Mass Transfer in Packed Liquid Extraction Towers

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FOR THE MOST part, studies on mass transfer for packed extraction towers have been confined to the effect of packing size and type for a single system, or the effect of temperature for a given system and packing. Experimental work has not been reported where effects of both system properties and packing characteristics have been considered. Therefore, the effect of physical properties on mass transfer were studied using different liquid systems and different packings—i.e., acetone dissolved in toluene, Pegasol or kerosine, dispersed in a continuous phase of water, using unglazed porcelain Raschig rings, copper rings, and glass beads. Propionic acid was also extracted from a dispersed methyl isobutyl ketone phase to a continuous water phase.

The effect of column height and direction of extraction on mass transfer have been studied. Photographic recordings of the column in operation were taken for a good number of runs, as these aided by visual observations proved to be helpful in interpreting the results on mass transfer.

APPARATUS

The extraction equipment consisted essentially of an extraction tower and the necessary accessories to maintain a steady flow of the two liquid phases. The design of the tower was based on the recommendations of Blanding and Elgin (*4*) with regard to the entrance sections. A schematic flow diagram of the packed tower and the related equipment is presented in Figure 1.

Two columns were utilized for the experimental work and each was constructed of a borosilicate glass tubing, 1.88 inches in i.d., as this facilitates visual observation of the column in operation. The short column used was 34 inches long (a 30-inch column packed with 6-mm. glass beads.) and the longer one was 56 inches in length.

The glass column with the end sections was fixed vertically in a frame built of Dexion angles. The frame carried two horizontal platforms for supporting the glass separating sections. The brass flange over the top end section carried connections for solvent outlet and water inlet. The latter consisted of four concentric ¼-inch i.d. copper tubes. The bottom brass plate underneath the lower settling section similarly had four ¼-inch i.d. copper tubes which serve as water outlets and a ½-inch i.d. copper tube for feeding the solvent phase. All connections to and from the column were made with ½-inch i.d. copper tube and brass fittings.

The upper flange of the bottom end section and the lower one of the top end section each have a gland and nut arrangement, and the extraction tower was fixed in position in them. The glass column was extended about 3 inches into the upper settling section. Water was introduced into the column through the four ¼-inch copper tubes extending into the annular space between the upper section and the 3-inch extension of the glass tube. Neoprene packings, ⅛-inch thick, were fitted between the brass plates and the end glass sections, and the latter were held tight by tie rods. Atmospheric vents are provided in the exit lines of water and solvent phases.

All flow lines were ½-inch i.d. copper tubing except for short sections of ¼-inch tubing on the rotameter control panel. The feed drums were standard 25-gallon drums, galvanized iron for water, and copper for solvent phase. Westco feed pumps (Fairbanks Morse and Co.) were used. The aqueous and solvent phases were pumped into the column from the respective storage (or feed) drums through the corresponding rotameters. Tap water formed the aqueous phase in all cases. Flow control valves of each liquid were paralleled by ¼-inch needle valves for fine control of the flow rates.

The tapered conical section which expanded from 2½ to 3½ inches was constructed of $\frac{1}{16}$-inch copper sheet, and was soldered to the top flange over the bottom glass section

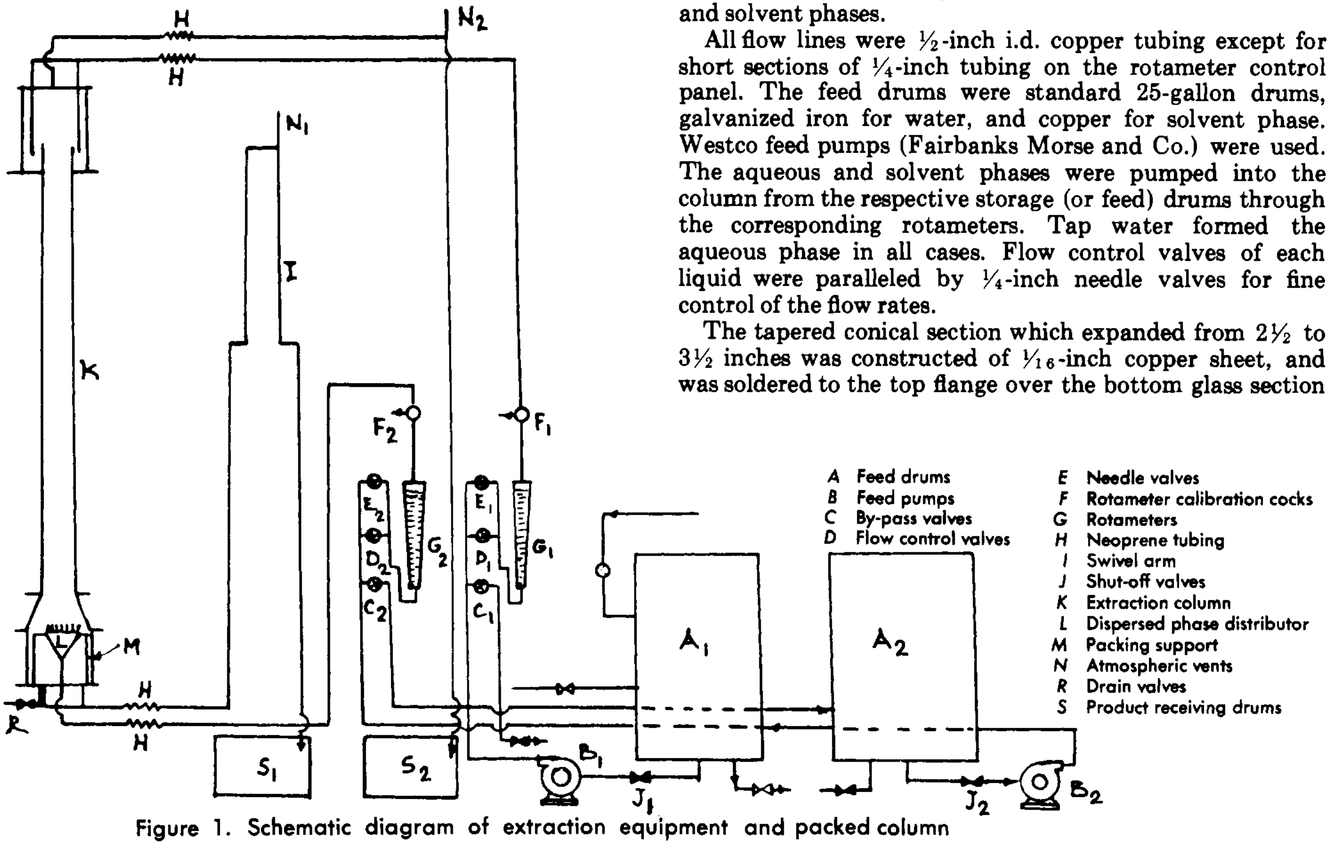

Figure 1. Schematic diagram of extraction equipment and packed column

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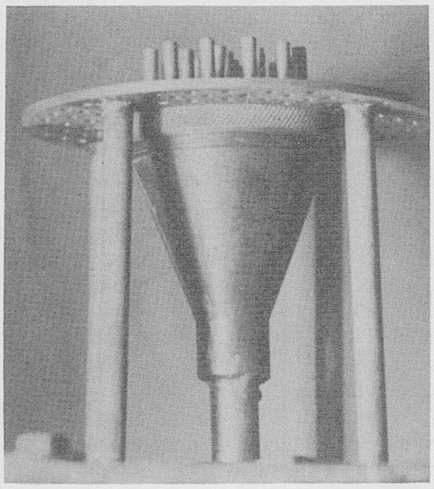

Figure 2. Packed tower distributor section, showing location of the packing support

on one side, and to the flange carrying the gland and nut arrangement on the other side. The dispersed phase distributor shown in Figure 2 consisted of a brass funnel, 3 inches long tapering from the outside of the ½-inch i.d. solvent feed tube to 3¼ inches in diameter at the discharge end. The distributor proper is essentially a brass plate, ⅛ inch thick, and 2¼ inches in diameter with 21 nozzles (⁵⁄₃₂ × 1 inch) equally spaced in the holes and soldered to the top of the plate. The latter is held tight on to the funnel by means of a neoprene gasket ring and a screwed cap. The packing support is a ⅛-inch thick ring of brass plate (2½ inch i.d. and 5½ inch o.d.) with ⁵⁄₃₂-inch perforations spaced at ½-inch centers all over the area, and it was held in position within the bottom separating section by the four supporting legs, built of ¼-inch o.d. and ¹⁄₁₆-inch thick copper tubing.

In view of the observations that in a packed extraction tower the drop size of the dispersed phase does not depend much on the nozzle size of the distributor (*4, 29*), but it is largely controlled by the size of the packing, no distributor other than the one described above was used in this work.

MATERIALS

Column Packings. The packing materials used were ⅜-inch nonporous and unglazed porcelain Raschig rings (Maurice A. Knight Co., Akron, Ohio), ¼-inch copper rings made from copper tubing, and 6 mm. glass beads [British Drug Houses (India) Ltd., Bombay]. The physical properties of the packings are described in Table I.

The column was packed with regular and unbroken pieces. The 6 mm. beads were graded and sorted from a mixture of 5 to 7 mm. glass beads. The column was filled with water, and the packing material was dropped in from the top, a few pieces at a time slowly and at random, to the desired height. The packing was not shaken or tamped. No settling of the packed bed during operation was observed for any of the packings used. Only the Raschig ring packing had fallen, but not more than ½ inch.

Liquid-Liquid Systems. The following systems were chosen because the distribution of the solute between solvent and water in each case obeyed the simple distribution law fairly well in the range of concentrations studied, and in view of the cheapness and availability of chemicals.

1. Acetone-water-toluene
2. Acetone-water-Pegasol
3. Acetone-water-kerosine
4. Propionic acid–water–methyl isobutyl ketone,

Chemicals of the following specifications have been used in the extraction work with the liquid systems.

WATER. Andhra University mains, Vizagapatam Municipality.

ACETONE. Cordite Factory, Aruvankadu, South India. Purity, 99.5% by chemical analysis. Specific gravity, 0.7793.

KEROSINE. Burmah-Shell Oil Co., India, Ltd. Boiling range, 160° to 285° C. Specific gravity, 0.778 (d^{30}_{30}). Viscosity, μ', at 30° C., 1.08 centipoises. Interfacial tension, σ, at 30° C., 39 dynes per cm.

PEGASOL. A petroleum ether of boiling range 60° to 80° C. from Standard Vacuum Oil Co. Specific gravity, 0.785. μ', 0.854 centipoises. σ, 29 dynes per cm.

TOLUENE. Nitration grade toluene (Bengal Chemical and Pharmaceutical works, Calcutta). Specific gravity, 0.859. μ', 0.556 centipoises. σ, 26 dynes per cm.

PROPIONIC ACID. E. Merck. Purity, 99.7% by chemical analysis.

METHYL ISOBUTYL KETONE. (4-Methyl-2-pentanone) Burmah Shell Oil Co. Specific gravity, 0.800. μ', 0.52 centipoises. σ, 10.0 dynes per cm.

In all cases, water formed the continuous phase and the solvent the dispersed phase. The direction of extraction was from solvent to water, except with the water-acetone-kerosine system where the solute was also extracted in the reverse direction.

Values of diffusivity, D, and Schmidt number for the systems studied are given in Table II. Literature data (*16, 17*) for the equilibrium distribution for the systems were used in the mass transfer calculations.

EXPERIMENTAL PROCEDURE

Before operation was started, the liquids in the feed drums were mutually saturated with the opposite solvent. This is particularly necessary when the solvent and water have any appreciable mutual solubilities, as in the case of

Table II. Values of Diffusivity and Schmidt Number

System No.	Solute in Solvent	D, Sq. Cm./Sec.	Schmidt No., $\mu/\rho D_v$, of Solvent Phase
1	Acetone in toluene	2.20×10^{-5}	309.2
2	Acetone in Pegasol	1.44×10^{-5}	750.0
3	Acetone in kerosine	1.10×10^{-5}	1260.0
4	Propionic acid in methyl isobutyl ketone	2.15×10^{-5}	310.0
5	Diethylamine in toluene data (22) at 26.8° C.	2.22×10^{-5}	285.0

Table I. Properties of Packing Materials

Packing	Average Size of Units, In. O.d.	I.d.	Length	Units per Cu. Ft.	a, (Sq. ft.)/(Cu. ft.)	ϵ Measured	Calcd.	a/ϵ^3
⅜-in. porcelain Raschig rings	0.375	0.273	0.384	28,680	178.1	0.610	0.667	750
¼-in. copper rings	0.250	0.175	0.250	81,060	215.4	0.635	0.692	841
6-mm. glass beads	6 mm. diam. sphere			163,000	198.0	0.381	0.396	3580

water and methyl isobutyl ketone. The continuous phase, water, was admitted at a predetermined rate until the column was about three-fourths full, and the dispersed phase—i.e., the solvent solution—having the desired solute concentration was then slowly introduced. The position of the two-phase interface in the top settling section was adjusted and maintained about 1 inch above the top of the packing by means of the overflow control valve in the exit water line (the inverted U-loop was used only for fine adjustments), and the flows were set and held at the desired rates by controlling the main control valves. The ¼-inch needle valves in parallel with the main control valves were used for final adjustment of the flow rates. As preliminary experiments have shown that the steady conditions were established when the column contents were displaced four times, the column was operated for 5 to 10 minutes more, before samples (250 ml. each) of exit water, exit solvent and inlet solvent streams were taken for analysis. Then the flows of both phases to and out of the column were stopped simultaneously and the column contents allowed to settle in order to measure the holdup of the dispersed phase. The holdup measurements were carried out only with the long column (56 inches of packing) for the ring packings.

All the runs were taken at ambient room temperature which varied from 29° to 32° C. in the course of the experimental work. This slight variation of temperature had a negligible effect on the equilibrium distribution curves of the systems studied. Only with the acetone-water-kerosine system, the effect of the reversed direction of extraction was studied. When the direction of extraction was from the continuous water phase to the dispersed kerosine, the aqueous acetone solution of the required concentration was first prepared in the water drum and agitated thoroughly to attain uniform concentration. The exit kerosine for each run was then depleted of its solute by scrubbing it with pure water, and was used for the subsequent runs.

About 220 runs were taken altogether with the four systems and three packing materials, which have been summarized in Tables III and IV.

CALCULATION OF MASS TRANSFER

Values of K_Wa, K_Sa, $(\mathrm{HTU})_{OW}$ and $(\mathrm{HTU})_{OS}$ (the over-all mass transfer coefficients and units respectively) were calculated on both the water (continuous) and solvent (dispersed) phases. The following relationships were employed for this purpose.

$$K_Wa = N/\theta/v(\Delta C)_{W\,\mathrm{l.m.}} \tag{1}$$

$$K_Sa = N/\theta/v(\Delta C)_{S\,\mathrm{l.m.}} \tag{2}$$

$$(\mathrm{HTU})_{OW} = V_W/K_Wa \tag{3}$$

$$(\mathrm{HTU})_{OS} = V_S/K_Sa \tag{4}$$

These equations hold where the system does not depart widely from the simple distribution law and where the volume changes, amount extracted, and concentrations involved are not large.

Individual film values are more useful and fundamental than the over-all mass transfer coefficients, but are difficult to determine experimentally except where the solute is reasonably dilute, and ideal relationships are closely approximated. The simplified forms of Colburn's equations (*7, 8*) relating over-all to individual transfer units are usually applied in practical extraction work.

$$(\mathrm{HTU})_{OW} = (\mathrm{HTU})_W + (\mathrm{HTU})_S\,(mV_W/V_S) \tag{5}$$

$$(\mathrm{HTU})_{OS} = (\mathrm{HTU})_S + (\mathrm{HTU})_W\,(V_S/mV_W) \tag{6}$$

where m is the slope of the equilibrium curve.

Equations 5 and 6 provide an interesting method for correlating extraction data under widely varying conditions of flow. Thus, if $(\mathrm{HTU})_{OW}$ plotted against mV_W/V_S is a straight line, the intercept of which represents the individual film unit HTU_W and the slope represents HTU_D. For the intercept and slope to be true values of the individual film units, the following conditions must be satisfied: The equilibrium line is straight—i.e., $m = dC_W/dC_S = C_W/C_S$. = constant. This condition is generally true for dilute solutions (*2*). The operating line is straight—i.e., the volumetric rate of each liquid phase is constant throughout the column (*3*). The individual film transfer units are constant and do not vary with flow rates.

For all the runs, material balance was checked, and those runs for which this was less than ±10% were used for evaluating the mass transfer coefficients and HTU values. The material balance error (MBE) was expressed as $(N_W - N_S)/N_{av} \times 100$. The over-all transfer coefficients and units were evaluated using average rates of mass transfer and ΔC values based on the experimental concentrations of the liquid streams.

VISUAL OBSERVATION OF FLOW CONDITIONS

To analyze and interpret the mass transfer data, several photographs were taken of the column during operation with the various packings and different liquid systems. In most cases, the top of each picture corresponded to a point approximately an inch or two below the top of the packings. Figure 3 is a typical photograph representative of the flow conditions of the two liquids and of drop size.

With the glass beads (and much less with the copper rings) the behavior of the liquid phases within the packing elements could not be studied in detail, as it was rather difficult to record the column operation by means of photographs, because no definite droplet flow of the dispersed phase occured.

The behavior of the column packed with the Raschig rings while extracting acetone from dispersed Pegasol phase (V_S = 23.5 feet per hour) to a continuous water phase (V_W = 13.3 feet per hour) was photographically recorded. An examination of this plate reveals that only few drops appeared to be moving through the packing, and thereby brings out the fact that at low flow rates, large portions of the packing surface are not utilized by the dispersed phase. The tortuous path of the drops, and their changing shape while passing through the interstices of the packing units

Table III. Summary of Over-all Mass Transfer Data

Series	No. of Runs	System[a]	Disp. Phase[b]	Packed Height, In.
		⅜-In. Raschig Rings		
A	22	2	Pegasol[c]	56
B_1	27	3	Kerosine[c]	56
B_2	9	3[d]	Kerosine	56
C	9	3	Kerosine	34
D	35	4	MIBK	56
E	18	4	MIBK	34
		¼-In. Copper Rings		
F	21	1	Toluene	30
G	14	2	Pegasol	30
H	19	3	Kerosine	30
I	11	4	MIBK	30
		6-Mm. Glass Beads		
J	8	1	Toluene	30
K	9	2	Pegasol	30
L	9	3	Kerosine	30
M	9	4	MIBK	30

[a] Systems defined in text under materials.
[b] Distributed with 21 × 5/32-in. nozzles except as noted.
[c] 19 × 5/32-in. nozzles.
[d] Direction of extraction reversed.

Table IV. Representative Runs of Mass Transfer Data

Series and Run No.	Flow Rates, Ft./Hr.		Concentration, Lb. Moles/Cu. Ft.			N_{av},	MBE	K_Wa	K_Sa	H_{OW}	H_{OS}
	V_W	V_S	C_{W2}	C_{S2}	C_{S1}						
	⅜-Inch Roschig Rings										
A	Actone-Water-Pegasol, 56-Inch Packed Height, Nozzles										
3	27.6	20.4	7.160	12.62	11.43	3.944	−7.56	1.32	7.382	20.82	2.76
6	27.0	43.0	14.80	12.95	3.435	7.777	−2.33	2.32	12.94	11.64	3.32
8	28.8	49.6	16.25	13.15	3.336	9.177	−4.00	2.78	15.51	10.35	3.20
17	50.7	34.4	9.974	18.96	4.525	9.637	+1.880	1.89	10.57	26.76	3.25
19	51.6	21.2	6.638	18.83	3.435	6.426	+4.97	1.38	7.69	37.40	2.75
21	50.9	10.7	3.514	17.700	1.767	3.363	+4.69	0.93	5.18	54.82	2.06
B	Acetone-Water-Kerosine, 56 Inch Packed Height, 19 × 5/32 Inch Nozzles										
1	24.2	16.0	10.17	18.70	2.250	4.90	−6.44	1.02	6.94	23.77	2.30
2	25.4	28.1	11.82	13.28	3.04	5.65	+4.05	1.36	9.22	18.70	3.05
6	25.4	44.6	14.40	11.10	3.27	6.80	+2.18	1.84	12.49	13.50	3.57
12	50.7	21:0	3.90	11.63	1.77	3.89	−4.46	1.17	7.98	43.21	2.63
14	49.6	45.2	9.91	13.94	3.73	9.16	+6.32	1.93	13.14	25.67	3.44
16	49.9	69.4	10.37	11.56	4.46	9.72	+4.87	2.16	14.72	23.07	4.72
B[a]	56-Inch Packed Height, 21 × 5/32-In. Nozzles;										
21[b]	25.6	19.3	8.124	1.59	12.88	4.21	+0.23	2.22	15.07	11.55	1.28
26[c]	25.9	23.7	7.067	1.27	9.02	3.49	−2.09	1.06	7.32	24.39	3.28
28[d]	24.2	36.4	6.737	1.27	6.90	3.97	+1.40	0.98	6.66	24.69	5.46
C	34 In Packed Height, 21 × 5/32-In. Nozzles										
4	26.5	23.7	8.233	12.39	2.861	4.267	−3.56	1.89	12.83	14.02	1.85
8	26.5	47.4	12.95	11.38	3.804	6.749	−4.73	2.93	19.95	9.03	2.38
9	26.5	65.6	15.71	11.38	4.873	8.100	−2.71	3.23	21.99	8.18	2.98
D	Propionic Acid–Water–Methyl Isobutyl Ketone, 56 In. Packed Height; 21 × 5/32-In. Nozzles										
2	24.8	20.1	2.092	5.666	3.060	1.024	−4.96	8.76	4.54	2.83	4.43
9	25.4	37.5	2.560	6.088	4.246	1.288	−6.15	10.84	5.62	2.34	6.68
10	23.1	50.1	2.919	6.088	4.621	1.358	−8.52	15.17	7.86	1.52	6.37
26	39.7	27.6	2.107	6.260	3.403	1.561	+6.00	11.37	5.89	3.49	4.68
28	12.7	25.4	3.184	6.291	4.589	0.803	−6.73	12.49	6.47	1.01	3.92
29	49.6	27.0	1.826	6.291	3.091	1.702	+4.69	11.62	6.02	4.27	4.48
E	34 In. Packed Height, 21 × 5/32-In. Nozzles										
2	26.5	20.7	1.748	5.307	2.966	0.912	−4.52	13.17	6.82	2.01	3.03
5	27.6	40.2	2.310	6.072	4.558	1.198	+4.42	14.96	7.75	1.84	5.19
7	26.0	63.9	2.779	6.260	5.120	1.396	−0.81	20.42	10.58	1.27	6.05
13	47.9	26.4	1.467	6.431	3.840	1.336	+2.59	12.73	6.60	3.76	4.00
	¼-Inch Copper Rings, 30-Inch Packed Height, 21 × 5/32-Inch Nozzles										
F	Acetone Toluene Water										
2	26.5	21.5	5.531	16.54	9.243	2.917	−6.99	4.13	5.63	6.41	3.82
6	25.9	43.0	8.673	18.11	12.39	4.525	−9.01	5.72	7.80	4.53	5.51
18	13.2	44.6	13.76	17.10	12.70	3.640	−7.66	5.63	7.68	2.35	5.81
G	Acetone-Pegasol-Water, 30-Inch Packed Height										
1	27.0	12.1	3.038	9.513	2.444	1.613	+4.35	1.20	6.69	22.50	1.81
4	25.4	26.5	5.680	9.513	4.096	2.765	+0.49	1.71	9.53	14.85	2.78
14	62.8	15.1	2.378	13.15	3.600	2.824	+3.61	1.47	8.21	42.67	1.84
H	Acetone-Kerosine-Water										
2	23.7	21.5	10.17	15.46	4.228	4.641	−0.21	1.74	11.84	13.61	1.82
17	23.1	15.4	4.888	11.03	3.666	2.182	−0.44	1.04	7.06	22.25	2.18
19	40.2	15.9	2.972	11.03	4.129	2.011	−9.57	0.89	6.09	44.87	2.61
I	Propionix Acid–Methyl Isobutyl Ketone–Water										
1	26.2	18.7	1.748	6.463	3.871	0.908	−5.72	10.47	5.43	2.50	3.44
4	25.4	36.4	2.123	6.338	4.870	1.031	+0.80	12.22	6.33	2.08	5.75
	6-Mm. Glass Beads, 30-Inch Packed Height, 21 × 5/32-Inch Nozzles										
J	Acetone Toluene Water										
2	26.5	14.9	6.077	19.16	7.927	3.153	−3.91	4.248	5.794	6.24	2.57
8	12.1	29.8	13.87	20.74	14.93	3.280	−2.82	3.92	5.53	3.09	5.57
K	Acetone-Pegasol-Water										
1	23.1	12.7	3.567	9.314	2.742	1.595	−0.88	1.15	6.44	20.02	1.97
9	15.6	30.1	9.511	10.31	5.054	2.948	−6.27	1.65	9.19	9.47	3.28
L	Acetone-Kerosine-Water										
4	26.5	12.7	4.491	16.19	6.001	2.384	−8.15	0.73	4.94	36.45	2.57
9	17.1	23.1	8.719	16.05	9.182	2.963	−6.56	0.76	5.14	22.62	4.49
M	Propionic Acid–Methyl Isobutyl Ketone–Water										
3	25.6	12.1	1.189	5.945	3.303	0.601	−5.19	6.93	3.59	3.69	3.37
9	15.1	18.7	1.982	5.978	4.294	0.591	−4.92	17.35	3.81	2.05	4.91

[a] Extraction direction reversed. [b,c,d] Initial water concentration [b]8.983, [c]7.761, [d]7.596 lb. moles/Cu. ft. × 10^3

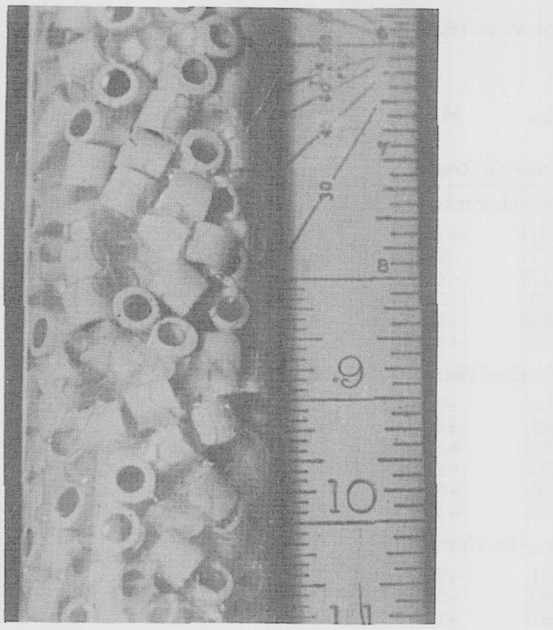

Figure 3. Conditions of the tower, packed with ⅜-in. Raschig rings

Extraction of acetone from dispersed Pegasol to water. V_W = 27.0 ft./hr. and V_P = 37.0 ft./hr.

can also be noted. With increased flow rate of the dispersed phase for a given rate of the continuous phase, the holdup gradually increased and the droplets showed a tendency to coalesce and form short slugs of the liquid. Figure 3 shows that at higher rates of the dispersed Pegasol phase, the drops tended to break down into a number of smaller drops. Some of these smaller drops might, in turn, coalesce to form larger drops. The units now seemed to be effectively utilized by the drops of the dispersed phase, and the increased holdup of the latter can be observed from the same figure. For a given system, the size of the droplets leaving the packing was observed to be nearly the same, and practically uneffected by increase in dispersed phase rate.

With the copper rings also, the drops of dispersed phase tended to coalesce and formed slugs at rates of flow lower than those for the Raschig rings.

With the glass beads, the column was densely packed, resulting in very low voids within the packing. The droplets inside the column could not maintain any definite shape, as they passed through the narrow interstices of the densely packed bed. Also, they coalesced even at fairly moderate rates of flow, depending on the nature of the system.

Glass beads gave approximately the same performance as the copper rings with acetone-water-kerosine, and acetone-water-Pegasol systems, whereas they gave transfer coefficients for acetone-water-toluene, and propionic acid–water–methyl isobutyl ketone systems, which were 10 to 15% lower than the coefficients with the copper rings. Somewhat large-sized drops were obtained with kerosine and Pegasol as dispersed liquids, while smaller and finer drops were formed with methyl isobutyl ketone. The droplets obtained with toluene dispersed were somewhat intermediate in size between kerosine and ketone droplets.

The above phenomena may be explained as follows: The increased coalescence of the dispersed phase due to the densely packed nature of the bed with 6 mm. glass beads, might more than offset the increase in interfacial area due to the smaller drops with methyl isobutyl ketone system, than with ¼-inch rings with the result that the transfer coefficients were reduced for the beads with the ketone system by about 15%. However, with kerosine and Pegasol as solvents, though the droplet size and holdup were observed to be somewhat different for ¼-inch rings and 6-mm. beads for similar flow rate conditions, the above factors might have so balanced each other, as to give nearly identical rates of mass transfer.

RESULTS AND DISCUSSION

Flow Rates. With the object of establishing the effect of flow rates on the efficiency of mass transfer, both dispersed and continuous phase flow rates were varied from 10 to 60 and 15 to 50 feet per hour, respectively, and the upper limits of these flow rates were within the flooding rates. For the system acetone-water-kerosine, the effect of the dispersed phase flow rate on mass transfer coefficients has been shown graphically in Figure 4 with continuous phase flow rate as parameter. It is clear from all these *Ka vs.* V_D plots (Figures 4 and 5) that the transfer coefficient, *Ka* increased with increased rate of the dispersed phase, owing to increased holdup of this phase. With further increase of V_D however, each of the curves showed a tendency to fall off, thereby indicating the less rapid rate of increase of coefficients with the flow rate, V_S.

This can be explained by the fact that the coalescence of the drops observed at high values of V_S might account for the less rapid increase in the mass transfer coefficients with increased dispersed phase flow rate.

At a constant flow rate of the dispersed phase, the effect of the continuous phase flow rate on the extraction coefficients was studied and a typical plot is shown in Figure 6. The extraction coefficient was almost constant, being very little influenced by the continuous phase flow rate for

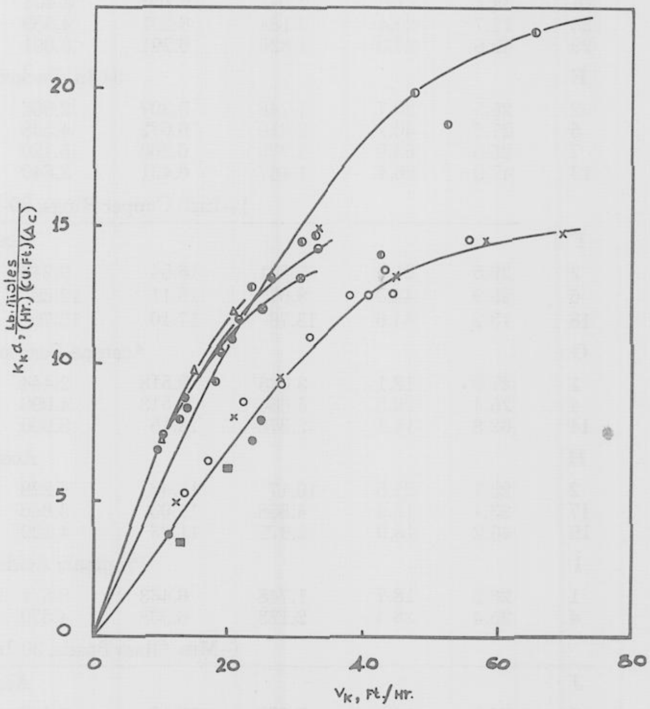

Figure 4. Effect of dispersed phase flow rate on over-all transfer coefficient, $K_K a$. Extraction of acetone between kerosine and water under different conditions of extraction

Symbol	Packed Height of Column, In.	Distributor Nozzels, In.	Packing, Rings	Extraction Direction	V_W, Ft. per Hr.
○	56	19 × 5/32	⅜-in.	K → W	25
×	56	19 × 5/32	⅜-in.	K → W	50
●	56	19 × 5/32	⅜-in.	W → K	25
■	56	21 × 5/32	⅜-in.	W → K	25
◐	34	21 × 5/32	⅜-in.	K → W	26
⊖	30	21 × 5/32	¼-in.	K → W	24
△	30	21 × 5/32	¼-in.	K → W	12
⊗	30	21 × 5/32	¼-in.	K → W	12

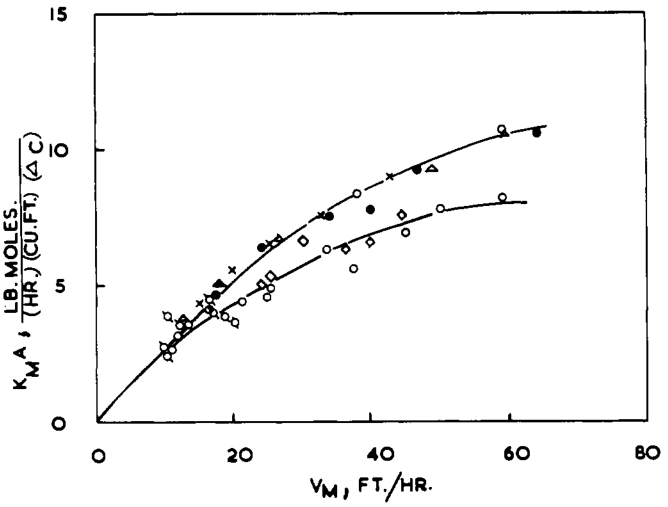

Figure 5. Effect of dispersed phase flow rate on over-all transfer coefficient, K_Ma. Extraction of propionic acid from dispersed methyl isobutyl ketone to a continuous water phase in a 1.88 in. i.d. tower

Symbol	Packing	Packed Height, In.	V_W, Ft./Hr.	V_D, Ft./Hr.
○	3/8-in. rings	56	25	10 to 60
×	3/8-in. rings	56	47	10 to 60
●	3/8-in. rings	34	25	10 to 60
▲	3/8-in. rings	34	50	10 to 60
◇	1/4-in. rings	30	25	10 to 45
○	6 mm. beads	30	15	10 to 20

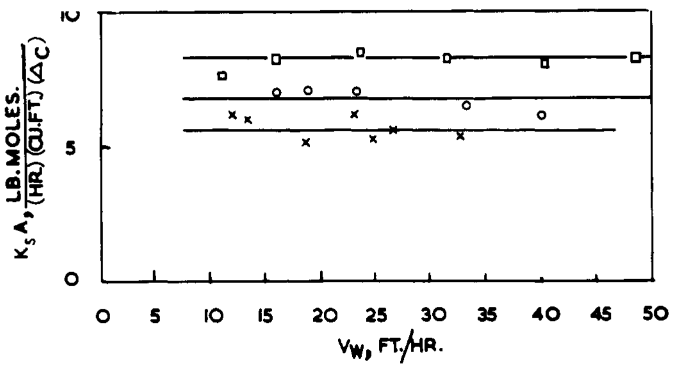

Figure 6. Effect of continuous phase flow rate on the over-all transfer coefficient, K_Da. Extraction of acetone from each of the dispersed solvents to a continuous water phase, with 1/4-inch copper rings as packing

Symbol	System	V_D, Ft./Hr.
×	1	24
□	2	15
○	3	16

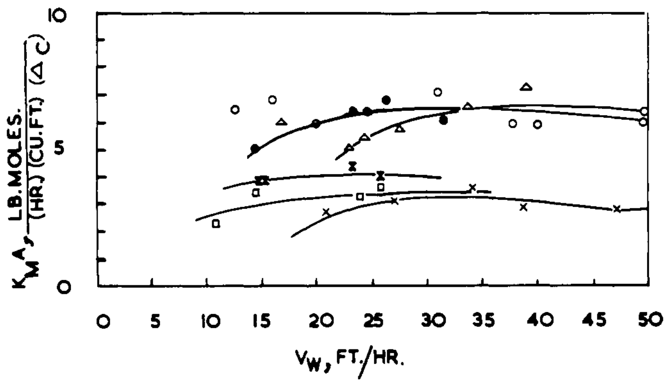

Figure 7. Effect of continuous phase flow rate on over-all transfer coefficient, K_Da. Extraction of propionic acid from methyl isobutyl Ketone (dispersed) to water

Symbol	Packing	V_D, Ft./Hr.
○	3/8-in. Raschig rings	25
●	3/8-in. Raschig rings	48
⊗	3/8-in. Raschig rings	12
△	1/4-in. copper rings	25
□	6-mm. glass beads	12
⧗	6-mm. glass beads	17

acetone-water-solvent systems. On the other hand, for propionic acid-water-methyl isobutyl ketone system (Figure 7), the continuous phase flow rate had practically no effect on the transfer coefficients above 25 feet per hour, while it slightly increased the coefficient below this flow rate. For the different conditions studied in the packed tower, the capacity coefficients were first correlated as functions of the flow rates, and the resulting empirical equations were presented in Table V for the ring packings.

Column Length. Figure 4 shows that the mass transfer efficiency in the extraction of acetone from kerosine to water was about 40% greater in the short tower (34 inches of packing) than in the long tower (56 inches of packed height) for the Raschig rings as packing material.

Appreciable end effects apparently existed in the short tower, and these became inappreciable as the tower height was increased. However, the performance of the column, packed with the Raschig rings for the extraction of propionic acid from dispersed methyl isobutyl ketone to a continuous water phase, was observed to be nearly the same for the two column heights (34 and 56 inches of packed heights) as was indicated by the almost equal transfer coefficients for identical conditions of flow rates.

The probable explanation seems to be that the delayed coalescence observed with the larger sized drops of kerosine must have caused an entrance effect at the continuous phase inlet, and this might explain the observed end effect with acetone-water-kerosine system.

While extracting propionic acid from dispersed methyl isobutyl ketone to water, however, the droplets of the ketone were smaller, and they passed up through the packing rather faster than the larger drops of kerosine. The moment these ketone droplets approached the interface, they readily coalesced with it.

Similar observations as regards the effect of end effects in reducing the efficiency of mass transfer in packed extraction towers have been made by Comings and Briggs (*10*), Pratt and Glover (*23*), and Leibson and Beckmann (*19*). They observed the effect to be rather small, if not entirely negligible, in a packed extraction tower. Also the

Table V. Empirical Correlation of Capacity Coefficients as Functions of Flow Rates for Packed Extraction Towers

System[a]	Packed Height, In.	Approximate Flow Rate Range, Ft./Hr. V_C	V_D	$Ka = \alpha V_S^n$ or V_W^m
		3/8-In. Raschig Rings		
2	56	15–60	12–60	$K_Pa = 0.8\ V_P^{0.74}$
3	56	15–60	12–60	$K_Ka = 0.8\ V_K^{0.74}$
3[b]	56	15–60	12–60	$K_Ka = 0.52\ V_K^{0.73}$
3	34	15–60	12–60	$K_Ka = 1.25\ V_K^{0.73}$
4	56	25–60	12–60	$K_Ma = 0.49\ V_M^{0.70}$
4	34	25–60	12–60	$K_Ma = 0.59\ V_K^{0.73}$
		1/4-In. Copper Rings		
1	30	12–50	12–50	$K_Ta = 1.3\ V_T^{0.48}$
2	30	12–60	12–50	$K_Pa = 1.8\ V_P^{0.52}$
3	30	12–60	12–50	$K_Ka = 2.2\ V_K^{0.53}$
4	30	12–60	12–50	$K_Ma = 2.3\ V_M^{0.30}$

[a] Systems defined in text under materials.
[b] Direction of extraction reversed.

recent studies of Gier and Hougen (*15*) on the end effects in spray and packed columns showed that the end effect was at the continuous phase entrance resulting from bulk mixing (or recirculation) in the continuous phase at its inlet. This bulk mixing effect was observed by these authors to be more significant in spray columns of high diameter to height ratio, whereas it should be restricted in packed extraction columns.

Direction of Extraction. An examination of Figure 4 makes it clear that the efficiency of mass transfer while extracting acetone from water to kerosine was approximately the same as while extracting it in the reverse direction.

It may be explained as follows: In a spray tower, the direction of extraction plays an important part in that coalescence and breaking up of drops at high flow rates are predominant, but extracting the solute from the dispersed phase in a packed tower, however, the packing more or less controls the drop size; hence, the mass transfer coefficients were approximately the same for both directions of extraction.

HTU CORRELATIONS

In view of the advantages of HTU's over *Ka*'s for the analysis of mass transfer data, the results were also expressed in terms of the over-all HTU's which were plotted according to Colburn's original Equations 5 and 6 in Figures 8 to 11. The slope and intercept relationships were reasonably satisfied in all cases. These slope and intercept values may be regarded as indicative of the film values of transfer units, and were listed in Table VI. Values of these individual film HTU's ranged from 0.85 to 2.85 feet for the dispersed phase, and zero to 5.60 feet for the continuous phase, depending on the nature of the system and type of packing used.

With a view to correlate these film HTU values with the system variables and packing characteristics, on the assumption that the film HTU's will not vary with flow rates (which was reasonably satisfied with the data in the present investigation), the film HTU values for the dispersed and continuous phases were plotted against the Schmidt group ($\mu/\rho D_v$) on log-log coordinates. Values of

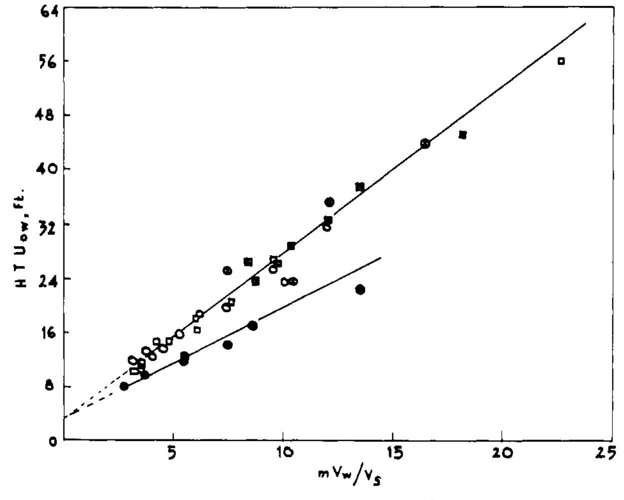

Figure 8. Over-all continuous phase transfer units: Extraction of acetone from dispersed solvent to continuous water phase in a 1.88-in. i.d. tower, packed with $\frac{3}{8}$-inch Raschig rings

Symbol	Solvent	Packed Height, In.	V_W, Ft./Hr.	V_D, Ft./Hr.
○	Kerosine	56	25	10 to 50 range
⊗	Kerosine	56	50	10 to 50 range
●	Kerosine	34	26	10 to 50 range
□	Pegasol	56	27	10 to 50 range
⊠	Pegasol	56	50	10 to 50 range

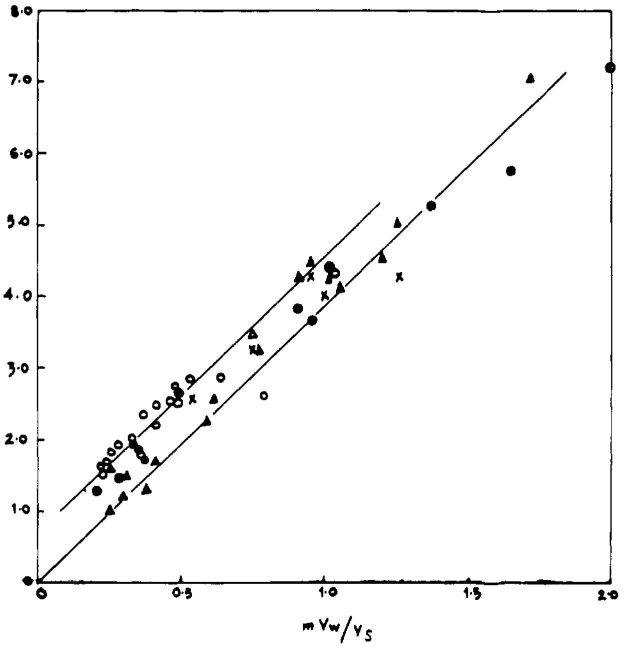

Figure 9. Over-all continuous phase transfer units. Extraction of propionic acid from dispersed methyl isobutyl ketone to continuous water phase in a 1.88-in. i.d. tower, packed with $\frac{3}{8}$-inch Raschig rings

Symbol	Solute	Dispersed Phase
○	Acetone	Kerosine
□	Acetone	Pegasol
×	Acetone	Toluene
△	Propionic acid	MIBK

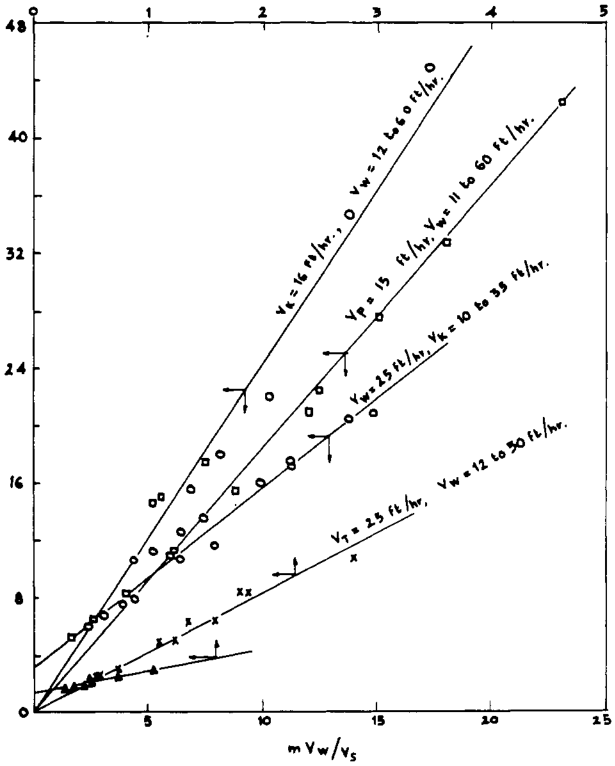

Figure 10. Over-all continuous phase transfer units: Extraction in a 1.88-in. i.d. tower, packed with $\frac{1}{4}$-inch copper rings, from solvent (dispersed) to water (continuous): A comparison of systems

Symbol	Solute	Dispersed Phase	Direction of Extraction
×	Acetone	Kerosine	K → W
⊗	Acetone	Pegasol	P → W
■	Acetone	Toluene	T → W
●	Propionic acid	MIBK	M → W

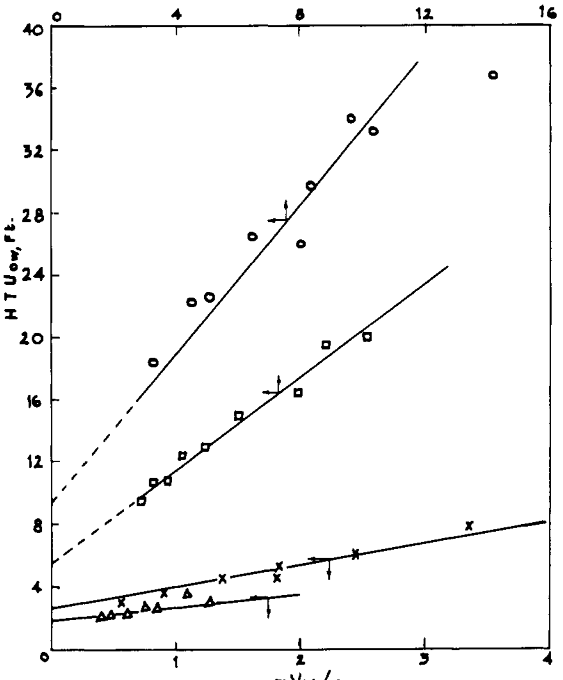

Figure 11. Over-all continuous phase transfer units: Extraction in a 1.88-in. i.d. tower, packed with 6-mm. glass beads, from solvent (dispersed) to water (continuous) phase. A comparison of systems

For legend, see Figure 10

diffusivity in the Schmidt group were calculated by the method of Wilke (*32*). Both HTU_W and HTU_S correlated satisfactorily with a slope of 0.68 for the Schmidt number. Having established the effect of $\mu/\rho D$, it was attempted to correlate the film HTU values with the packing characteristics. A plot of $\text{HTU}_S/(\mu/\rho D_v)^{0.68}$ against (a/F^3) was made for the present data on the ring packings. A slope of -0.8 was obtained from the above plot. $(\text{HTU})_W$ values could not, however, be correlated satisfactorily with the packing characteristics. The same value of the exponent was obtained on the graph for the data of Leibson and Beckmann (*19*) on ring packings of different sizes. The functional relationship between $(\text{HTU})_S$, $\mu/\rho D_v$ and a/F^3 could now be written as

$$(\text{HTU})_S = K_1\,(\mu/\rho D_v)^n\,(a/F^3)^m \qquad (7)$$

where the constants m and n were found to be -0.8 and 0.68 respectively.

Table VI. Correlations for Packed Extraction Tower

Packing	System	Packed Height, In.	HTU_S Ft.	HTU_W Ft.
3/8-in. Raschig rings	Acetone-water Pegasol	56	2.00	3.0
	Acetone-water-kerosine	56	2.45	3.0
		34	1.65	3.0
1/4 inch copper rings	Acetone-water-Pegasol	30	1.25	3.0
	Acetone-water-kerosine	30	1.30	3.0
	Acetone-water toluene	30	1.75	3.25
		30	1.75	1.62
	Propionic Acid–water–methyl isobutyl ketone	30	1.10	1.60
6 mm. glass beads	Acetone-water-toluene	30	1.40	2.60
	Acetone-water-Pegasol	30	1.48	5.60
	Acetone-water-kerosine	30	2.40	9.30
	Propionic Acid–water–methyl isobutyl ketone	30	0.85	1.80

In order to evaluate K_1 in the above equations, a plot of $(\text{HTU})_S$ against $(\mu/\rho D_v)^{0.68} \times (a/F^3)^{-0.8}$ has been shown in Figure 12, for the present data on ring packings, the data of Leibson and Beckmann on the system, diethylamine-water-toluene; and the data of Pratt and Glover (*23*) on the system acetone–water–vinyl acetate for ring packings. The data of Leibson and Beckman were represented by the above equation with a value of $K_1 = 10.3$ while the present data and that of Pratt and Glover were represented by $K_1 = 4.4$. The variation of K_1 from 4.4 to 10.3, therefore, showed that there are other factors like interfacial tension etc., which also appeared to influence the extraction coefficients. Since the present study included a variation in the system properties ($\mu/\rho D_v$ ranging from 295 to 1260) a value of 4.4 for K_1, satisfying the above equation for a number of different systems, seems to be more justified for design purposes than the value of 10.3 for K_1, resulting from the data on the single system, diethylamine-water-toluene.

Hence for design purposes, a suitable value of K_1 must be selected to calculate HTU_S for any given system, thereby giving due allowance for variation in the other properties of the system.

In view of the differences in void spaces of the solid bead packings as compared with the more open ring packings, marked difference in behavior were observed between solid beads and the more open rings. Hence, it seemed reasonable to treat the data for the beads separately from the ring-type packings.

Due to the densely packed nature of the bed with 6-mm. beads, the effect of packing variables on mass transfer could not be established for these packings. Hence, the constant, K_1 and the packing factor (a/F^3) in Equation 7

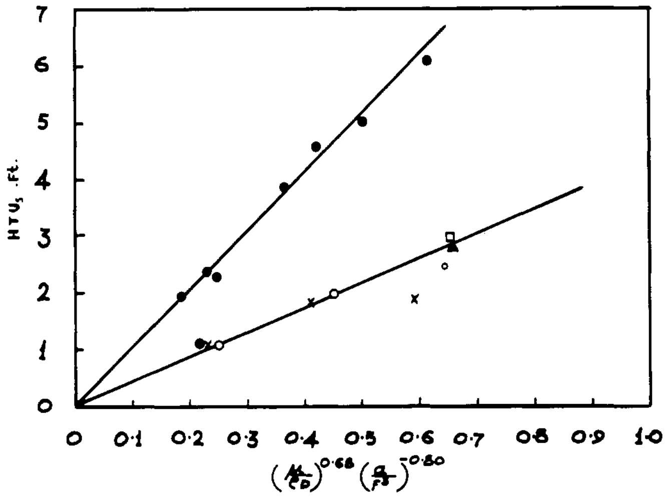

Figure 12. Final correlation of dispersed phase film transfer unit with Schmidt number and packing factor

Symbol		System	Source
○	3/8-in. Raschig rings	Acetone-water-kerosine Acetone-water-Pegasol Propionic acid–water–MIBK	This work
×	1/4-in. copper rings	Propionic acid–water–MIBK	This work
⊗	1/4-in. copper rings	Acetone-water-toluene	
▲	3/8-in. Raschig rings	Acetone–water–vinyl acetate	23
●	1/4, 1/2, and 1-in. ring packings	Diethylamine-water-toluene	19

were grouped together as K_2 and the equation for $(\text{HTU})_S$ for the solid beads could be written as

$$(\text{HTU})_S = K_2(\mu/\rho D_v)^{0.68} \tag{8}$$

with a value of K_2 ranging from 0.017 to 0.03 for the different systems reported in this investigation.

Further work will be necessary on more systems before a more general correlation can be developed. A more extensive study with systems and packings, varied over a wide range of properties, is in progress in these laboratories, and a more general correlation is close at hand.

NOMENCLATURE

a = interfacial contact area per unit tower volume, surface area of packing per cu. ft.
C = concentration in lb. moles/cu. ft.
ΔC = over-all concentration difference in lb. moles/cu. ft.
D_v = diffusivity
g = acceleration due to gravity
x = fractional volumetric holdup of dispersed phase, cu. ft./cu. ft.
HTU_{OX} = height of over-all transfer unit based on X-phase in ft.
HTU_X = height of individual transfer unit of X-phase in ft.
ka = individual volumetric mass transfer coefficient in lb. moles/(hr.)(cu. ft.)(ΔC)
m = slope of equilibrium line = dC_c/dC_d or dC_w/dC_s
N = N'/θ = rate of solute transfer in lb. moles/hr.
Re = Reynolds' number
U = superficial velocity in ft./hr., (flow rate of phase referred in cu. ft./(hr.)(sq. ft.)
v = volume of tower
ϵ or F = void fraction
σ or γ = interfacial tension in dynes/cm.
ρ = density in lb./cu. ft.
ρ' = specific gravity
μ = viscosity in lb./ft.(sec.), μ' = viscosity in centipoises
K_1 = constant in the correlation for ring packings
K_2 = constant in the correlation for bead packings
θ = time in hours

Subscripts

W or C = water, continuous phase
S or D = solvent, dispersed phase
K,M,P,T = kerosine, methyl isobutyl ketone, Pegasol, and toluene, respectively
l.m. = logarithmic mean
av = average

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LITERATURE CITED

(1) Allerton, J., Strom, B.O., Treybal, R.E., *Trans. Am. Inst. Chem. Engrs.* **39**, 361 (1943).
(2) Appel, F.J., Elgin, J.C., *Ind. Eng. Chem.* **29**, 451 (1937).
(3) Berg, C., Manders, M., Switzer, R., Paper presented to the Am. Inst. Chem. Engrs., Los Angeles, Calif. March 9, 1949.
(4) Blanding, F.H., Elgin, J.C., *Trans. Am. Inst. Chem. Engrs.* **38**, 305 (1942).
(5) Chu, J.C., Taylor, C.C., Lerry, D.J., *Ind. Eng. Chem.* **42**, 1157 (1950).
(6) Claffey, J.B., Badgett, C.O., Skalamera, J.J., Phillips, G.W.M., *Ibid.*, **42**, 166 (1950).
(7) Colburn, A.P., *Trans. Am. Inst. Chem. Engrs.* **35**, 211 (1939).
(8) *Ibid.*, **39**, 383 (1943).
(9) Colburn, A.P., Welsh, D.G., *Ibid.*, **38**, 179 (1942).
(10) Comings, E.W., Briggs, S.W., *Ibid.*, **31**, 639 (1935).
(11) Elgin, J.C., Browning, F.M., *Ibid.*, **31**, 639 (1935).
(12) Gaylor, H., Pratt, H.R.C., *Trans. Inst. Chem. Engrs. (London)* **29**, 110 (1951).
(13) *Ibid.*, **31**, 69 (1953).
(14) Gaylor, H., Roberts, N.W., Pratt, H.R.C., *Ibid.*, **31**, 57 (1953).
(15) Gier, T.E., Hougen, J.O., *Ind. Eng. Chem.* **45**, 1362 (1953).
(16) Jayarama Rao, D.Sc. Thesis, Andhra University, December 1955.
(17) Johnson, F.H., Bliss, H., *Trans. Am. Inst. Chem. Engrs.* **42**, 331 (1946).
(18) Laddha, G.S., Smith, J.M., *Chem. Eng. Progr.* **46**, 195 (1950).
(19) Leibson, I., Beckmann, R.B., *Ibid.*, **49**, 405 (1953).
(20) Lewis, J.B., Jones, I., Pratt, H.R.C., *Trans. Inst. Chem. Engrs. (London)* **29**, 126 (1951).
(21) Meissner, H.P., Stokes, C.A., Hunter, C.M., Morrow, G.M., *Ind. Eng. Chem.* **36**, 917 (1944).
(22) Morello, V.S., Beckmann, R.B., *Ibid.*, **42**, 1078 (1950).
(23) Pratt, H.R.C., Glover, S.T., *Trans. Inst. Chem. Engrs. (London)* **24**, 54 (1946).
(24) Raja Rao, M,D.Sc. thesis, Andhra University, June 1956.
(25) Row, S.B., Koffolt, J.H., Withrow, J.R., *Trans. Am. Inst. Chem. Engrs.* **37**, 559 (1941).
(26) Rushton, J.H., *Ind. Eng. Chem.* **29**, 309 (1937).
(27) Sherwood, T.K., Evans, J.E., Longcor, J.V.A., *Trnas. Am. Inst. Chem. Engrs.* **35**, 597 (1939).
(28) Smythe, R.W., Senior thesis, Carnegie Institute of Technology, Pittsburgh, Pa., 1950.
(29) Treybal, R.E., "Liquid Extraction," p. 335, McGraw Hill, New York, 1951.
(30) Varteressian, K.A., Fenske, M.R., *Ind. Eng. Chem.* **22**, 928 (1936).
(31) Wicks, C.E., Beckmann, R.B., *A.I.Ch.E. Journal* **1**, 426 (1955).
(32) Wilke, C.R., *Chem. Eng. Progr.* **45**, 218 (1949).

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CORRECTION

In "Fall of Liquid Drops in Water. Drag Coefficients, Peak Velocities, and Maximum Drop Sizes" [P.M. Krishna, D. Venkateswarlu, and G.S.R. Narasimhamurty, *J. Chem. Eng. Data* **4**, 340 (1959)] Equations 5, 6, 8, and 9 on page 343 should read as follows

$$(\text{Re}/\text{We})_P = 0.3592\, Sd^{0.8260} \tag{5}$$

$$U_P = 2.784/Sd^{0.8260} \tag{6}$$

$$(\text{Re}/\text{We})_M = 0.6883\, Sd^{0.7567} \tag{8}$$

$$U_M = 1.453/Sd^{0.7587} \tag{9}$$

The constants in the equations may be rounded off as suggested by Markowitz (*3*) to have the correlations workable and meaningful.

Regarding the Weber number, this dimensionless group is stated as $D\rho U^2\sigma$ by Hu and Kintner (*1*) and Johnson and Braida (2).. Other workers have defined the Weber number as $D\rho U^2\sigma g_c$. If it is necessary to include g_c to be consistent with the definition of the *Sd* group, it can be defined as $D\rho U^2/\sigma g_c$.

(1) Hu, S., Kintner, R.C., *A. I. Ch. E. Journal* **1**, 42 (1955).
(2) Johnson, A.I., Braida, L., *Can. J. Chem. Eng.* **35**, 165 (1957).
(3) Markowitz, Joel, 9 Wainwright Ave. Yonkers, New York, Private communication.

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