Liquid-liquid Extraction in a Pulsed Perforated-plate Column

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Liquid extractions of benzoic acid–toluene solutions by means of water were studied under varying flow rates of both solvents in a 2-in. perforated-plate column to which pulsations of different frequencies and amplitudes were applied. Studies on reversal of the phase of the dispersion were made. With fixed flow rates of both solvents, the rate of extraction increased much more rapidly when the flow through the perforation became turbulent. The results are correlated by means of Reynolds number based on the flow through the perforations and are calculated from the product of the frequency and amplitude of the pulsations.

Since Sherwood, Evans, and Longcor (*14*) found in their study of extraction from single drops that 40 to 50% of the extraction is accomplished before a drop leaves the nozzle, frequent reformation of droplets of the dispersed phase should greatly improve the efficiency of extraction. A perforated-plate column should achieve this result, and a series of investigations on extraction in perforated-plate column was made by several investigators (*1*, *11*, *12*, *13*, and *16*).

Later, extraction from single drops was further studied. In a consideration of the mechanism of solute transfer in spray towers Licht and Conway (*10*) reported that the amounts of acetic acid extracted during drop formation by use of isopropyl ether, methyl isobutyl ketone, and ethyl acetate as solvent are 5, 8, and 17% respectively. These values are, however, less than those reported by Sherwood et al. Data on the extraction of acetic acid from benzene drops of known volume by water have been reported by West et al. (*17*), whose results have shown that approximately 14 to 20% extraction was obtained during drop formation.

The rate of emission of droplets through the perforations of the plate is limited by the physical properties of both phases and the size of perforations. Furthermore, each phase passes only once through each plate on its way through the column. It is thus believed that if pulsations were applied to the fluids in the column, so that the fluids would pass back and forth through the same plate before they leave for the next one, not only the frequency but also the speed of reformation of droplets through each plate would be greatly increased, depending on the frequency and amplitude of the pulsation applied. Dijck (*6*) was granted a patent on an agitated perforated-plate column for liquid-liquid extraction based on this principle. In Dijck's column the pulsation was achieved by a reciprocating mechanism which lowered and raised the perforated plates in relation to the column and its liquid contents. Alternatively, the plates or the packing may be made stationary and the liquids reciprocated by means of an external piston and cylinder. A preliminary investigation on performance of a pulsed packed column for liquid-liquid extraction was reported by Feick and Anderson (*8*).

From the results reported by the previous investigators on perforated-plate towers and pulsed columns, it seems desirable to investigate a pulsed perforated-plate column for liquid-liquid extraction. Improvement of the performance of such a column should be expected.

The variables investigated in this work were flow rates of both phases, frequency and amplitude of pulsation, and the reversal of phase of dispersion. The effects of these variables on the performance of a pulsed perforated-plate column were studied.

EQUIPMENT AND MATERIALS

A schematic diagram of the extraction unit consisting of perforated-plate column, separators, rotameter, flow controls, pumps, storage tanks, and other auxiliary equipment is shown in Figure 1.

Extraction Column. The column consisted of eleven segments of Pyrex pipe spacers, 2 in. I.D., 2⅝ in. O.D., and 2 in. long, which were manufactured by the Corning Glass Company. The disengaging chambers at the top and bottom of the column were standard 2-in. cast iron crosses, between which the perforated plates and glass spacers were all held together by four ⅝-in. tie rods.

Perforated Plates. The brass plates were 5/32 in. thick and perforated with eighty-five holes of 1/16-in. diameter. The fraction of cross-sectional area of the column covered by these eighty-five holes was 8.51%. The center-to-center distance between the holes was 3/16-in., and the holes were arranged in an equilateral-triangular pattern. In order to reduce the pressure drop through the plates, all the holes on each plate were countersunk on both sides to a depth of 1/16 in. Sample above each plate was withdrawn from the column for analysis through a hole drilled horizontally through the plate and connected with a cock valve which was directly attached to the plate outside the column. Between the plate and the glass spacer a ⅛-in. asbestos gasket was used at each

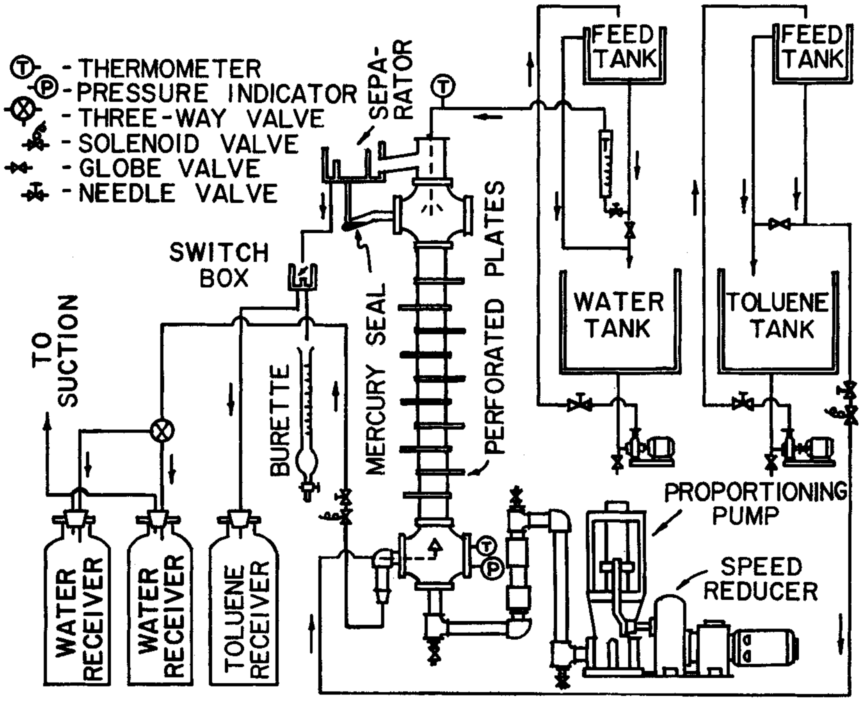

Fig. 1. Flow diagram of equipment.

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joint. A thin layer of graphite was pasted over the surfaces of the gaskets before they were assembled. In all this work ten plates were used, with a plate spacing of $2\frac{1}{4}$ in.

Separators. Two separators were used, one at each end of the column, to facilitate coalescence of all the droplets so as to separate completely the two liquid phases before they were withdrawn from the column. The top separator consisted of a rectangular brass box with a width, length, and height of 5, 7, and 3 in. respectively. The weir, with a height and width of $1\frac{1}{2}$ and 5 in. respectively, was located at a distance of 1 in. from the outlet end. A baffle was used near the entrance of the separator to prevent the emulsion from flowing straight through the separator. In the return line from the separator to the column for the heavier phase (water) a mercury seal was used to restrict the flow to one direction only. The bottom separator was made of a piece of 1-in. pipe, 4 in. long.

Feed Distributors. The distributor for toluene feed at the bottom of the column was made of 16-gauge aluminum sheet in the shape of a cone with a vertical angle of 60 deg. and a base $1\frac{1}{2}$ in. in diameter. On the surface of the cone seventy-eight holes 1/16 in. in diameter were spaced in such a way that their projections on the base would give the same number of holes per unit area. In the center of the bottom disengaging chamber the cone was mounted so that its presence did not restrict the flow of the liquids through the column. A piece of $\frac{3}{8}$-in. stainless pipe projecting down to the center of the top disengaging chamber was used to introduce the water feed.

Pulsing Unit. The pulsations were generated by a proportioning pump which was connected with the motor through a variable-speed reducer covering an output speed range from 0 to 110 rev./min. The plunger diameter was $2\frac{1}{2}$ in.

In the U bend of the 1-in. line connecting the pump and the column a mercury seal was used to separate the fluid in the proportioning pump, water in this case, from the fluids in the column. A surge section was also provided in the pulsing line to prevent the possibility of mercury leaking into the pump. The amount of mercury used was carefully adjusted so that it was enough to stop the possible circulation of fluid from pump to the column or vice versa, but not sufficient to fill up the lower part of the bottom disengaging chamber. A vent was provided to purge any gas entrained in the pulsing line.

Feed Controls. Two $\frac{1}{2}$-in. solenoid valves, full port type and normally closed, were used to control the streams fed into or taken out of the column. Two sets of switches for these solenoid valves were mounted on the revolving arm of the proportioning pump so that they were open only when the plunger of the pump came to its top and bottom positions respectively. The opening of the valves was thus synchronized with the strokes of the pump. The duration of opening each valve, as fixed by the length of contact of the switch, was about 7% of the time for a complete revolution. In other words, the volume of displacement by the pump was not appreciably affected by the number of streams added to or withdrawn from the column.

Feed Supplies and Other Accessories. All the lines for toluene solution containing benzoic acid were $\frac{3}{8}$-in. stainless steel except the valves. Glass tanks were used for toluene–benzoic acid solution.

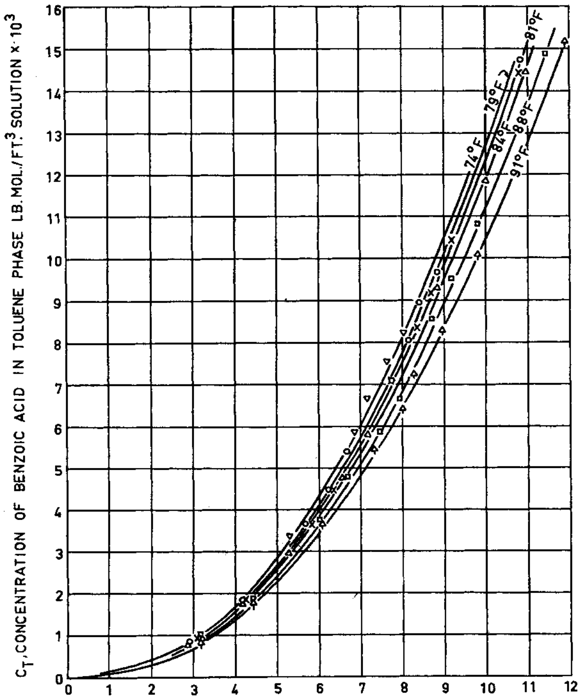

Fig. 2. Equilibrium distribution of benzoic acid–toluene–water system.

A constant head for both water and toluene feed was maintained by circulating the feeds from the storage to two constant-level tanks, which were located 20 ft. above the top of the column.

At the higher water flow rates it was found necessary to apply suction to the water-receiving bottles in order to maintain the desired flow rate of water through the solenoid valve, which was closed 93% of the time.

A pressure indicator was made of a piece of rubber tubing on which a slot approximately $\frac{3}{8}$ in. long was cut with a sharp blade. One end was plugged up, and the other was connected by glass tubing to the bottom disengaging chamber. This apparatus amounts to a Bunsen valve commonly used on laboratory wash bottles. The rubber tubing itself was enclosed in a glass tube, which was provided with a long glass tubing to indicate the height of the water column for pressure determination. The recorded pressure, therefore, was the maximum pressure at the center of the bottom disengaging chamber, including both the pressure to overcome the friction through the plates and the static head of the fluid mixture in the column. Actually the pressure-drop relationship was more or less sinusoidal and had both positive and negative peaks. The technique reported here was a measure of the peak pressure drop on the upstroke. Since some finite pressure differentials might have to be maintained for periods of time in order to open the split-tube valve to permit it to communicate pressure from inside to outside, the pressure drop obtained by this method might be slightly lower than the actual value. However, this method was used because of its simplicity.

Materials. Merck's reagent-grade toluene, Baker's C. P. grade benzoic acid, and Atlanta city water were used in determining the equilibrium-distribution curve and making the runs. The toluene solution after more benzoic acid had been added to make up the proper concentration was used over and over again; the water extract was discarded. The city water was saturated with toluene before use throughout this work.

EXPERIMENTAL

Equilibrium-distribution Curve

Equilibrium distribution of the system benzoic acid–toluene–water was determined for a temperature range from 74° to 91°F. The results were plotted in Figure 2. In the analysis an alcoholic solution of sodium hydroxide, $0.01N$, was used for titration with phenolphthalein as the indicator.

Procedure

In starting up a run the column was filled with a water solution of benzoic acid from the previous run up to the top plate and the rest was filled with fresh water. The exact concentration of the initial water solution was immaterial because the purpose of charging the column with this water solution was to hasten the attainment of steady state. Both the fresh water and the toluene solution were then pumped into the overhead constant-level tanks, and the regulating valves were set for desired flow rates. The proportioning pump and the switches for the solenoid valves were turned on. By regulating the suction applied to the water-receiving bottles and the valve setting on the exit-water line, one could set the level of the interface and maintain it constant in the top separator for runs with water as the *continuous* phase. Constancy of the exit composition of both streams was tested by frequent titration of samples withdrawn at 5-min. intervals. A steady state was reached after a total throughput of three to four times the column volume, depending on the flow rates used. For the case with water as the discontinuous phase, the interface was set in the bottom-disengaging chamber.

When steady state was obtained, the by-pass for each stream was closed, the electric time recorder was started, and the exit streams were collected in the receivers provided for that purpose. At the conclusion of the run the time on the recorder was noted and the by-pass valves were opened.

During the run the exit toluene solution was periodically led off through the switch box into a graduated burette, where its flow was measured within a fixed time interval. The water rate was indicated by the rotameter.

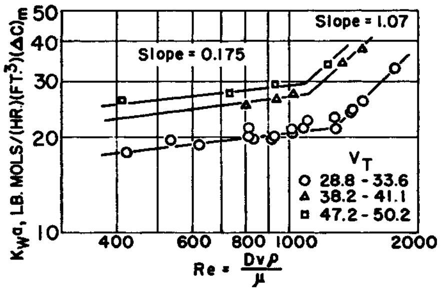

Fig. 3. Effect of pulsation on over-all transfer coefficient with constant flow rates, toluene dispersed.

At the end of a run the proportioning pump, solenoid valves, and water feed were turned off simultaneously. The thickness of toluene layer separated under each plate was measured and the water samples were withdrawn from each plate for titration. After all the toluene in the top separator had been decanted, the proportioning pump was run again with the water feed on, until all the toluene solution in the column rose and overflowed into a graduated cylinder, where its total volume in the column was measured as holdup. This total volume could be checked with the sum of all layers previously measured under each plate before they were combined. Holdup is therefore defined as the volume percentage of the dispersed phase in the liquid mixture which is present in the column during a steady operation.

Concentrations of both exit streams were analyzed by titrating the samples taken from the receivers with $0.01N$ alcoholic solution of sodium hydroxide with phenolphthalein as indicator. Blanks on fresh water which had been saturated with toluene were also determined. Plate concentrations of toluene layers were not analyzed but calculated by material balance after the concentrations of water layers and flow rates were known.

The toluene solution containing benzoic acid was replaced by a fresh batch after every five or six runs, and the viscosity and surface tension of the used batch were determined. Throughout this investigation the viscosity of the toluene feed varied between 0.6230 and 0.6575 centipoise and the surface tension between 29.5 and 30 dynes/cm.; both properties were measured at 80.5°F.

The length of stroke traversed by the plunger of the proportioning pump was recorded by a tracing device attached to the upper end of the plunger.

CALCULATIONS

In this study all runs of which the error in material balance was greater than 5% were discarded.

The effective volume used in the calculation of $K_w a$ values from Equation (1) was 0.1015 cu. ft., which included the disengaging chambers, the top separator, and the glass column. The slope dC_T/dC_W of equilibrium curve at different temperatures was taken at an average toluene concentration. For each run this slope was determined according to the temperature.

The Reynolds number through the perforations was calculated by use of the density and viscosity of a two-phase liquid mixture containing 16% toluene by volume, which was the average analysis of the holdup of all the runs. Although a rigorous calculation of ρ and μ by use of the actual holdup analysis for each run was possible, nevertheless the variation in holdup analysis had little effect upon the ratio ρ/μ since both ρ and μ decreased with increasing toluene holdup. The average values of ρ and μ of the liquid mixture were calculated by taking the sum of individual density and viscosity of each pure component in proportion to their respective volumes. It might also be pointed out that during each flow wave the initial movement of liquid mixture through the perforations was probably streamline although the major portion of the flow was turbulent. With this fact that the viscosity of the liquid mixture varied not only with the composition but also with the manner of dispersion, this method of averaging the kinematic viscosity was only an approximation. For the same reason, an average of room temperature of 80°F. was used to calculate ρ/μ for all the runs. The term v, the arithmetic average velocity of liquid mixture through the perforations, was calculated from the volume of displacement of the plunger in the pump and the area of perforations on the plate. Table 1* summarizes the experimental data and calculated results.

THEORY

Colburn's H.T.U. method of correlation (*4*) has been applied by the previous authors (*1, 13, 16*) to their data on liquid-liquid extraction in perforated-plate column. The equations may be summarized as follows:

$$K_W a = \frac{N/\theta}{(\Delta C)_m V'} \qquad (1)$$

$$(\text{H.T.U.})_{OW} = \frac{V_W}{K_W a} \qquad (2)$$

$$(\text{H.T.U.})_{OW} = H \frac{V_W}{V_T} (\text{H.T.U.})_T + (\text{H.T.U.})_W \qquad (3)$$

In this investigation with the superimposition of pulsation on a perforated-plate column, the Reynolds number through the perforations was used to characterize the pulsation in the column.

Mass transfer film coefficients in wetted-wall columns have been shown by Gilliland and Sherwood (*9*) and Chilton and Colburn (*3*) to be functions of Reynolds and Schmidt numbers. Dodge and Dwyer (*7*), Comings and Briggs (*5*), and Brinsmade and Bliss (*2*) reported that mass transfer film coefficients are affected by the flow rates of both phases. The mass transfer film coefficients for liquid-liquid extraction in a given pulsed column under constant temperature and certain-sized perforations can be expressed through dimensional analysis as

$$k_T a = \phi_T (V_T)^n (V_W)^m (Re)^d \qquad (4)$$

$$k_W a = \phi_W (V_T)^t (V_W)^s (Re)^r \qquad (5)$$

Brinsmade and Bliss (*2*) pointed out that their method of correlation could be applied if either individual coefficient is independent of one flow rate. The exponent t in Equation (5) can be assumed to be zero. The effect of pulsation on both film coefficients is assumed to be same, that is, d equal to r. Equations (4) and (5) are combined to give the over-all coefficient

$$\frac{(Re)^d}{K_W a H} = \frac{1}{\phi_T (V_T)^n (V_W)^m} + \frac{1}{H \phi_W (V_W)^s} \qquad (6)$$

or

$$(\text{H.T.U.})_{OW} (Re)^d = \frac{H (V_W)^{1-m}}{\phi_T (V_T)^n} + \frac{1}{\phi_W (V_W)^{s-1}} \qquad (7)$$

If the water film resistance is small—as is the case with the benzoic acid-toluene-water system—the second term on the right of Equation (6) can be omitted, and the following equation is obtained:

$$\frac{K_W a H}{(Re)^d} = \phi_T (V_T)^n (V_W)^m \qquad (8)$$

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*Table 1 has been deposited as document 5121 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D.C., and may be obtained for $1.25 for photoprints or $1.25 for 35-mm. microfilm.

DISCUSSION OF RESULTS

Unique Characteristics of a Pulsed Column with Toluene Dispersed

With constant flow rates of both phases, the effect of pulsation expressed in terms of Reynolds number *Re* on the rate of mass transfer is shown in Figure 3. Each curve shows a break in slope at a Reynolds number between 1,050 and 1,200 which divides the curve into two regions. The lower region, where the effect of Reynolds number on $K_W a$ value is small, is called the *streamline* region and the other the *turbulent* region.

With varying flow rates of both phases, the slopes of both streamline and turbulent regions remain unchanged, as shown by curves *A*, *B*, and *C* in Figure 3. The critical Reynolds number, however, decreases slightly with increasing flow rates.

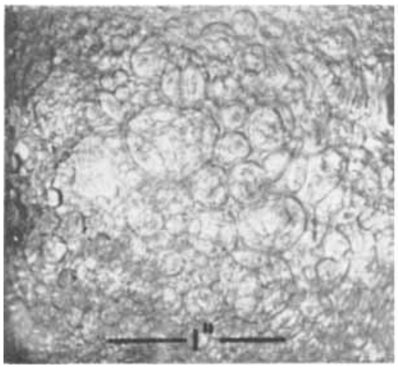

Fig. 4. Dispersion of toluene, Re = 417.

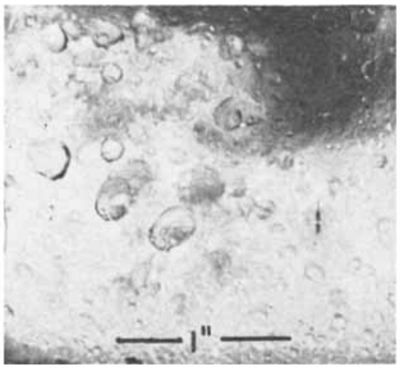

Fig. 5. Dispersion of toluene, Re = 1029.

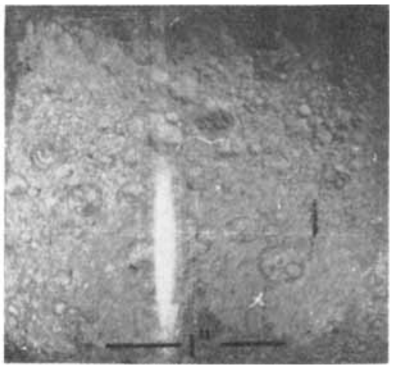

Fig. 6. Dispersion of toluene, Re = 1475.

When the column was operating near the lower end of the streamline region with low Reynolds number, a cell-like formation with large drops of water phase surrounded by toluene films was observed as shown in Figure 4. The reason for this cell-formation phenomenon is that during the suction stroke a water layer which had been accumulated above the plate by settling during the short interval of stroke reversal was slowly sucked downward through the perforations against a thin layer of toluene accumulated under the plate and then expanded into drops surrounded by a toluene film which has the lower surface tension. This phenomenon of forming cell-like structure by dispersion of large water drops between toluene films is similar to the formation of foams by blowing of air into a soap solution.

The toluene phase in the cell-like mixture began to coalesce into droplets after leaving the top plate and rose as a dispersed phase in the top disengaging chamber. In the lower end of the streamline region, therefore, the water phase was actually discontinuous in the space between plates. To avoid confusion with the case to be described later where water was the true dispersed phase throughout the column, the water phase, despite its cell-like formation, was still considered as continuous in this lower part of the streamline region.

As the Reynolds number increased in the streamline region, the cell size became smaller and the cell-formation phenomenon gradually disappeared (Figure 5). Upon further increase of Reynolds number beyond 1,200, the toluene phase became clearly dispersed with vigorous turbulence in the water phase, as shown in Figure 6.

As the Reynolds number was further increased, the size of the dispersed droplets became finer and finer, until with a Reynolds number around 1,800 the fine toluene droplets in the exit water stream could not be settled out in the bottom separator. With all toluene and water rates investigated, flooding occurred at a Reynolds number of approximately 1,800. Curves in Figure 3, therefore, should not be extrapolated at the upper end.

When operating at the lower end of the streamline region with a Reynolds number around 400, the column was only partially filled with cell-like dispersions, the lower space of each column segment between plates being left mainly full of continuous water phase. The column seemed to be underloaded. For this reason, runs with Reynolds number less than 400 were not attempted.

When pressure drop across the column was plotted vs. Reynolds number in Figure 7, the same shape of curve was obtained as in Figure 3, with the same critical Reynolds number around 1,200.

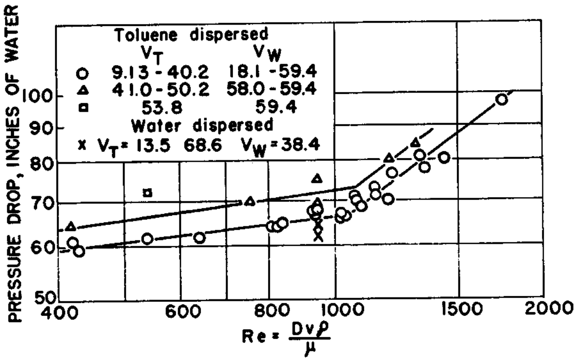

Fig. 7. Effect of pulsation on pressure drop.

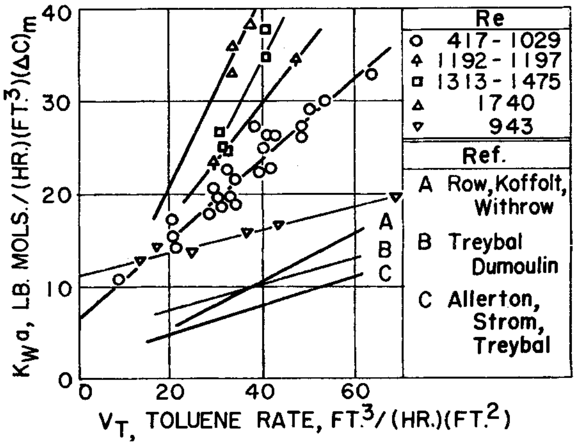

Fig. 8. Extraction coefficient in pulsed (curves with data points) and nonpulsed (curves *A*, *B*, and *C*) perforated-plate columns.

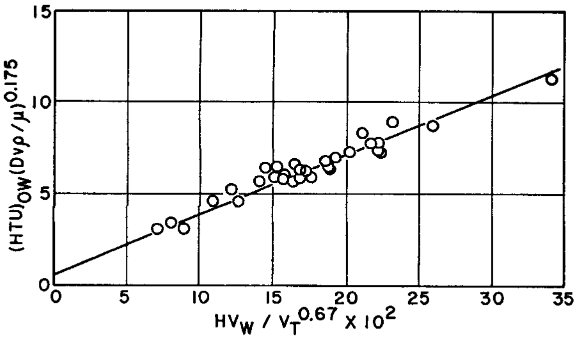

Fig. 9. Over-all $(H.T.U.)_{OW}$ in the streamline region, toluene dispersed.

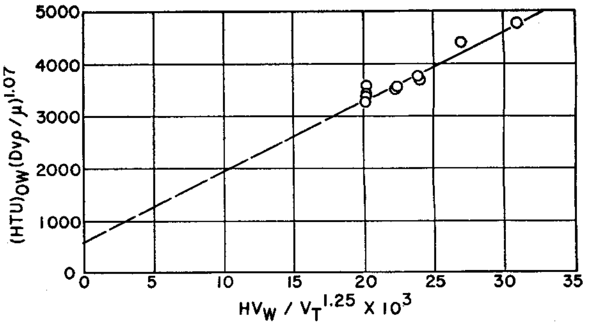

Fig. 10. Over-all $(H.T.U.)_{OW}$ in the turbulent region, toluene dispersed.

To study the cause of the break of the curves in Figure 3 and 7, the column was operated with water alone under the same conditions as with both water and toluene. Water fed into the bottom of the column through the toluene feed line was kept at a rate of 40.2 cu. ft./(sq. ft.)(hr.) for all the runs. A set of pressure-drop data was taken with varying Reynolds numbers and recorded in Table 2. These data, if plotted in Figure 7, showed the same shape of curve with a break at a Reynolds number around 1,200. It is evident that the break of curves in Figures 3 and 7 is due to the change of nature of flow of liquid mixture in the column, that is, from streamline to turbulent flow.

TABLE 2. PRESSURE DROP WITH WATER PHASE ALONE

Stroke length, in.	Frequency strokes/ min.	Re	Pressure drop, in. of water
1.44	9.75	425	61.4
1.44	27.50	1197	75.3
1.44	30.40	1325	80.5
1.44	37.10	1615	89.3

Effect of Varying Pulsation

It may be seen from Figure 3 that the slopes are 0.175 and 1.07 for the streamline and turbulent regions respectively. The mass transfer coefficients were greatly increased by operation in the turbulent region. Similarly, the pressure drop across the column was increased in the same manner as the mass transfer coefficients, with slopes 0.155 and 0.85 for streamline and turbulent regions respectively.

For the data shown in Figure 3, different stroke length and frequencies were used. The effect of pulsation on transfer coefficients was found to be dependent on the product of the stroke length and frequency of the pulsation. For practical consideration, low frequency and long stroke length seem to be preferable; however, a constant interphase was more easily maintained during operation under short strokes. In this work a stroke length of 1.44 in. or a height of 2.25 in. of displacement in the column was found to be satisfactory; in other words, there was a linear displacement of one plate spacing.

Effect of Varying Flow Rates

With a constant Reynolds number, a series of runs was made to study the effect of flow rates of both phases on over-all mass transfer coefficients. In the streamline region the over-all mass transfer coefficient was only slightly affected by the water rate $[K_Wa = \alpha(V_W)^{0.11}]$ with toluene rate held constant, but considerably more by the toluene rate $[K_Wa = \beta(V_T)^{0.55}]$ with water rate held constant. In the turbulent region, the effect of water rate on the over-all mass transfer coefficient was still small $[K_Wa = \gamma(V_W)^{0.23}]$, and the toluene rate had a large influence upon this coefficient $[K_Wa = \delta(V_T)^{1.2}]$.

In Figure 8 K_Wa is plotted vs. V_T with Reynolds number as the parameter. The over-all transfer coefficient increases more rapidly with toluene rate as the Reynolds number increases.

With water as the dispersed phase, the rate of increase of K_Wa values with increasing toluene rate is small. It seems that with either phase being dispersed, the over-all transfer coefficient is affected mainly by the flow rate of the dispersed phase and slightly by the continuous phase.

H.T.U. Correlations

In the investigations (*1*, *13*, *16*) reported in the literature, the results on liquid-liquid extraction in a perforated-plate column were correlated by Equation (3) and values of $(H.T.U.)_W$, the intercept, and $(H.T.U.)_T$, the slope, were obtained from the plot of $(H.T.U.)_{OW}$ vs. $H(V_W/V_T)$. $(H.T.U.)_W$ and $(H.T.U.)_T$ values obtained from a similar plot by use of the present results are given in Table 3. Both $(H.T.U.)_W$ and $(H.T.U.)_T$ values or both film resistances decreased as the Reynolds number increased.

When $(H.T.U.)_{OW}$ was plotted vs. $H(V_W/V_T)$ with the present results, runs 5, 6, and 30, where the flow-rate ratio of water to toluene was high, did not fall on the straight line. This fact agrees with the finding of Row, Koffolt, and Withrow (*13*) that there was marked deviation from straight line with low toluene rate and high water rate.

The correlation of results by Equation (7) is more satisfactory. By trial and error, the best straight line was obtained when a value of n equal to 0.67 for the streamline region was used as shown in Figure 9. The intercept $1/(\phi_W(V_W)^{n-1})$ is 0.55 and the slope $1/\phi_T$ is 33.1. All the data fell on a straight line. Similarly for the turbulent region, the value of n was found to be 1.25, and the results are shown in Figure 10, giving an intercept

TABLE 3 COMPARISON OF H.T.U.'S IN PERFORATED-PLATE COLUMNS

Plate characteristics Spacing, in.	Hole Diam., in.	Reference	Continuous phase	Dispersed phase	Flow rates, Cu. ft./(hr.)(sq. ft.) V_W	V_T	$(H.T.U.)_W$	$(H.T.U.)_T$	*Re*, $Dv\rho/\mu$
6	3/32	*13*	Water	Toluene	11.8–37.1	12.5–47.7	0.25	106	
6	1/8	*13*	Water	Toluene	11.8–37.1	11.8–47.7	0.25	115	
3	3/16	*16*	Water	Toluene	15.1–32.5	9.7–81.8	0.65	30	
6	3/16	*16*	Water	Toluene	22.9–41.5	30.2–76.8	0.25	63	
9	3/16	*16*	Water	Toluene	23.2–38.9	35.8–78.6	0.25	68	
4¾	3/16	*1*	Water	Kerosene	24.7–13.6	33.8–16.2	0.65		
2¼	1/16	Present	Water	Toluene	18.1–38.4	9.13–63.6	0.50	26.6	417–1,150
2¼	1/16	data, pulsed	Water	Toluene	38.4–58.0	30.7–41.1	0.42	20.4	1,300–1,500
2¼	1/16	column	Toluene	Water	38.4	13.5–68.6	1.85	10.0	943

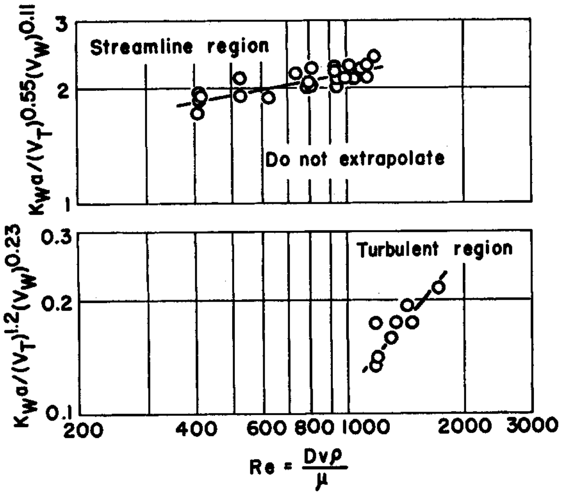

Fig. 11. Correlation of over-all transfer coefficient with Reynolds number, toluene dispersed.

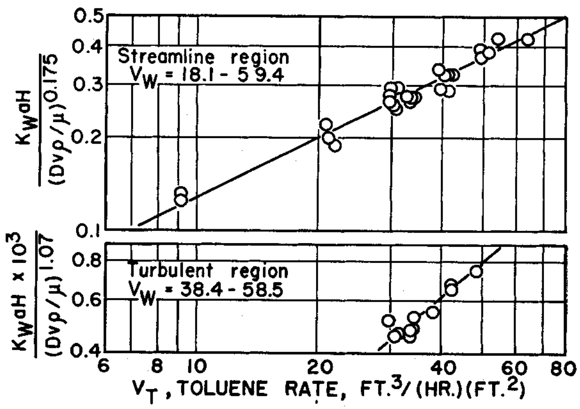

Fig. 12. Effect of toluene rate on $K_W a$, toluene dispersed.

550 and a slope 1.36×10^5. Since the value of m is small, it is neglected in plotting Equation (7) on Figures 9 and 10.

According to Equation (7), a series of lines should be drawn in Figures 9 and 10 with each line representing one constant water rate. However, since all points within the range of the water rates studied fell so close to the same straight line, it is obvious that such procedure is not necessary. The intercept and the slope of the curves in Figures 9 and 10 were, therefore, obtained by use of an average water rate.

Approximate Correlation

The results are correlated by Equation (8) and plotted in Figure 11. It is seen that the value of exponent m is small compared with that of n, and so Figure 11 can be replotted without much error, as shown in Figure 12 by the following equation:

$$\frac{K_W aH}{(Re)^d} = \phi_T'(V_T)^n \qquad (9)$$

Most of the data on liquid-liquid extraction in perforated-plate columns with the benzoic acid–toluene–water system reported in the literature were correlated fairly well by a log-log plot of H.T.U. vs. V_W/V_T as shown by Treybal (*15*). A similar plot was made in Figure 13 with the present results and the correlation was not so satisfactory as that obtained by using Equation (7).

Water as Dispersed Phase

In the operation with water as discontinuous phase, the interface was maintained in the bottom disengaging chamber, and all the other operating conditions remained the same. The droplet size of the dispersed water phase was found to be small, as shown in Figure 14.

The results with water as dispersed phase were plotted in Figure 8, which shows that with a high toluene rate above 15 cu. ft./(hr.)(sq. ft.) a lower transfer coefficient was obtained by dispersing water in toluene, but at low toluene rate below 15 cu. ft./(hr.)(sq. ft.) a higher transfer coefficient was obtained. Whether the water or toluene phase should be dispersed in order to obtain the highest transfer coefficient will consequently depend on the toluene rate used. The low values of transfer coefficients with water as discontinuous phase and a toluene rate above 15 cu. ft./(hr.)(sq. ft.) were probably due to the fact that the increase of water film resistance more than offsets the decrease of toluene film resistance caused by the phase reversal.

Comparison of Results from the Literature

The results on perforated-plate columns using the system benzoic acid–toluene–water were taken from the literature for comparison. But owing to wide variation of conditions under which the results were obtained by different investigators, only a rough comparison of results to show relative magnitudes was possible.

Since the values of $(\text{H.T.U.})_W$ and $(\text{H.T.U.})_T$ reported in the literature were obtained as intercepts and slopes by plotting $(\text{H.T.U.})_{OW}$ vs. HV_W/V_T according to Equation (3), the same procedure was followed in obtaining $(\text{H.T.U.})_W$ and $(\text{H.T.U.})_T$ values for the present work. The results are shown in Table 3.

The values of $(\text{H.T.U.})_W$ reported by various investigators are not much different from those in this study, and they are all much smaller than the $(\text{H.T.U.})_T$ values. The difference between $(\text{H.T.U.})_T$ values reported in the literature and this study, however, was appreciable, and the $(\text{H.T.U.})_T$ values from the present work are the lowest. Large increase of over-all transfer coefficients by superimposition of pulsation is evident from comparison of the present results with those of previous investigators, as shown in Figure 8.

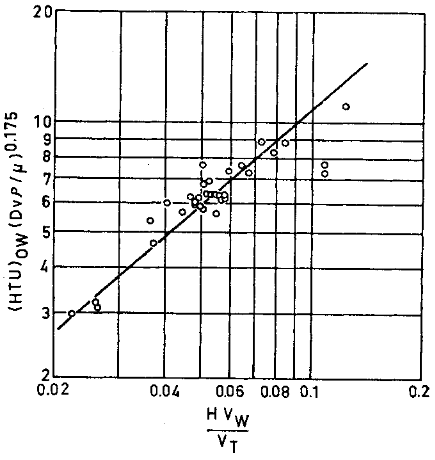

Fig. 13. Extraction in a pulsed column, toluene dispersed in streamline region.

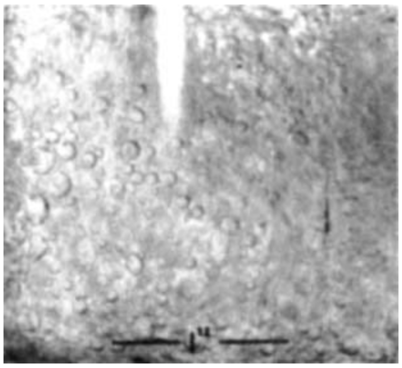

Fig. 14. Dispersion of water, Re = 943.

Holdup

Under constant pulsation the toluene holdup was increased by increasing the toluene rate and only slightly affected by the water rate. The results are plotted in Figure 15.

The effect of pulsation on holdup, shown in Figure 16, was large in both streamline and turbulent regions. As the Reynolds number increased, the size and the rising velocity of dispersed toluene in the column decreased, and so the toluene holdup increased. In transition from streamline to turbulent region

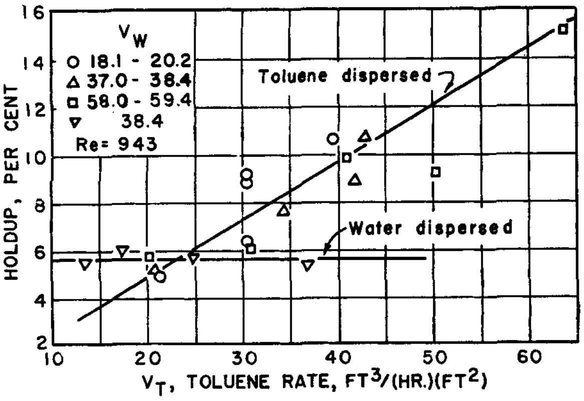

Fig. 15. Effect of toluene rate on holdup with constant pulsation, Re = 943.

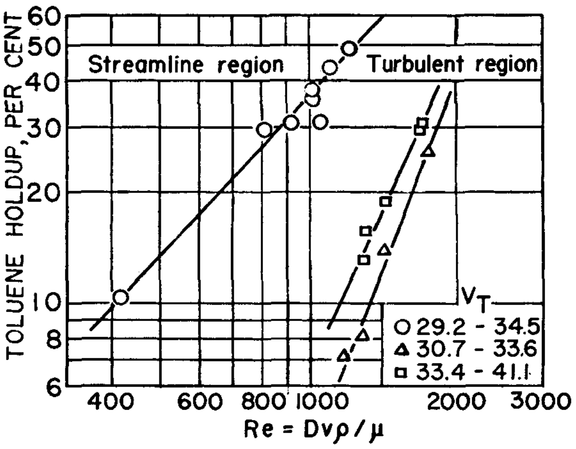

Fig. 16. Effect of pulsation on toluene holdup.

the toluene holdup suddenly dropped from 48 to 7%, as shown in Figure 16. This sudden drop of toluene holdup was due to the fact that the dispersed toluene changed its form from cell-like films into drops upon transition from the streamline to the turbulent region. Since the toluene drops would rise faster in the continuous-water phase than the toluene films, the toluene holdup in the column became less for the turbulent region than for the streamline region.

With water as the dispersed phase, the water holdup was found to be unaffected by the toluene rate. It seemed that the holdup was affected mainly by the flow rate of the discontinuous phase and was practically independent of the flow rate of the continuous phase.

CONCLUSIONS

The performance of the perforated-plate column for liquid-liquid extraction was greatly improved by superimposing pulsations. As compared with the results in literature, transfer coefficients found in this work are considerably higher.

A critical Reynolds number based on the average velocity through the perforations was found to be around 1,200, above which the rate of increase of the transfer coefficient was much more rapid with an increase in the Reynolds number. Pressure drop across the column increased with the Reynolds number in a similar manner.

Toluene-film resistance was found to be the controlling factor for the mass transfer.

Lower transfer coefficients were obtained with water as the discontinuous phase. This decrease was probably due to the increase of water film resistance, which was more than enough to offset the decrease of toluene film resistance.

A high flow-rate ratio of water to toluene gave abnormally low $(H.T.U.)_{OW}$ values, which could not be correlated by plotting $(H.T.U.)_{OW}$ vs. HV_W/V_T.

A modified H.T.U. method of correlation is recommended and general equations including the effect of pulsation are presented.

The holdup increased with increasing flow rate of the discontinuous phase and independent of flow rate of the continuous phase. With constant flow rates holdup increased exponentially with an increase in the Reynolds number.

NOTATION

a = interfacial area, sq. ft./cu. ft. of effective column volume
C = concentration of benzoic acid, lb. moles/cu. ft. of solution
$(\Delta C)_m$ = logarithmic mean of concentration-difference driving force at the extremities of the column, lb. moles/cu. ft. of solution
D = diameter of the perforations on the plate, ft.
H = reciprocal of the slope of the equilibrium-distribution curve equal to dC_W/dC_T
H.T.U. = height of transfer unit, ft.
k = film transfer coefficient, lb. moles benzoic acid transferred /(hr.)(sq. ft.)$(\Delta C)_m$
K = over-all transfer coefficient, lb. moles benzoic acid transferred /(hr.)(sq. ft.)$(\Delta C)_m$
N = number of moles of benzoic acid transferred
Re = Reynolds number through the perforations on the plate, equal to $Dv\rho/\mu$
v = velocity of toluene-water mixture through the perforations, ft./sec.
V' = effective volume of the column, cu. ft.
V = flow rate, cu. ft./(hr.)(sq. ft.)

$$\left.\begin{matrix} d \\ m \\ n \\ r \\ s \\ t \end{matrix}\right\} = \text{exponents} \qquad \left.\begin{matrix} \alpha \\ \beta \\ \gamma \\ \delta \\ \phi \\ \phi' \end{matrix}\right\} = \text{constants}$$

θ = time, hr.
μ = viscosity of the toluene-water mixture, lb./(sec.)(ft.)
ρ = density of the toluene-water mixture, lb./cu. ft.

Subscripts

O = over-all
T = toluene phase
W = water phase

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