Dispersed-phase Holdup in Packed, Countercurrent Liquid-liquid Extraction Columns

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This investigation is a study of the effect of flow rates and packing size and column diameter upon the holdup of a toluene dispersed phase, flowing countercurrent to a continuous water phase in packed liquid-liquid extraction columns. Six different packings were used: ¼-, ⅜-, ½-, ⅝-, ¾-, and 1-in. nonporous, unglazed-porcelain Raschig rings. Three extraction columns, 3, 4, and 6 in. I. D., were used in the experimental work.

Three types of dispersed-phase holdup, free, operational, and total, have been investigated. An empirical correlation is presented for the total holdup data below the loading point. A correlation of the effect of packing size on the exponential term r and the coefficient A_1 is developed for packing sizes ½ in. or larger when the column-diameter-to-packing-size ratio is at least 8 to 1. The term $A_1(V_D)^r$ in the equation accounts for at least 90% of the total holdup. The small magnitude of the residual term $B_1(V_D)(V_C)^s$ did not permit a definite correlation of the coefficient B_1 or the exponent s.

Observation of the dispersed-phase holdup during column operation revealed a transitional behavior of the ⅜-in. rings as compared with that of the ¼- and ½-in. or larger packing. Two, and sometimes three, regimes of flow occur in packed extraction columns. The increase in holdup with increasing continuous-phase flow rate differed for each zone. In the two zones below the loading zone the holdup was found to increase linearly with the dispersed-phase flow rate for a constant continuous-phase flow rate. A new method of randomly packing an extraction column has been found to give reproducible holdup data.

During recent years the widespread use of packed, dispersed-solvent, extraction columns as a means of separating the components of solutions has made apparent the need of fundamental information on the extraction mechanism of the column in order to improve the operational efficiency. A consideration of the problem of predicting the height of packing required for any given separation suggests the following five basic factors which need to be taken into consideration: (1) the resistance to diffusion in the continuous phase adjacent to the interface, (2) the resistance to transfer across the interface, (3) the resistance to transfer in or out of the body of the droplet, (4) the interfacial area of contact of the phases, and (5) the limitations on flow rates for a given packing. It was realized at the outset that in order to provide this fundamental information each of these factors will probably require individual investigations. This paper is concerned with the effect of flow rates and the effect of packing size and column diameter upon the dispersed-phase holdup within a randomly packed extraction column. Holdup information, along with drop-size information, for a given system and packing provides a means for calculating the interfacial contact area when it is below the loading rates.

Very little information has appeared in the literature on the determination of holdup* in packed extraction columns. In general, the available holdup data can be classified into two principal categories, depending upon the manner in which the data were obtained: (1) holdup data in which the volume of holdup was measured by draining the column and separating the desired phase and (2) holdup data obtained by simultaneously closing all streams to and from the column and then measuring the volume of continuous phase needed to restore the altered two-phase interface to its initial position. The relationship, if any, existing between these two forms of holdup has not yet been clarified.

Row, Koffolt, and Withrow(*10*) obtained holdup data during study of the rate of mass transfer of benzoic acid from water to toluene by use of an 8.75-in. I. D. packed column. The tower was packed with ½-in. unglazed Berl saddles, ½-in. unglazed Raschig rings, and knitted copper cloth packing. The hold-

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*When liquid-liquid extraction columns are dealt with, the term *holdup* will be used to designate the fractional holdup of the dispersed phase in that portion of the total column volume which may be occupied by liquid.

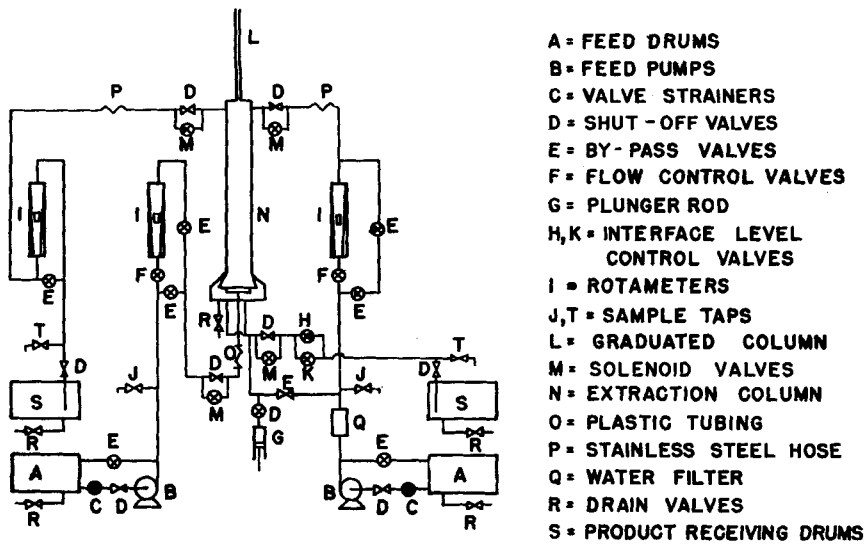

Fig. 1. Schematic flow diagram of extraction equipment.

up data, measured by correcting the interfacial level, indicated a sharp increase in the liquid holdup as flooding was approached. There also appeared to be more or less rough correlation between holdup values and capacity coefficients *Ka*. Allerton, Strom and Treybal(*1*) measured holdup in a 3.56-in. I. D. column packed with ½-in. carbon rings with benzoic acid as the solute in the kerosene-water system. Their results indicated an increase in holdup with increasing rates of both phases. Gier and Hougen(*6*) reported similar results in their holdup measurements made in a 6.0-in. I. D. column packed with ½- and ⅜-in. unglazed Raschig rings. The holdup data which they obtained by determining the volume alteration in the interfacial level lowering were found to be a linear function of the dispersed-phase rate when the continuous-phase rate was constant.

Gayler and Pratt(*4*) and Gayler, Roberts, and Pratt(*5*) conducted an extensive study of dispersed-phase holdup using 3-, 6-, and 12-in. I. D. columns with a packed height of 10 ft. The packing used was ¼-, ⅜-, ½-, ¾-, 1-, and 1½-in. ceramic Raschig rings and ¼-, ½-, and 1-in. ceramic Berl saddles. The continuous phase in each of the systems studied was water; methyl isobutyl ketone, butyl acetate, dibutyl carbitol, benzene, and isooctane were each used as the dispersed phase. The normal holdup data used by the authors in developing their conclusions were obtained by determining the volume alteration in the interfacial level lowering. During the course of the

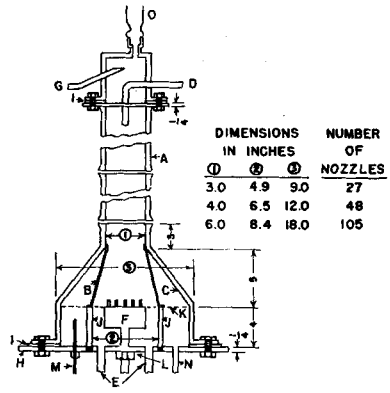

Fig. 2. Extraction-column details.

investigation it was found that an additional volume of the dispersed phase was always present in the column. This was presumably present in the form of droplets which were trapped in the packing. The quantity of permanent holdup present was determined by draining the column after removing the normal holdup from the column. The authors concluded that only the normal holdup was involved in the mass transfer operation; consequently, the normal holdup data were correlated by means of the following equation:

$$V_D + \frac{X}{1-X} V_C = F\overline{v_o} X (1 - X) \qquad (1)$$

where V_D and V_C are superficial velocity of the dispersed phase and the continuous phase respectively, X is the fractional holdup, F is the fractional voidage of packing, and $\overline{v_o}$ is a velocity, characteristic of a given packing. A plot of $V_D + (X/1-X)V_C$ against $X(1-X)$ for a given packing should produce a straight line through the origin. However, the present authors have found that the data for the 3-in. I. D. column and for the majority of the 6-in. I. D. column require separate lines for each different continuous-phase flow rate. Gayler and Pratt(*4*) also presented the following equation for normal holdup at flooding:

$$X_{flood} = 0.62 \frac{V_D^2 a_p}{gF^3} \qquad (2)$$

which must be in error, for a substitution of the proper values for the variables into the equation resulted in values of X_{flood} greater than one, which is an impossibility.

APPARATUS

A schematic flow diagram of the extraction equipment is presented in Figure 1. The material of construction for all lines, valves, valve strainers, and column parts (unless otherwise stated) was type-304 stainless steel. All lines were ½-in. N.P.S. pipe with the exception of short sections of ¼-in. N.P.S. pipe on the rotameter control panels in the line to the plunger rod, and in the drain lines. The feed drums and the receiver drums were standard 55-gal. drums, black iron for toluene and galvanized iron for water. The feed pumps were Eastern, type 2-J, three-stage centrifugal pumps.

Flow rates in the system were regulated by ¼-in. needle valves with 40 threads to the inch. The flow of the water entering, and the toluene entering and leaving, the column was measured by rotameters covering a flow range of 0.01 to 0.89 gal. of water/min. Two 3-ft. lengths of stainless steel hose were employed to provide flexibility in the elevation of the lines at the top of the column. This arrangement permitted a variation in the column height without changing the flow system. The level of the two-phase interface was regulated by means of two parallel valves in the water exit line, a ½-in. globe valve for coarse adjustments, and a ¼-in. needle valve for fine control.

The extraction column is shown in detail in Figure 2. Three extraction columns, 3, 4, and 6 in. I. D., were used in the experimental work. The structural features of each of the columns used were identical. Pertinent dimensions are given in Figure 2. The extraction column consisted of

two 2-ft. flanged sections of Pyrex-glass pipe supported on a 20-gauge stainless steel bell section. A ¼-in.-thick stainless steel top section was fitted with an aqueous feed line, an extract exiting line, and a ½-in. pipe, threaded into the top of the section, which connected a small graduated column to the extraction column.

The aqueous phase entered the column through a ½-in. pipe which ends 1½ in. below the two-phase interface. The aqueous raffinate was removed from the column through two ½-in. pipes threaded into the base flange of the column. Toluene entered the column through the dispersed-phase distributor and was dispersed by a number of sharp-edged nozzles (0.106 in. I. D., 0.014 in. wall thickness and 1¼ in. long) made from stainless steel hypodermic tubing. These nozzles were spaced in concentric circles with a minimum circumferential distance of ½ in. between nozzles. The number of nozzles used on each of the dispersed-phase distributors for the various sizes of the columns was maintained approximately proportional to the column cross-sectional area. The number of nozzles per distributor for each column is given in Figure 2. The distributor was flush with the bottom of the packing with the nozzles embedded 1¼ in. into the packing. Thus the cross-sectional area of the annular space between the dispersed-phase distributor and the stainless steel cone at this point was equal to the cross-sectional area of the column, as recommended by Blanding and Elgin (*2*). The drops of toluene rose through the column packing into the separating section where they coalesced at the interface, forming a layer of toluene above the two-phase interface. The toluene extract overflowed the column through ½-in. pipe welded into the top section.

The support screen for the column packing is a type-304 stainless steel screen, 3¾ mesh with 0.054-in. wire and openings 0.213 in. square. This screen was held in place by the dispersed-phase distributor and by four ½-in. stainless steel rods, which were threaded into the base flange of the column. This bottom flange was bolted firmly to the structural steel support through Dravo Elasto-rib, vibration-insulating material.

All gaskets used between glass sections of the column were fabricated from ⅛-in. Johns Manville 84-S asbestos sheet. The gasket material used between the glass section of the column and the top or bottom sections was ¼-in. hard-cork sheet.

COLUMN PACKING

Six different packings were used: ¼-, ⅜-, ½-, ⅝-, ¾-, and 1-in. nonporous unglazed porcelain Raschig rings. The actual dimensions of these packings are summarized in Table 2. The column was packed by slowly dropping several rings at a time into the empty column filled with water. Initially no attempt was made to obtain the densest possible arrangement of the packing, thus following general procedure used in extraction investigations. After repeated attempts to obtain reproducible data, however, it was decided that a new, more satisfactory means of packing was needed. Only when the random-packed column was settled by passing an air stream up through the column at a rate high enough to flood the column, i.e., by using the method generally accepted for a packed absorption column, was reproducible data obtained. The procedure adopted for the measurement of the superficial area a_p and the void fraction was as follows. The number of pieces of packing dumped into the column was carefully counted and the volume of the column to be packed was accurately measured. Since the surface area and the volume per piece of packing were known, the superficial area a_p and the dry void fraction F of the packing can be calculated. These are reported in Table 3.

LIQUID-LIQUID SYSTEM

The binary solvent system of nitration-grade toluene-distilled water was studied. Prior to this investigation mass transfer in the toluene water-diethylamine system had been studied by Leibson and Beckmann (*7*), using similar equipment. Accurate values for the equilibrium distribution constant for the toluene-water-diethylamine system as a function of temperature and solute concentration were available from the work of Morello and Beckmann (*9*) and Wehn and Franke (*11*). From the mutual-solubility data of Wehn and Franke, it is evident that at 0 wt. % concentrations of diethylamine for either the water or the toluene phase, the solubility of one phase in the other is negligible. This validates the simplifying assumption that the volumetric flow rate of each phase throughout the column is constant.

EXPERIMENTAL PROCEDURE

The holdup of the dispersed-solvent phase, toluene, passing countercurrent to the continuous aqueous phase, was investigated. Before operation was started, the liquids in the feed drums were mutually saturated with the opposite solvent. During operation the two-phase interface was located at an established mark, 3 in.

TABLE 1.—COEFFICIENTS AND EXPONENTS FOR CORRELATION OF TOTAL HOLDUP DATA

$$X_T = A_1 (V_D)^r + B_1 (V_D)(V_C)^s$$

Column diameter I. D., in.	Packing size, in.	A^1	r	B_1	s
3	¼	35.2	0.0556	4.40×10^{-3}	1.850
	⅜	19.6	0.304	4.25×10^{-3}	2.003
	½	6.5	0.428	4.00×10^{-3}	1.318
	¾	2.18	0.482	7.15×10^{-8}	3.252
	Spray	0.175	0.635	8.15×10^{-8}	2.266
4	⅜	26.3	0.250	5.30×10^{-4}	2.142
	½	16.0	0.260	5.60×10^{-4}	2.350
	¾	3.7	0.481	4.70×10^{-5}	1.783
	Spray	0.425	0.643	5.38×10^{-8}	2.590
6	⅜	23.7	0.437	5.25×10^{-2}	1.796
	½	12.5	0.311	1.55×10^{-3}	1.600
	⅝	6.03	0.445	1.55×10^{-3}	1.445
	¾	3.08	0.513	1.55×10^{-3}	1.211
	1	1.42	0.580	1.55×10^{-3}	1.154
	Spray	0.364	0.610	5.2×10^{-6}	2.173

TABLE 2.—RASCHIG-RING PACKING PROPERTIES

Nominal packing size, in.	Outside diameter, in.	Wall thickness, in.	Length, in.	Weight per piece, g	Volume per piece $\times10^5$, cu. ft.	Surface area per piece $\times10^2$, sq. ft.
¼	0.243	0.035	0.244	0.308	0.458	0.245
⅜	0.384	0.080	0.397	1.19	1.76	0.633
½	0.538	0.105	0.517	2.88	4.27	1.18
⅝	0.652	0.099	0.667	4.39	6.64	1.85
¾	0.756	0.129	0.772	7.75	11.3	2.46
1	1.14	0.136	1.14	15.0	28.3	5.59

above the packing. Operation was held constant and the flow rates were observed on the rotameters. After constant operations had been maintained for an hour, all the streams to and from the tower were simultaneously closed by means of rapid-shut-off solenoid valves.

The free holdup, the volume of dispersed toluene which is able to rise to the interface freely owing to density differences, was allowed ample time to rise, and in turn lower the interface. Additional aqueous phase returned the interface to its initial mark and in turn forced the excess toluene out through the graduated column. The differential volume of toluene in the graduated column was recorded as free holdup volume. By means of a pulsating action on the continuous phase, produced by the plunger rod, additional loosely entrapped droplets were freed from the interstices of the packing. After the interface was returned once again to its initial mark, the new differential volume of toluene in the graduated column was recorded as operational holdup. The manual pulsations were continued until further pulsations caused no increase in the differential volume. The column was then drained. After repeated washings of the packing, the collected drainage was separated and the total volume of toluene was measured. The total volume, corrected for the volume of toluene existing between the interface mark and the operating mark in the graduated column, was recorded as the total holdup volume.

The experiments were carried out at ambient room temperature which varied from 24° to 32°C. as extreme limits. It is evident from the results of Morello and Beckmann(*9*) that an over-all variation in temperature of this magnitude is not important for this system in this temperature range.

TYPES OF DISPERSED-PHASE HOLDUP OBSERVED

In the only previous extensive study on dispersed-phase holdup in a packed extraction column, Gayler and Pratt(*4*) presented data on two different types of holdup, normal and permanent. In a study of droplet behavior within a packed column, Lewis, Jones, and Pratt(*8*) observed that with ¼- and ⅜-in. packings the dispersed-phase droplets were normally retained within the interstices of the packing. The vertical motion of the entrapped drops resulted from continual impacts from the succeeding drops. On the basis of these results of Lewis, Jones, and Pratt(*8*) it would appear, for at least the ¼- and ⅜-in. packings, that a larger volume than the free-rising, normal holdup volume participates in the extraction operation.

Inasmuch as neither the normal nor the total holdup (i.e., sum of the normal and permanent holdup) can actually describe the volume of dispersed phase which is needed for active interfacial-area calculation, the present authors concluded that a third holdup, operational, must be actually involved in the mass transfer. Operational holdup includes the normal holdup volume plus an additional volume to account for the droplets temporarily retained by the packing, but having an over-all net movement through the packing. The latter droplets are normally freed during the course of the operation by impact from succeeding drops and by longitudinal thrusts from the descending continuous phase. As a result, holdup measurements were carried out to study the following three types of holdup:

1. *Free holdup,* the volume of dispersed phase which rises freely to the interface, owing only to the differences in density.
2. *Operational holdup,* the volume of dispersed phase which is considered to be the active portion of the dispersed phase taking part in the mass transfer.
3. *Total holdup,* the total volume of dispersed phase within the effective packing volume at any time.

EXPERIMENTAL HOLDUP RESULTS

Value of free, operational, and total holdups, reported as volume percentage of the total voids, are for the 3-in. I. D. column packed with ¼-, ⅜-, ½-, and ¾-in. Raschig rings and unpacked as a spray column for the 4-in. I. D. column packed with ⅜-, ½-, and ¾-in. Raschig rings and unpacked as a spray column, and for the 6-in. I. D. column packed with ⅜-, ½-, ⅝-,

TABLE 3.—PROPERTIES OF THE PACKED SYSTEMS

Nominal packing size, in.	Run series	Ratio of column diameter to packing size	Number of packing pieces per cu. ft.	Superficial area of packing a_v, (sq. ft.)/(cu. ft.)	Void fraction F, (cu. ft.)/(cu. ft.)	Void volume, cc.	$\frac{a_p}{F^3}$
¼	3-1	12.0	90,566	222	0.585	3,512	1,110
⅜	3-2	8.0	23,490	148	0.587	3,514	732
⅜	3-2A	8.0	23,470	148	0.587	3,514	732
⅜	4-2	10.7	25,469	161	0.552	5,831	957
⅜	6-2	16.0	23,973	152	0.578	13,586	787
½	3-3	6.0	8,656	102	0.630	3,782	392
½	3-3A	6.0	8,678	102	0.630	3,782	392
½	3-3B	6.0	8,726	103	0.628	3,770	416
½	3-3C	6.0	8,585	101	0.633	3,800	398
½	4-3	8.0	9,517	112	0.594	6,275	534
½	6-3	12.0	8,795	104	0.625	14,691	426
⅝	6-4	9.6	4,692	86.8	0.688	16,161	267
⅝	6-4A	9.6	4,639	85.7	0.692	16,266	260
¾	3-5	4.0	2,547	62.7	0.712	3,843	174
¾	4-5	5.3	2,748	67.6	0.690	7,289	206
¾	6-5	8.0	2,771	68.2	0.687	16,148	215
1	6-6	6.0	1,111	62.1	0.686	16,125	201
Spray	3-0	...	....	....	....	6,004	
Spray	4-0	...	....	....	....	10,564	
Spray	6-0	...	....	....	....	23,506	

Note:—Packed volumes: 3-in. I. D. column = 0.212 cu. ft., 4-in. I. D. column = 0.373 cu. ft., 6-in. I. D. column = 0.830 cu. ft.

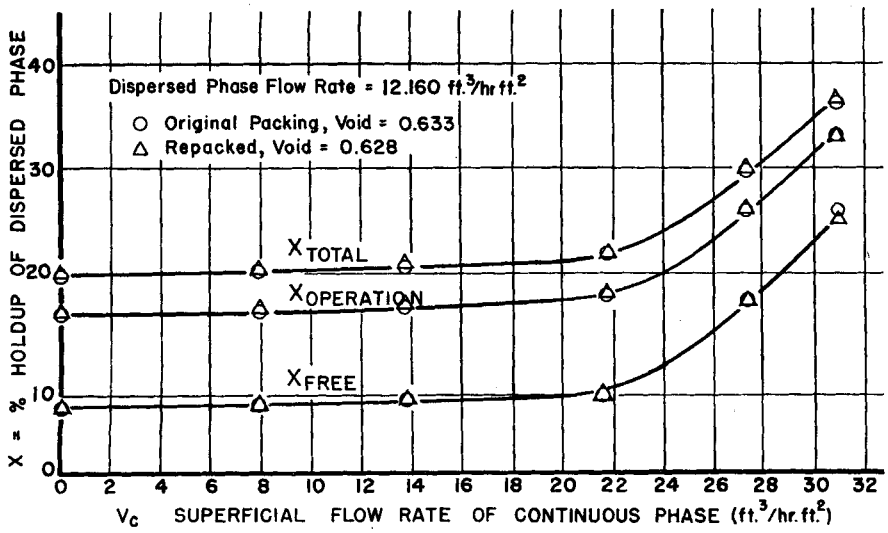

Fig. 3. Reproducibility of experimental data for measuring total holdup.

¾-, and 1-in. Raschig rings and unpacked as a spray column. Table 4 lists typical experimental results obtained for ¾-in. rings in the 6-in. I. D. column.*

REPRODUCIBILITY OF EXPERIMENTAL RESULTS

The total holdup results obtained for a specific packed bed are reproducible within 2% on a relative basis.† The operational holdup, being in the same range of magnitude, also showed a reproducibility within 2%. The free holdup results obtained for the packed beds are reproducible within 6%. The holdup results for the spray column are reproducible within 4%. This slightly higher percentage of reproducibility for the free and spray holdup is due primarily to the low numerical values of these results. The reproducibility of the data, as well as the internal consistency of the data, is illustrated in Figure 3.

In the initial phase of this investigation the column was packed randomly with no attempt being made to obtain the densest possible arrangement of the packing. This procedure followed the previously established manner of packing an extraction column. The results obtained for the unsettled ½-in. rings in the 3-in. I. D. column showed, upon repacking, that the total holdup data were reproducible within ± 15%; the free holdup data were reproducible within ± 19%. Dell and Pratt(3), Gayler and Pratt(4), and Leibson and Beckmann(7) encountered similar repacking difficulties in the reproducibility of flooding data, free holdup data, and mass transfer data, respectively.

The initial data obtained from a column packed to operate initially as a flooded absorber were generally erratic. This was undoubtedly due to the presence of air bubbles in some of the favorite droplet resting spots within the interstices of the packing. However, after several preliminary runs, during which the air was gradually removed, consistent results were obtained. When the 3-in. column was repacked in this manner with ½-in. rings, the total holdup data were reproducible to within 1.2%; the free holdup data to within 1.5%. For the ⅜-in. rings repacked in the 3-in. column the total holdup data were reproducible to within 2%; the free holdup data to within 5%. The total holdup data for the 6-in. column when it is repacked with ⅝-in. rings were reproducible to within 1.5%; the free holdup data to within 1.5%. Since all previous investigations in liquid-liquid extraction have failed to find a reproducible method of packing a column, it is suggested that this new procedure of settling the packing be used in mass transfer studies wherever it is particularly desirable to obtain the effect of packing on the variables.

CORRELATION OF HOLDUP DATA

An initial insight into the behavior of a physical system can often be obtained by a dimensional analysis of the variables that characterize the definite physical system. Dimensional analysis entails certain assumptions and limitations

*See footnote on page 426.
†The reproducibility of results will always be compared on a relative basis in this paper.

TABLE 4.—SAMPLE OF EXPERIMENTAL HOLDUP DATA

6-in. I. D. column — ¾-in. rings
Packed height: 48 in. — Void fraction: 0.687

Run	Flow rates, cu. ft./(hr.) (sq. ft.) Toluene Inlet	Toluene Exit	Water Inlet	Stream temperatures, °F. Toluene Inlet	Water Inlet	Holdup data, vol. % of total voids Free	Operation	Total
6-5-2	5.374	5.394	0.0	90	80	3.73	6.21	7.31
-3	5.374	5.394	0.0	87	80	3.72	6.16	7.36
-4	5.374	5.394	9.172	91	89	3.87	6.35	7.47
-5	5.374	5.394	15.009	87	83	4.07	6.56	7.64
-6	5.374	5.394	21.042	89	86	4.27	6.71	7.82
-7	5.374	5.394	27.124	91	88	4.44	6.86	7.98
-8	5.374	5.394	33.599	92	89	4.63	6.95	8.22
-9	5.374	5.394	40.220	94	91	4.81	7.13	8.43
-10	8.774	8.646	0.0	89	80	5.97	8.07	9.66
-11	8.774	8.646	9.184	91	88	6.21	8.32	9.83
-12	8.774	8.646	15.009	92	89	6.44	8.55	9.99
-13	8.774	8.646	21.042	90	86	6.71	8.85	10.19
-14	8.774	8.646	27.124	94	89	6.90	9.12	10.38
-15	8.774	8.646	33.599	87	87	7.10	9.40	10.57
-16	8.774	8.646	0.0	80	80	6.04	8.10	9.59

inherent in this method of correlation; and although it does not give the investigator any new information about the exact behavior of the situation, it is a powerful preliminary tool. In the case of dispersed-phase holdup in a randomly packed liquid-liquid contacting tower, the following factors may be considered to be of impartance:

$$X, d_o, V_D, \rho_C, \rho_D, V_C, \mu_C, \mu_D, D, h,$$

$$F, a_p, \sigma \text{ and } g_C$$

An analysis of these variables produces the following general equation:

$$\left(\frac{d_o}{D}\right)^a \left(\frac{h}{D}\right)^b \left(\frac{\rho_D}{\rho_C}\right)^c \left(\frac{\mu_D}{\mu_C}\right)^d \left(\frac{D\rho_C \sigma g_C}{\mu_C^2}\right)^e (a_p D)^i \left(\frac{D\rho_C V_D}{\mu_C}\right)^g \left(\frac{D\rho_C V_C}{\mu_C}\right)^h (X)^i + \ldots = 0 \quad (3)$$

This equation can be simplified, when only one system, one column diameter, one droplet diameter, and one packed height are dealt with, to the following equation:

$$A\,(a_p)^f \left(\frac{D\rho_C V_D}{\mu_C}\right)^g \left(\frac{D\rho_C V_C}{\mu_C}\right)^n (X)^i + \ldots = 0 \quad (3a)$$

Equation (3a) can be rearranged to be explicit in holdup:

$$X = \phi\left[A\,(a_p)^f \left(\frac{D\rho_C V_D}{\mu_C}\right)^g \left(\frac{D\rho_C V_C}{\mu_C}\right)^i\right] \quad (3b)$$

where ϕ signifies some function. Of course, if the equation were used for only one size and type of packing, the $(a_p)^f$ term could be grouped with the constant A term. From observation of the experimental data, the following final equation was chosen, which is directly obtainable from the foregoing equation:

$$X = \phi\,[C\,(V_D)^{g-y}\,(V_D)^y\,(V_C)^i] \quad (3c)$$

where C is a new constant, including A, $(a_p)^f$, and $(D\rho_c/\mu_c)^{g+i}$

Utilizing Equation (3c) and the data below the loading point yielded the following correlation for the total holdup in a specific packing and column:

$$X_T = A_1\,(V_D)^r + B_1\,(V_D)\,(V_C)^s \quad (4)$$

Table 1 presents the coefficients and the exponents for the packings and columns studied. It was generally found that the correlation permitted calculating total holdup to within ± 1.5% of the observed value. A few exceptional points, predicted to within ± 4%, were points in the vicinity of the loading point. The correlation presented here is applicable only to the system studied in this investigation. Further work with other systems is necessary before a more general correlation can be developed.

For a specific system a specific type of packing, the availability of a correlation between extraction variables, and some factor characterizing the size of the packing would extend the applicability off experimentally obtained data. Equation (3b) suggests the possibility of correlating holdup with an a_p term. A correlation of this type for the height of individual transfer unit for the toluene-diethylamine-water system was presented by Leibson and Beckmann(7) for Raschig rings ½ in. or larger. A similar correlation for the r exponent is presented on Figure 4 for the toluene-water system for packing sizes ½ in. or larger. This exponential term r was found to be proportional to $(a_p/F^3)^{-0.8}$. The work of Leibson and Beckmann(7) is also shown on Figure 4. Figure 5 reveals that the first coefficient, A_1, also has a dependency on the (a_p/F^3) term.

It is interesting to observe in the total holdup correlation the importance of the first term, $A_1(V_D)^r$. This term represents the total holdup obtained for zero continuous-phase rates. For all runs in which the continuous-phase rate differed from zero, the first term represents at least 90% of the numerical total holdup value. As a direct consequence of the magnitude of the first term, the evaluation of the residual term $B_1(V_D)\,(V_C)^s$, was made with relatively small numbers; a slight error in one or two of these numbers could possibly alter the numerical value of the coefficient and/or exponent of the term. The small magnitude of the residual term, $B_1(V_D)\,(V_C)^s$, did not permit a definite correlation of the coefficient B_1 or the exponent s.

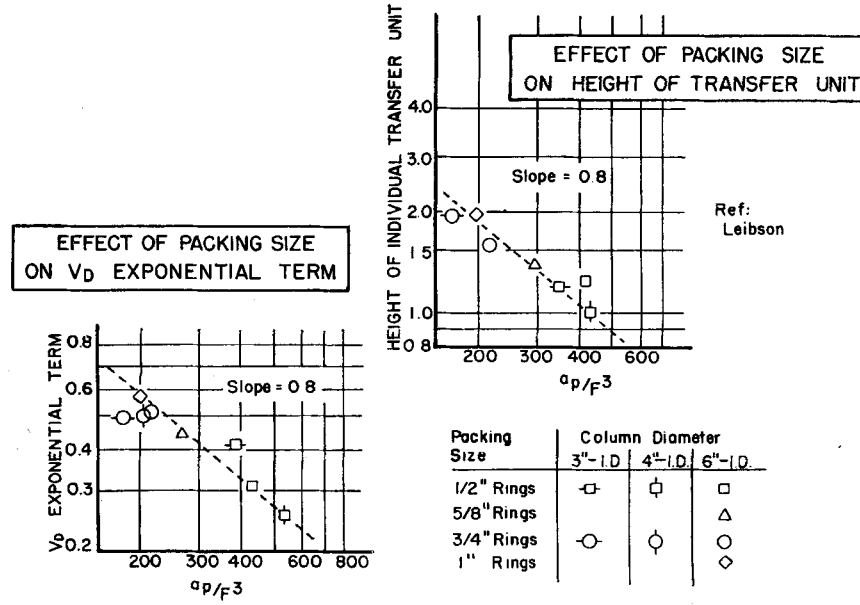

Fig. 4. Effect of packing size and column diameter on holdup and mass transfer.

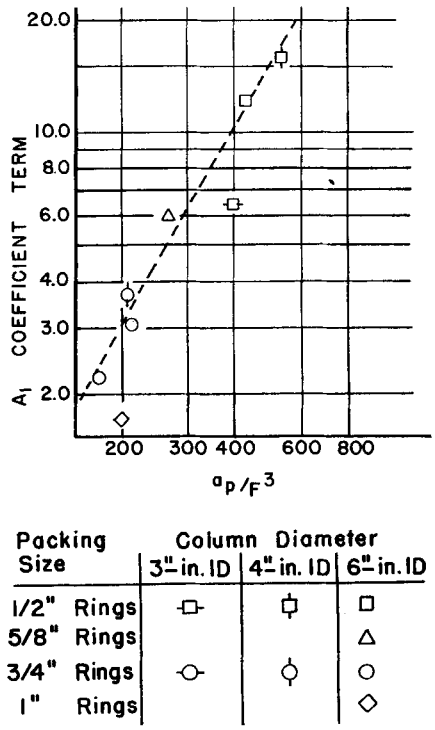

Fig. 5. Effect of packing size and column diameter on holdup.

A number of attempts to find a correlation between the phase flow rates and the free holdup were made, with only partial success. Figure 6 illustrates the linear dependency of the free holdup on the dispersed-phase rate when the continuous-phase rate was constant. Similar plots were obtained for the 3- and 4-in. I. D. columns. As with the total holdup data, these zero continuous-phase rate data account for 75 to 90% of the free holdup data in which the continuous-phase rate differs from zero. Owing to the relatively small magnitude of the actual free holdup values, the accuracy of the data did not allow a proper evaluation of the relationship of the continuous-phase rate to the free holdup.

As neither the normal nor the total holdup actually described the volume of dispersed phase which is involved in the over-all net movement through the packing, an attempt was made to measure the operational holdup. The freeing of the temporarily detained droplets was brought about by a manual pulsating of the continuous phase. This is undoubtedly only an approximation of what actually occurs during the column's operation; consequently, the obtained operational holdup data must be evaluated as only approximate operational holdup data. At the present time additional investigations are being made on this important holdup. Owing to the uncertainty of the relationship between the pulsating and the correct operational holdup, no separate correlation was developed for this pseudooperational holdup.

EFFECT OF PACKING SIZE ON HOLDUP

The appearance and behavior of the column with respect to variation in the packing size was quite similar to that observed by previous investigators (*4, 7, 8*). The column operation with ring sizes ½ in. and larger is markedly different from that with ¼-in. rings. The ⅜-in. rings behave as a critical or a transition packing between the ¼-in. rings and the ½-in. or larger sizes of packing.

In the case of the ½-in. and larger packing, the droplets of the dispersed phase visually appeared to be well defined and of a relatively constant size distribution independent of packing size. With these packings the operation of the column became steady early in the run and no difficulty was encountered in controlling the interface. However, severe channeling of the rising droplets was observed when the column diameter to packing size was less than 6.0.

The type of flow observed with the ¼-in. rings differs greatly from that described above. The droplets rise through the packing as irregularly shaped globules. Each droplet hangs in the interstices of the packing until impacted by the succeeding droplet. The drops emerge from the top of the packing at only a few different points. The position of these "active points" at the top of the packing varied with time. The droplets, leaving the packing, were well-defined drops. The length of time required to steady the operation of the column was considerably longer for the ¼-in. packing than for any other packing. Channeling of the droplets near the wall was not particularly noticeable.

The type of flow observed with the ⅜-in. rings may be classified as a transition between droplet behavior in the ¼-in. rings and that in the ½-in. or larger. At low flow rates the droplet flow in the ⅜-in. rings appeared to be the same as that observed for the ½-in. and larger rings. As the flow rates were increased, the droplet size increased markedly and the type of flow became similar to the flow in the ¼-in. rings. Erratic results were sometimes encountered in

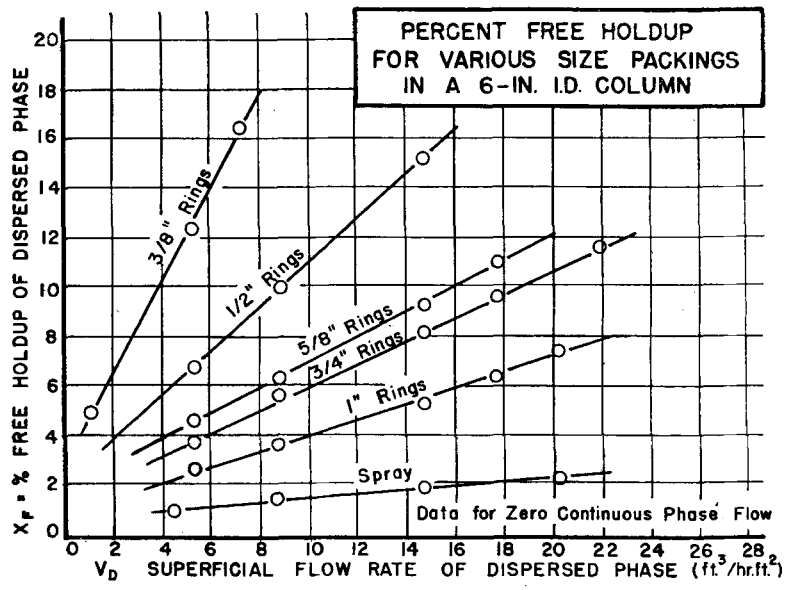

Fig. 6. Effect of flow rate (dispersed phase) on the free holdup for Raschig rings in a 6-in. column.

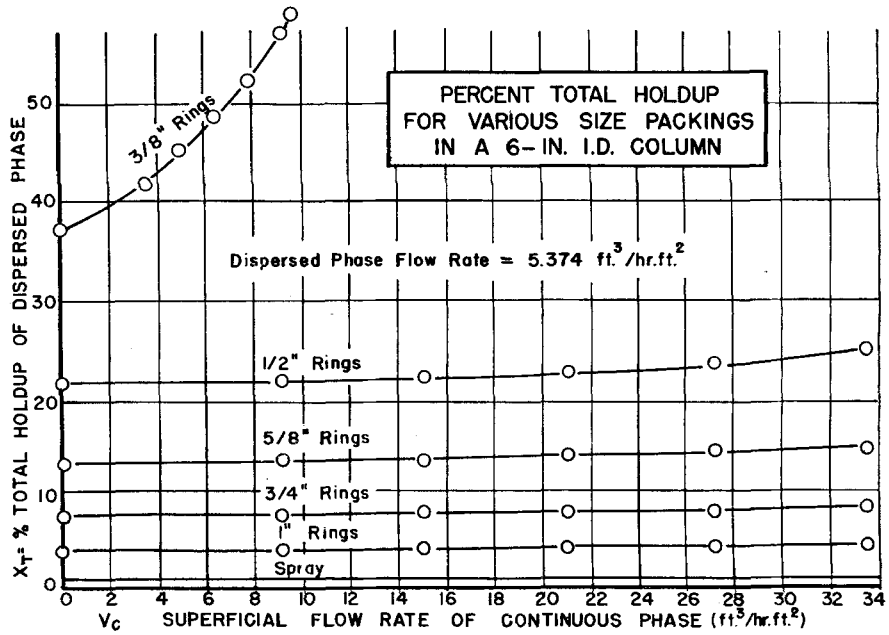

Fig. 7. Effect of continuous-phase flow rate on total holdup for Raschig rings in a 6-in. column.

columns packed with 3/8-in. rings. Possibly this may have been due to the transitional behavior of the packing.

For a given tower the holdup increases as the packing size is decreased. The effect upon the 6-in. I. D. column may be observed in Figure 7. This behavior is to be expected, as one of the main functions of the packing is to produce a series of baffles within the column. Thus, as the packing size decreases and the "baffling effect" increases, the holdup should increase.

FLOW-RATE EFFECTS ON HOLDUP

Although the correlation for the total holdup gives a mathematical relationship for the effect of the flow rates on total holdup, an additional important observation can be made from the data of this investigation. In plotting on logarithmic coordinates the total holdup vs. the continuous-phase flow rate for the various dispersed-phase flow rates, three zones of flow were observed. With the spray column, and also with some of the larger packing sizes, a zone of constant holdup occurred at low continuous-phase velocities. A zone, or zone of flow, as herein referred to is specifically concerned with the variation of holdup with either continuous- or dispersed-phase flow rate (the other phase flow rate being held constant) when the data are plotted in the conventional manner. These terms have no relationship to the visual behavior of the column. As the continuous-phase flow rate was increased, a secondary zone of gradually increasing holdup resulted. This increase in holdup appeared due to the drag placed on the droplets by the increasing flow of the continuous phase. The third zone, where the holdup increases exponentially with the increasing continuous-phase velocity, occurred after the loading point was reached. In the smaller packings the first zone was not observed, probably because of the relatively higher local velocities of the continuous-phase produced by the smaller voids in the packed sections.

SUMMARY

The effect of flow rates and of packing size and column diameter upon the holdup of a toluene dispersed phase, flowing countercurrent to a continuous water phase in packed columns, was investigated, and the results follow.

1. Consistent data were obtained for three different types of holdup: free, operational, and total. The free holdup includes the dispersed-phase drops that rise freely to the interface. The operational holdup includes the free-holdup volume plus the volume of the dispersed-phase droplets freed from the interstices of the packing by pulsations delivered to the continuous phase. The total holdup is the total volume of dispersed phase within the effective packing volume at any time.

2. The total holdup, below the loading point, has been correlated by the equation

$$X_T = A_1 (V_D)^r + B_1 (V_D) (V_C)^s$$

Tabulated values are given for the coefficients and the exponents. A dimensional analysis of the variables involved presents an equation in agreement with the correlating equation. For packing sizes 1/2 in. or larger, a graphical correlation of the effect of packing size on total holdup suggests that the exponential term r and the coefficient A_1 are related to a_p/F^3. The small magnitude of the residual term $B_1(V_D)(V_C)^s$ did not permit a definite correlation of the coefficient B_1 or the exponent s.

3. The procedure adopted for randomly packing of the column gives reproducible results upon repacking of the column. Prior to this paper no satisfactory reproducible method of packing a column was observed.

4. The effect of packing size on holdup is in agreement with previous investigators. The column operating with ring sizes 1/2 in. and larger was observed to be markedly different from that with 1/4-in. rings. The 3/8-in. rings behaved as a critical or transitional packing between 1/4-in. rings and the sizes of packing 1/2-in. or larger. The holdup of the dispersed phase increased as the packing size decreased.

5. Two, and sometimes three, distinct zones of flow were found to occur. The increase in holdup with the increase in continuous-phase rate was found to differ in each zone.

NOTATION

A_1 = coefficient to first term in total holdup correlation
a = interfacial contact area/cu.ft. of column volume, sq.ft./cu.ft.
a_p = packing surface area/cu.ft. of column volume, sq.ft./cu.ft.
B_1 = coefficient to second term in total holdup correlation
D = inside diameter of the column, ft.
d_o = droplet diameter, ft.
D_N = nozzle diameter, ft.
F = void fraction of packing
g_C = gravitational constant, ft./sq.hr.
h = height of packing, ft.
H_t = height of transfer unit, ft.
V = superficial liquid velocity, cu. ft./(hr.)(sq.ft. of column cross section)
X = fractional holdup of the dispersed phase in that portion of the column volume which may be occupied by liquid
Δ = differential amount
μ = absolute viscosity, lb./(hr.)(sq.ft.)
ρ = density, lb./cu.ft.
σ = interfacial tension, lb./ft.
ϕ = any function

Subscripts

C = continuous phase
D = discontinuous phase
F = free
$flood$ = flooding
Op = operational
T = total

Exponents

r = exponent to first term, i.e., to (V_D), of total holdup correlation
s = exponent to second term, i.e. to (V_C), of total holdup correlation

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