Hence effect ABC + D is due to D as well as interaction between Factors A, B, and C. The extent of leakage was quantified by L_f, the value of leakage at steady state obtained by extrapolation of a semilog plot of percent leakage versus time. Figure 2 shows a representative plot for a run with Soltrol 220. Complete design runs were carried out for emulsions prepared with the Soltrol 220, S100N, and S500N oils.

Liquid Membrane Emulsion Preparation

All the membrane emulsions were prepared in 60 mL batches in either a single speed Waring blender with a stirring speed of 21,000 rpm or an

TABLE 1
Factor Levels Used in the Experimental Runs

	Soltrol 220		S100N		S500N	
Variable	+	−	+	−	+	−
A: Agitator speed, rpm	500	275	500	370	540	370
B: Emulsifying device[a]	B	U	B	U	B	U
C: Microdroplet volume fraction	0.5	0.2	0.4	0.2	0.17	0.05
D: Percent surfactant	4	1	4	1	4	1

[a] B = Waring blender, U = Ultrasonic Dispenser.

TABLE 2
Level Choices for the Half-Fractional Factorial Design[a]

		Variables							
Run	Experiment	A	B	C	D = ABC	AB	AC	BC	Effects
1	(1)	−	−	−	−	+	+	+	Total + ABCD
2	a(d)	+	−	−	+	−	−	+	A + BCD
3	b(d)	−	+	−	+	−	+	−	B + ACD
4	ab	+	+	−	−	+	−	−	AB + CD
5	c(d)	−	−	+	+	+	−	−	C + ABD
6	ac	+	−	+	−	−	+	−	AC + BD
7	bc	−	+	+	−	−	−	+	BC + AD
8	abc(d)	+	+	+	+	+	+	+	ABC + D

[a] For details on level choice in a half-fractional experimental design the reader is referred to G. P. Box, W. G. Hunter, and J. S. Hunter, *Statistics for Experimenters*, Wiley, New York, 1978.

Ultrasonic emulsifier (Heat Systems Ultrasonics, Inc.) with an maximum power output of 475 W. The internal and external phases were emulsified in both cases for 1 min. The energy dissipation in the ultrasonicator was constant for all emulsions and was 10% of full output power. The membrane phase was 1 or 4% by weight Span 80 and the balance was an oil. Span 80 (sorbitan mono-oleate, a product of ICI America) is a surfactant with an HLB value of 4.3. Experimental runs were carried out for three different oils: Soltrol 220, a isoparaffinic solvent manufactured by Phillips Chemical Co., S100N (L.P.) and S500N, solvent-extracted neutral oils made by Exxon. S100N (L.P.) is a low pour point oil. Table 3 lists the physical properties of these oils. The two components of the membrane phase were well mixed before adding to the blender. The internal phase was 0.2 *N* NaOH prepared from NaOH pellets and deionized water. The volume fraction of internal phase varied from 0.05 to 0.5, depending upon the oil being used.

PROCEDURE

For each run, liquid membranes were made by dispersing 50 mL emulsion in 950 mL deionized water in an agitated vessel with 11 cm inner diameter and four baffles 1 cm in width. A marine-type 2-blade propeller of 5 cm diameter was driven by a motor equipped with a speed scale. This scale was calibrated using a Strobotac Type 151 stroboscopic tachometer. The agitator speed was set from 275 to 540 rpm depending on

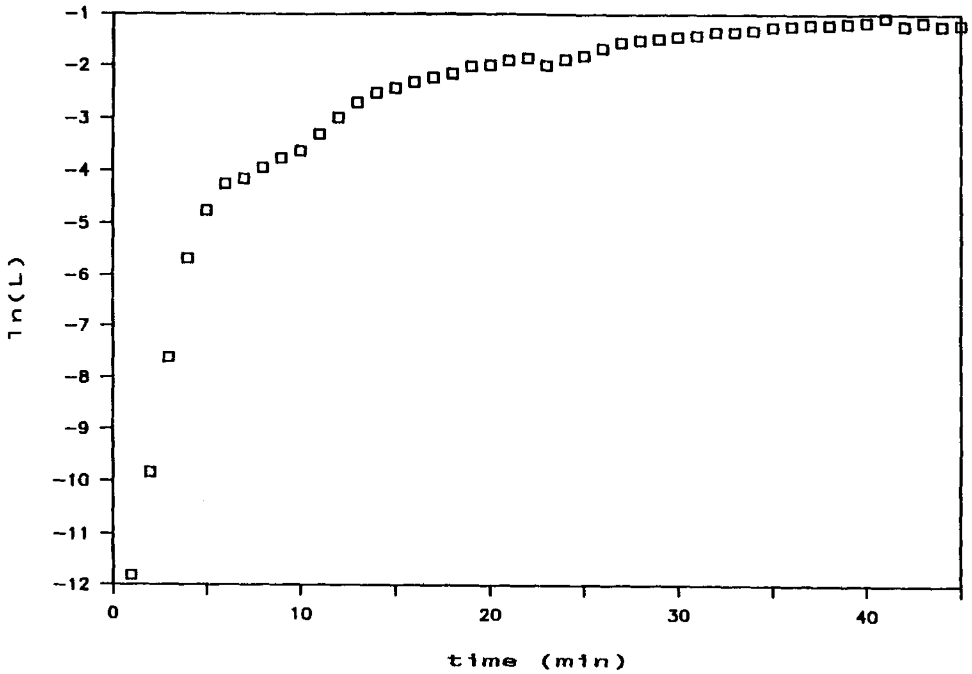

FIG. 2. Logarithm of the leakage as a function of time for Soltrol 220 Run 3.

TABLE 3
Physical Property Data for the Oils Used

Property	S100N (L.P.)	S500N	Soltrol 220
IBP, °F	550	650	452
Specific gravity 60/60 °F	0.86	0.88	0.803
Molecular weight (average)	370	500	NA
Viscosity, cP	36.9	197.5	3.7

the experiment. The pH of the external phase was noted at intervals of 1 min starting from the point of addition of the emulsion to the vessel. The pH was measured using a digital pH meter manufactured by Cole-Parmer Instrument Co.

The pH electrode immersed in the vessel was a combination electrode with calomel reference, ideal for use in organic environments without getting contaminated. The blank runs made using deionized water instead of NaOH did not indicate any effect of the organic phase on the pH reading. The experiment was terminated when the change in pH became about 0.005 pH units per minute. All experiments were conducted at 298 K. Experiments were repeated for each oil to check the reproducibility of the results obtained, which were found to be consistent.

The droplet size estimates were obtained by a photographic technique. Photographs of the emulsion slides were taken using ISO 400 black and white film by a 35mm Nikon camera attached to the microscope with a magnification of 100×. The developed film was projected on the screen of a Nikon projector with a magnification of 20×. Fifty microdroplet diameters were measured to calculate the Sauter mean diameter. The Sauter mean diameter, d_{32}, is defined as

$$d_{32} = \sum d^3 / \sum d^2 \tag{2}$$

The viscosity of the emulsion was measured using a Brookfield Viscometer. Measurements using the small sample adapter required 8 mL emulsion. The interfacial tension between the emulsion and deionized water was measured by a ring-type tensiometer. Measurements were also made for the pure oil phase and deionized water.

RESULTS AND DISCUSSION

The responses from each set of experiments were the limiting leakage value, L_f, and the time required to reach half this value, T. The former signifies the extent of leakage while the latter quantifies the rate of leakage. Tables 4 and 5 list the results for the Soltrol 220 and S100N runs. The values of L_f and T were obtained from semilog plots of leakage versus time. The total effects for the variables under study were obtained using Yates's algorithm (*28*). A partial check on this calculation is that the sum of squares increases by a factor of 2 (for two-level experiments) for each sum calculated. The mean effect for all the effects shown in Tables 4 and 5, except the mean, is the total effect divided by 2^{n-k-1}, where n is the number of variables and k is the fraction being used in the experimental design. In the present case the divisor is 2^{4-1-1} (= 4) for each effect and 8 for the mean. The sum of squares for each effect, for error variance analysis, was obtained by division of the square of the corresponding total effect by 2^{n-k} (= 8). The insignificant mean squares were averaged to determine the error variance with degree of freedom equal to the number of mean squares pooled together. The F ratio, which is the ratio of the

TABLE 4

Factor Analysis of Response L_f for Soltrol 220 Runs[a]

Run	Response	Sum 1	Sum 2	Total effect	Effects	Mean effect
(1)	3.37E-04	0.37	0.82	3.18	Mean	0.40
a(d)	0.368	0.45	2.37	0.21	A/BCD	0.05
b(d)	0.368	1.21	0.08	0.03	B/ACD	0.01
ab	0.082	1.16	0.13	−2.91	AB/CD	−0.73
c(d)	6.74E-03	0.37	0.08	1.55	C/ABD	0.39
ac	1.2	−0.29	−0.05	0.05	AC/BD	0.01
bc	1.11	1.19	−0.65	−0.13	BC/AD	−0.03
abc(d)	0.0498	−1.06	−2.25	−1.60	ABC/D	−0.40
Sum of squares	2.95	5.90	11.81	23.62		

Effects	Sum of squares	Mean square	Degrees of freedom	F ratio	Significance level
Mean	5.76E-03				
A/BCD	1.51E-04				
B/ACD					
AB/CD	1.06E+00	1.06	1	508.41	0.01
C/ABD	3.00E-01	0.30	1	144.19	0.01
AC/BD	3.30E-04				
BC/AD	2.07E-03				
ABC/D	3.20E-01	0.32	1	153.96	0.01
	Error	0.002	4		

[a] $F(0.01,4,1) = 21.2$.

TABLE 5
Factor Analysis of Response T (min) for S100N 220 Runs[a]

Run	Response	Sum 1	Sum 2	Total effect	Effects	Mean effect
(1)	34	61.00	88.5	175.50	Mean	21.9
a(d)	27	27.50	87.00	−4.50	A/BCD	−1.1
b(d)	17	52.00	−13.5	−50.50	B/ACD	−12.6
ab	10.5	35.00	9.00	21.50	AB/CD	5.4
c(d)	29	−7.00	−33.50	−1.50	C/IABD	−0.4
ac	23	−6.50	−17.00	22.50	AC/BD	5.6
bc	10	−6.00	0.50	16.50	BC/AD	4.1
abc(d)	25	15.00	21.00	20.50	ABC/D	5.1
Sum of squares	4379.3	8759	17517	35034		

Effects	Sum of squares	Mean square	Degrees of freedom	F ratio	Significance level
Mean					
A/BCD	2.53				
B/ACD	318.78	318.78	1	226.69	0.01
AB/CD	57.78	57.78	1	41.09	0.05
C/ABD	0.28				
AC/BD	63.28	63.28	1	45.00	0.05
BC/AD	34.03	34.03	1	24.00	0.05
ABC/D	52.53	52.53	1	37.36	0.05
	Error	1.41	2		

[a] $F(0.01,2,1) = 98.49$. $F(0.05,2,1) = 18.51$.

mean square of the other effects to the error variance, was evaluated to determine the confidence levels of those effects.

The effects of runs with Soltrol 220 are listed in Table 4. Assuming the ternary interactions to be negligible, we find that the effect of microdroplet volume fraction (Variable C) and percent surfactant (Variable D) are significant. Interactions AB (speed-device) and CD (volume fraction-percent surfactant) together are also significant at the 0.01 level. No information about independent effects of the interactions AB and CD can be obtained.

Table 6 gives an account of the experimental conditions for each run with Soltrol 220. Table 7 lists the values of some important parameters for these runs. Tables 8 and 9 contains similar information for S100N. The Weber number listed in these tables is the Weber number of the impeller based on the impeller diameter. Table 4 indicates that the effect of

TABLE 6
Experimental Conditions for Soltrol 220 Runs

Run[a]	Energy density, (mW/cm^3)	Emulsifying device[b]	φ_m	Percent surfactant
1	7.46	U	0.2	1
2	44.90	U	0.2	4
3	7.46	B	0.2	4
4	44.90	B	0.2	1
5	7.46	U	0.5	4
6	44.90	U	0.5	1
7	7.46	B	0.5	1
8	44.90	B	0.5	4

[a] See Tables 1 and 2 for the experimental conditions corresponding to the listed run numbers.

[b] B = High-speed blender, U = ultrasonic emulsifier.

microdroplet volume fraction (Variable C) is large. An increase in the volume fraction increases leakage. As can be seen from Tables 6 and 7, emulsion viscosity increases with volume fraction. Intuitively, one might expect this to stabilize the liquid membranes, but this effect is overpowered by the decrease in stability of liquid membranes due to the decrease in thickness of the oil phase encapsulating the microdroplets. Careful study of Tables 6 and 7 indicates that microdroplet size increases in some cases and decreases in others with the other conditions held constant, but these deviations in diameters are small and will not affect leakage to a great extent. It is natural to expect an increase in size with an

TABLE 7
Parameter Values for Soltrol 220 Runs

Run[a]	γ(dyn/cm)	We	$d_{32_{(\mu m)}}$	Emulsion viscosity (cP)
1	25.3	104	3.19	6.6
2	15.5	560	3.6	7.1
3	15.0	175	4.16	8.46
4	25.4	342	3.66	8.46
5	11.2	234	2.94	70.0
6	16.2	535	2.88	36.0
7	17.5	150	4.52	34.3
8	12.2	711	3.04	75.0

[a] See Tables 1 and 2 for the experimental conditions corresponding to the listed run numbers.

TABLE 8
Experimental Conditions for S100N Runs

Run[a]	Energy density (mW/cm^3)	Emulsifying device[b]	φ_m	Percent surfactant
1	18.20	U	0.2	1
2	44.90	U	0.2	4
3	18.20	B	0.2	4
4	44.90	B	0.2	1
5	18.20	U	0.4	4
6	44.90	U	0.4	1
7	18.20	B	0.4	1
8	44.90	B	0.4	4

[a] See Tables 1 and 2 for the experimental conditions corresponding to the listed run numbers.

[b] B = High-speed blender, U = ultrasonic emulsifier.

increase in volume fraction of microdroplets, but size is also dependent on the time of emulsification.

The percent surfactant effect (Variable D) indicates that an increase in the surfactant concentration of the oil phases causes a decrease in leakage. Higher surfactant concentration lowers the interfacial tension at the macrodroplet surface. It appears at first sight that this should result in more leakage because a lower surface tension value means easier rupture of macrodroplets. But the main stabilizing feature is the increase in the number of monolayers adsorbed at the microdroplet interface. It is known that an increase in these layers leads to increased stability of the droplet up to a limiting value. It should be noted that the higher the

TABLE 9
Parameter Values for S100N Runs

Run[a]	γ(dyn/cm)	We	$d_{32(\mu m)}$	Emulsion viscosity (cP)
1	23.7	201	2.12	123
2	17.1	507	1.73	146
3	17.6	270	1.77	205
4	23.9	363	2.32	200
5	6.0	793	2.54	571
6	18.2	477	2.10	200
7	16.0	298	3.37	257
8	8.1	1071	2.88	1333

[a] See Tables 1 and 2 for the experimental conditions corresponding to the listed run numbers.

energy dissipation, the less is the effect of surfactant on microdroplet size, as is observed in our case from Tables 6 and 7.

The analysis of effects for S100N with response *T* is given in Table 5. The composite binary interactions shown in this table are significant at the 0.05 level. Variable D, that is, surfactant concentration, is significant between the 0.01 and 0.05 level. This variable causes a positive effect on *T*. This means the leakage is slower for higher surfactant concentration. This is consistent with the observed surfactant effect for the Soltrol 220 runs.

The Effect B (emulsifying device) is significant at the 99% confidence level. The use of a high-speed blender rather than an ultrasonic disperser for making the emulsion causes faster leakage. It is noted from Tables 8 and 9 that the viscosity of the emulsion increases if a high-speed blender is used, but the stability acquired due to this is offset by the increase in the size of the microdroplets with the use of the blender. The larger size is mainly due to the lower energy density in a blender as compared to a ultrasonic disperser. The speed of agitation and volume fraction of microdroplets are not significant for S100N runs. This is mainly due to the high viscosity of S100N oil. These factors produce significant effects only in coordination with other factors.

For experimental runs made with S500N, none of the four factors was found to be significant at less than the 0.05 level. Unlike emulsions prepared with Soltrol 220, those prepared with the solvent-extracted neutral oils, S100N and S500N, were highly viscous. This limited the influence of the factors under study on the extent of leakage.

CONCLUSIONS

The percentage of liquid membrane leakage, which is indicative of the stability of liquid surfactant membranes, was measured as a function of time by using sodium hydroxide as tracer. The water-in-oil emulsions were prepared with SOLTROL 220, an isoparaffinic solvent, and solvent-extracted neutral oils, S100N and S500N. The influence of microdroplet volume fraction, weight percent surfactant, agitation speed, and emulsifying device on percent leakage was studied using a half fraction of 2^4 experimental design.

Microdroplet volume fraction and percent surfactant showed significant effect on the extent of leakage in the case of SOLTROL 220 runs at the 99% level or better. For runs with S100N, the effect of emulsifying device and that of percent surfactant on the rate of leakage was found to be significant at the 99 and 95% level or better, respectively. Several interactions between variables were also significant. Emulsions prepared with solvent-extracted neutral oils, S100N and S500N, were quite viscous, which limited the influence of the factors under study on the extent of leakage. Hence, emulsions prepared with high viscosity oils will be, in general, more stable. It is also found that high viscosity oils allow a smaller amount of internal reagent to be emulsified, but high viscosity oils are still desirable for low leakage rates since a microdroplet volume fraction of 0.2 is usually sufficient for extraction applications. We also conclude that for low viscosity oils the leakage can be decreased by decreasing the microdroplet volume fraction or increasing the surfactant weight percent.

SYMBOLS

C_i	concentration of NaOH in microdroplets (mol/L)
C_0	initial concentration of NaOH in the external phase (mol/L)
C_t	concentration of NaOH in the external phase at time t (mol/L)
d	diameter of microdroplets (m)
d_{32}	Sauter mean diameter (m)
L	percentage of internal reagent leaked out from liquid surfactant membranes (%)

Greek Letters

φ_m	volume fraction of microdroplets in liquid membranes
φ_M	volume fraction of macrodroplets in the overall system

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