Mass Transfer and Axial Dispersion in a Kühni Extraction Column

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Concentration profiles were determined in both phases for the system methyl isobutyl ketone (MIBK)—water-acetic acid in a Kühni 72.45-mm-dia. extraction column of 25 stages. Mass transfer and backmixing parameters were estimated numerically from the profiles in terms of the backflow model. The results indicated that backmixing of the continuous phase was appreciable, while that of the dispersed phase was negligible. Independent values of the continuous-phase backmixing coefficient, α_c, were also determined by both unsteady-state and continuous tracer injection, in the former case both with and without mass transfer. The results indicated that α_c is increased by mass transfer, and that the tracer values, both with and without mass transfer, are greater than those obtained from the profile data.

Introduction

An important step in the design of liquid-liquid extraction columns is to determine the column height in the case of differential contactors such as spray and packed columns or the number of stages in stagewise contactors (e.g., the rotary agitated type) such as the Kühni column.

In early design procedures, ideal plug-flow conditions were assumed in both phases for the estimation of height or number of stages required for the desired separation. However, it is now widely accepted that axial dispersion in one or both phases can lead to considerable deviations from ideal plug flow, resulting in an increase in the height or number of stages required to achieve the design specification.

Two models are available (Sleicher, 1959, 1960) to accommodate the effect of axial dispersion: the diffusion and the backflow model. In the diffusion model, applicable to differential-type contactors such as the packed column, nonideality is expressed in terms of an axial dispersion coefficient, E. In contrast, the backflow model is basically a stagewise model, in which total mixing is assumed within each stage, and the deviation from plug flow is represented by a backmixing coefficient, α, between adjacent stages. Thus, in the design of mechanically agitated stagewise columns, it is more appropriate to use the backflow model. Analytical solutions are available for simplified cases of both models, and numerical techniques are also applicable for design and scale-up purposes (Pratt and Baird, 1983).

Another form of axial dispersion, known as forward mixing, can also occur in the dispersed phase. This is a result of the distribution of droplet sizes and velocities within the column, which leads to an additional reduction in performance, independent of that due to backmixing, if any, in that phase (Olney, 1964). As a consequence, the models are increased considerably in complexity and require numerical techniques, together with appropriate experimental data, for the design and simulation of columns.

These models all require an accurate knowledge of both backmixing and mass transfer parameters, together with additional information on drop size and velocity distribution in the case of the forward mixing model, at the specified design conditions for the given system and type of column.

In the past, much research has been dedicated to the understanding of continuous-phase axial mixing and to the estimation of the appropriate backmixing parameters. Most of the earlier studies were performed on single-phase systems using tracer injection techniques in the absence of mass transfer, and the results were extrapolated to two-phase systems. It was later realized that such extrapolation does not include a realistic contribution of the dispersed phase to such backmixing, and research has therefore since been directed mainly to the studies of backmixing in two-phase systems. However, since such studies cannot quantify the effect of mass transfer

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on continuous-phase backmixing, it may not be appropriate to use parameters obtained from tracer studies without modification for design purposes. Thus, it is necessary to develop both a procedure and an experimental technique for the estimation of backmixing parameters in the two phases together with the mass transfer parameter. To date, however, little research has been done in this area.

The aim of this work is to determine both mass transfer and backmixing parameters simultaneously from solute concentration profiles. For this purpose, a laboratory-scale, Kühni-type, mechanically-agitated stagewise column was employed, and was operated with methyl isobutyl ketone (MIBK) and deionized water as solvents and acetic acid as solute. Concentration profiles of the solute along the column were measured in both phases, and used to estimate the mass transfer and backmixing parameters in terms of the backflow model. Independent determinations of the backmixing parameters were also conducted using both transient and continuous tracer injection methods.

Previous Studies

Kühni column

The Kühni contactor represents a logical development of the rotary disc and multiimpeller (Oldshue-Rushton) columns, in which the baffled impellers of the latter are replaced by unbaffled double-entry turbines, and the annular partition plates between stages in the former are replaced by perforated stator plates. Although several hundred Kühni columns are reported to be in use for research, process development and industrial processing (Mögli and Bühlman, 1983), data for use in design and scale-up are limited.

A review of published work up to 1983 on various aspects of throughput and mass transfer for performance has been given by Mögli and Bühlman (1983). Since then, data on single- and two-phase axial mixing have been reported by Elasser and Bühlmann (1983), and on single-phase axial mixing by Vatai and Tolic (1983) and by Breysse et al. (1983). Bailes et al. (1986) presented data on holdup and drop size, and further holdup data were given by Kumar et al. (1986). Hydrodynamic and mass transfer data have been reported by Goldmann and Blass (1983), while Steiner et al. (1988) have presented axial dispersion data obtained with simultaneous mass transfer.

The most recent study of mass transfer in the Kühni column has been described by Kumar and Hartland (1988), who obtained mass transfer and backmixing parameters from experimental solute concentration profiles for a 150-mm-dia. column. In this work, which was published toward the end of the present study and to which it is somewhat parallel, the concentration profiles were simulated assuming 36 hypothetical stages instead of the 18 actual compartments in the column so that the physical significance of the results is not clear.

Axial dispersion and mass transfer

The most common methods of obtaining data on axial dispersion make use of tracer injection. Such tracers generally take the form of dyes or inorganic salts, although fluorescent and radioactive compounds are sometimes used. Two methods that employ tracers are available: continuous tracer injection with monitoring of the response at several downstream locations, and unsteady-state injection with upstream monitoring.

The continuous injection method, while somewhat more complex to apply, is simple to interpret and gives a measure of true backmixing. The unsteady-state method, on the other hand, is easier to apply, especially if a simple pulse (Dirac delta function) of tracer is used. However, the resulting data are more difficult to interpret and give strictly a measure of the residence time distribution (RTD). The solution for a delta function injection has been given by Levenspiel and Smith (1957) in terms of the Peclet number, *Pe*, for the diffusion model; they also showed that *Pe* can conveniently be obtained from the mean and variance of the output curve. Errors are introduced by the practical impossibility of producing a perfect delta function and distributing it over the column cross-section; these can be overcome by measuring the responses at two downstream locations, as shown by Aris (1959), Van der Laan (1960), and Bischoff (1960); see also Bischoff and Levenspiel (1962).

Unfortunately, in the past, data for stagewise contactors have been interpreted in terms of the diffusion model, i.e., as *Pe*, rather than the backflow model, for which the backmixing ratio, α, is the appropriate parameter. Solutions for this model, which are more complex, have been obtained by Roemer and Durbin (1967) and Sawinsky and Hunek (1981); they have been generalized by Dongaonkar (1990) in a form that is used in the present work.

The use of tracer methods, especially in the absence of mass transfer, has been criticized on the grounds that they do not necessarily apply to the practical operation of columns. Rod (1965) and Mecklenburgh and Hartland (1967) have described graphical methods for the determination of both mass transfer and backmixing parameters from measured concentration profiles for each phase along a column. Such methods, however, are subject to the normal inaccuracies of graphical procedures, and both Spencer et al. (1981) and Rod et al. (1983) have described numerical procedures based on matrix formulations by means of which the various parameters can be determined from measured concentration profiles. The latter workers also derived important conclusions as to the influence of sample point locations on the accuracy of the derived parameters by a numerical study of the influence of perturbations introduced into exact, computed profiles. Such a procedure is used in the present work to obtain the mass transfer parameter, together with the backmixing parameters for comparison with those obtained by tracer injection techniques.

Experimental Studies

Main column

The main section of the column was constructed from two 1.0-m-long precision bore glass sections with an internal diameter of 72.45 mm; this encloses 25 50-mm-high compartments separated by stainless steel partition plates. Three 72.45-mm-dia. stainless steel sleeves were used to enclose three of the compartments in the region of the buttress joint between the glass sections to avoid bypassing of liquid in the expanded ends of the precision bore glass tubes. A 250-mm-long glass tee section with a 25-mm outlet for the dispersed phase was attached to the top of the column; this, together with the upper 0.56 m of the main column, formed the coalescence and interface section.

A 76- to 101-mm-ID, 150-mm-long glass expansion piece,

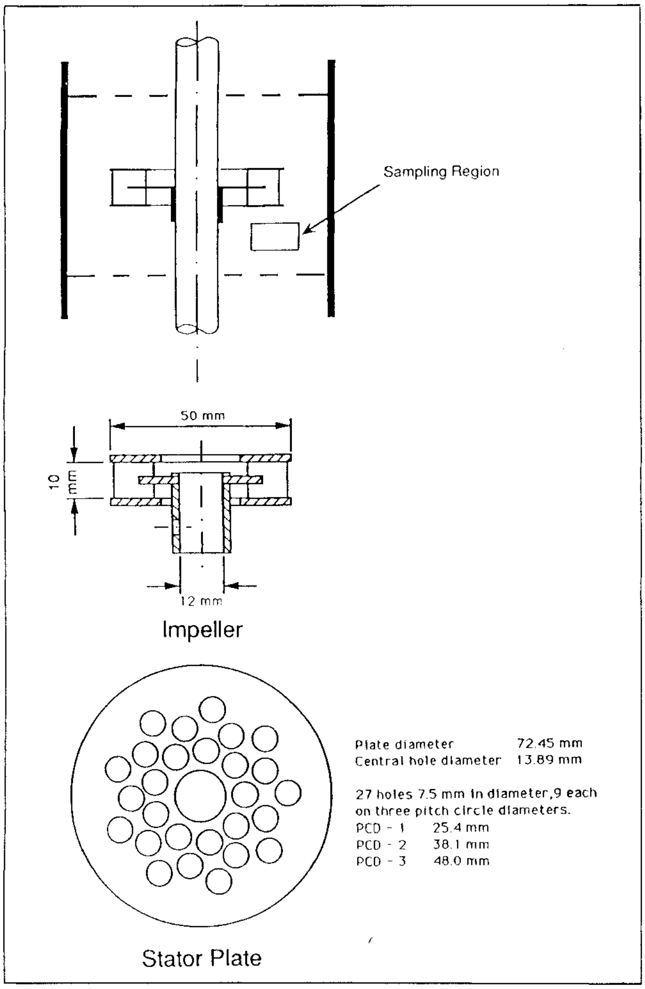

Figure 1. Stages, impellers, and stator plates.

with a 150-mm stainless steel conical adapter below, was attached to the column base. This contained the dispersed-phase distributor, which consisted of a stainless steel cone into which were fitted seven 2.64-mm-dia. nozzles. The continuous-phase distributor consisted of two 4.76-mm stainless steel tubes with closed ends passing through the top plate. These were drilled each with 3-3.2-mm holes which distributed the continuous phase radially about 50 mm above the top stator plate.

The impellers, supplied by Kühni A.G., were of 50-mm-dia. and were mounted on a 12-mm-dia. shaft passing through the column and provided with Teflon bearings in the 1st, 13th and 25th stator plates. This was driven by an eight-vane air motor mounted above the column. The rotor speed was measured to an accuracy of ±1 rpm using a portable noncontact digital tachometer. The 1.57-mm stainless steel stator plates were drilled each with 27 holes of 7.5 mm dia., together with a 14.5-mm-dia. central hole, giving a free area of 30%. Details of the compartments, impellers, and stator plates are shown in Figure.

Auxiliary equipment

The general arrangement of the equipment is shown in Figure 2. The continuous and dispersed phases were contained in 250-L stainless steel tanks and were pumped by magnetically-driven centrifugal pumps made of Ryton (polyethylene sulphide) with fully enclosed 0.25-hp (0.19-kW) flameproof motors. Piping of 6.35 and 9.53 mm OD, and compression fittings were of Type 316 stainless steel. Flows were controlled by diaphragm valves with Teflon inserts; stop valves were provided on both phase inlets and on the continuous-phase outlet, arranged so that all three could be closed simultaneously for holdup measurement. Interface control was effected by means of a needle valve on the continuous-phase outlet.

Spectrophotometers (Pye Unicam SP6450, UV/VIS) were connected on line at the 17th and 24th stages for measurement of tracer concentrations. The responses were recorded continuously on a two-pen chart recorder. Droplet size was measured photographically using an Olympus OM-4 35-mm camera with an extension tube and two close-up lenses together with the standard 50-mm lens. To avoid distortion, the column was surrounded by two water-filled viewing boxes made of Perspex, one on each side of the joint between the column sections, that could be moved up and down the column subject to restrictions due to the sample probes, as will be discussed later. These were blackened except for a 1.5-mm slit on one surface for the light source and a square window for the photography (Figure 3), enabling photographs to be taken of droplets illuminated by the plane of light within the column. The light source was a Sunpak MX124 flash, and Kodak Tmax 400 ASA black and white film was used, with a f16 stop and 1/60-s shutter speed.

Sample points

Eleven 12-mm-dia. sampling points were provided on the column in stages 1, 2, 4 and 8 of the upper section (i.e., above the stainless steel sleeve), and 11, 14, 17, 21, 24, 26 and 27 of the lower section. These were located approximately halfway between the stator plates and the impellers immediately above (Figure 1); they were drilled in the glass column sections using a Sonorode 120 ultrasonic drill.

Sample head probes of stainless steel and Teflon for the aqueous and solvent phases of the type described by Bonnet and Jeffreys (1983) were first tried but were replaced by simple conical cups which were equally effective (Figure 4). A hole was provided in the Teflon cup to allow the excess organic phase to escape. The sample heads were connected to hypodermic needles, one of each of which passed through tapered Teflon plugs which were inserted in the holes in the column wall and held in place with worm-drive clamps The needles were connected externally to 3.18-mm-OD stainless steel tubes, 150 and 200 mm long, for the two types of head, and sample flow rates were regulated using screw clamps on flexible tubes leading to the sample bottles.

It was found that uncontaminated samples of the aqueous phase were obtainable without difficulty at rates up to 3 cm^3/min. Some difficulty was encountered with the dispersed phase; however, satisfactory samples at up to the same rates were obtainable from more than five points in 80% of the experimental runs and from more than seven in 60%. The resulting solvent concentration profiles were therefore interpolated for the stages giving contaminated samples.

Materials used

Deionized water was used as the continuous phase, together with technical grades of methyl isobutyl ketone (MIBK) and

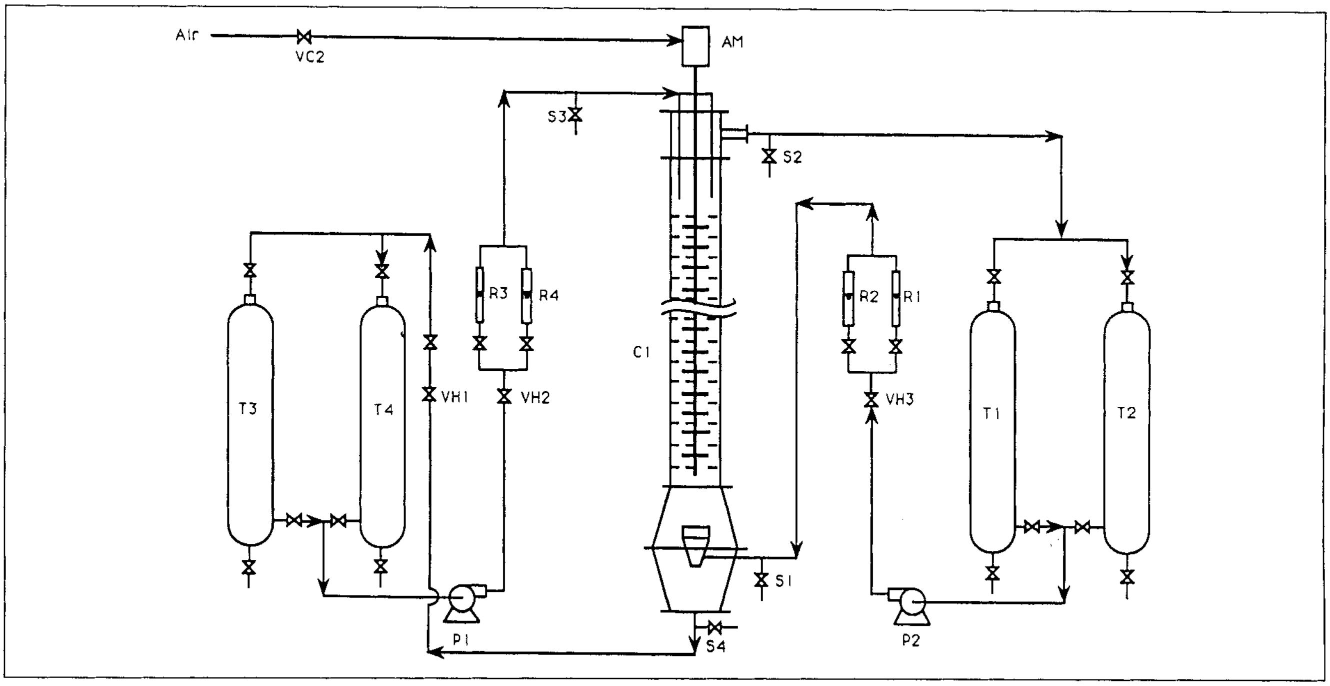

Figure 2. General arrangement of equipment.

acetic acid. The mutually saturated phases had densities of 997 and 809 kg/m^3, and viscosities of 0.97 and 0.69 mPa·s at 20°C; the interfacial tension for the solute-free system was 10.1 mN/m, measured both by du Noüy ring and drop weight methods (Prvcic, 1987), falling to 6.3 mN/n with 6% of acetic acid present in the aqueous phase at equilibrium.

Tartrazine (Acid Yellow 23) was used, as a 1.0% solution in water, as tracer in the measurements of backmixing coefficients. This was found by Fitzpatrick et al. (1986) to give satisfactory results in a pulsed column, free from the major effects on holdup and flood-point encountered with salt tracers.

Experimental Procedures

Concentration profile measurement

In all cases, the solvents were mutually saturated before the experiments by repeated circulation through the column. For the concentration profile experiments, acetic acid was added to the feed phase (generally the aqueous phase) to give an initial concentration of 5 to 7 mass %. The flows were then brought up to the desired rates and the column was allowed to operate under steady-state conditions for about two hours, corresponding to four to six changes of continuous phase. Samples were collected for 1.5 to 2.0 hours at flows of 0.5 to

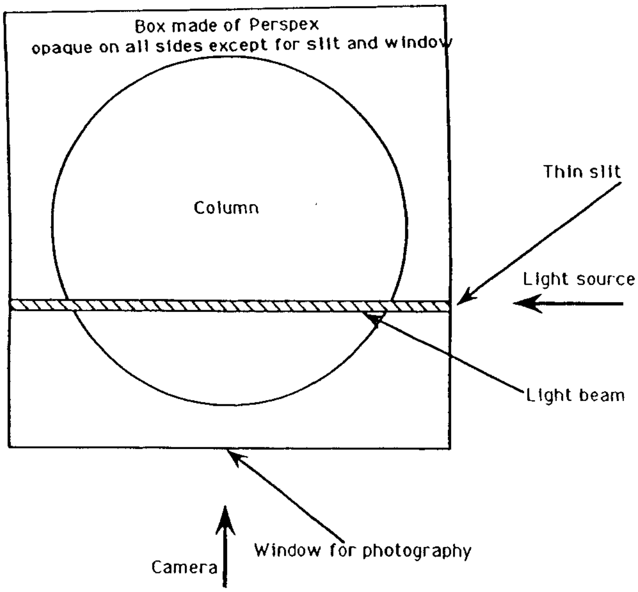

Figure 3. Photographic technique for droplet size measurement.

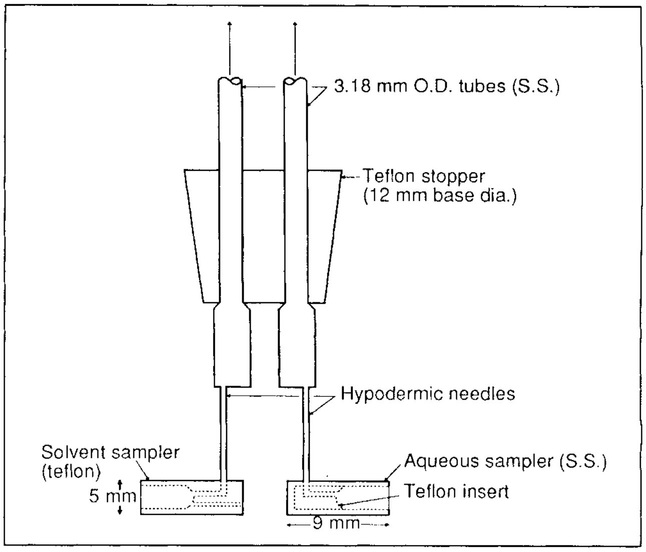

Figure 4. Sample probes.

Although shown in line for clarity, in use both faced the flow direction, inclined at 30° to 45° to the vertical.

1.5 cm³/min. Photographs of the droplets were taken during steady-state conditions and used to determine the drop size distribution and Sauter mean droplet size, d_{32}.

At the end of the experiment, the dispersed-phase holdup was determined using the standard drainage method (Gayler et al., 1953). As it was necessary to reuse the solvent, it was backwashed with water after the runs with transfer into solvent to reduce its solute content to 0.5 to 1.0%. The loaded aqueous phase was rejected after runs with transfer from solvent into water.

The samples were analyzed by titrating known masses against 0.1N NaOH, using phenolphthalein as indicator. Each sample was titrated three or more times, and the results, expressed as mass fractions, were averaged.

Backmixing experiments using tracer

For the steady-state method, tracer solution was injected continuously into the 24th stage by peristaltic pump. When steady state had been attained, samples were taken from the feed stage and from three upstream stages. These were then analyzed for tracer by measuring the absorption at 425 nm in a spectrophotometer.

Using the unsteady-state method, a pulse of tracer solution was injected into the continuous-phase inlet pipe through the sample valve. The responses at the 4th and 11th stages were measured on-line by removing samples of the continuous phase at rates of 1.0 to 1.5% of the total flow and passing them to flow cells within the spectrophotometers, the outputs of which were plotted on the chart recorder. Some of the runs were conducted simultaneously with the profile measurements, i.e., with mass transfer taking place.

A few qualitative tests were also carried out in which a pulse of tracer solution consisting of Waxoline Blue dissolved in MIBK was injected into the dispersed-phase distributor. They showed a marked spread of the band of color through the column, indicating that forward dispersion of the droplets was appreciable. There was, however, no indication of backmixing of droplets below the bottom stage, which would have resulted in an inlet concentration "jump."

Results

Concentration profiles

Six preliminary mass transfer runs were carried out: three each with mass transfer from the continuous phase into the dispersed phase and *vice versa*. These established that the most favorable rotor speed range was 110 to 130 rpm for transfer from the aqueous phase, and 160 to 200 rpm for transfer in the opposite direction. However, it was decided to concentrate mainly on the former conditions, as the results are influenced greatly by surface tension gradient (i.e., Marangoni) effects for transfer out of the dispersed phase, and in fact only one final experiment was run under these conditions.

A total of 16 experiments in all was then carried out, the results of which (excluding detailed concentration profiles) are summarized in Table 1. The results were interpreted in terms of various combinations of mass transfer and backmixing parameters using a computer program written to minimize an objective function Φ_1, defined as the sum of the squares of the differences between the experimental solute concentration profiles and those predicted for trial values of the parameters. This was given by:

$$\Phi_1 = \sum_{i=1}^{N_P} (\overline{X}_{\text{calc}} - \overline{X}_{\text{exp}})_i^2 W_{x,i} + (\overline{Y}_{\text{calc}} - \overline{Y}_{\text{exp}})_i^2 W_{y,i} \qquad (1)$$

where $\overline{X}$ and $\overline{Y}$ refer to dimensionless solute concentrations in feed and extractant phases, and W_x, W_y are nonnegative weighting factors.

The calculated values of $\overline{X}$ and $\overline{Y}$ in Eq. 1 were obtained for assumed values of the parameters, using the matrix formulation of Ricker et al. (1981). This can be written in the form:

$$[\underset{\sim}{M}](\underset{\sim}{\overline{X}}) + (\underset{\sim}{Z}) = 0 \qquad (2)$$

where $[\underset{\sim}{M}]$ is a block tridiagonal matrix with nonzero elements represented by 4×4 submatrices $[\underset{\sim}{A}_j]$, $[\underset{\sim}{B}_j]$ and $[\underset{\sim}{C}_j]$ which contain the coefficients of the material balance equations for

Table 1. Concentration Profile Measurements

Expt. No.	Phase Flow Rates: Water cm³/s	Phase Flow Rates: MIBK cm³/s	Rotor Speed rpm	Conc. of Acetic Acid in Mass %: Water-Phase Inlet	Water-Phase Outlet	MIBK-Phase Inlet	MIBK-Phase Outlet	Dispersed-Phase Holdup x_d, %	Extraction Factor E	d_{32}, cm	Direction of Mass Transfer
P-07	3.76	8.20	120	6.74	1.11	0.48	3.53	7.33	0.86	0.091	$c \to d$
P-08	9.58	4.27	160	0.68	3.01	7.42	0.39	4.96	0.21	0.119	$d \to c$
P-09	3.76	4.27	120	6.76	3.93	1.58	4.50	3.87	1.64	0.110	$c \to d$
P-10	3.76	6.16	120	6.63	2.58	1.20	4.06	6.72	1.13	0.111	$c \to d$
P-11	6.34	8.20	120	5.46	2.33	0.68	3.52	8.06	1.47	0.113	$c \to d$
P-12	4.78	10.34	120	5.95	1.59	0.97	3.23	9.56	0.88	—	$c \to d$
P-13	4.78	8.20	120	7.21	2.30	1.08	4.35	7.77	1.07	—	$c \to d$
P-14	4.78	9.26	120	6.60	2.10	1.10	3.70	7.80	0.97	0.105	$c \to d$
P-15	3.76	6.16	110	6.61	1.85	0.68	3.80	4.53	1.17	—	$c \to d$
P-16	3.76	6.16	130	6.61	1.70	0.57	3.93	6.66	1.17	—	$c \to d$
P-17	3.76	8.20	120	6.40	1.44	0.71	3.14	7.20	0.86	0.084	$c \to d$
P-18	4.78	10.34	120	5.98	1.85	1.02	3.18	9.91	0.88	0.088	$c \to d$
P-19	4.78	8.20	120	6.19	2.40	1.16	3.73	7.21	1.07	0.092	$c \to d$
P-20	4.78	9.26	120	6.60	2.18	1.14	3.78	8.43	0.97	0.084	$c \to d$
P-21	3.76	9.26	120	6.06	1.24	0.64	2.75	8.43	0.78	0.087	$c \to d$
P-22	6.34	9.26	120	5.66	2.21	0.87	3.52	9.94	1.30	0.088	$c \to d$

each phase with respect to the variables for stages $j-1$, j and $j+1$, together with the mass transfer rates and the equilibrium relationship for stage j.

The vector $(\overline{\underline{X}})$ is composed of subvectors $(\overline{\underline{X}}_1)$ to $(\overline{\underline{X}}_N)$ representing the dependent variables, e.g.,

$$(\overline{\underline{X}}_j) = (\overline{X}_j,\ \overline{Y}_j,\ \overline{X}^i_j,\ \overline{Y}^i_j)^T \tag{3}$$

where superscript i refers to the interfacial values. The vector $(\underline{Z})$ contains the corresponding subvectors $(\underline{Z}_j)$ for $j=1, \ldots, N$; these are (0) in the present case except for the subvectors $(\underline{Z}_m)$ and $(\underline{Z}_n)$ representing the feed and extractant inlet stages, as follows:

$$(\underline{Z}_m) = (0,\ \overline{Y}^0,\ 0,\ 0)^T \quad \text{and} \quad (Z_n) = (1,\ 0,\ 0,\ 0)^T \tag{4}$$

In formulating the equations, it is assumed that fictitious end stages 0 and $N+1$ are provided, in which no mass transfer occurs, and also that no mass transfer occurs in the feed stages 1 and N, which are of negligible volume.

The solution to Eq. 2 is obtained by inverting $[\underline{M}]$ and postmultiplying the result by $(\underline{Z})$, assuming a constant distribution coefficient,

$$(\overline{\underline{X}}) = -[\underline{M}]^{-1}(\underline{Z}) \tag{5}$$

However, an iterative procedure must be used if the distribution coefficients are functions of concentration. In such cases, a constant distribution coefficient is assumed initially, and more accurate values are obtained from the predicted solute concentrations in subsequent iterations. For very complex equilibrium relationships, a Newton-Raphson iterative technique can be used (Ricker et al., 1981).

The following procedure was used in the computer program to solve Eq. 1 in terms of the present data:

Step 1. Assume initial values of the backmixing and mass transfer parameters.

Step 2. Assume values for the distribution coefficients on each stage.

Step 3. Calculate the bulk and interfacial concentrations using Eq. 5.

Step 4. Calculate new values of the distribution coefficients for each stage from the resulting solute concentrations.

Step 5. Return to step 3 and iterate until successive concentration profiles converge.

Step 6. Calculate the magnitude of objective function Φ from Eq. 1.

Step 7. Generate a new set of parameters via the optimization routine used.

Step 8. Repeat steps 2 to 7 until the objective function is minimized.

Both the Levenburg-Marquardt derivative-free method (Levenberg, 1944; Marquardt, 1933; Dennis and Schnabel, 1983) and the complex Box method (1966) were used to minimize the objective function. Initially, both methods were used as the mass transfer, and backmixing parameters were found to be highly interactive; however, the Levenberg-Marquardt method converged successfully in most cases. Subprograms for matrix inversion using the LU factorization technique (Cline et al., 1979; Dongarra et al., 1979) and for the foregoing optimization techniques were obtained from the IMSL (1987) library of mathematical routines.

Equilibrium data for the present system, reported by Misek (1978), are practically temperature-independent in the range $283\text{ K} < T < 303\text{ K}$. The data, expressed in mass fraction rather than mass-volume concentration units as they are more nearly linear in this form, were represented as follows:

$$X^* = (1.7428 - 0.0701\,Y + 0.0027\,Y^2)\,Y \tag{6}$$

In analyzing the data, it was assumed that the column had 27 rather than 25 stages, the two extra stages being included to allow for the contact that occurred at the phase inlets above and below the end stages. The conditions in these stages were assumed to be the same as in the other stages. The flows at each sample point were corrected for the volumes of samples withdrawn as the run progressed. Weighting factors of $W_x = W_y = 3.0$ were assumed for each stage except for stage numbers 1, 2, 26 and 27, where values of 6.0 were assumed, in view of the importance of the inlet "jumps" (Rod et al., 1983).

The results of the three-parameter estimation, in terms of N^1_{od}, α_d and α_c are summarized in Table 2. This indicates that, with three exceptions, the values of α_d were very low and it was concluded that backmixing of the dispersed phase was negligible, unlike that of the continuous phase. The results are therefore also given for a two-parameter estimation, with α_d set equal to zero. Figure 5 shows a typical plot of the profiles with a marked "jump" at the continuous-phase inlet due to backmixing and the absence of a jump at the dispersed-phase inlet.

Unsteady-state tracer injection

The results of a typical experiment are shown in Figure 6. The two output curves, giving the residence time distributions, were digitized to obtain concentration vs. time data, and the concentrations were then input to a computer program that normalized them in terms of the areas under the curves:

$$C_{n,o}(t) = C_n(t)\,V\left[F\int_0^\infty C_n(t)\,dt\right]^{-1} \quad \text{for } n = 1, 2 \tag{7}$$

The program then performed the numerical integration and convolution to obtain the backmixing coefficient, α_c and the delay time Δt, i.e., the difference between the sample transit times from column to spectrophotometer, by minimizing the following objective function:

$$\Phi_2 = \int_0^\infty \left[C_2(\theta) - \int_0^\theta f(\tau,\alpha_C)\,C_1(\theta-\tau)\,d\tau\right]^2 W(\theta)\,d\theta \tag{8}$$

where C_1, C_2 are the responses at the first and second sample points, $f(\tau,\alpha_c)$ is the response function in terms of the backmixing parameter, and $W(\theta)$ is a nonnegative weighting factor.

The function $f(\tau,\alpha_c)$, giving the transient response at any stage n with respect to that at stage m for an arbitrary tracer input at the inlet, has been obtained by Dongaonkar (1990):

Table 2. Overall Mass Transfer and Backmixing Parameters

Expt. No.*	Three-Parameter Estimation N^1_{od}	α_d	α_c	Two-Parameter Estimation $\alpha_d = 0$ N^1_{od}	α_c	α_c by Transient Tracer Injection With Solute Transfer	Without Solute Transfer
P-07	1.33	0.01	1.13	1.35	1.07	—	—
P-08**	1.08	0.00	1.21	1.09	1.29	—	—
P/Tt-09	0.84	0.07	0.88	0.87	1.01	—	1.50
P/Tt-10	1.00	0.01	1.11	1.01	1.08	—	1.37
P/Tt-11	0.66	0.00	0.49	0.65	0.50	—	1.60
P-12	1.01	0.01	0.23	1.08	0.20	—	—
P-13	1.33	0.00	0.33	1.37	0.29	—	—
P/Tt-14	1.32	0.58	0.08	1.01	0.48	1.35	1.15
P/Tt-15	0.76	0.00	1.50	0.89	1.39	2.20	1.37
P/Tt-16	1.13	0.01	1.15	1.05	1.09	2.10	1.63
P/Tt-17	1.36	0.00	1.01	1.34	1.01	1.35	1.45
P/Tt-18	0.76	0.07	0.41	0.76	0.53	1.77	1.20
P/Tt-19	0.96	0.21	0.52	0.89	0.55	1.90	0.90
P/Tt-20	0.93	0.41	0.04	0.78	0.32	1.87	1.15
P/Tt-21	1.68	0.92	0.14	1.14	0.98	2.35	1.37
P/Tt-22	0.69	0.01	0.53	0.71	0.56	1.55	0.97

**P* refers to concentration profile, and *Tt* to transient tracer injection runs with and without mass transfer.

**Transfer $d \rightarrow c$ (remainder $c \rightarrow d$).

$$C_n(\theta) = \int \sum_{i=1}^{N-m} A_i \exp(s_i\theta) C_m(\theta - \tau) d\tau \tag{9}$$

where

$$a = [(1+\alpha_c)/\alpha_c]^{1/2} \tag{9a}$$

$$s_i = N\{[\alpha_c(1+\alpha_c)]^{1/2} \cos(\psi_i) - (1+2\alpha_c)\} \tag{9b}$$

$$A_i = \frac{(-2Na^{n-m+1}) \sin(\psi_i)[\sin((N-n)\psi_i) - a\sin((N-1-n)\psi_i)]}{d[\sin((N-m)\psi) - a\sin((N+1-m)\psi)]_{\psi=\psi_i}/d\psi}. \tag{9c}$$

and the N values of ψ are the zeros of the denominator DEN in the interval of $0 < \psi < \pi$ as follows:

$$\mathrm{DEN} = a\sin[(N+1)\psi] - 2\sin(N\psi) + \frac{1}{a}\sin[(N-1)\psi] \tag{9d}$$

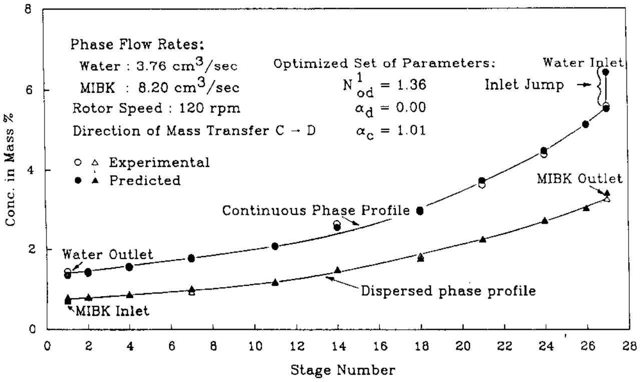

Figure 5. Experimental concentration profiles for run *P*-17.

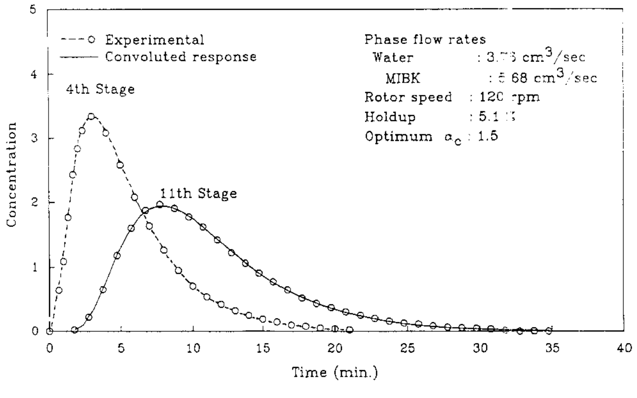

Figure 6. Experimental and convoluted tracer responses for run *Tt*-24.

The solution procedure for Eq. 7 is as follows:

1. Assume a value for the delay time, Δt.
2. Assume an initial value for α_c.
3. Evaluate Φ_2, given by Eq. 8.
4. Increment α_c.
5. Repeat steps 3 and 4 until Φ_2 is minimized.
6. Assume another value for Δt.
7. Repeat steps 2 to 6 until the lowest value of Φ_2 is obtained.

It should be noted that α_c and Δt are independent parameters and hence did not require simultaneous estimation. A typical example comparing the experimental with the convoluted response is shown in Figure 6.

Values of α_c obtained in the presence of mass transfer are included in Table 2, together with runs conducted under identical conditions in the absence of mass transfer. Further data obtained without mass transfer are given in Table 3, together

Table 3. Comparison of Continuous-Phase Backmixing Coefficients by Transient and Steady-State Tracer Injection without Mass Transfer

Expt. No.*	Phase Flow Rates Water cm³/s	MIBK cm³/s	Rotor Speed rpm	Holdup x_d, %	α_c Transient Tracer	Steady-State Tracer	% Error Steady-State Values
T-23	3.76	4.27	120	3.4	1.50	1.53	−2.0
T-24	3.76	5.68	120	5.1	1.55	1.51	+2.7
T-25	3.76	7.68	120	7.6	1.27	1.47	−13.6
T-26	3.76	9.26	120	9.7	1.27	1.39	−8.6
T-27	5.29	4.27	120	4.1	1.00	0.97	+3.1
T-28	7.41	4.27	120	4.2	0.90	0.69	+30.4
T-29	9.07	4.27	120	4.0	1.00	0.53	+88.7
T-30	3.76	4.27	140	5.9	1.50	1.67	−10.2
T-31	3.76	4.27	160	8.3	1.67	1.91	−12.6
T-32	3.76	4.27	180	10.8	1.80	2.22	−18.9
T-33	3.76	4.27	200	12.8	2.00	2.69	−25.7

**T* refers to *Tt* (transient tracer) and *Tc* (continuous tracer injection) runs.

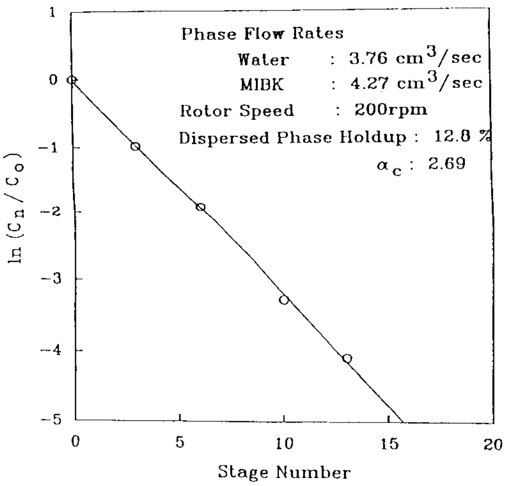

Figure 7. Steady-state tracer response for run *Tc*-33.

with corresponding values obtained by the steady-state tracer method.

Table 4. Comparison of Experimental and Predicted Overall Mass Transfer Coefficients

Expt. No.	k_{od}, 10^4 m/s				
		Predicted Values*			
	Exp.	$k_{od(1)}$**	% Error	$k_{od(2)}$**	% Error
P-07	1.19	0.87	−26.8	1.04	−12.6
P-08	0.97	0.77	−20.6	1.09	+12.4
P-09	0.92	0.90	−2.2	1.15	+25.0
P-10	0.89	0.76	−14.6	1.06	+19.1
P-11	0.65	0.87	+25.3	1.16	+78.5
P-14	1.11	0.96	−13.5	1.18	+6.3
P-17	1.13	0.88	−22.1	1.00	−11.5
P-18	0.59	0.83	+40.7	0.98	+66.1
P-19	0.81	0.90	+11.1	1.06	−30.9
P-20	0.63	0.85	+34.9	0.98	+55.6
P-21	0.95	0.86	−9.5	1.00	+5.1
P-22	0.51	0.76	+49.0	0.93	+82.4

*Based on Higbie model for continuous phase, and extended Handlos and Baron model for dispersed phase.

**$k_{od(1)}$, based on slip velocity of droplet; $k_{od(2)}$, based on terminal velocity of droplet.

Steady-state tracer injection

The tracer concentration C_n at any upstream stage n is related to that at the injection stage, C_o (Pratt and Baird, 1983):

$$C_n = C_o\left(\frac{\alpha_c}{1+\alpha_c}\right)^n \tag{10}$$

A plot of $\log(C_n/C_o)$ against n should therefore give a straight line of slope $\alpha_c/(1+\alpha_c)$ passing through the origin, from the slope of which α_c can be obtained. A typical experimental result, plotted in this way in Figure 7, shows that this in fact is the case, indicating the applicability of the backflow model with each compartment represented as a stage.

The experimental results are summarized in Table 3 for various combinations of flow rates and rotor speeds. Those obtained for the same conditions using the transient tracer method in the absence of mass transfer are also included for comparison.

Interpretation of Results

Mass transfer parameter

Values of the overall mass transfer coefficient k_{od} were calculated from those of N^1_{od} obtained by two-parameter estimation (Table 2). For this purpose, the interfacial area per unit volume, a, was expressed as $6x_d/d_{32}$, giving:

$$k_{od} = N^1_{od} d_{32} F_d N/(6x_d V) \tag{11}$$

The experimental values of d_{32} obtained from the droplet photographs, listed in Table 1, were used in Eq. 11.

Values of k_{od} were predicted from those of the individual-phase coefficients, k_c and k_d, for comparison with the present values. As no reliable method is available to predict the drop rise velocities in a droplet swarm produced by mechanical agitation, it was assumed that the effective velocity lies between the terminal velocity of freely moving droplets, as given by the correlation of Klee and Treybal (1956) and the mean slip velocity obtained from the present holdup data as follows:

$$\bar{v}_s = U_c/(1-x_d) + U_d/x_d \tag{12}$$

Values of the continuous-phase coefficient were calculated in terms of both velocities from the models of Higbie (1935), Garner et al. (1954) and Griffith (1960), and those for the dispersed phase from the models of Newman (1931), Rozen and Bezzubova (1968) and the rigorous solution of the Handlos and Baron model (1957) described by Korchinsky and Cruz-Pinto (1979); see also Cruz-Pinto and Korchinsky (1983). It was clear from the results that the only combination of models that gave values of k_{od} approximating those obtained in the present work were the Higbie with the extended Handlos and Baron models. A comparison of experimental with predicted values based on both slip and terminal droplet velocities is given in Table 4. This shows that the use of the slip velocity gives lower predicted coefficients than the terminal velocity, with a mean overall error of 4.6% as compared with 23.0%. It is to be noted that the forward dispersion of droplets, reported earlier, would have the effect of reducing the predicted values of k_{od} somewhat.

Continuous-phase backmixing

It is clear from Table 3 that the values of α_c in the absence of mass transfer obtained by the two tracer techniques are in reasonable agreement, with an average error relative to the steady-state method of 19.7% (12.8% excluding run 29). However, the results obtained by the transient (i.e., residence time distribution) method, given in Table 2, show a significant increase, averaging 51%, in α_c in the presence of mass transfer. Such an increase is predicted by a model proposed by Korchinsky (1988), which assumes that an additional contribution to dispersion is provided by the entrained wake behind the droplets; evidence for this was provided by data for a rotary disc column. However, the values of α_c obtained from the present profile data simultaneously with the mass transfer pa-

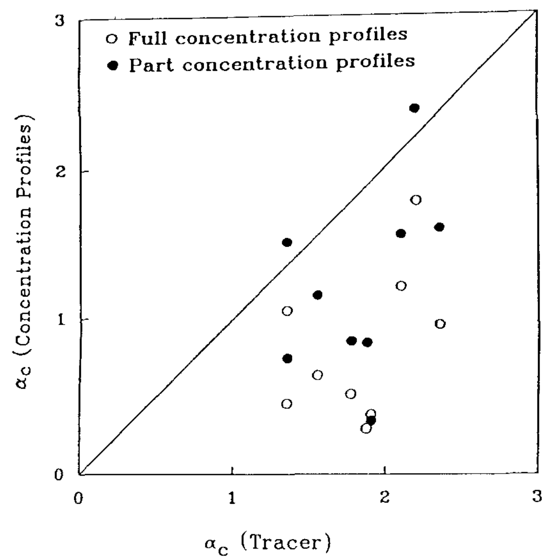

Figure 8. Continuous-phase backmixing coefficients, α_c: concentration profiles vs. transient tracer response in the presence of mass transfer.

rameters are considerably lower than those obtained by tracer injection, with or without mass transfer, as shown in Table 2.

The discrepancy between the profile and tracer experiments may be explainable from the fact that α_c is estimated from the profile over the whole length of the column in the former case, but only from the flow behavior in the upper part of the column toward the continuous-phase inlet in the latter. Thus, in the former case the lower section of the column, in which the droplet size distribution is changing rapidly, and the settling zone above the top plate, where the continuous phase is introduced, are likely to differ in performance from the central region where the tracer method is applied. To overcome this, the two-parameter estimation of the profile data was repeated, giving zero weighting to four sample points, two at each end, thus reducing their significance.

The results of this study showed an average increase of 54% in the values of α_c obtained from the concentration profiles. These are, therefore, closer to the values obtained by tracer injection in the presence of mass transfer, although, as shown in Figure 8, they are still lower than the latter. The effect of solute transfer in increasing continuous-phase backmixing does require further study; however, Steiner et al. (1988) in a somewhat similar investigation reported little or no effect.

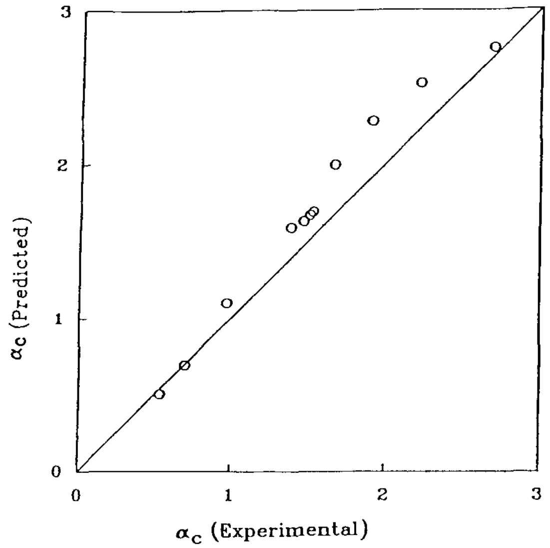

Figure 9. Steady-state tracer values: α_c vs. Eq. 15.

Correlation of continuous-phase backmixing coefficient

Single-phase axial dispersion data, using water, have been reported by Breysse et al. (1984), Ingham et al. (1974), and Vatai and Tolic (1983) for a wide range of column geometries and operating conditions. The results were all correlated in terms of the diffusion model using expressions of the form

$$1/Pe_c = A_1 + A_2 \frac{nd_i}{U_c} f(G) \qquad (13)$$

where $f(G)$ is an unknown function of compartment geometry. To quantify this, 250 values of α_c were computed from the various correlations over their full ranges and used to obtain a unified relationship by dimensional analysis. This led to the following expression, after determining that the exponent on the group (D_c/H_c) was zero:

$$1/Pe_c = (\alpha_c + 0.5) = 0.154 + 0.063 \left(\frac{nd_i}{U_c}\right) \left(\frac{h_i}{d_i}\right)^{0.067} \left(\frac{d_i}{D_c}\right) S_\phi^{0.587} \qquad (14)$$

Equation 14 indicates that α_c increases with n, i.e., the agitation rate, and decreases with continuous-phase velocity, U_c. The present results indicate that α_c tends to decrease with increasing flow of dispersed phase, apparently due to the effect of increased holdup of droplets in increasing the continuous-phase velocity. Applying this concept to Eq. 14 results in the following expression:

$$\alpha_c = -0.346 + 0.063 \left[\frac{nd_i(1 - x_d)}{U_c}\right] \left(\frac{h_i}{d_i}\right)^{0.067} \left(\frac{d_i}{D_c}\right) S_\phi^{0.587} \qquad (15)$$

Figure 9 shows that the present steady-state tracer results obtained in the absence of mass transfer are correlated by Eq. 15 with a mean error of + 9.7%. However, the predicted values of α_c show an increasing deviation, up to 20% above the measured values, with increase in α_c, suggesting that the influence of the dispersed phase may not be fully taken into account.

Conclusions

- Average values over the whole column of the overall mass transfer parameter and the continuous- and dispersed-phase backmixing coefficients have been derived from measured concentration profiles for a Kühni extraction column. A high degree of precision was required in measuring the profiles and interpreting the data, as the mass transfer and backmixing parameters were highly interactive.
- The results indicated that dispersed-phase backmixing was negligible, although appreciable forward dispersion was detected in separate experiments.

- Derived values of the overall mass transfer coefficient, k_{od}, were in moderate agreement with those predicted from a combination of the extended Handlos-Baron model for k_d, and the Higbie model for k_c, assuming droplets of the measured mean size d_{32} moving with the measured slip velocity.
- Continuous-phase backmixing coefficients determined by both the continuous and transient tracer injection methods in the absence of mass transfer were in reasonable agreement. However, values obtained using the transient method in the presence of solute transfer were some 50% higher, in qualitative agreement with Korchinsky's recent model (1988).
- Continuous-phase backmixing coefficients, α_c, obtained from concentration profiles, were 59% lower on average than those obtained by tracer injection with accompanying mass transfer. The agreement was somewhat better, to within 36%, when they were obtained from the central part of the column. Tracer methods, preferably in the presence of solute transfer, are therefore to be preferred for the determination of continuous-phase backmixing coefficients.
- A correlation of published single-phase α_c values, obtained by dimensional analysis, has been extended to allow for the effect of the dispersed phase in two-phase solute-free operation. The correlation predicted the experimental α_c values to within ±20%, but in practice allowance should be made for the effect of solute transfer.

Notation

a = interfacial area per unit volume, m^{-1}
a_i = parameter defined by Eq. 9a
A_i = parameters defined by Eqs. 9 and 9c
$[\underset{\sim}{A}_j]$, $[\underset{\sim}{B}_j]$, $[\underset{\sim}{C}_j]$ = submatrices of $[\underset{\sim}{M}]$
$\underline{C}_n$ = solute concentration in stage j, $kg \cdot m^{-3}$
$\overline{C}_n$ = Laplace transform of C_n
C_x, C_y = solute concentrations in phases X and Y, $kg \cdot m^{-3}$
$C_1(\theta)$, $C_2(\theta)$ = tracer concentration responses at first and second sample points as a function of time, $kg \cdot m^{-3}$
d_{32} = Sauter mean droplet diameter, m
D_i = column diameter, m
d_i = impeller diameter, m
$\underset{\sim}{E}$ = extraction factor, mU_c/U_d
E_c = axial dispersion coefficient for continuous phase, $m^2 \cdot s^{-1}$
F = net forward flow rate, $m^3 \cdot s^{-1}$
F_d = volumetric flow rate of dispersed phase, $m^3 \cdot s^{-1}$
h_i = impeller height, m
H_c = compartment height, m
k_{od} = overall mass transfer coefficient based on dispersed phase, $m \cdot s^{-1}$
i = stage number counted from feed phase inlet
$[\underset{\sim}{M}]$ = block tridiagonal matrix containing submatrices $[\underset{\sim}{A}_j]$, $[\underset{\sim}{B}_j]$ and $[\underset{\sim}{C}_j]$
n = rotor speed, s^{-1}
N = number of stages in column
N_{od}^1 = number of overall transfer units for single stage based on dispersed phase
N_p = number of sample points in concentration profiles
Pe_c = Peclet number for continuous phase, U_cH_c/E_c
s_i = parameter defined by Eq. 9b
S_f = fractional free area of stator plates
t = time, s
Δt = difference in sample transit times between column and spectrophotometers, s
U_c, U_d = superficial velocities of continuous and dispersed phases, $m \cdot s^{-1}$
$\bar{v}_s$ = droplet slip velocity, $m \cdot s^{-1}$
V = overall volume of extractor stages, m^3
W_x, W_y = nonnegative weighting factors for phases X and Y
$W(\theta)$ = nonnegative weighting factor in Eq. 8
X_j = $C_{x,j}/C_x^o$, dimensionless concentration of solute in feed phase
$(\underset{\sim}{X})$ = vector of subvectors given by Eq. 3
Y_j = $C_{y,j}/C_x^o$, dimensionless concentration of solute in extract phase
$(\underset{\sim}{Z})$ = vector of constant terms in Eq. 2

Greek letters

α_j = backmix ratio for phase j, ratio of backflow to net forward flow
Φ_1, Φ_2 = objective functions in Eqs. 1 and 8
τ = dummy variable
θ = dimensionless time, Ft/V
ψ = zeros of Eq. 9d

Superscripts

o = feed-phase inlet
T = transpose
$*$ = value at equilibrium

Subscripts

c = continuous phase
d = dispersed phase
j = stage number counted from feed-phase inlet
X = feed phase (continuous in present case)
Y = extractant phase (dispersed in present case)
calc = calculated
exp = experimental

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Manuscript received Oct. 23, 1990, and revision received Feb. 25, 1991.