cause on that basis the 1-in.-diameter column should give the lowest MCR and the 6-in. column the highest. Since that is not the case, a more plausible explanation is that time is required for the large polar molecules to enter the porous structure of the clay adsorbent. Thus, up to some limiting value, the longer the saturates take to pass through the adsorbent column, the more completely the high microcarbon polars should be removed.

The data (Table IV) show a regular reduction in microcarbon residue of the saturate fraction as the residence time of the solvent is increased. Further evidence from the laboratory experience of the authors has shown that about 1-h solvent residence time is the minimum for effective separation and that longer times given even better separations. The reason that short residence times are not effective is bypassing of the internal surface area of the adsorbent. This problem can be serious during loading of the feed sample on the column if the sample is highly diluted with solvent and the solvent is drawn down rapidly to compensate for the feed addition. This was not a problem in the chromatographic runs work performed during this study. The easiest way to identify this problem is by dark color in the separated fraction. It should normally be from water white to pale straw colored. When present in the saturates fraction, the dark color is provided by polar molecules that have bypassed the adsorbent.

To our knowledge, this is the first reported large-scale separation of whole crudes and residua other than those recent processes that use adsorption in conjunction with catalytic cracking. ¹⁴ Because of the higher molecular weight polar constituents in these feedstocks, the molecules require time to reach the internal surface of the adsorbent particles. This accounts for (a) the large pore size requirement for the adsorbent and (b) the influence of residence time.

Conclusions

The large 12-in.-diameter adsorber designed for this work is capable of giving the same separations over Attapulgus clay as smaller laboratory units. Care must be taken when loading the unit not to cause feed bypassing of the adsorbent bed due to rapid drainage of solvent from the bottom of the adsorber into the solvent-recycle boiler.

Such bypassing shows up as a black contamination of the normally clean saturate fraction.

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Prediction of Continuous-Phase Axial Mixing Coefficients in Pulsed Perforated-Plate Extraction Columns

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The correlation of axial mixing in the continuous phase of pulsed perforated-plate extraction columns is presented. Published experimental results obtained with both the steady-state and the dynamic tracer injection techniques are considered. On the basis of 992 data points with and without mass transfer for 28 liquid/liquid systems from 13 different sources, a single equation is determined that reproduces the continuous-phase axial mixing coefficient in the mixer-settler, transition, and emulsion regions of operation over a wide range of variables. This may be used for design purposes under mass-transfer conditions.

Introduction

A knowledge of the extent of axial mixing in liquid/ liquid extraction columns is necessary with regard to design and for understanding the fundamental processes involved in the mass transfer. The presence of axial mixing results in a concentration "jump" at the inlet of each phase. The concentration driving potential for mass transfer is therefore reduced, and hence, the extraction efficiency is adversely affected. The factors that contribute to axial

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mixing are discussed by Pratt and Baird (1983).

The most common methods for obtaining data on axial mixing use tracer injection. The experimental results obtained are usually interpreted in terms of an axial dispersion model which assumes plug flow with disturbances caused by circulatory flow, molecular diffusion, small eddies, channeling, and a liquid velocity profile lumped in an axial mixing coefficient. The axial mixing coefficient, obtained with steady injection of a tracer near the phase outlet, gives a measure of backflow only and does not include the effect of residence-time distribution caused by nonuniform forward flow. However, the dynamic tracer method, in which a pulse or step change is introduced near the phase inlet, gives a measure of the residence-time distribution and hence of the total axial mixing. Since the actual flow patterns in an extractor are complex, the use of tracer methods in conjunction with the axial dispersion models may have limitations. It may therefore be desirable to measure the concentration profiles of a transferring solute during the steady-state operation and evaluate the axial mixing by solution of the transport equations (see, e.g., Hartland and Mecklenburgh (1966) and Pratt and Baird (1983)). The experimental difficulty of carrying out the profile studies together with the insensitivity of profiles to axial dispersion in many cases (Pratt and Baird, 1983) make it difficult in practice to determine the axial mixing coefficients in this way. This insensitivity is actually advantageous since the predicted separation when mass transfer occurs is not usually sensitive to the values of the mixing coefficients.

Several studies on pulsed perforated-plate extraction columns (see, e.g., Cohen and Beyer (1953) and Sege and Woodfield (1954)) have shown that column HTU passes through a minimum value with increasing intensity of agitation, an effect that could be due to the increasing magnitude of axial mixing. Although several experimental investigations have been made to determine the continuous-phase axial mixing coefficients, the data on dispersed-phase axial mixing are scanty and contradictory. In the present work, published data on continuous-phase axial mixing coefficients with and without mass transfer are analyzed, and an empirical correlation of the data in terms of the physical properties of the liquid/liquid systems, column geometry, and operating parameters is suggested.

Previous Work

Although a number of correlations for the prediction of continuous-phase axial mixing coefficients are available, most of them are either approximate or based on limited experimental ranges of operating variables. These correlations should not, therefore, be extrapolated beyond the quoted ranges of application. Moreover, as will be discussed below, there is a lack of agreement between various correlations regarding the form of the dependence of axial mixing coefficient upon the different variables involved.

A summary of the published correlations predicting the continuous-phase axial mixing coefficients is given in Table I which also lists the conditions for which the use of a correlation has been recommended together with an identification of the regions of operation. Furthermore, the techniques to measure the data on axial mixing, used in the formulation of a correlation, are also given. It must be emphasized that SI units are consistently used and $E_{\rm c}$ represents the actual axial mixing coefficient. Care is therefore required since some of the original equations were given in terms of the superficial axial mixing coefficient, $e_{\rm c}$, related to $E_{\rm c}$ by

$$e_c = E_c(1 - \epsilon) \tag{1}$$

The major correlations (eq TI.1–TI.6) listed in Table I are based on data from more than one liquid-liquid system and/or one column geometry, whereas the minor correlations (eq TI.7–TI.12) only apply to specific situations. On the basis of these correlations, the effects of different variables on $E_{\rm c}$ may be summarized as follows.

- (1) Generally, the axial mixing coefficient, $E_{\rm c}$, increases with increased agitation, although the effects of amplitude, A, and frequency, f, are not necessarily accounted for by their product, Af (Mar, 1958; Mar and Babb, 1959; Sehmel, 1961; Sehmel and Babb, 1964; Rama Rao et al., 1983). According to Sehmel (1961), Sehmel and Babb (1964), and Reissinger (1985), only in the mixer-settler region of operation does $E_{\rm c}$ increase with agitation until a maximum is reached, corresponding to the beginning of the transition (dispersion) region, beyond which $E_{\rm c}$ decreases as agitation increases.
- (2) Correlations due to Mar (1958) and Mar and Babb (1959), Sehmel (1961) and Sehmel and Babb (1964), and Kagan et al. (1965) show that $E_{\rm c}$ varies inversely with the continuous-phase superficial velocity, $V_{\rm c}$. Reissinger's (1985) formula indicates no dependence, whereas Vassallo's (1983) correlations show that the effect of $V_{\rm c}$ may be negative or positive depending upon the value of $V_{\rm c}$. The remaining correlations show that $E_{\rm c}$ increases with an increase of $V_{\rm c}$.
- (3) Most of the published correlations do not show any effect of the dispersed-phase superficial velocity, $V_{\rm d}$, on $E_{\rm c}$. Mar (1958) and Mar and Babb (1959), Rozen et al. (1970), Vassallo (1983), and Reissinger (1985) suggest an increase of $E_{\rm c}$ with increasing $V_{\rm d}$, and Kagan et al. (1965) find the opposite effect.
- (4) Large values of the plate spacing, h, and hole size, $d_{\rm o}$, invariably result in higher axial mixing. Only the correlation of Rama Rao et al. (1983) indicates a decrease of $E_{\rm c}$ with h, and an unrealistic negative index on $d_{\rm o}$ determined by Reissinger (1985) may be due to the fact that $d_{\rm o}$ did not vary in his experiments. An increase of the plate free area, ϕ , usually results in lower axial mixing coefficients. Rozen et al. (1970), however, suggest that $E_{\rm c}$ should increase with ϕ .
- (5) The effect of column diameter, D, on E_c is insignificant (Kagan et al., 1973; Tung, 1984; Tung and Luecke, 1986). The correlations of Miyauchi and Vermeulen (1963), Miyauchi and Oya (1965), and Miao (1978) indicate a decrease and that of Rozen et al. (1970) indicates an increase of E_c with increasing D.
- (6) It is not possible to assess the effects of continuous-phase density, ρ_c , and viscosity, μ_c , on E_c since water was the continuous phase in all the cases. Mar (1958) and Mar and Babb (1959) reported that the density difference, $\Delta\rho$, had little effect on E_c and did not include it in their final correlation; the correlation of Sehmel (1961) and Sehmel and Babb (1964) shows that E_c would increase with $\Delta\rho$. The axial mixing also increases with the interfacial tension, σ (Mar, 1958; Mar and Babb, 1959). Reissinger (1985) finds it necessary to include the ratio $\sigma_{\rm top}/\sigma_{\rm bottom}$ for his case of mass transfer from the heavy continuous phase to the lighter dispersed phase in which σ increases down the column.

Published Data Sources

The axial mixing data of the continuous phase in pulsed columns are available for both single- and two-phase flows. However, not only do the values of axial mixing coefficient for the two cases differ significantly (see, e.g., Kagan et al. (1965) and Nemecek and Prochazka, 1974), the varia-

correlation	range of applic		
Major Cor	relations		method
Mar (1958) and Mar and Babb (1959): $\frac{E_{c}(1-\epsilon)}{V_{c}d_{0}} = 0.171 \left(\frac{\mu_{c}}{\rho_{c}V_{c}t}\right)^{1.45} \left(\frac{t}{d_{0}}\right)^{0.70} \left(\frac{h}{t}\right)^{0.88} \left(\frac{V_{d}\rho_{c}t}{\mu_{c}}\right)^{0.30} \times$	(TI.1)	mixer-settler, transition	а
$\begin{pmatrix} \sigma \rho_{\rm c} t \\ \mu_{\rm c}^2 \end{pmatrix}^{0.42} \begin{pmatrix} f \rho_{\rm c} t^2 \\ \mu_{\rm c} \end{pmatrix}^{0.36} \left(\frac{A}{t}\right)^{0.07}$			
Sehmel (1961) and Sehmel and Babb (1964): $E_c(1-\epsilon) = 1.08 \times 10^{-4} - 1.80 \times 10^{-2} V_c (f-f_{\rm m})^2 + 3.06 \times 10^{-7} \Delta \rho$ $E_c(1-\epsilon) = 1.80 \times 10^{-4} - 1.39 \times 10^3 V_c^3 (f-f_{\rm m})^2 + 2.67 \times 10^{-7} \Delta \rho - 4.88 \times 10^{-3} A + 9.96 \times 10^{-2} A^2$	(TI.2a) (TI.2b)	$f < f_m$, mixer-settler $f > f_m$, transition, emulsion	a a
$f_{\rm m} = 258.9 \mu_{\rm d} \sigma \Delta \rho - 0.667 \ln{(A)} - 2.249$ Miyauchi and Vermeulen (1963) and Miyauchi and Oya (1965):	(TI.2c)		
$\frac{E_{\rm c}}{Afh} = \frac{V_{\rm c}}{Af(1-\epsilon)(2-1/N)}$	(TI.3a)	mixer-settler	b
$\frac{E_{\rm c}}{Afh} = \frac{V_{\rm c}}{Af(1-\epsilon)(2\beta-1/N)} + \frac{1}{\beta}$	(TI.3b)	transition, emulsion	b
$\beta = 0.57(D^2h)^{1/3}\phi/d_0$ Miao (1978):	(TI.3c)		
$\frac{E_{\rm c}}{Afh} = \frac{V_{\rm c}}{Af(1-\epsilon)(2\beta-1/N)} + \frac{1}{\beta}$	(TI.4a)	mixer-settler, transition, emulsion	a, b
$\beta = 0.765 (D^2 h)^{1/3} \phi / d_0$ Tung (1984) and Tung and Luecke (1986):	(TI.4b)		
$\frac{E_{\rm c}(1-\epsilon)}{hV_{\rm c}} = 0.250\phi^{-1.30} \left(\frac{d_{\rm o}}{h}\right)^{0.565} \left(\frac{Af}{V_{\rm c}}\right)^{0.606}$	(TI.5)	$ ho_{\rm c}(Af)^3/(2\phi^2) > 0.06 \ { m kg/s^3}, \ { m emulsion}$	a, b
Reissinger (1985): $\frac{E_{c}}{V_{c}h} = 6.26 \left(\frac{\mu_{c}}{\rho_{c}V_{c}d_{o}}\right)^{1.00} \left(\frac{\rho_{c}V_{d}d_{o}}{\mu_{c}}\right)^{0.25} \left(\frac{\sigma\rho_{c}d_{o}}{\mu_{c}^{2}}\right)^{0.10} \times$	(TI.6)	0.012 m/s < Af < 0.031 m/s, transition (mass transfer c \rightarrow d; $\rho_{\rm c}$ > $\rho_{\rm d}$)	b
$\left(rac{ ho_{ m c}Afd_{ m o}}{\mu_{ m c}} ight)^{\!-0.40}\!\!\left(rac{\sigma_{ m top}}{\sigma_{ m bottom}} ight)^{\!1.40}$			
Minor Co	rrelations		
Kagan et al. (1965): $E_{\rm c}(1-\epsilon)=1.26\times 10^{-4}A^{1.2}f^{1.35}/(V_{\rm c}+V_{\rm d})^{1.4}$ Rozen et al. (1970):	(TI.7)	mixer-settler, transition	а
$E_{\rm c}(1-\epsilon) = 3.0[\phi h/(1+h/D)]^{2/3}d_{\rm o}^{1/3}(V_{\rm c}+0.5V_{\rm d}+2Af)$ Kagan et al. (1973):		mixer-settler, transition, emulsion	b
$\tilde{E}_{c}(1-\epsilon) = 0.16h^{0.76}(Af + V_{c})$ Ugarcic (1981):	(11.9)	$(Af + V_c) > 0.018 \text{ m/s}$, transition, emulsion	. в
$\frac{E_{\rm c}}{V_{\rm c}h} = \frac{8.06}{N(1-\epsilon)} + \frac{3.77Af}{NV_{\rm c}}$	(TI.10)	transition	а
Rama Rao et al. (1983): $E_{\rm c}(1-\epsilon) = 6.64 \times 10^{-3} A^{0.5} f^{0.33} V_{\rm c}^{0.8} d_{\rm o}^{0.2} \phi^{-0.8} h^{-0.69}$ Vassallo (1983):	(TI.11)	mixer-settler, transition	b
$E_{c} = 0.009 A f V_{d}^{0.4} / V_{c}^{0.5}$ $E_{c} (1 - \epsilon) = 0.037 [2A f (1 - \epsilon) + V_{c}] V_{d}^{0.2}$	(TI.12a) (TI.12b)	$V_{\rm c}$ < 0.0008 m/s, transition, emulsion $V_{\rm c}$ > 0.0012 m/s, transition, emulsion	<i>b</i> <i>b</i>
and plate-free area	ly. ^c Recipro	ocating-plate column fitted with plates having	small hole

and plate-free area.

tion in the coefficient with Af may also have different form (Nemecek and Prochazka, 1974). Therefore, the data used to determine the correlation for E_c are restricted to twophase flow.

The axial mixing data on 28 liquid/liquid systems correlated in this work were taken from 13 different data sources: Mar (1958), Claybaugh (1961), Sehmel (1961), Kagan et al. (1965), Rozen et al. (1970), Barame (1972), Kagan et al. (1973), Arthayukti (1975), Ugarcic (1981), Niebuhr (1982), Britsch (1983), Vassallo (1983), and Reissinger (1985). The steady-state or dynamic tracer injection technique was used to measure the axial mixing coefficient in all the cases. In 24 systems, no mass transfer took place, but it occurred in the remaining 4 from the continuous to the dispersed phase. Since the holdup data for 13 systems were not available, these were predicted with the correlation proposed by Kumar and Hartland (1988). Furthermore, the experimental or predicted values of the holdup were used to calculate E_c in cases where the original data were given in terms of the superficial axial mixing coefficient, e_c , with eq 1. The ranges of physical properties, operating conditions, and column dimensions

were $\rho_{\rm c}=994-1092~{\rm kg/m^3},~\rho_{\rm d}=652-1590~{\rm kg/m^3},~\Delta\rho=102-591~{\rm kg/m^3},~\mu_{\rm c}=0.84-7.86~{\rm mPa}~{\rm s},~\mu_{\rm d}=0.29-1.99~{\rm mPa}~{\rm s},~\sigma=3.7-50.0~{\rm mN/m},~V_{\rm c}=0.22-10.14~{\rm mm/s},~V_{\rm d}=0.14-9.78~{\rm mm/s},~A=5.00-51.56~{\rm mm},~f=0.32-4.25~1/{\rm s},~Af=2.10-102.70~{\rm mm/s},~\epsilon=0.004-0.675,~E_{\rm c}=16-1792~{\rm mm^2/s},~D=50.0-300.0~{\rm mm},~h=37.5-300.0~{\rm mm},~d_{\rm o}=1.6-5.0~{\rm mm},~{\rm and}~\phi=0.082-0.32.$ Detailed information on the results for different liquid/liquid systems reported by the investigators, whose data are covered in the present analysis, is given in Tables II and III of the supplementary material.

Development of a New Correlation for $E_{\rm c}$

The published results on $E_{\rm c}$ detailed above are correlated in terms of the physical properties of the liquid/liquid systems, the operating parameters, and the column geometry on the following basis.

- (1) The continuous-phase density, ρ_c , varies only from 994 to 1092 kg/m³, so it is not considered to be a variable.
- (2) The pulsation velocity, Af, is the correlating parameter for the amplutide and frequency effects.
- (3) As observed by previous authors (Sehmel, 1961; Niebuhr, 1982; Reissinger, 1985), $E_{\rm c}$ is assumed to increase with increasing Af in the mixer-settler region of operation until a maximum is reached, corresponding to the beginning of the transition region. Thereafter, $E_{\rm c}$ decreases slowly with Af since the drop size is uniform and the holdup is low. However, in the emulsion region of operation, $E_{\rm c}$ increases once again with Af due to the turbulence in the continuous phase, high population density of small drops, and high values of holdup.
- (4) The value of the amplitude–frequency product at the boundary between the mixer–settler and transition regions of operation is given by the following equation proposed by Kumar and Hartland (1988):

$$(Af)_{\rm m} = 9.69 \times 10^{-3} (\sigma \Delta \rho^{1/4} \phi / \mu_{\rm d}^{3/4})^{0.33} \quad [\rm m/s] \quad (2)$$

(5) For $Af < 2(Af)_m$, a translated agitation group is defined by

$$\psi = \left[\frac{Af - (Af)_{m}}{(Af)_{m}} \right]^{3} - \left[\frac{Af - (Af)_{m}}{(Af)_{m}} \right]^{2}$$
(3)

whereas for $Af > 2(Af)_{m}$, the group

$$\psi = [Af - 2(Af)_{m}]/(Af)_{m} \tag{4}$$

may be used to describe the shape of $E_{\rm c}$ data with Af. It is postulated next that $E_{\rm c}$ is only a function of ψ , $V_{\rm c}$, $V_{\rm d}$, $\Delta \rho$, $\mu_{\rm c}$, $\mu_{\rm d}$, σ , D, h, $d_{\rm o}$, and ϕ . The dimensionless correlation expressing $E_{\rm c}$ as a product of an exponential function for ψ and the simple power functions for each of the other variables can be written as

$$\frac{E_{c}\Delta\rho}{\mu_{c}} = k_{1} \exp\left[k_{2}\psi\right] \left(\frac{V_{d}\mu_{c}}{\sigma}\right)^{p_{1}} \left(\frac{\mu_{c}}{\mu_{d}}\right)^{p_{2}} \times \left(\frac{\mu_{c}}{(\sigma\Delta\rho h)^{1/2}}\right)^{p_{3}} \left(\frac{d_{o}}{h}\right)^{p_{4}} \left(\frac{V_{c}}{V_{d}}\right)^{p_{6}} \left(\frac{D}{h}\right)^{p_{6}} \phi^{p_{7}} (5)$$

On data from 992 measurements with and without mass transfer from 28 liquid/liquid systems obtained with the steady-state and the dynamic tracer injection methods (Tables II and III of the supplementary material), Marquardt's (1963) algorithm was performed to calculate the unknown parameters of the above equation. Preliminary calculations showed that it was necessary to introduce the density of water at 20 °C, ρ_* = 998 kg/m³, and the standard plate spacing, h_* = 0.05 m, as reference variables to correctly allow for the effects of $\Delta\rho$ and h on E_c . Furthermore,

Table IV. Values of k_1 and k_2 Determined under Different Conditions

			k			
tracer injection techn	no. of data pts	k_1	$Af < 2(Af)_{\mathbf{m}}$	$Af > 2(Af)_{\mathbf{m}}$	s, %	
no mass transfer						
steady state	361	46.64	0.90	0.17	32.5	
dynamic	556	49.50	0.43	0.44	34.2	
$\begin{array}{c} \text{mass transfer} \\ \text{c} \rightarrow \text{d} \end{array}$						
steady state, dynamic	75	43.17	0.43	0.36	23.2	
all data steady state, dynamic	992	46.15	0.80	0.34	33.8	

the indices on groups $V_{\rm c}/V_{\rm d}$ and D/h were close to zero, and the index on ϕ was statistically insignificant. Therefore, in eq 5, the last three terms were deleted and the groups $\Delta\rho/\rho_*$ and h/h_* added. A subsequent regression analysis gave

$$\frac{E_{c}\Delta\rho}{\mu_{c}} = k_{1} \exp\left[k_{2}\psi\right] \left(\frac{V_{d}\mu_{c}}{\sigma}\right)^{0.11} \left(\frac{\mu_{c}}{\mu_{d}}\right)^{-0.37} \times \left(\frac{\mu_{c}}{(\sigma\Delta\rho h)^{1/2}}\right)^{-0.61} \left(\frac{d_{o}}{h}\right)^{0.36} \left(\frac{\Delta\rho h}{\rho \cdot h_{\bullet}}\right)^{1.05}$$
(6)

with $k_1 = 46.15$ and

$$k_2 = \begin{cases} 0.80 & \text{for } Af < 2(Af)_{\text{m}} \\ 0.34 & \text{for } Af > 2(Af)_{\text{m}} \end{cases}$$

Since the indices on groups $\Delta\rho/\rho_*$ and h/h_* were almost identical, these groups were combined in eq 6. The value of the standard deviation, s, in the predicted values of the continuous-phase axial mixing coefficient, $E_c(\text{pred})$, from the experimental points, $E_c(\text{exp})$, defined for M pairs and m parameters by

$$s = 100(\sum_{1}^{M} [\{E_{c}(\text{pred}) - E_{c}(\text{exp})\}/E_{c}(\text{exp})]^{2}/(M-m))^{1/2}$$
(7)

was 33.8%

As mentioned in the Introduction, the values of E_c obtained with the two tracer injection techniques may well differ from each other. Moreover, the value of E_c may also be affected by the transfer of solute from one phase to the other (Reissinger, 1985). Separate equations are therefore needed to describe the data obtained with different measurement techniques and when mass transfer occurs. However, the same values of the indices were employed, and only the constants k_1 and k_2 in eq 6 were optimized for the different cases. For no mass transfer, the steadystate and dynamic tracer injection data were treated separately. The mass-transfer data measured with both methods were considered together since only 34 data points on a single liquid system (Ugarcic, 1981) corresponding to the steady-state method are available (Tables II and III of the supplementary material). The best values of the constants k_1 and k_2 , together with those of s for the cases just mentioned, are given in Table IV. Although optimizing the values of the constants k_1 and k_2 reduces the value of s only by about 1%, comparison of these values for the case of no mass transfer shows that the values of $E_{\rm c}$ determined with the steady-state technique are smaller than those obtained with the dynamic tracer injection method as expected, since the former technique only measures the actual backmixing, whereas the latter method determines the total axial mixing.

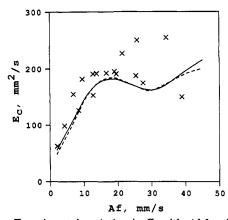


Figure 1. Experimental variation in $E_{\rm c}$ with Af for the system benzene (dispersed)/water (continuous) with $V_{\rm c}=V_{\rm d}=2.61~{\rm mm/s}$ measured by Sehmel (1961) (indicated by the symbol ×) compared with that predicted by eq 6 (full line). The dashed line gives the variation with the values of k_1 and k_2 listed in Table IV determined using steady-state data with no mass transfer.

Results and Discussion

Equation 6 shows that $E_{\rm c}$ is independent of the continuous-phase superficial velocity, $V_{\rm c}$, in agreement only with the work of Reissinger (1985). As discussed in the Previous Work section, the opposite directional effects of V_c were observed by different authors. The cause of this discrepancy may lie in the choice of the fitting equation and the ranges of variables covered.

The effect of column diameter on E_c turns out to be insignificant. Many other authors (Kagan et al., 1973; Rouyer et al., 1974; Tung, 1984; Tung and Luecke, 1986) have found similar results. The correlations of Miyauchi and Vermeulen (1963) and Miyauchi and Oya (1965), however, show that E_c should decrease sharply with column diameter. This difference may probably be due to the fact that these authors worked with small diameter columns of 32 and 54 mm. It should be noted, however, that E_c does depend upon the plate-free area since this is included in the definition of (Af)_m given by eq 2.

Equation 6 predicts the experimental data with a standard deviation of 33.8%. Such a reproduction of data is satisfactory, considering that the experimental results scatter widely. The values of E_c from repeat runs measured by Niebhur (1982) often differ by a factor of 2 and sometimes by an order of magnitude. Vassallo (1983) attributed the large scatter of his data to the difficulty in maintaining the stable liquid-liquid interface and the assumptions of uniform holdup and drop size. An additional error might have also been introduced in eq 6 since predicted values of holdup were used to calculate $E_{\rm c}$ when the original data were given in terms of e_c and the holdup values were not measured or quoted by the original authors.

Unfortunately, only a few attempts have been made to experimentally examine the effect of mass transfer upon axial mixing. Additional data are needed for c -> d direction of mass transfer to confirm the values of the constants k_1 and k_2 in eq 6. To our knowledge, no data are available on systems with mass transfer taking place from the dispersed to the continuous phase $(d \rightarrow c)$.

Figure 1 compares the experimental variation in E_c with Af in the mixer-settler, transition, and emulsion regions of operation measured by Sehmel (1961) by using the steady injection of a tracer for the benzene (dispersed)/ water (continuous) system with flow rates $V_c = V_d = 2.61$ mm/s in a column of 50-mm diameter with that predicted by eq 6 (full line). In Figure 2, eq 6 (full line) is compared with the experimental data of Reissinger (1985) in the

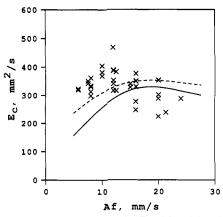


Figure 2. Experimental variation in E_c with Af for the system toluene (dispersed)/water (continuous) with $V_c = V_d = 2.78 \text{ mm/s}$ measured by Reissinger (1985) (indicated by the symbol ×) compared with that predicted by eq 6 (full line). The dashed line shows the variation with the values of k_1 and k_2 given in Table IV for dynamic tracer injection data with no mass transfer.

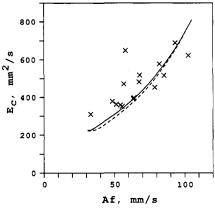


Figure 3. Experimental variation in E_c with Af for the system carbon tetrachloride (dispersed)/iodine (solute)/water (continuous) when iodine is transferred from the continuous to the dispersed phase with $V_c = 5.66$ mm/s and $V_d = 0.14$ mm/s measured by Barame (1972) (indicated by the symbol ×) compared with that predicted by eq 6 (full line). The dashed line shows the variation with the values of k_1 and k_2 listed in Table IV obtained by both steady and unsteady-state injection data during mass transfer.

mixer-settler and transition regions for the toluene (dispersed)/water (continuous) system with $V_c = V_d = 2.78$ mm/s obtained with the dynamic tracer injection technique in columns of 72- and 152-mm diameter. Figure 3 compares eq 6 (full line) with the data of Barame (1972) in the emulsion region for the carbon tetrachloride (dispersed)/water (continuous) system during mass transfer of iodine from the continuous to the dispersed phase with $V_c = 5.66 \text{ mm/s}$ and $V_d = 0.14 \text{ mm/s}$ obtained with the dynamic tracer injection technique in a column of 50-mm

Figure 1 also shows the variation in E_c with Af predicted by eq 6 (dashed line) with the particular values of k_1 and k_2 , determined with steady-state data with no mass transfer, given in Table IV. Figure 2 shows the variation predicted by eq 6 (dashed line) with the values of k_1 and k_2 listed in Table IV for dynamic tracer injection data with no mass transfer, and Figure 3 shows the variation predicted by eq 6 (dashed line) with the values of k_1 and k_2 listed in Table IV obtained by both steady- and unsteady-state injection data during mass transfer. It can be seen from Figures 1 and 3 that the full and dashed lines lie close together, indicating that eq 6 is not sensitive to the relevant values of k_1 and k_2 given in Table IV. Only in Figure 2 is there some divergence between the lines.

Table V. Values of the Standard Deviation, s, in Predicted Values of Ec from Experimental Points As Given by Various Correlations

	no mass transfer				$\frac{\text{mass transfer } c \rightarrow d}{\text{steady-state and}}$			
	steady-state tracer technique (no. of data pts = 361)		dynamic tracer technique (no. of data pts = 556)		dynamic tracer techniques (no. of data pts = 75)		all data	
correlation	NDAª	s, %	NDA	s, %	NDA	s, %	NDA	s, %
1. Mar (1958) and Mar and Babb (1959)	350	178.0	343	42.5	75	81.5	768	125.1
2. Sehmel (1961) and Sehmel and Babb (1964)	350	204.2	343	165.7	75	259.4	768	191.8
3. Miyauchi and Vermeulen (1963) and	361	168.7	556	58.8	75	82.2	992	113.1
Miyauchi and Oya (1965)								
4. Miao (1978)	361	115.5	556	56.9	75	58.2	992	83.1
5. Tung (1984) and Tung and Leucke (1986)	184	44.3	195	48.6	44	34.1	423	45.1
6. Reissinger (1985)	226	50.5	359	52.3	51	36.2	636	50.2
7. present work (eq 6)	361	33.2	556	35.4	75	25.8	992	33.8

^a NDA = number of data points for which correlation is applicable.

The data of Reissinger (1985) are somewhat unusual since they lie mainly in the mixer-settler region close to the value of $(Af)_{m}$, below the optimum conditions for good mass transfer as explained in the Introduction (Cohen and Beyer, 1953; Sege and Woodfield, 1954). It would appear that, in practice when mass transfer invariably occurs, the general form of eq 6 with the constants k_1 and k_2 determined from all the data points may be used to predict the values of E_c for design purposes (or for particular operating conditions in a given column).

The values of the continuous-phase axial mixing coefficient predicted by the proposed correlation (eq 6) are compared with those predicted by published formulae in Table V in terms of the standard deviation, s. Only the major correlations lited in Table I are included. It should be noted that the published correlations have only been applied within the ranges calculated by criteria (wherever given) used in the work of original authors. The calculations were performed by using the data summarized in Tables II and III of the supplementary material, and the following additional assumptions had to be made. Although some of the published equations are only based on the results obtained with either the steady-state or the dynamic tracer injection method, these are used to predict E_c for both types of data. Moreover, these equations are also used to calculate E_c when mass transfer occurs. The value of the plate thickness, t, required in the correlation of Mar (1958) and Mar and Babb (1959), when not quoted by the original authors, was taken as 1 mm. The term 1/N, which is usually small, was neglected in comparison with 2 or 2β in eq TI.3a, TI.3b, and TI.4a due to Miyauchi and Vermeulen (1963), Miyauchi and Oya (1965), and Miao (1978). The break point between eq TI.3a and TI.3b for the mixer-settler, transition, and emulsion regions of operation, respectively, was predicted with eq 2. For the case of mass transfer, the term $\sigma_{\rm top}/\sigma_{\rm bottom}$ in eq TI.6 suggested by Reissinger (1985) was taken as unity due to the lack of sufficient data. Table V shows that, out of the literature correlations, the equation of Tung (1984) and Tung and Luecke (1986) reproduces the data with a minimum s value of 45.1%. However, their equation is only valid in the emulsion region of operation. Similarly, the formula of Reissinger (1985) is also not applicable over the entire range of agitation. Since separate values of A and f are not available for all the data points, the correlations of Mar (1958) and Mar and Babb (1959) and Sehmel (1961) and Sehmel and Babb (1964) could not be applied to all the data points.

Conclusions

(1a) On the basis of published data with and without mass transfer for 28 liquid/liquid systems from 13 data sources, eq 6 has been determined for the prediction of continuous-phase axial mixing coefficients for design purposes (or under particular operating conditions in a given column). The value of the standard deviation in the predicted values of the axial mixing coefficient from the experimental points is 33.8%.

(1b) The continuous-phase density was not included in the correlation since it did not vary appreciably in the experiments. Although the continuous-phase superficial velocity and column diameter varied over wide ranges, their effects were found to be insignificant.

(2) The axial mixing increases with agitation in the mixer-settler region until the onset of transition region. Then it decreases until a minimum at $Af = 1.67(Af)_{m}$ is reached, beyond which it once again increases with agitation.

(3) The value of the axial mixing coefficient determined with the dynamic tracer injection method is larger than that measured with the steady injection of a tracer. Since the former method measures all the deviations from plug flow (such as circulation, channeling and dead zones) in addition to turbulent diffusion, it is more likely to give a better estimate of the axial mixing coefficient for design

(4) Additional data under both directions of mass transfer are required to further investigate the effect of mass transfer upon axial mixing.

Nomenclature

A = length of pulsation stroke (twice the wave amplitude),

D = column diameter, m

 d_o = diameter of holes in perforated plate, m

 $E_{\rm c}$ = continuous-phase axial mixing coefficient, m²/s

 e_c = continuous-phase superficial axial mixing coefficient, m²/s f = pulse frequency, Hz

h = center-to-center plate spacing, m

 $h_* = \text{standard plate spacing} = 0.05 \text{ m}$

 k_1, k_2 = constants, see eq 6, dimensionless

M = number of data points, dimensionless

m = number of parameters, dimensionless

N = number of stages in column, dimensionless

 p_1-p_7 = indices, dimensionless

s = standard deviation defined by eq 7, dimensionless

t =plate thickness, m

 $V_{\rm c}$ = continuous-phase superficial velocity, m/s

 $V_{\rm d}$ = dispersed-phase superficial velocity, m/s

 β = number of perfectly mixed stages in series between each pair of plates, see eq TI.3c and TI.4b, dimensionless

 $\Delta \rho$ = density difference between phases, kg/m³

 ϵ = volume fraction holdup of dispersed phase, dimensionless

 μ_c = continuous-phase viscosity, Pa s

 $\mu_{\rm d}$ = dispersed-phase viscosity, Pa s

- ρ_c = continuous-phase density, kg/m³ $\rho_{\rm d}$ = dispersed-phase density, kg/m³
- ρ_* = density of water at 20 °C = 998 kg/m³
- σ = interfacial tension, N/m
- ϕ = fractional free area of perforated plate, dimensionless
- ψ = agitation group, see eq 3 and 4, dimensionless

Subscripts

bottom = value at bottom of column m = value corresponding to minimum in holdup curve

top = value at top of column

Supplementary Material Available: Table II listing the physical properties of 28 liquid/liquid systems taken from 13 different data sources and Table III listing the ranges of axial mixing coefficients, operating variables, and column dimensions (7 pages). Ordering information is given on any current masthead page.

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