Axial Dispersion in Pulsed-, Perforated-Plate Extraction Columns

Continuous-phase backmixing coefficients have been determined by continuous tracer injection for pulsed-plate extraction columns of 72-, 152-and 300-mm-diameter. Both single-phase water and two-phase kerosene-water were studied in all three columns, together with four other solvent-water systems in the smallest column; in all cases, water was the continuous phase. Evidence of maldistribution of the phases was obtained in the largest column. The backmixing coefficient was found to increase with column diameter in single-phase operation, but was diameter-independent in the two-phase case. Alternative correlations of the data were based on dimensional analysis and on a physical model, respectively. The influence of different systems was accounted for in the latter in terms of droplet size and terminal velocity, dispersed phase holdup, and density difference. Qualitative color tracer tests on the dispersed phase gave no evidence of backmixing of this phase, although strong forward dispersion was observed in the emulsion regime.

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Introduction

In the design of liquid extraction columns, particular difficulty is encountered in the prediction of the height required to achieve a specified degree of separation of the feed components. This is a direct result of the complex hydrodynamics of two-phase droplet systems, in which axial dispersion leads to departures from ideal countercurrent plug flow. Such axial dispersion takes the form of backmixing, particularly of the continuous phase, and of forward dispersion of droplets due to the differing velocities and mass transfer rates of the various drop sizes. Of these, continuous-phase backmixing is of major importance as it reduces the driving force for mass transfer.

Extraction columns are designed generally by scale-up from pilot-plant data obtained for the actual system to be used; a convenient analytical method for this purpose has been described by Pratt (1983). However, data on the effect of column diameter on the backmixing parameters are seldom available. There is, therefore, an urgent need for such data, especially for currently-used columns of mechanically-agitated type.

In 1935, Van Dijck proposed that the efficiency of perforated-plate columns could be improved by pulsing the fluids or by applying a reciprocating motion to the plates. This led to the development of pulsed-and reciprocating-plate liquid extraction columns. Pulsed columns were initially developed for use in the nuclear industry, where their relatively high efficiency and absence of internal moving parts were seen to be advantageous. Since then, these columns have found applications in a variety of other industries.

Many studies have been undertaken with pulsed-plate columns to determine their hydrodynamic behavior, mass transfer performance, and axial dispersion characteristics. It is generally agreed that dispersed-phase backmixing is small and that forward mixing, due to the range of drop sizes present, has a relatively small influence on the mass transfer rate (Garg and Pratt, 1984; Reissinger et al., 1982). The many correlations presented for predicting continuous-phase axial dispersion parameters are of limited use, as they apply only to the particular systems and column geometries for which they were obtained. As a consequence, the effects of the various design parameters on continuous-phase backmixing is still uncertain.

Various workers have found that the performance of pulsed columns decreases with increasing diameter (e.g., Woodfield and Sege, 1954; Logsdail and Thornton, 1957; Clappier et al., 1983). Woodfield and Sege (1954) considered that this was due to maldistribution and channelling of the phases, while Garg and Pratt (1981) suggested that it might be due to increased backmixing. To date, however, no studies have been reported

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which compare the effect on continuous-phase backmixing of either column diameter or system physical properties.

In the present work, continuous-phase backmixing coefficients have been obtained for pulsed-plate columns of 72-, 152- and 300-mm-diameter, both for single-phase operation with water and for two-phase operation with kerosene-water to determine the effect of column diameter. Data for four other solvent-water systems were also obtained in the smallest column to ascertain the effect of system physical properties. A single, commonly-used plate geometry was used in all three columns, and backmixing was determined by the continuous tracer injection method.

Literature Survey

Continuous-phase axial dispersion

In most studies of continuous-phase axial dispersion, the data have been presented generally in terms of the diffusion model, although the stagewise model is more appropriate for such extractors, particularly when the plate-free area is small. Kagan et al. (1965) employed both impulse and steady-state tracer injection techniques in a 56-mm-diameter column, obtaining values of the dispersion coefficient, which in both cases were higher for single-phase than for two-phase operation. Considerable differences between the results obtained from the two experimental methods were observed, but no attempt was made to explain them. The steady-state data for the kerosene-water system were correlated in terms of a continuous-phase axial dispersion coefficient, E_c, thus

$$E_c = [(1.2 \times 10^5 A^{1.2} f^{1.35})/(U_d + U_c)^{1.4}] \text{ cm}^2/\text{s} \qquad (1)$$

Miyauchi and Oya (1965) correlated their single-phase and two-phase data for a 50-mm-dia. column in terms of a parameter β_m, the number of "perfectly-mixed stages" in series per compartment. This was expected to have values between 1 and 2; however, for large d_c, Logsdail and Slater (1983) showed that β_m assumes unrealistic values. As a single correlation was found to describe both the single-phase and two-phase data of these workers, Ingham (1971) concluded that there was no significant difference between the flow mechanisms for the two cases.

Baird (1974) developed an acid-base indicator technique applicable to both steady-state and transient tracer injection which gave consistent results for single-phase operation of a 150-mm-dia. reciprocating-plate column. An approximate square law dependence of E_c on pulse amplitude was observed, with less sensitivity to frequency. For two-phase operation, the transient method was unsuccessful as the color change of the indicator occurred preferentially in a narrow stream up one side of the column.

Kim and Baird (1976) investigated the influence of plate material for single-phase operation of a 50-mm-dia. reciprocating-plate column and presented alternative correlations of E_c for SS (stainless steel) and Teflon plates. A comparison of their SS plate data with the foregoing data of Baird (1974) showed good agreement, suggesting that column diameter did not influence single-phase axial dispersion. For two-phase emulsion operation using the kerosene-water system, a small effect of U_d on E_c was observed, the direction of which depended upon the level of agitation.

Hafez et al. (1979) found, however, that E_c was larger for single-phase operation at low agitation levels in a 150-mm-diameter reciprocating-plate column with low free area, and was influenced by U_c. In two-phase operation, E_c exhibited maxima at an agitation rate of $Af = 0.25$ cm/s, whose size increased with decreasing U_d. Circulation effects due to hydraulic nonuniformity of the dispersed phase were also described and were most severe at low agitation rates and low values of U_d. It was, therefore, suggested that E_c would tend to increase with increasing column diameter due to such circulation effects.

The influence of scale-up on continuous-phase axial dispersion is still uncertain. Rouyer et al. (1974) observed no increase in E_c for pulsed-plate columns in two-phase operation on scaling up from 45- to 600-mm-diameter; on the other hand, Aufderheide and Vogelpohl (1986b) noted a large influence of diameter when scaling up the toluene-water system, but only slight increases for butyl-acetate-water, although similar increases were observed at low volumetric flow rates.

The effect of mass transfer was studied by Bensalem et al. (1983) and Bensalem (1985), who obtained different correlations for E_c with the toluene-acetone-water system according to the direction or absence of mass transfer. A significant increase in E_c was observed with mass transfer from dispersed to continuous phase which was attributed to the larger drop size.

Despite the extensive work reported, correlations for predicting E_c tend to be limited to the particular system and column, from which they were obtained. Mar and Babb (1959) attempted to provide a single correlation to predict E_c for different organic-water systems with water continuous. In a later study, however, Sehmel and Babb (1963) observed that E_c passed through a maximum as the column behavior changed from mixer-settler to emulsion operation. They, therefore, recorrelated their results accordingly, introducing a transition frequency to distinguish between mixer-settler and emulsion behavior; a correlation was also presented to predict the transition frequency.

Finally, Tung and Luecke (1986) recorrelated published data to obtain a generalized expression for the prediction of E_c for two-phase emulsion operation. This gave

$$E_c = 0.250 \left(\frac{h_c U_c}{\epsilon^{1.30}}\right) \left(\frac{d_h}{h_c}\right)^{0.565} \left(\frac{Af}{U_c}\right)^{0.606} \qquad (2)$$

for water continuous; column diameter was found not to be a significant variable.

Physical model

As an alternative to empirical correlations, Novotny et al. (1970) and Nemecek and Prochazka (1974) proposed a physical model to predict continuous-phase axial dispersion in reciprocating- and pulsed-plate columns. This model, unlike those of earlier workers, was based on the backflow model and assumed that axial mixing resulted from two factors, *viz.*

- Backflow of liquid through the plate holes when it moves in the opposite direction to its net flow
- Axial mixing between two neighboring plates

As a consequence, the column was assumed to consist of well-mixed regions in the vicinity of and on each side of the plates in which the backflow model applies, connected by a diffusion region between these in which the axial dispersion model applies (see Figure 1). If a nontransferring tracer is injected continuously into the continuous phase at some downstream stage, the tracer concentrations upstream were shown to be related as fol-

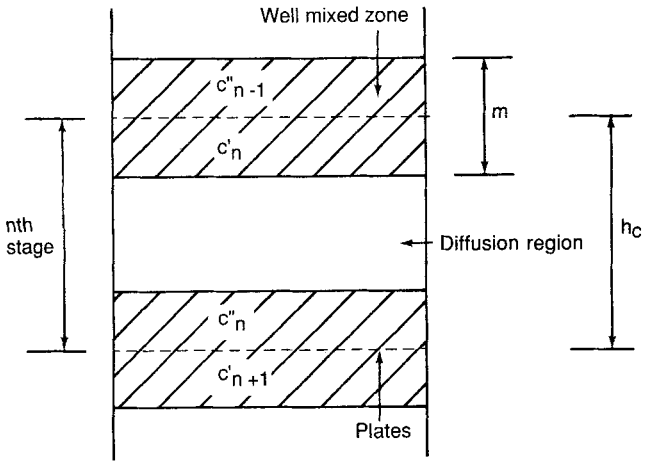

Figure 1. Continuous-phase backmixing model (Novotny et al., 1970).

lows for the fully-mixed region adjoining upstream stages n and $n-1$

$$\frac{c''_{n-1}}{c'_n} = \frac{q_c}{1 + q_c} \tag{3}$$

where $q_c = U_b/U_c$, the ratio of backflow to net forward flow. For the diffusion region in stage n, solution of the appropriate differential equation gave

$$\frac{c'_n}{c''_n} = \{\exp [\, U_c(h_c - m)/E_c]\}^{-1} = [\exp Pe_c]^{-1} \tag{4}$$

The backflow ratio between stages n and $n-1$ is obtained as follows in terms of the backflow model (see Eq. 14 with $n = 1$)

$$\frac{c''_{n-1}}{c''_n} = \frac{\alpha_c}{1 + \alpha_c} \tag{5}$$

Hence combining Eqs. 3–5,

$$\alpha_c = [(1 + 1/q_c) \exp (Pe_c) - 1]^{-1} \tag{6}$$

Integration of the simple harmonic motion over a full pulse cycle gave finally

$$q_c = [(R - 1)/(R\pi)][\phi + \cot \phi - (\pi/2)] - [(1 + q_d)/R] \tag{7}$$

where $R = U_c/U_d$ and

$$\phi = \sin^{-1}[U_d(R - 1)/\pi Af] \tag{8}$$

Equation 7 becomes indefinite when $R = 1$, but in the limit as $R \to 1$, q_c is given by

$$q_c = [(Af)/U_c] - (1 + q_d) \tag{9}$$

The solution of Eqs. 7 and 9 require values of q_d, the dispersed-phase backmixing across the plates. For the mixer-settler region, $q_d = 0$ and for dispersion (i.e., emulsion) operation Eqs. 8 and 9 become

$$q_c = (1/\pi)[\phi + (1 - x_d)(R - 1)/R] \cot \phi - (\pi/2)], R \neq 1 \tag{10}$$

$$q_c = [Af(1 - x_d)/U_c] - 0.5, R = 1 \tag{11}$$

For the single-phase operation, Eqs. 7 and 8 become

$$q_c = [\phi + \cot \phi - (\pi/2)]/\pi \tag{12}$$

$$\phi = \sin^{-1} [U_c/\pi Af] \tag{13}$$

Expressions for m and E_c in Eq. 4 were obtained semiempirically in the original papers and will be discussed later. The model was applied to both single-phase and two-phase operation of a reciprocating-plate column, with average errors between predicted and measured backmixing coefficients of ±12.0 and 12.5%, respectively.

Dispersed-phase axial dispersion

Transient tracer injection methods have been used by a number of workers to study dispersed-phase axial dispersion in pulsed- and reciprocating-plate columns (e.g., Miyauchi and Oya, 1965; Bensalem et al., 1983; Niebuhr and Vogelpohl, 1980). In all such cases, the results were interpreted in terms of backmixing coefficients; however, the existence of a droplet-size distribution renders the resulting dispersion coefficient physically meaningless (Levenspiel and Fitzgerald, 1983), although the measured residence time distribution curves can be fitted to such a model. True backmixing of the dispersed phase is, in fact, unlikely in pulsed- and reciprocating-plate columns, and the spread of residence times is a result of forward dispersion due to differences in velocity of the various droplet sizes (Rod, 1966; Aufderheide and Vogelpohl, 1986a).

Scope of Present Study

The aims of the present study are as follows:

- To measure continuous-phase backmixing coefficients for columns of three different diameters in order to quantify the effect of diameter on backmixing.
- To investigate backmixing for five organic (d)-water (c) systems, to determine the effect of varying physical properties on the backmixing coefficients.
- To determine whether single-phase axial dispersion measurements are relevant to two-phase operation of these columns.
- To make a preliminary study of the relevance of dispersed-phase backmixing in these columns.

The steady-state tracer injection method was selected as the measurement technique. This has the advantage of being simpler and easier to interpret than the transient method, and also permits a direct comparison between the present results and those of Novotny et al. (1970) and Nemecek and Prochazka (1974) in reciprocating-plate columns.

Equipment

Columns and auxillaries

The columns investigated, numbered 1, 2 and 3, had diameters of 72.45, 151.2 and 299.6 mm, respectively; the basic geo-

metric data are summarized in Table 1. A schematic diagram of column 1 is presented in Figure 2. Full details of the equipment have been given by Prvcic (1987).

Column 1. The main column section, *A*, consisted of a 1.0 m QVF precision bore glass tube with an internal diameter of 72.45 ± 0.08 mm, enclosing a stack of 16 perforated plates, (Figure 2). This was surmounted by a coalescence section consisting of a 300-mm-long QVF glass section, *C*, and a 250-mm-long QVF T-piece, *D*, with a 25-mm outlet, *E*, both of 76-mm nominal ID. Below the plate section was a 75- to 100-mm glass converter, *F*, enclosing the SS dispersed-phase distributor, *G*. The latter was integral with an 18 swg SS conical adaptor, *H*, which reduced the diameter to 50 mm, where it was mounted on the pulsing unit.

The aqueous phase was fed through distributor *J*, located approximately 60 mm above the top plate. The distributor consisted of two 9.5-mm-dia. SS tubes passing through top plate *K*. The outlet for the continuous phase, *L*, was located in the SS adaptor at the base of the column. The dispersed-phase distributor, *G*, comprised a 40-mm length of 75-mm-OD SS tube closed at the top by a plate containing 44 × 2.64-mm-ID SS nozzles. The base was fitted with a cover carrying the dispersed-phase inlet tubes.

The SS sieve plates were spaced 50 mm apart by means of three 3.2-mm-OD SS tie rods with 6.4-mm spacer sleeves located on a 56.5-mm triangular pitch. The plates were machined to closely fit into the precision bore tube, and the stack was supported between the *T* section, *D*, and the 300-mm glass section, *C*.

A flat paddle stirrer, *M*, and tracer inlet tube, *N*, were located between the 6th and 7th plates from the top of the stack. The 9.5-mm-dia. shaft, *P*, of the stirrer passed through a sleeve down the center of the column and was driven by a variable speed air motor, *Q*. The tracer inlet tube, a 3.2-mm-OD SS hypodermic tube, also passed through the plate stack to the top plate, where it was fixed with a brass collar. The five 3.2-mm-OD SS sample tubes were supported on the top plate and passed through the plates terminating as close as possible below the 2nd, 3rd, 4th, 5th and 9th plates from the top of the stack. These tubes were blocked at the bottom and had 1.4-mm-dia. holes drilled in the side to minimize dispersed phase carry-over.

Table 1. Column Details

	Column		
	1	2	3
Column Dia., mm	72.45	151.2	299.6 (Avg.)
Height of Main Col. Sect., m	1.0	2.0	2.0
Plate Spacing, mm	50	50	50
Plate Thickness, mm	1.575	3.2	2.6
Hole Dia., mm	3.2	3.2	3.2
Hole Pitch (Triang.), mm	6.0	6.0	6.1
Fractional Free Area	0.213	0.227	0.24 (Avg.)
Number of Plates	16	27	26
Plates above Tracer Inlet	5	13	11
Number of Sample Points	5	6	6
Number of Holes	113	509	2,079 (Avg.)
Method of Hole Formation	Drilled	Drilled	Punched
Stirrer Size (Length × Width), mm	35 × 9	75 × 13	150 × 13
Dispersed-Phase Distributor OD, mm	75	120	200
No. and Dia. of Nozzles, mm	44 × 2.64	72 × 6.4	310 × 6.4

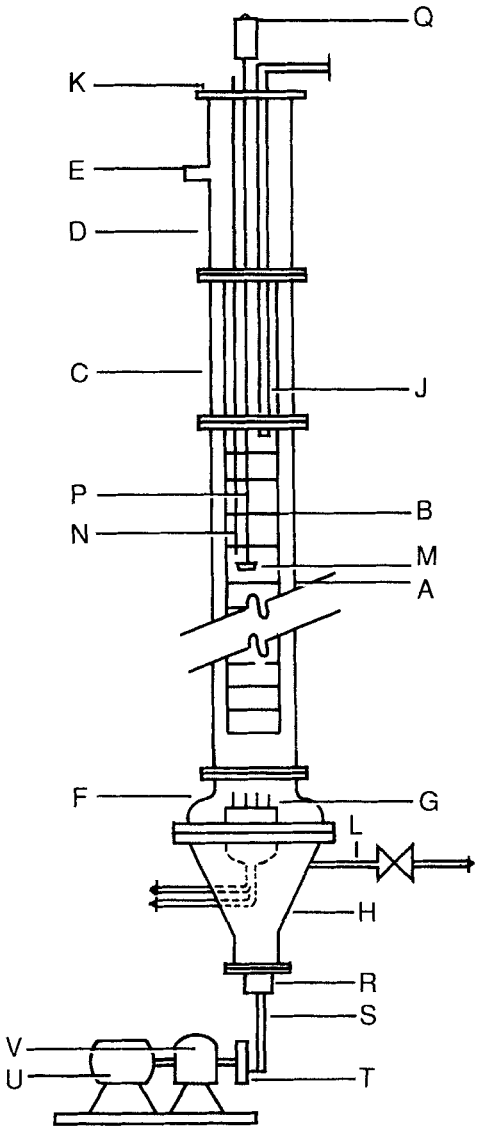

Figure 2. Schematic arrangement of column no. 1.

The pulsing unit consisted of a piston fitted with a cup leather, within a 51.8-mm-ID stainless-steel cylinder, *R*. The piston was connected by a crank arm, *S*, to a variable eccentric drive head, *T*, driven by a flameproof 0.5-hp (0.4-kW) electric motor, *U*, and variable speed unit, *V*. The maximum stroke length available in the column was 4.7 cm, with a frequency range from 0 to 6 Hz. The pulsation within the column was effectively sinusoidal. The pulsing unit was supported on a heavy base plate anchored firmly to the concrete floor.

The general arrangement of the column and auxilliary equipment is shown schematically in Figure 3. All tanks, rotameters, and piping were of SS, and flow control valves were of SS dia-

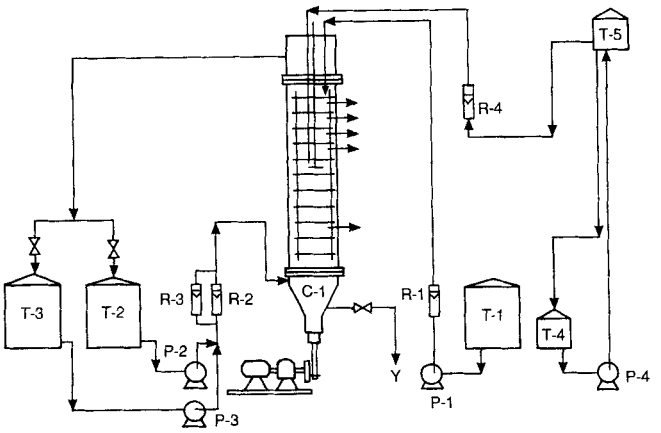

Figure 3. Diagrammatic arrangement of auxiliary equipment for column no. 1.

phragm type with Teflon inserts. The pumps were magnetically driven with totally enclosed motors; the main constructional material was Ryton (polyphenylene sulfide).

The interface level was controlled manually using a needle valve located in the continuous-phase outlet line. Samples were siphoned from the column through 3.2-mm-ID. Nylon tubing connected to the SS sample tubes outside the column.

Columns 2 and 3. Both these columns were mounted on the same pulsing unit, which consisted of a 150-mm-dia. SS cylinder fitted with a piston sealed by a cup leather. It was driven via a variable-stroke eccentric by a 5.4-hp (0.37 kW) motor and variable speed gearbox, giving a frequency range of 0.56 to 3.6 Hz and maximum stroke heights of 15.0 and 7.5 cm in columns 2 and 3, respectively. Column 2 was mounted directly on to the top of the pulse cylinder, and column 3 was mounted via a suitable conical SS adaptor.

Column 2 comprised a 2.0-m-long main section of QVF glass tube with an average diameter of 151.2 mm, surmounted by a 300-mm-long coalescence section and a 250-mm T-piece with a 50-mm outlet for the dispersed phase. Although not of precision bore, the plates were of a close fit in the main section.

The main section of column 3 consisted of a 2.0-m length of QVF glass tube of 300-mm nominal diameter, above which was mounted a 400-mm-long glass T-piece of the same diameter, with a 50-mm side branch for the dispersed-phase offtake. The main column section was constructed from 300-mm-long pressed glass sections fused together, giving an irregular wall which varied in inside diameter between 295.8 and 303.4 mm. To avoid bypassing at the wall, therefore, 12.7-mm-wide annular rings of nitrile rubber were fitted to each of the plates to provide seals.

The auxilliary equipment for these columns was similar to that used for column 1, suitably scaled up. The interface level was, however, controlled automatically using a differential pressure transmitter and controller to maintain a predetermined pressure difference across two air bubbler tubes 350 mm apart, located at each side of the interface.

Materials used

Solvent Systems. Either deionized or mains water was used as the continuous phase, according to the criteria described below.

Kerosene (BP Chemicals K24) was used in all three columns; in addition, four other solvents were used in column 1. The physical properties of the various mutually-saturated phases are given in Table 2.

Tracers. The following tracers were used:

Sodium Chloride	5.5 wt. %
Potassium Dichromate	1.5 to 4.5 wt. %
Tartrazine (Acid Yellow 23)	0.08 to 0.8 wt. %

Methylene blue was also tried, but rejected as it was found to leave a blue coating on the plates which appeared to alter the wetting characteristics. The inlet concentrations of the potassium dichromate and Tartrazine were adjusted to improve the ease and accuracy of the measurements in each experiment.

Sodium chloride was used as tracer for the kerosene-water system in column 1, using a conductivity meter (Philips PR9501) to determine its concentration. Deionized water was used necessarily in view of the very low tracer concentrations in the upper sample points, when backmixing was low. As large volumes of water were required for columns 2 and 3, it was impractical to use deionized water and tap water was therefore used together with potassium dichromate as tracer for column 2 and Tartrazine for column 3; in these cases, the tracer concentrations were measured with a UV/Vis spectrophotometer (Philips SP6-450) at wavelengths of 350 and 425 nm, respectively.

Tartrazine was also used in column 1 with the butyl acetate and MIBK systems, after discovering that the presence of ioniz-

Table 2. Physical Property Data

Continuous Phase	Dispersed Phase*	ρ_c kg/m^3	ρ_d kg/m^3	μ_c cp	μ_d cp	γ** dyne/cm	Tracer
Deionized Water	*n*-Butyl Acetate	999	883	0.96	0.72	14.5	Tartrazine
Mains Water	Heptane	999	682	0.98	0.40	51.7	Potassium Dichromate
Mains Water	Kerosene (BP Chemicals Grade K.24)	998	783	1.00	1.70	44.0	NaCl Tartrazine Potassium Dichromate
Deionized Water	Methyl Isobutyl Ketone	997	809	0.97	0.69	10.1	Tartrazine
Mains Water	Toluene	998	866	0.95	0.59	35.3	Potassium Dichromate

*All solvents used were of technical grade.
**Measured by drop weight (Harkins and Brown, 1916) and du Noüy ring (Zuidema and Waters, 1941) methods.

ing salts increased holdup and reduced throughout with these solvents ("salt effect," Fitzpatrick et al., 1986). Tests conducted with the different tracers gave the same backmixing results within the expected accuracy.

Procedure

Prior to a run, the phases were first circulated through the column to ensure mutual saturation. The pulse amplitude and frequency were then set at the desired values, as were the flow rates of the two phases, and the interface was set at the required level. When conditions were steady, samples were withdrawn from the sample tubes at rates of about 8 mL/min, and the stirrer was turned on and set to a speed of approximately 80 rpm, the maximum possible without causing apparent droplet breakup. The tracer flow rate was then set at 10% of the continuous-phase flow rate (5% in some cases in column 3). When steady state had been reached, two to four samples of 80 mL each were collected from each sample tube and analyzed for tracer content. All runs were conducted at ambient temperature, which ranged between 15 and 25°C. The range of variables investigated is summarized in Table 3.

A qualitative study of axial dispersion in the dispersed phase was made in column 1 by injecting a pulse of Waxoline Blue in kerosene into the dispersed-phase distributor, when operating with the kerosene-water system. The degree of spread of color within the column was observed visually.

Results

Interpretation of data

The results were interpreted in terms of the backflow model, for which the concentration, $c_{c,n}$, of nontransferring tracer at the nth stage above the injection point, stage 0, is given as follows (Pratt and Baird, 1983)

$$c_{c,n} = c_{c,o}[\alpha_c/(1 + \alpha_c)]^n \qquad (14)$$

Hence, a plot of log $[c_{c,n}/c_{c,o}]$ vs. n should give a line of slope $\log[\alpha_c/(1 + \alpha_c)]$, from which α_c can be calculated.

Typical data for columns 1 and 2, plotted in this way, gave a linear relationship; however, as shown in Figure 4, the line of best fit did not pass through the origin (i.e., $n = 0$) and further, a discontinuity occurred between stages 1 and 2. Photographs and video recordings with injected dye present showed that the colored regions appeared uniform over the cross section in the injection stage and those above (Prvcic, 1987). An oscillating color "interface," with a sharp change of color, was also present within each stage, confirming the theory of Prochazka et al. (Novotny et al., 1970; Nemecek and Prochazka, 1974) that fully-mixed regions occur on each side of each plate: i.e., the true demarcation between stages occurs in-between, not at the plates themselves. In consequence, values of α_c for these columns were obtained from plots of log c_n against n, where $\log[\alpha_c/(1 + \alpha_c)]$ is the slope of the line of best fit, i.e., disregarding the tracer concentration below or at the injection point. The discontinuity observed in Figure 4 appeared to result from a difference in the distribution of tracer within the first stage due to the influence of the stirrer; in consequence, only the data for $n > 1$ were used in interpreting the results.

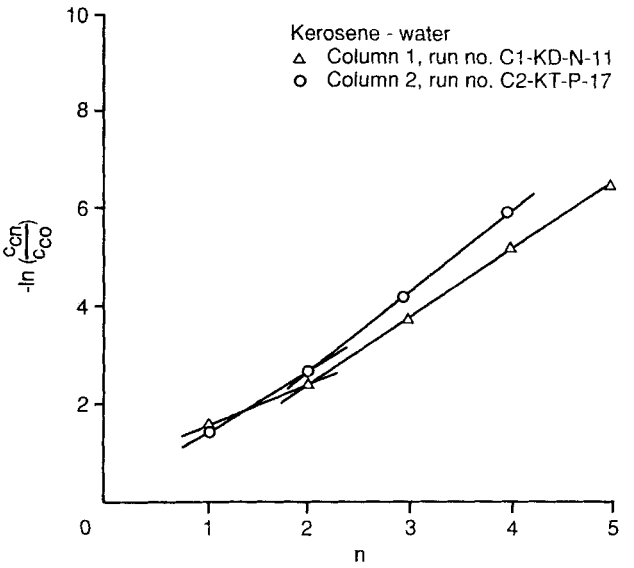

Figure 4. Typical experimental results for column nos. 1 and 2.

In column 3, however, radial variations in color tracer concentration were observed in all stages above the injection stage. To study this further, seven sample tubes were installed in the fourth stage above the injection stage. These revealed that considerable variations in tracer concentration occurred within each stage in single-phase operation; these were reduced, but not eliminated, in two-phase operation and were not affected by improved initial distribution of the continuous phase.

It appeared, therefore, that radial mixing within stages was poor, leading to maldistribution of the phases of the type reported by Woodfield and Sege (1954) in large-diameter columns of this type. To overcome this problem for the present purpose, α_c was, therefore, determined for this column from samples taken from tubes located almost directly above one another. A typical result obtained in this way is shown in Figure 5.

Experimental results

The single-phase data for columns 1 to 3 are summarized in Tables S1-S3 of the Supplementary Material. The corresponding two-phase kerosene-water data are given in Tables S4-S6, and those for the other four solvent-water systems in column 1 in Tables S7-S10. (Tables S1-S10 are available as Supplementary Material in microfiche form.) In all cases, the derived values of α_c are listed; in the two-phase case, values are also given of the dispersed-phase holdup, obtained by the drainage method and smoothed by means of a refitted Bell and Babb correlation (1969), and of the Sauter mean diameter, predicted using the Pietzch-Pilhofer correlation (1984) as corrected by Prvcic (1987). Estimated maximum errors in the backmixing data

Table 3. Ranges of Variables Investigated

Variable	Range
f, s^{-1}	0.5–2.4
A, cm	1.1–2.3
U_c, cm/s	0.09–0.63
U_d, cm/s	0.09–0.60
$R = U_c/U_d$	0.25–2.5
x_d, %	2.0–20.0

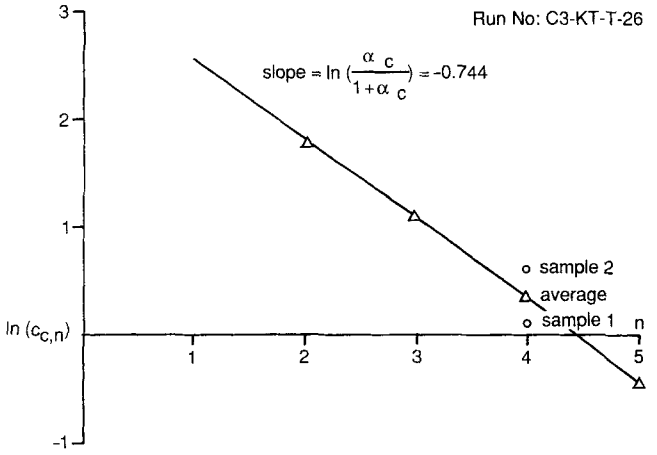

Figure 5. Typical plot of data from column no. 3.

ranged from ±8 to ±36% for column 1 (six data sets), ±9 to ±16% for column 2, and ±6 to ±17% for column 3 (two data sets each).

Typical data for the kerosene-water system are plotted in Figure 6 in the form α_c vs. R, with holdup as parameter. This shows clearly the important effect of increased holdup in increasing the backmixing, especially for values of R below about 1.0.

Modeling of Data

A preliminary study of the α_c values indicated that the single-phase, but not the two-phase, data were dependent upon column diameter. Separate correlations were, therefore, derived for the two cases, based on the physical model of Prochazka et al. (Novotny et al., 1970; Nemecek and Prochazka, 1974). Alternative correlations, obtained by dimensional analysis, were also obtained.

Physical model

In the physical model of Prochazka et al. described earlier, the backmixing coefficient given by Eq. 6 is a combination of a

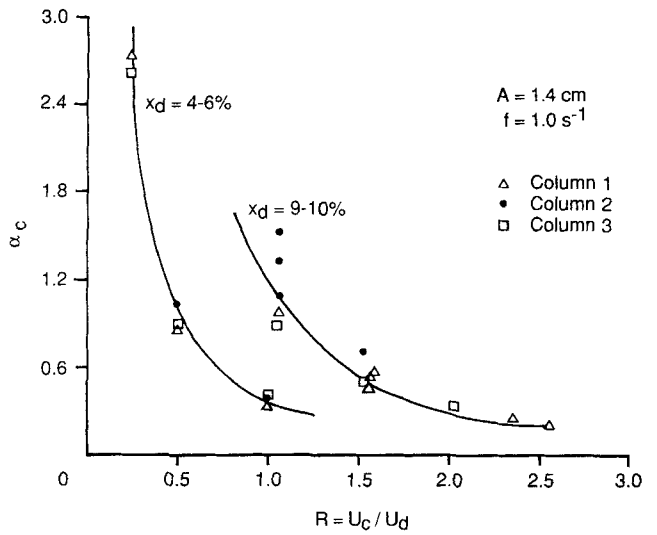

Figure 6. Two-phase backmixing data for kerosene water.

theoretical expression for the backflow ratio, q_c, in the fully-mixed regions on each side of the plates, with an exponential term describing the backmixing in the intervening diffusion regions. This second term requires a knowledge of the length of diffusion path, $(h_c\text{-}m)$, and the effective axial diffusivity, E_c, which were derived empirically by Prochazka et al. for their reciprocating-plate column. The resulting expressions were found not to be applicable to the present pulsed-plate column data, and alternative correlations were therefore sought.

Single-Phase Case. The height of the well-mixed regions, m, may be assumed to be proportional to the mean displacement of fluid from the plate during up- and down-stroke, i.e.

$$m = K_4 (m_d + m_u) \tag{15}$$

The fluid displacement is

$$x(t) = U_c t + (A/2) \sin (2\pi f t) \tag{16}$$

and integration between $t = 0$ and $1/(4f)$ for m_d, and between $t = 1/(2f)$ and $3/(4f)$ for m_u gives for the present case, in which $(A/\pi) > [U_c/(8f)]$

$$m = K_4 (2A/\pi) \tag{17}$$

The axial diffusion term may be assumed to comprise the sum of a Taylor diffusion term and a turbulent diffusion term, *viz.*

$$E_c = E_{Ta} + E_t \tag{18}$$

where for the turbulent case (Taylor, 1954).

$$E_{Ta} = K_5 d_c (\tau_w/\rho_c)^{0.5} \tag{19}$$

For turbulent flow,

$$\tau_w = 0.0396 \rho_c U_c^2 / Re^{0.25} \tag{20}$$

and hence

$$E_{Ta} = K_6 (\mu_c/\rho_c)\, Re^{0.875} \tag{21}$$

Novotny et al. (1970) expressed E_c as the product of a characteristic eddy size l and a fluctuating velocity v', from which they derived

$$E_t = K_2 (2Af) d_h / \epsilon^{1.5} \tag{22}$$

Substitution of m and E_c from Eqs. 17, 18, 21 and 22 then gives Pe_c in Eq. 6, *viz.*

$$Pe_c = \frac{U_c [h_c - K_4 (2A/\pi)]}{K_6 Re^{0.875} (\mu_c/\rho_c) + K_2 (2Af d_h)/\epsilon^{1.5}} \tag{23}$$

Best estimates of the constants, obtained using a nonlinear regression package (Dixon, 1983), were $K_2 = 0.067$, $K_4 = 0.764$, and $K_6 = 0.271$, with correlation coefficient $r^2 = 0.9628$. Values of α_c predicted by Eqs. 6, 12 and 13, using Pe_c values from Eq. 23, are compared with the experimental results in Figure 7.

Two-Phase Case. In their study of two-phase operation of a reciprocating plate column, Nemecek and Prochazka (1974)

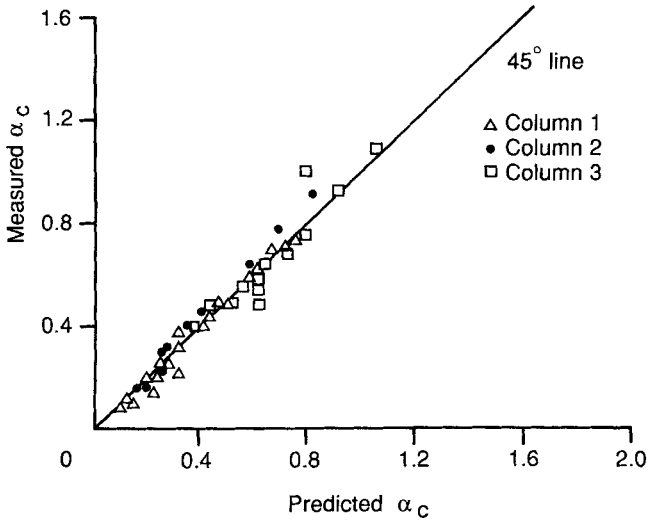

Figure 7. Comparison of single-phase backmixing data with model predictions (Eqs. 6, 12, 13 and 23).

correlated their data on the basis that the axial diffusion coefficient, E_c, could be treated as the sum of the single-phase value plus an increment due to the presence of the second phase. Their resulting correlations did not fit the present data, and an alternative model was developed assuming that E_c is the sum of three components: 1. turbulence due to pulsing through the plates, E_t; 2. induced circulation due to the droplet motion, E_{cir}; and 3. entrainment in the droplet wakes, E_e. However, E_{cir} and E_e both result from dissipation of energy to the continuous phase by the droplets and can be combined in a single term, E'_e ($= E_{cir} + E_e$). Therefore

$$E_c = E_t + E'_e \tag{24}$$

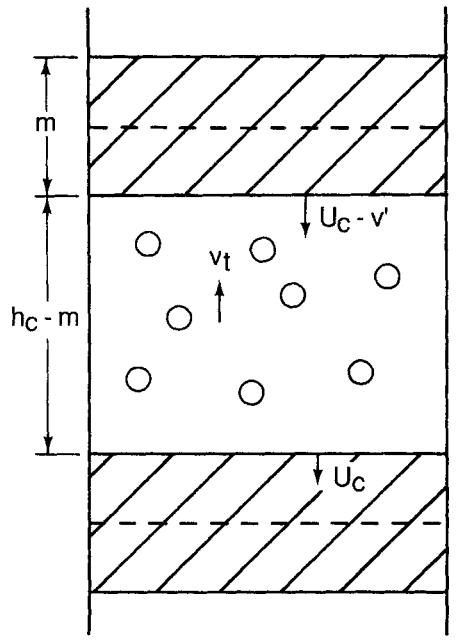

Figure 8. Flows within diffusion region.

where E_t is given by Eq. 22 with a new value of K_2, estimated for the two-phase data.

As proposed by Wijffels and Rietema (1972a, b) and Rietema (1982), E'_e may be assumed to be given by

$$E'_e = K_3 v' l \tag{25}$$

where the mixing length, l, is assumed to be proportional to droplet size, and v' is a characteristic velocity proportional to the maximum velocity induced by the droplets on the continuous phase in their direction of motion. This velocity can be obtained by equating the rate of change of continuous-phase kinetic energy to the rate of energy dissipation by the droplets, both expressed per unit volume of continuous phase.

Thus, the net resistive force, F_r, acting on a single droplet in the diffusion region is

$$F_r = (\pi/6)\, d_{32}^3\, g\, \Delta\rho \tag{26}$$

and the potential energy dissipated per unit time by a swarm of droplets in this region is

$$(\Delta PE)_t = F_r (v_t - U_c) N_d \tag{27}$$

where N_d, the number of droplets in the diffusion region, is

$$N_d = 6S\,(h_c - m)\, x_d/(\pi d_{32}^3) \tag{28}$$

Hence the rate of energy dissipation per unit volume of continuous phase in this region is given by

$$(\Delta PE)_v = g\Delta\rho\,(v_t - U_c)\, x_d/(1 - x_d) \tag{29}$$

The change in kinetic energy per unit volume of continuous phase between top and bottom of the diffusion region is as follows (see Figure 8)

$$\Delta KE = (KE)_2 - (KE)_1 = \tfrac{1}{2}\rho_c\,[(v' - U_c)^2 - U_c^2] \tag{30}$$

and the rate of change is therefore

$$d(\Delta KE)/dt = \tfrac{1}{2}\rho_c\,[(v' - U_c)^2 - U_c^2]\, v_m/(h_c - m) \tag{31}$$

where v_m, the average continuous phase velocity in the diffusion region, is assumed to be

$$v_m = \tfrac{1}{2}\,[(v' - U_c) + (-U_c)] = \tfrac{1}{2}\,(v' - 2\,U_c) \tag{32}$$

Equating Eqs. 29 and 31 and putting $v' \gg U_c$, which was found to be a reasonable assumption, gives

$$v' = \left[\frac{4g\Delta\rho(v_t - U_c)\, x_d(h_c - m)}{\rho_c(1 - x_d)}\right]^{1/3} \tag{33}$$

Substitution of Eq. 33, together with $l = Kd_{32}$ into Eq. 25, and hence with Eq. 22 into Eq. 24, gives finally, on combining the constants

$$E_c = \frac{K_2(2Af)d_h}{\epsilon^{1.5}} + K_3 d_{32}\left[\frac{4g\Delta\rho(v_t - U_c)\, x_d(h_c - m)}{\rho_c(1 - x_d)}\right]^{1/3} \tag{34}$$

The expression for m obtained for the single-phase case (Eq. 17), was found to provide the best fit to the data. Hence, α_c can be calculated from Eqs. 6-8, using this expression for m together with E_c from Eq. 34 to obtain the value of Pe_c. The best estimates of the constants in Eqs. 17 and 34 for the two operating regions, obtained using the nonlinear regression package together with predicted values of x_d, d_{32} and v_t (Prvcic, 1987), are given in Table 4. Comparisons of experimental and predicted results for the mixer-settler and dispersion regions are shown in Figures 9 and 10. The transition between these two regions may be considered to occur at the minimum in the plot of dispersed-phase holdup against fA.

Dimensional analysis

It is apparent that the foregoing models provide satisfactory representations of the data. However, they have the disadvantage of requiring *a priori* knowledge in the two-phase case of the dispersed-phase holdup and Sauter mean droplet diameter. As these at present cannot be predicted with sufficient accuracy (Prvcic, 1987), alternative correlations of the data were obtained by dimensional analysis. These are described in the Supplementary Material; the result for the important case of two-phase operation in the emulsion region is a follows:

$$\alpha_c = 0.125 \left(\frac{U_c}{fA}\right)^{-0.45} \left(\frac{h_c}{A}\right)^{-0.89} \left(\frac{U_d}{U_c}\right)^{0.21} \left(\frac{\gamma}{AU_c^2\Delta\rho}\right)^{0.37} \left(\frac{\Delta\rho}{\rho_c}\right)^{0.55} \quad (39)$$

with $r^2 = 0.9505$.

Discussion

The steady-state tracer injection method appears to be satisfactory for the measurement of continuous-phase backmixing in pulsed-plate columns. Some modifications to the normal procedure, however, were necessary. These included the provision of a stirrer in the injection stage, which appeared to give good radial mixing in that stage in the two smaller diameter columns. There was, however, some evidence in the largest column that mixing may not have been complete at the maximum stirring rate possible without inducing extra droplet breakup; this could have been responsible in part for the segregation noted, which was overcome by withdrawing the samples from a near-vertical plane. It, however, appears for columns of 300-mm-diameter and more that it would be desirable to employ multipoint tracer injection in addition to agitation.

An important conclusion from the results is that the derived α_c values were unaffected by column diameter in two-phase operation, unlike those for the single-phase case; the latter are,

Table 4. Constants Used in Eqs. 17 and 34 for Two-Phase Operation

Constant	Region	
	Mixer-Settler	Emulsion
K_2	0.080	0.029
K_3	0.082	0.274
K_4	2.28	1.40
r^2	0.9586	0.9424

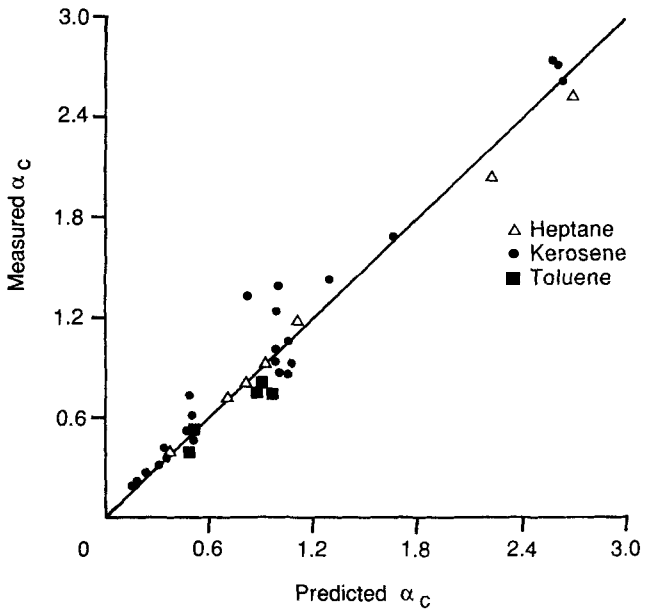

Figure 9. Comparison of model predictions for mixer-settler regime (Eqs. 6-8, 17 and 34).

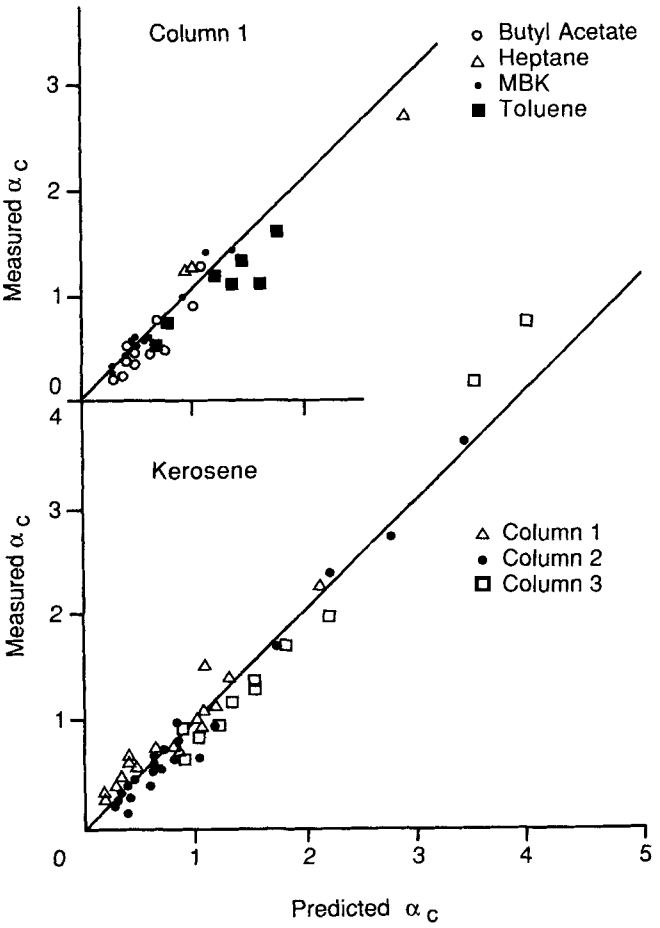

Figure 10. Comparison of model predictions for emulsion regime (Eqs. 6-8, 17 and 34).

therefore, of little use in predicting two-phase performance. It was evident, however, that phase maldistribution of the type reported by Woodfield and Sege (1954) becomes important with the larger diameters, requiring the provision of radial mixing devices such as the swirl plates described by these workers. Such maldistribution would also have a major influence on the continuous-phase residence time distribution, in addition to that due to backmixing, so that backmixing coefficients estimated using the well-established transient tracer injection method would be expected to differ from those given by the present continuous injection method.

In terms of correlation coefficients, there is little difference in accuracy between the dimensional analysis and the physical models proposed as representations of the data. In general, however, the physical model is to be preferred on grounds of realism, especially for extrapolation purposes, provided that the necessary holdup and droplet size data are available. In this regard, account must be taken of the major effect reported by Fitzpatrick et al. (1986) of small concentrations of ionizable salts on the holdup and size of polar solvent droplets.

It may be concluded, from the qualitative tests reported, that backmixing of dispersed phase is almost certainly negligible. In addition, forward dispersion of this phase is negligible in the mixer-settler region, but must be taken into account for operation in the emulsion region.

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Notation

A = peak-to-peak amplitude of pulsation, m
c = tracer concentration, kg · m^{-3}
d_c = diameter of column, m
d_h = diameter of holes in plates, m
d_{32} = Sauter (volume-surface) mean diameter of droplets, m
E_c = axial turbulent diffusivity of continuous phase, m^2 · s^{-1}
f = frequency of pulsation, s^{-1}
g = acceleration due to gravity, 9.814 m · s^{-2}
h_c = plate spacing, m
K_2-K_6 = constants
m = height of fully-mixed regions, m
n = stage number
Pe_c = continuous-phase Peclet no., defined by Eq. 4
q = ratio of backflow of a phase to net forward flow
R = U_c/U_d
Re = Reynolds no., $(d_c U_c \rho_c/\mu_c)$
S = cross-sectional area of column, m^2
t = time, s
U = superficial velocity of phase, m · s^{-1}
v_t = terminal velocity of isolated droplet, m · s^{-1}
x_d = fractional holdup of dispersed phase
$x(t)$ = displacement of fluid as a function of time, m

Greek letters

α_c = backmixing ratio (ratio of backflow to net forward flow) of continuous phase
γ = interfacial tension, N · m^{-1} = kg · s^{-2}
ϵ = fractional free area of plates
μ = viscosity, kg · m^{-1} · s^{-1} = Pa · s
ρ = density of phase, kg · m^{-2}
$\Delta\rho$ = $|\rho_c - \rho_d|$, kg · m^{-3}
ϕ = parameter defined by Eq. 8

Subscripts

b = backflow
c = continuous phase
d = dispersed phase
n = plate no.

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