Back Mixing in a Reciprocating Plate Column with Stable Density Gradients

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Axial dispersion coefficients were measured in an aqueous phase flowing downward through a 5.08-cm-dia. Karr-type reciprocating plate column. The steady tracer injection method was used, in which the concentration profile was measured upstream of tracer injection. The tracer solution was a strong (150–170 g/L) solution of sodium chloride, whereby a significant stable density gradient (density decreasing with height) was created in the upstream section. The operating variables included frequency (0 to 7.1 Hz), amplitude (0.7 to 1.4 cm), and plate spacing (2.65 to 7.65 cm). Some data were also obtained under two-phase conditions with a refined kerosene (Isopar M) dispersed in the aqueous phase as droplets. Some control experiments were done using a neutrally buoyant tracer solution containing sodium chloride and methanol. There was no evidence that the stable density gradient had any effect in reducing the values of the axial dispersion coefficient, although earlier published work had shown that an unstable density gradient did increase axial mixing.

Introduction

Axial dispersion plays an important role in the design and operation of extraction columns. Its effect in the continuous liquid phase has been studied extensively and reviewed by Pratt and Baird (1983), among others. Dispersion in differential columns such as packed or open-structure plate columns is characterized by the continuous-phase axial Peclet number ($Pe = u_c L/E$). If this is less than about 0.05, the continuous phase can be regarded as "well-mixed" and the equivalent number of transfer units in countercurrent flow is greatly reduced. It is desirable that the continuous phase should move in "plug flow" ($Pe > 20$) or as close as possible to that condition. Since the active height of the column, L, and the continuous-phase superficial velocity, u_c, are usually fixed, it is desirable that the axial dispersion coefficient, E, should be reduced to as low a value as possible.

The value of E, typically in the order of 1 to 10 cm^2/s, is determined by the flow rates of each phase; the design of the column, including internals; the level of mechanical agitation; and the properties of the liquid–liquid system such as interfacial tension and density difference. The physical mechanisms by which these variables affect E include turbulence, circulation, and the wake effects of liquid drops. Because of the complexity of these mechanisms operating together, there is no generally reliable equation to predict axial dispersion. There has been much research in the area, however, and there are many equations for E that are specific to particular types and sizes of equipment (Godfrey and Slater, 1994).

Recently Holmes et al. (1991) identified a new contributing factor to axial mixing, namely the "unstable density gradient." This occurs when the continuous-phase density increases with height in the column, as, for example, in the case of a heavy metal solute being extracted from an aqueous continuous phase moving down the column, or a light solute being extracted from an organic continuous phase moving up the column. Several studies (see Table 1) have confirmed that extremely small unstable density gradients can significantly increase axial dispersion coefficients. While it may be seen from Table 1 that this effect has only been studied in reciprocating plate columns and open columns (with no internals), there is no reason to suppose that similar effects do not occur in other types of extraction columns.

The effects of unstable density gradients have been modeled on the basis of a turbulent mechanism involving the mixing length, as originally proposed by Kolmogoroff (1941) and extensively used by subsequent workers:

$$E = l^{4/3}\epsilon_t^{1/3}. \tag{1}$$

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Table 1. Previous Work on Effects of Unstable Density Gradient on Axial Mixing in the Continuous Phase

Reference	Type of Column and d_c	Conditions
Holmes et al. (1991)	Karr column, 7.62 cm	Single phase. $CaCl_2$/water system used to maintain density gradient
Baird and Rama Rao (1991)	Karr column, 5.08 cm	Single phase. NaCl/water and hot/cold water used for density gradient
Baird et al. (1992)	Open column, 1.48, 1.91 and 2.63 cm	Single phase. NaCl/water system, unsteady pulse injection
Aravamudan and Baird (1996)	Karr column, 5.08 cm	Single- and two-liquid phases. NaCl/water used to maintain density gradient

The total specific energy dissipation rate, ϵ_t, expressed in W/kg, is taken as the sum of terms due to mechanical agitation, the agitation due to the dispersed phase drops, and a third term for energy dissipation due to the unstable density gradient in the continuous phase:

$$\epsilon_t = \epsilon_m + \epsilon_d + \epsilon_b. \tag{2}$$

The term ϵ_b is much smaller than the other two terms on the right of Eq. 2 and usually contributes relatively little to ϵ_t. However, it can have a significant effect on the mixing length in Eq. 1, which is expressed as an energy-weighted sum of mixing lengths:

$$l = l_m + (l_d - l_m)(\epsilon_d/\epsilon_t)_d^n + (l_b - l_m)(\epsilon_b/\epsilon_t)_b^n. \tag{3}$$

Typically in the Karr column, the mechanical mixing length l_m is in the order of 3 mm, while the density gradient mixing length l_b was about 35 mm in the work of Aravamudan and Baird (1996). These authors found that a small value of ϵ_b had a significant effect on l, and therefore upon E, and they fitted their data to a modified form of Eq. 3 by means of parameter adjustment.

As a result of the previous studies, which are summarized in Table 1, it has been recommended that designers of extraction columns should avoid conditions where an unstable density gradient can be set up, in order to prevent increases in axial dispersion. It was also recommended to study the effects of a *stable* density gradient, with which the continuous-phase density decreases with height, to find out whether this would lead to a reduction in axial dispersion. The present article addresses that objective.

Experimental Studies

Axial dispersion was measured in a 5.08-cm (2-in.) nominal diameter pilot scale Karr reciprocating plate extraction column, as shown in Figure 1. This type of column, which is widely used in the industry (Godfrey and Slater, 1994), essentially consists of a vertical cylindrical shell, in which an assembly of perforated plates is reciprocated vertically to provide controlled agitation of the liquid–liquid dispersion. The principal dimensions and operating conditions of the present column are summarized in Table 2.

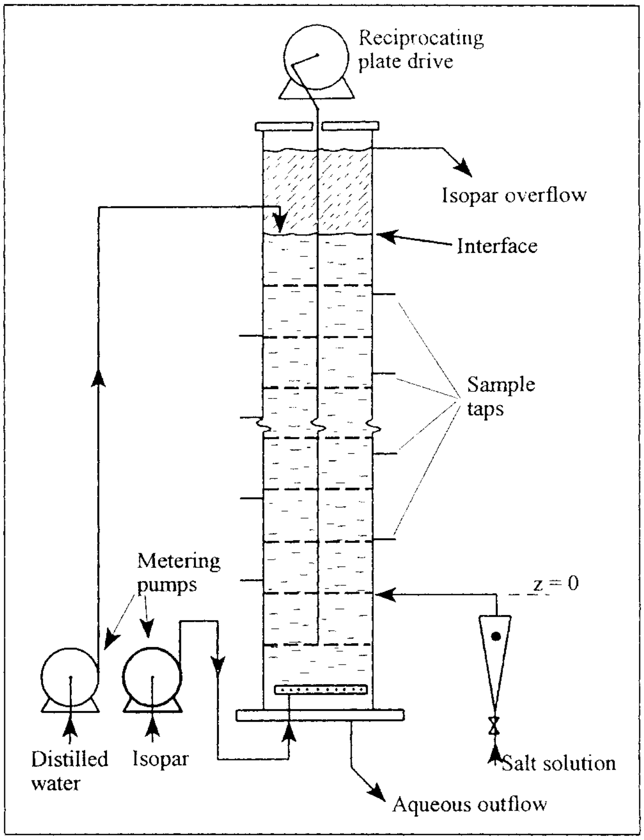

Figure 1. The apparatus.

For dimensions, see Table 2.

Although the column is the same one that was used in earlier work with unstable density gradients (Baird and Rama Rao, 1991; Aravamudan and Baird, 1996), the flow arrange-

Table 2. Experimental Conditions

Column and plate dimensions			
Column ID	5.08 cm		
Column overall hgt.	2.3 m		
Plate stack hgt.	1.4 m		
Plate perforation dia.	12.7 mm		
Plate fraction open area	0.57		
Plate spacings used	2.65 cm	5.15 cm	7.65 cm
No. of plates in stack	53	28	17
Salt injection point	24 cm above the bottom of the plate stack		
Sample points*	8, 15.5, 23, 33.5, 51, 68, 87.5, 105 cm above salt injection point		
Operating conditions			
Amplitude of reciprocation	0.7 to 1.4 cm (center to peak)		
Freq. of reciprocation	0 to 7.1 Hz		
Superficial vel. of continuous phase (water)	0.695 cm/s		
Superficial vel. of dispersed phase (Isopar M)	0 to 0.61 cm/s		
System properties (24°C)			
	Dens., kg/m^3	Visc., Pa·s	
Distilled water	997	1.0	
Isopar M	785	2.4	
See Eq. 4 for salt solution dens.			

*Located on alternating sides of the column.

ment has been changed in this work to provide stable density gradients for back mixing. Distilled water enters the top of the column as the main continuous phase, while a stream of concentrated sodium chloride (typically 150–170 g/L) is continuously added at the side wall of the column near the base of the plate stack and mixes with the distilled water. In most of the experimental work, the flow of distilled water was 14 mL/s and the flow of concentrated sodium chloride solution was 10 mL/s, resulting in a mixed exit solution with salt concentration of typically about 65 g/L. The concentrated sodium chloride entered from the side wall through an inset tube with internal diameter about 4 mm, and the jet moving at about 80 cm/s had enough lateral momentum to ensure good radial mixing at the injection point.

The density of aqueous sodium chloride is a linear function of concentration:

$$\rho = \rho_0 + 0.70c. \qquad (4)$$

Therefore the mixed solution density below the sodium chloride injection point is 1,043 kg/m^3, but at higher points (upstream of salt injection) the density decays to that of the distilled water, 997 kg/m^3.

A few additional tests were carried out in which the sodium chloride solution feed contained sufficient methanol to bring its density to the same value as the distilled water and thereby eliminate axial density gradients. Some tests were also carried out under two-phase conditions using Isopar M, a purified aliphatic kerosene manufactured by Imperial Oil of Canada (Sarnia, Ont.). Its properties are given in Table 2.

In each experiment, the column was operated for long enough (2–3 residence times) for steady state to be reached. Then, 20-mL samples of the aqueous phase were withdrawn at 5-min intervals from various sample points located alternatively on either side of the column at axial positions indicated in Table 2. The samples were analyzed using a precalibrated conductivity meter. The order of taking the samples was varied randomly to ensure that no bias was created due to a fixed sampling sequence.

Back-mixing concentration profiles

Steady-state unidirectional back mixing in the continuous phase is represented by

$$u_c c - E\,dc/dz = 0. \qquad (5)$$

In previous work with unstable density gradients (Table 1), it could not be assumed that E was constant, and it had to be evaluated as

$$E = u_c c/(dc/dz). \qquad (6)$$

The local slope of the concentration profile was difficult to obtain manually on the basis of only a few data points. This led Aravamudan and Baird (1996) to fit their profiles to polynomial equations, which could then be differentiated to obtain the slope and hence E at any point.

In the special case where E is constant, Eq. 5 can be solved to give the well-known expression:

$$c = c_0 \exp(u_c z/E). \qquad (7)$$

The distance z is measured in the direction of bulk flow, relative to $z = 0$ at the tracer injection point. Back mixing occurs only in the upstream region in which $z < 0$. It follows that

$$\ln(c) = \ln(c_0) + u_c z/E. \qquad (8)$$

Therefore, a semilogarithmic plot of tracer concentration vs. upstream distance $(-z)$ will be linear if E is constant, and the value of E is obtainable from the slope of the plot. This method of measuring E was originally reported by Mar and Babb (1959), and has been used by many subsequent workers. This approach could not be used in cases where there was an unstable density gradient (for example, Holmes et al., 1991), that is, where the density increased with height.

Some typical data from this work are shown in Figure 2, plotted semilogarithmically. The profiles become progressively steeper as frequency is reduced; a test at zero frequency (not shown in Figure 2) indicated that no perceptible back mixing occurred. It can also be seen that, for a given frequency, the plots are linear for concentrations greater than about 0.05 g/L, which was the lower limit of accuracy of the conductivity meter. The linearity confirms Eq. 8 which is based on the assumption that E is independent of salt concentration. The linearity of the plots in Figure 2 also confirms that the sodium chloride is well-mixed laterally; if there was poor lateral mixing, there would have been fluctuations due to the locations of the sample ports at alternate sides of the column (Figure 1).

The "well-mixed" exit concentration (c_0) in these particular tests was 66.7 g/L, and therefore ideally all the lines in Figure 2 should converge at that value when they are extrapolated to $z = 0$. That does not appear to be exactly the case. It is likely that the frequency of reciprocation had a slight effect on the local cross-mixing of the added salt at $z = 0$. Therefore, analysis is based on the measured tracer concen-

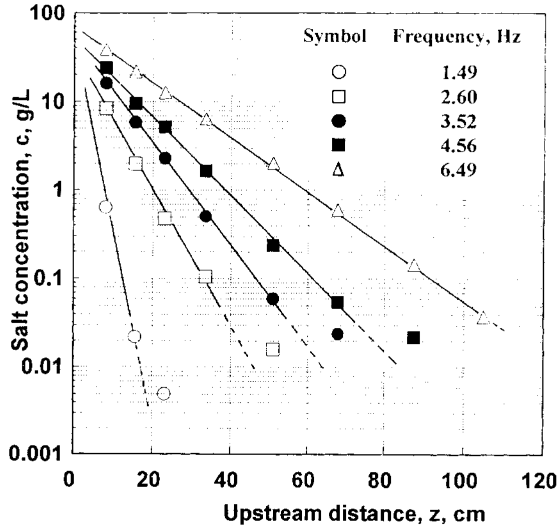

Figure 2. Semilogarithmic plots of sodium chloride concentration vs. upstream distance at various reciprocation frequencies.

$a = 1.05$ cm; $u_c = 0.695$ cm/s; $u_d = 0$ cm/s; $H = 2.65$ cm.

tration c_8 at the first sample point ($z = -8$ cm). It can be shown from Eq. 8 that

$$E = u_c(z + 8)/\ln(c/c_8), \tag{9}$$

where z is the negatively signed upstream distance. Values of E calculated in this way from different parts of a given profile were found to be essentially constant and therefore independent of the salt concentration gradients.

Axial dispersion coefficients in single-phase conditions

Baird and Rama Rao (1988) measured axial dispersion coefficients in this column earlier, using the pulse-response method. Their results under single-phase conditions were represented approximately as

$$E/af \approx 1.15 \text{ cm}. \tag{10}$$

This relationship held at different amplitudes and frequencies, and at a plate spacing of 2.7 cm, but at higher plate spacings E was reduced.

The present data for single-phase conditions are shown on Figure 3 with E plotted against af. The present data, including some results with a neutrally buoyant tracer mixture of methanol and brine, are correlated by the dashed straight line shown as

$$E/af = 1.21 \text{ cm}, \tag{11}$$

which is reasonably close to the previous correlation (Baird and Rama Rao, 1988).

It should be noted that in the present work the correlation holds for all three plate spacings studied: 2.65, 5.15 and 7.65 cm. For example at $af = 4$ cm/s and $H = 5.15$ cm, E is found to be 4.8 cm^2/s in this work. However, Baird and Rama Rao (1988) reported $E = 3.5$ cm^2/s at the same conditions. This is not easy to explain because a stable density gradient might be expected to reduce E or have no effect, but it would not be expected to increase E.

Axial dispersion coefficients in two-phase conditions

The present data are plotted in Figure 4 on the same coordinates as in Figure 3, with Eq. 11 being shown for reference as a dashed line. It can be seen that at plate spacings of 2.65 cm, the results are not greatly different from those for single-phase flow. At the higher plate spacing of 5.15 cm, however, the two-phase data (denoted by filled triangles) are significantly reduced. This effect of plate spacing was observed in single-phase flow by Baird and Rama Rao (1988), but not in the present work.

If the reduction in E at higher plate spacings was associated with the stable density gradient, it would be expected that the reduction would be greatest near the salt injection point where the density gradient is greatest; this would then result in nonlinear plots of the concentration profile on semilogarithmic coordinates. Such plots were found to be essentially linear, however, as illustrated in Figure 5. Therefore, it cannot be concluded that the reduced values of E at $H = 5.15$ cm were due to the stable density gradient.

Discussion

Although a few anomalies have been observed, it can be generally concluded that there is no significant effect of a stable density gradient in reducing the axial dispersion coefficients. This is in contrast to the previously established effect of an unstable gradient in increasing the values of E, as discussed earlier in this article.

In order to explain this, it is necessary to review the reasoning behind Eqs. 1 to 3, which represent the enhancement of axial mixing by an unstable density gradient. In the recip-

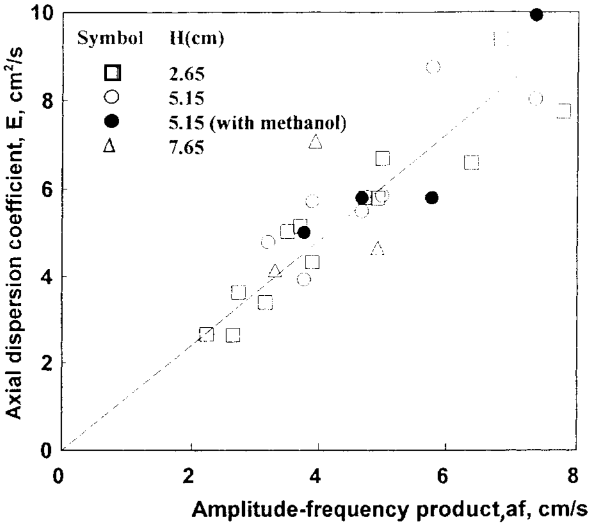

Figure 3. Axial dispersion coefficients in single-phase flow $u_c = 0.695$ cm/s.

--- Line denoting $E/af = 1.21$ cm.

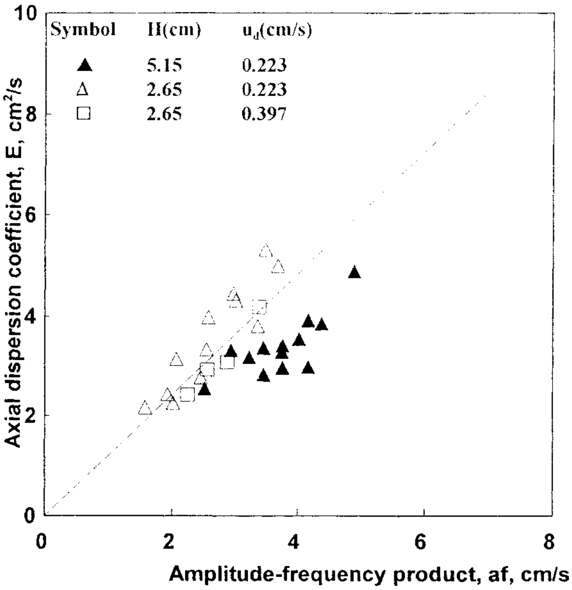

Figure 4. Axial dispersion coefficients in liquid–liquid flow, $u_c = 0.695$ cm/s.

--- Line denoting $E/af = 1.21$ cm.

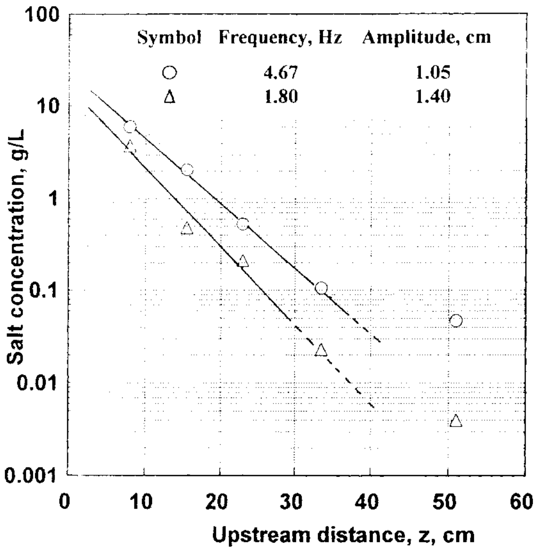

Figure 5. Semilogarithmic plots of sodium chloride concentration vs. upstream distance for two-phase flow.

$u_c = 0.695$ cm/s; $u_d = 0.223$ cm/s; $H = 5.15$ cm.

rocating column, agitation is normally provided from two sources: mechanical agitation of the plates, and the buoyancy-driven action of the droplets of the dispersed phase. The former mode of agitation approximates to isotropic turbulence with a primary scale of about 3 mm corresponding to the strips of metal (webs) between the perforations on the plate. The effect of the rising drops is more complex, but the model equations simplify it as a larger-scale turbulence. The same argument is applied in regard to an unstable density gradient in the continuous phase. Although the energy input from such a gradient is very small, the primary eddy scale is in the same order as the column diameter. This scale, superimposed on the smaller eddy scales due to mechanical and droplet effects, can significantly increase axial dispersion, as has been shown.

The preceding argument cannot be applied in reverse to the case of a stable density gradient. In this case no extra energy is contributed to the system by the density gradient and there is no analogy to the "eddy scale" l_b induced by the unstable density gradient. Rather, there is a resistance to the transmission of the eddies due to mechanical or droplet agitation. Mixing studies with stratified liquids in large tanks (Ahmad et al., 1985) have indeed shown that mixing rates are reduced by density stratification. In the present work, however, the scale across which mixing is occurring is small (plate to plate) and the specific mechanical energy input ϵ_m is relatively large, on the order of 1 W/kg. It appears that any retardation of axial dispersion due to the stable density gradient is too small to be measurable in this work.

Conclusions and Suggested Further Work

It is concluded that axial mixing in a reciprocating plate column, in the presence of a *stable* density gradient in the continuous phase, is not significantly different from axial mixing in the absence of any density gradient. This does not alter the conclusion from earlier work that *unstable* density gradients should be avoided in Karr extraction columns, as they can lead to increased axial mixing. Until now, all the experimental work on density gradient effects on mixing has been done either in open columns or in Karr reciprocating-plate columns. It is likely that such effects may also exist in other types of commercially used extraction columns, and further work in this area is recommended.

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Notation

a = reciprocation amplitude (center to peak), m
c = salt concentration, kg/m^3
d_c = internal diameter of column, m
f = reciprocation frequency, Hz
H = plate spacing, m
n = exponent in mixing-length equation
ρ_0 = density of pure water, kg/m^3

Subscripts

b = associated with unstable density gradient
d = dispersed phase

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