$$\begin{aligned}T_{m2}(\tau) = \sum_{k=0}^{\infty}\sum_{n=0}^{\infty} C_n\{&u_kC_m[u_nC_kG(\lambda_m^2,\lambda_m^2 + \lambda_k^2,\lambda_m^2 + \lambda_k^2 \\ &+ \lambda_n^2,\tau) - U_{kn}G(\lambda_m^2,\lambda_m^2 + \lambda_k^2,\lambda_m^2 + \lambda_n^2,\tau)] \\ &+ u_kC_k[u_nC_mG(\lambda_k^2,\lambda_m^2,\lambda_m^2 + \lambda_n^2,\tau) \\ &- U_{mn}G(\lambda_k^2,\lambda_m^2,\lambda_m^2 + \lambda_n^2,\tau)] \\ &- U_{km}[u_nC_kG(\lambda_m^2,\lambda_k^2,\lambda_k^2 + \lambda_n^2,\tau) - U_{kn}G(\lambda_m^2,\lambda_k^2,\lambda_n^2,\tau)]\} \\ &+ \sum_{n=0}^{\infty} C_n[D_{nm}F(\lambda_k^2,\lambda_m^2,\tau) - d_nC_mF(\lambda_m^2 + \lambda_k^2,\lambda_k^2,\tau)]\end{aligned}$$

where

$$d_n = \langle \boldsymbol{X}_n^T \boldsymbol{W}\boldsymbol{P}_\zeta^{-1}\boldsymbol{X}_0\rangle$$
$$D_{nm} = \langle \boldsymbol{X}_n^T \boldsymbol{W}\boldsymbol{P}_\zeta^{-1}\boldsymbol{X}_m\rangle$$

and

$$G(a,b,c,\tau) = \frac{1}{(c-b)}\left[\frac{e^{-a\tau} - e^{-b\tau}}{b-a} - \frac{e^{-a\tau} - e^{-c\tau}}{c-a}\right]$$

which has the following limiting forms when two or more of the parameters a, b and c are equal:

(i) $a = b, a \neq c$

$$G(a,a,c,\tau) = \frac{1}{(c-a)}\left[\tau e^{-a\tau} - \frac{e^{-a\tau} - e^{-c\tau}}{c-a}\right]$$

(ii) $a \neq b, a = c$

$$G(a,b,a,\tau) = \frac{1}{(a-b)}\left[\frac{e^{-a\tau} - e^{-b\tau}}{b-a} - \tau e^{-a\tau}\right]$$

(iii) $b = c, b \neq a$

$$G(a,b,b,\tau) = \frac{1}{(b-a)}\left[\frac{e^{-a\tau} - e^{-b\tau}}{b-a} - \tau e^{-b\tau}\right]$$

(iv) $a = b, a = c$

$$G(a,a,a,\tau) = \tau e^{-a\tau}$$

Dispersion, Mass Transfer and Chemical Reaction in Multiphase Contactors

Part II: Numerical Examples

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A numerical study, based on the theoretical developments of Part I of this series, has demonstrated that the chemical activity of a solute can have a significant effect on the convective dispersion behavior of multiphase reactors, and that the dispersion parameters estimated using inert tracers for the individual phases may not be representative of the parameters that should be used in the design of reactive systems. Moreover, the use of different transverse averaging procedures for the individual phases can give rise to additional interaction effects between the phases which are not accounted for in the traditional axial dispersion models. The predicted differences in dispersion parameter values depending on whether the solute is inert or not, are in qualitative agreement with published experimental results.

SCOPE

Procedures for the rational design of multiphase contactors should allow for the reduction in mass transfer efficiency which invariably accompanies axial dispersion of the phases. However, a unique characterization of the dispersion processes for any given hydrodynamic situation using the axial dispersion model does not appear possible, since it has been shown that the rate of interphase mass transfer can modify the effective dispersion processes to a significant degree (Hatton and Lightfoot, 1982). In Part I, an extension of the generalized dispersion theory of Gill and Sankarasubramanian (1970, 1971) to multiphase, reactive problems suggested that further modifications to the convective dispersion processes can be anticipated if the solute is chemically active. Moreover, the theoretical results indicate that the use of arbitrary transverse-averaging procedures for the individual phases may give rise to off-diagonal terms in the dispersion parameter matrices, thereby introducing additional interaction effects between the phases not normally accounted for in the axial dispersion model. The purpose of this paper is to report on a numerical study of these effects, for selected parameter values, and thus to complete the study initiated in Part I.

CONCLUSIONS AND SIGNIFICANCE

The numerical results presented here confirm that the chemical activity of a solute within a given phase can have a significant effect on the dispersion characteristics of multiphase contactors, and suggest that more attention should be paid to large-scale channelling effects in the design of these contactors. This is anticipated to be particularly important for reactors operating under nonisothermal conditions, or for systems ex-

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hibiting nonlinear reaction kinetics.

In any analysis of the dispersion behavior of mass transfer systems, note should be taken of the effects that different transverse concentration averaging procedures can have on the values estimated for the dispersion parameters. It has been demonstrated that different radial averaging of the concentrations in the individual phases can cause an unusual coupling of the species balance equations, in which the off-diagonal terms of the dispersion coefficient matrix now take on finite values, comparable in magnitude to the diagonal entries.

INTRODUCTION

The extensive use of multiphase, column-type contactors in the process industries attests to their effectiveness as mass transfer devices, both for separations processes and for reaction engineering operations. However, the performance of these contactors can be particularly susceptible to the adverse effects of axial dispersion, and this has prompted a large number of studies, based on the axial dispersion model, of such equipment in an effort to characterize their behavior more effectively. There is both experimental (Hatton and Woodburn, 1978; Linek et al., 1978) and theoretical (Hatton and Lightfoot, 1982) evidence that the dispersion characteristics of the individual phases can be affected significantly by the presence of interphase mass transfer, which suggests that the use of dispersion parameters estimated from intraphase or residence time distribution measurements may lead to unacceptable errors in the design of mass transfer equipment. In the above works, the question as to whether the chemical activity of the transferable solute could have a further influence on the dispersion properties of multiphase contactors was ignored. This situation has since been redressed, and a theoretical analysis of the convective dispersion behavior of reactive, multiphase processes is now available (Hatton and Lightfoot, 1984). The purpose of this paper is to present some specific numerical examples examining the predicted effects of not only chemical reactions, but also of arbitrary transverse concentration averaging policies on the dispersion characteristics of multiphase contactors.

Various aspects of convective dispersion phenomena in two-phase, mass transfer operations were discussed in some detail in our earlier communication (Hatton and Lightfoot, 1982). It was shown, for instance, that the transient period required for the development of an asymptotic state, characterized by constant dispersion parameter values, may be comparable to the phase residence times, and under such conditions the transient behavior of these parameters should not be ignored. This is, however, a factor not normally accounted for in the analysis of experimental response curves, and one that can lead to wide variations in the estimated values for the dispersion coefficients depending on the distance between detection points in the experimental set-up. Indeed, this poses the question as to what parameters are actually determined in dispersion experiments.

The generalized convective dispersion equation is of the form

$$\epsilon \frac{\partial \boldsymbol{c}_m}{\partial \tau} + \boldsymbol{V}(\tau) \frac{\partial \boldsymbol{c}_m}{\partial \zeta} - \boldsymbol{P}^{-1}(\tau) \frac{\partial^2 \boldsymbol{c}_m}{\partial \zeta^2} - \boldsymbol{R}(\tau) \boldsymbol{c}_m = \boldsymbol{0} \tag{1}$$

which, when subjected to a Fourier transform, is

$$\epsilon \frac{d \boldsymbol{c}_m^*}{d\tau} = [-i\lambda \boldsymbol{V}(\tau) - \lambda^2 \boldsymbol{P}^{-1}(\tau) + \boldsymbol{R}(\tau)] \boldsymbol{c}_m^* \tag{2}$$

where

$$\boldsymbol{c}_m^* = \int_{-\infty}^{\infty} \boldsymbol{c}_m e^{i\lambda\zeta} d\zeta \tag{3}$$

Equation 2 has the solution

$$\boldsymbol{c}_m^* = \exp\left[-i\lambda\epsilon^{-1} \int_0^\tau \boldsymbol{V}(\tau)d\tau - \lambda^2\epsilon^{-1} \int_0^\tau \boldsymbol{P}^{-1}(\tau)d\tau + \epsilon^{-1} \int_0^\tau \boldsymbol{R}(\tau)d\tau\right] \boldsymbol{c}_{m0}^* \tag{4}$$

For the constant coefficient model, the corresponding solution is

$$\boldsymbol{c}_m^* = \exp[\epsilon^{-1}(-i\lambda \boldsymbol{V}_c \tau - \lambda^2 \boldsymbol{P}_c^{-1} \tau + \boldsymbol{R}_c \tau)] \boldsymbol{c}_{m0}^* \tag{5}$$

where $\boldsymbol{V}_c$, $\boldsymbol{P}_c$ and $\boldsymbol{R}_c$ are the parameters normally determined in tracer studies, and are generally treated as constants in the model formulation. Now, if the pulse peak reaches the detection station at a time T following its injection, clearly the parameters that are measured must be

$$\boldsymbol{V}_c = \frac{1}{T} \int_0^T \boldsymbol{V}(\tau)d\tau \tag{6}$$

$$\boldsymbol{P}_c^{-1} = \frac{1}{T} \int_0^T \boldsymbol{P}^{-1}(\tau)d\tau \tag{7}$$

and

$$\boldsymbol{R}_c = \frac{1}{T} \int_0^T \boldsymbol{R}(\tau)d\tau \tag{8}$$

respectively, which shows, very reasonably, that the measured coefficients are the time-averaged quantities. It must be remarked, however, that these relationships are approximate only, since they ignore the fact that the actual measurements are made over a time interval around T, during which time, of course, the parameters are varying. However, if the width of the residence time distribution curve is much smaller than the mean residence time, the approximations obtained will be valid. For large values of T, the parameters $\boldsymbol{V}_c$, $\boldsymbol{P}_c^{-1}$ and $\boldsymbol{R}_c$ will approach the asymptotic values obtained for $\boldsymbol{V}$, $\boldsymbol{P}^{-1}$ and $\boldsymbol{R}$ respectively, and under such conditions, use of a constant coefficient model can be justified. These coefficients will, however, be sensitive to their mode of detection, and to the prevailing mass transfer conditions in the system, thus diminishing the utility of the axial dispersion model as it is usually formulated for the design of countercurrent contactors.

Other factors affecting the development of the overall dispersion characteristics of multiphase contactors, such as lateral dispersion rates in the individual phases and interphase mass transfer rates, need not be considered here, since they have been dealt with adequately in our earlier paper. Indeed, in what follows, we restrict our attention only to the effect of chemical reaction, and of more flexible transverse-averaging procedures, on the parameters appearing in the axial dispersion model, little discussion being devoted to the transient qualities of these parameters.

MASS TRANSFER WITH CHEMICAL REACTION

To illustrate the effect of a chemical reaction on the convective dispersion parameters $\boldsymbol{P}^{-1}$ and $\boldsymbol{V}$, we consider the physical absorption of a soluble gas by a liquid, with a subsequent conversion of the solute to reaction products. The reactions may be either reversible or irreversible.

Irreversible Reactions

In the case of an irreversible reaction, the situation corresponds to the following "reaction" scheme

$$C_y \underset{K_y a}{\overset{K_y a}{\rightleftarrows}} mC_x \xrightarrow{k'} \text{Products}$$

where $K_y a$ is the volumetric overall mass transfer coefficient, k' is the reaction rate constant, m is the equilibrium constant ($C_y^* = mC_x$), and C_y and C_x are the solute concentrations in the gas and liquid phases, respectively. The parameter values selected for the purposes of this study are shown in Table 1, and are discussed at greater length elsewhere (Hatton and Lightfoot, 1982). In the

TABLE 1. PARAMETER VALUES USED IN NUMERICAL STUDIES

Irreversible Reactions

$$V(\eta) = \begin{pmatrix} 2(1-\eta^2) & 0 \\ 0 & -2\eta^2 \end{pmatrix}; \quad P_\eta = \begin{pmatrix} 100 & 0 \\ 0 & 50 \end{pmatrix}$$

$$R = \begin{pmatrix} -1 & 1 \\ 1 & -(1+k) \end{pmatrix}; \quad \epsilon = \begin{pmatrix} 0.6 & 0 \\ 0 & 0.15 \end{pmatrix}$$

Reversible Reactions

$$V(\eta) = \begin{pmatrix} 2(1-\eta^2) & 0 & 0 \\ 0 & -2\eta^2 & 0 \\ 0 & 0 & -2\eta^2 \end{pmatrix}; \quad P_\eta = \begin{pmatrix} 100 & 0 & 0 \\ 0 & 50 & 0 \\ 0 & 0 & 50 \end{pmatrix}$$

$$R = \begin{pmatrix} -1 & 1 & 0 \\ 1 & -2 & k \\ 0 & 1 & -k \end{pmatrix}; \quad \epsilon = \begin{pmatrix} 0.6 & 0 & 0 \\ 0 & 0.15 & 0 \\ 0 & 0 & 0.15 \end{pmatrix}$$

Transverse Averaging

$$V(\eta) = \begin{pmatrix} 2(1-\eta^2) & 0 \\ 0 & -2\eta^2 \end{pmatrix}; \quad P_\eta = \begin{pmatrix} 100 & 0 \\ 0 & 50 \end{pmatrix}$$

$$R = \begin{pmatrix} -1 & 1.2 \\ 1 & -1.2 \end{pmatrix}; \quad \epsilon = \begin{pmatrix} 0.6 & 0 \\ 0 & 0.15 \end{pmatrix}$$

$$\omega(\eta) = \begin{pmatrix} 2(1-\eta^2) & 0 \\ 0 & 2\eta^2 \end{pmatrix}$$

calculations that follow, only $k = k'/K_y a$ is permitted to vary; the other model parameters are held constant.

The transient evolution of the convective dispersion coefficients for both phases, and for an irreversible reaction, is shown in Figure 1, whereas the behavior of the solute velocities with time is illustrated in Figure 2. The parameter on these curves is k, the reaction rate constant, and, for comparison purposes, the results pertaining to the intraphase studies are shown as broken lines.

It is clear that the presence of the reaction term can have a major influence on the development of the dispersion processes in both phases. Of particular interest here is that when the reaction is in the x-phase, an increase in k reduces the effect of interphase mass transport on the dispersion properties of the y-phase. In fact, for sufficiently large k (greater than about 5, say), the y-phase dispersion behavior is indistinguishable from that shown for the intraphase (no mass transfer) case. On reflection, such behavior is not unexpected, since the effect of the reaction is to remove the solute as it is absorbed, and hence to reduce the solute back pressure. Consequently, the amount of x-phase information transmitted to the y-phase is reduced, thereby diminishing the importance of the mass transfer interactions in determining the y-phase characteristics. At the high reaction rates, where the solute is depleted rapidly by the chemical reaction, there can be essentially no reverse mass transfer, and the absorption process can be described simply as a first-order, irreversible removal of the solute from the y-phase, i.e., the presence of the second phase can be ignored. As shown by Subramanian et al. (1974), a homogeneous, irreversible chemical reaction has no effect whatsoever on the dispersion characteristics of the reactive solute, and hence, for rapid reactions, the intraphase results for the y-phase should be recovered, as indeed they are.

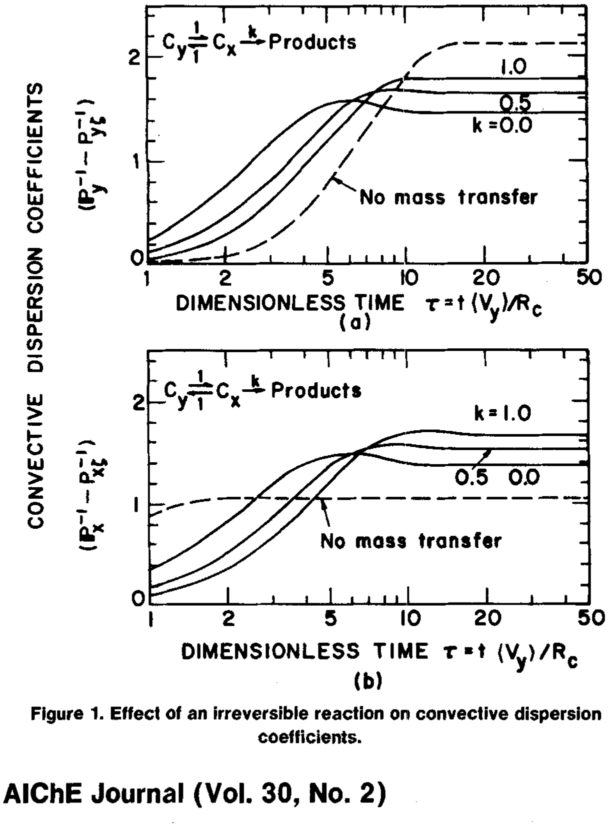

Figure 1. Effect of an irreversible reaction on convective dispersion coefficients.

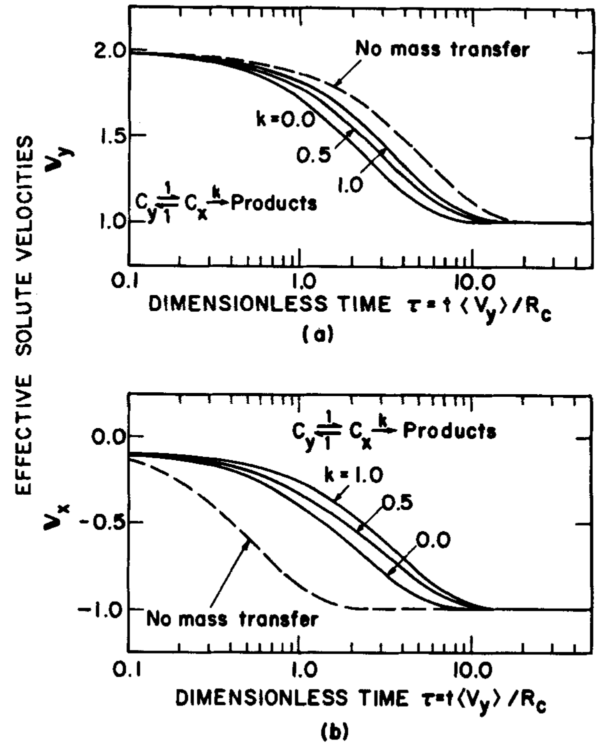

Figure 2. Effect of an irreversible reaction on effective solute velocities.

The effect of the reaction term on the x-phase dispersion characteristics is entirely different. Here, the differences in the dispersion behavior observed when there is interphase mass transfer, relative to that which results in the absence of such mass transfer, are accentuated as k is increased. The transverse concentration profiles in the x-phase will, for large k, be of similar form to those in the y-phase. This is because removal of the solute by reaction is sufficiently rapid to ensure that the effect of the x-phase lateral dispersion processes in smoothing out the concentration profiles is negligible. (The reaction time constants are considerably smaller than the lateral diffusion characteristic times.) Under these conditions, the x-phase can be expected to reflect the dispersion characteristics of the y-phase, as it is seen to do.

Reversible Reactions

Reversible reactions are also common in industrial multiphase processes, and we consider now the effect of introducing a reversibility into the reaction scheme discussed above. For the purposes of this example, we assume the forward reaction to proceed at the same rate as the mass transfer processes, and investigate the effect of the reverse reaction rate on the dispersion characteristics of all species considered. The pertinent results are shown in Figures 3 and 4. The broken line indicates those results that should be obtained in the total absence of chemical reaction in the x-phase.

As the rate of the reverse reaction is increased, the dispersion

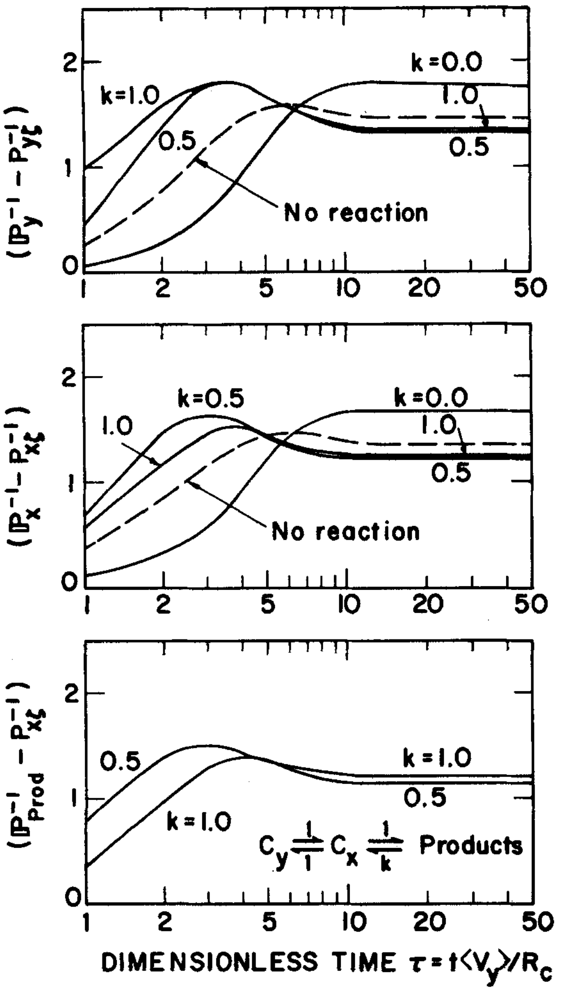

Figure 3. Effect of a reversible reaction on convective dispersion coefficients.

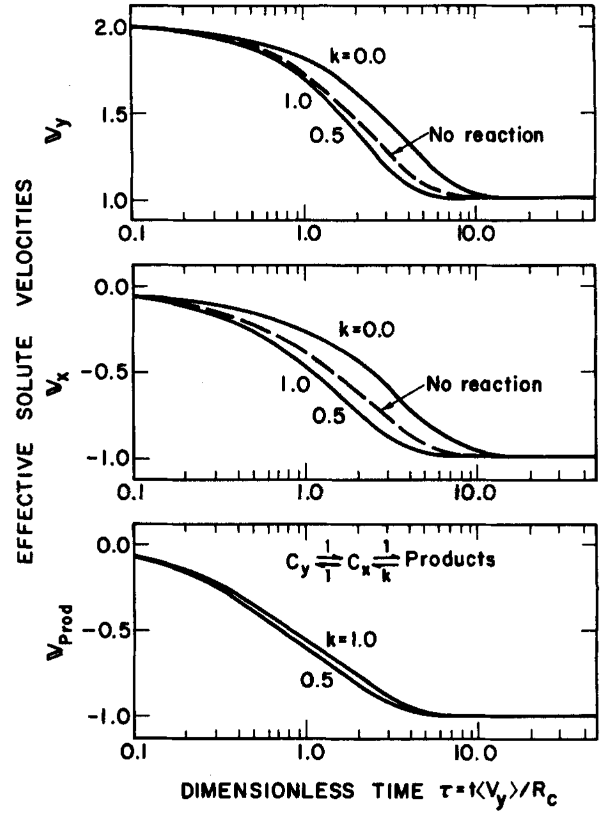

Figure 4. Effect of a reversible reaction on effective solute velocities.

coefficients for the solute in both the x- and the y-phase initially decrease past those values obtained in the absence of chemical reaction. However, with further increases in the reaction rate constant, the dispersion characteristics approach those that would be observed when the process is one of pure physical absorption with no chemical reaction. This is because any product formed immediately undergoes the reverse reaction, and, for all practical purposes, there is no depletion of the absorbed solute by reaction.

TRANSVERSE-AVERAGING PROCEDURE

The numerical results presented here and in our earlier paper (Hatton and Lightfoot, 1982) have all been calculated on the basis of the area-averaging problem. In many situations, however, such averaging procedures may be experimentally inaccessible or inconvenient, and other measurement techniques may be more practical. In process applications, for instance, where interest is usually in the prediction of process yields or effluent concentrations, it is the flow average or cup-mixing concentration within each phase that must be considered. It does not follow, however, that the results relating to the dispersion characteristics for the area-averaging problem will necessarily carry over to problems dealing with the flow-averaging conditions. Indeed, consistent with the earlier findings of DeGance and Johns (1978) for the scalar, single-solute problem, the mathematical analysis of Part I indicates that such a correspondence between the results for the two averaging procedures cannot, in general, be expected. In particular, it is evident that when the transverse averaging applied to the individual phases is different for each phase, additional interaction effects between the phases become apparent, manifested as nondiagonal solute velocity and convective dispersion matrices, respectively. Whether or not these interaction effects will be important, however, will depend on the magnitudes of the off-diagonal terms in these matrices relative to the diagonal entries. We investigate this aspect of multiphase, convective dispersion here, and show that these interaction effects can certainly be of some significance in countercurrent separations processes.

The transient development of the convective dispersion coefficient matrix is shown in Figure 5 for a selected set of parameter values, and for a central injection of a pulse of solute in the y-phase. Corresponding results are shown for the effective solute velocity and rate constant matrices in Figure 6 and 7, respectively. The two sets of curves shown in each figure correspond to the two different averaging procedures used in the preparation of these graphs, and indicate clearly the major differences in the dispersion parameters that can occur when dissimilar transverse averages are to be entertained for the different phases.

It is of particular importance to note that, whereas the off-diagonal terms in the $\boldsymbol{P}$ and $\boldsymbol{V}$ matrices are identically zero when area averaging is used, for the flow-averaging problem these terms can be significant. In fact, for the velocity profiles and model parameters used here, the "interaction" components in the velocity and dispersion matrices are comparable in absolute magnitude with the diagonal elements, especially at the larger times. Hence it would seem that the off-diagonal elements should not be ignored in the analysis and interpretation of mass transfer experiments, nor in the prediction of mass transfer performance. However, we may legitimately ask what effect neglect of these interactions would have on the estimation of the effective dispersion parameters from experimental response curves.

Let V_{ye} and P_{ye}^{-1} be the respective velocity and dispersion parameters for the y-phase that would appear to be in effect at any given time if the phase interactions were to be ignored. Similar quantities can be defined for the x-phase. The convective and dispersive fluxes in the y-phase are then

$$V_{ye}c_y = V_{yy}c_y + V_{yx}c_x \tag{9}$$

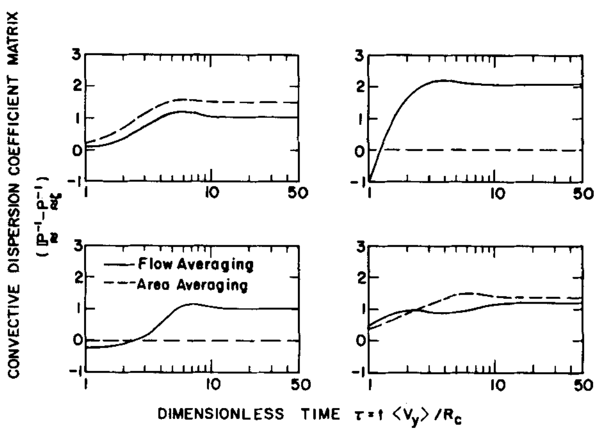

Figure 5. Effect of transverse-averaging procedure on convective dispersion coefficient matrix.

and

$$P_{ye}^{-1}\frac{\partial c_y}{\partial \zeta} = \mathbf{P}_{yy}^{-1}\frac{\partial c_y}{\partial \zeta} + \mathbf{P}_{yx}^{-1}\frac{\partial c_x}{\partial \zeta} \tag{10}$$

respectively. The effective parameters that will be estimated are therefore

$$V_{ye} = \left(\mathbf{V}_{yy} + \mathbf{V}_{yx}\frac{c_x}{c_y}\right) \tag{11}$$

and

$$P_{ye}^{-1} = \left(\mathbf{P}_{yy}^{-1} + \mathbf{P}_{yx}^{-1}\frac{c_x}{c_y}\right) \tag{12}$$

respectively, which, we note depend explicitly on the ratio of the solute concentrations in the two phases. Thus it would seem that neglect of the interaction components will introduce into the one-dimensional axial dispersion model a nonlinearity effect, since now the effective velocities and dispersion coefficients depend explicitly on the solute concentration level within each phase.

The reason for the presence of the off-diagonal terms in the parameter matrices is directly related to the different weighting attached to the solute depending on whether it is in the y-phase or the x-phase. A material balance for the one phase (the y-phase, say) over a small region of column will consist of two terms: the intraphase flux, determined by the convective and diffusive processes; and the mass transfer flux, which depends on the actual concentration driving force. The first of these is, of course, unaffected by the x-phase and can have no contribution to the off-diagonal elements in the matrices. The mass transfer term, on the other hand, reflects the interaction between the two phases and can influence the off-diagonal terms in these matrices. As the solute transfers from one phase to the other, its weighting changes, and there is a residual concentration term which reflects the difference in the weight attached to the solute within the two phases. This "residual solute" is also carried over from one phase to the other, and must appear as a source (or sink) term in the material balance equation for the receiving phase. Moreover, this source term will reflect the convective-dispersive behavior of the donor phase, and will thus depend on both the convection and the axial dispersion of the solute in this phase. Consequently, the velocity and dispersion coefficient matrices will have nonzero off-diagonal elements.

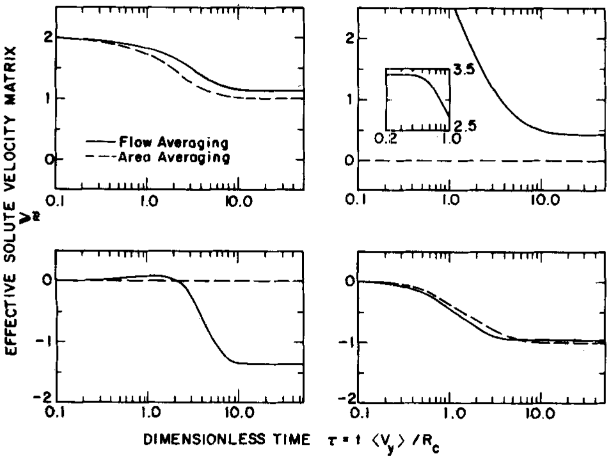

Figure 6. Effect of transverse averaging procedure on effective solute velocities.

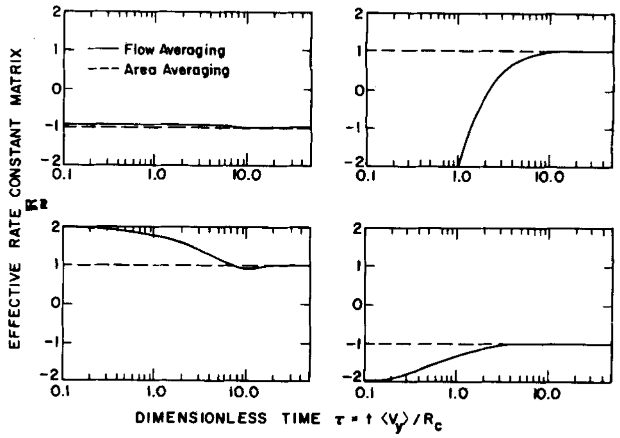

Figure 7. Effect of transverse averaging procedure on effective rate constant matrix.

COMPARISON OF PREDICTED EFFECTS WITH PUBLISHED EXPERIMENTAL RESULTS

A direct quantitative comparison between the model predictions and reported experimental data is not possible because available information on the flow characteristics of the columns used in the experimental studies is incomplete. Nevertheless, some qualitative observations can be made based on the predicted and observed effects of mass transfer on the dispersion parameters, and two sets of results are available for this purpose; those of Hatton and Woodburn (1978) and Linek et al. (1978), respectively. Since the operating conditions and column configurations differed significantly in these two studies, we consider them separately in the discussion that follows.

Hatton and Woodburn (1978) investigated the dispersion and mass transfer characteristics of a large absorption column operating under extremely high irrigation rates. Steady-state carbon dioxide absorption profiles were measured for the gas phase, and the relevant model parameters were estimated by nonlinear regression of the dispersion model on these profiles. The gas-phase mixing data were compared with those obtained earlier on the same column by Woodburn (1974) using an insoluble gas tracer. These results have been replotted as shown in Figure 8, where the ordinate is the ratio of the Peclet number obtained under mass transfer conditions, Pe_G^*, relative to that determined in the absence of such mass transfer, Pe_G^0. The Pe_G^* results are tabulated by Hatton (1976), and the Pe_G^0 values were estimated from the curves given by Woodburn (1972, 1974). It would seem that a unique curve, independent of liquid Reynolds number, is obtained, and that the Peclet number ratio varies linearly with gas rate, at least over the flow rate ranges studied.

The column used in these studies had a length to diameter ratio of approximately 38, although the distance between detection points in Woodburn's (1972, 1974) study was somewhat less than this. Hence it is reasonable to assume that, relative to the gas hold-up time, the transient development of the dispersion processes was sufficiently rapid to ensure that the asymptotic long-time dispersion coefficients are representative of the dispersion behavior of this column. The numerical study given earlier (Hatton and

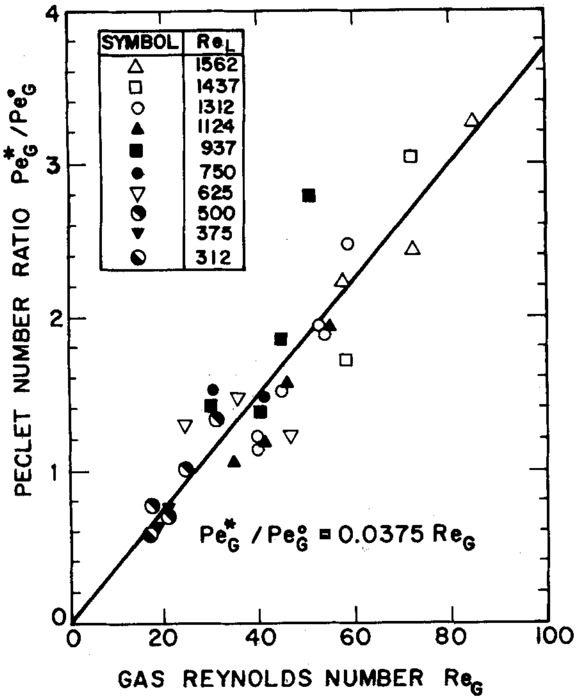

Figure 8. Peclet number ratio as a function of gas rate.

Lightfoot, 1982) and our present results all indicate that under these conditions mass transfer can modify the effective dispersion coefficients to the extent observed in Figure 8, i.e., the increases in the experimental Peclet numbers owing to mass transfer effects are in line with those predicted by our dispersion analysis.

Linek et al. (1978) also clearly demonstrate that dispersion coefficients measured during intraphase tests are inappropriate for use in the prediction of mass transfer performance of industrial installations. They used a short column, equivalent to about five column diameters, and operated at very low liquid rates. Under their flow conditions, the axial dispersion in the liquid is known to be more important than that in the gas (Dunn et al., 1977). It is, therefore, not surprising that Linek et al. found there to be no effect of mass transfer on their gas-phase axial mixing coefficients. The liquid-phase results, however, showed a marked increase in the effective dispersion of the liquid phase in the presence of interphase mass transfer, the changes being by as much as an order of magnitude or more. Such large differences between the dispersion parameters for the inert and soluble tracer studies, respectively, have been predicted by our model only for the transient period of the dispersion process, and it is likely that since Linek et al. used a short column, it was this period that was dominant in their studies. Thus, again, experimental observations are not inconsistent with the model predictions, at least as far as the magnitudes of the predicted effects are concerned.

DISCUSSION

The numerical results presented here have clearly demonstrated the inadequacy of the axial dispersion model as it is usually formulated for the description of mass transfer and chemical reaction in interacting, multiphase flows. The inconstancy of the dispersion coefficients and solute velocities with time is a factor not normally accounted for in the analysis of experimental response curves, and one that can lead to wide variations in the estimated values for the dispersion coefficients depending on the distance between detection points in the experimental set-up. Moreover, the dependence of the dispersion coefficients for any one phase on the hydrodynamic characteristics of the other is a cause for concern, as is the intrinsic effect of the solute solubility on these parameter values.

This lack of unique dispersion parameters for any given set of flow conditions undermines the effectiveness of present day design procedures based on the constant coefficient axial dispersion model, and suggests the need for a more faithful representation of the hydrodynamic and mass transfer interactions in processing equipment. Thus more attention should be given to the analysis of large-scale channelling effects in multiphase contactors if more reliable predictions of mass transfer performance are to be realized, and if a better understanding of the often anomalous behavior of these contactors is to be gained.

While the results of the formal mathematical analysis have indeed given us a good insight as to the complexities of the hydrodynamic and mass transfer interactions in processing equipment, it is apparent that we are not yet in a position to provide quantitative predictions of these effects. The primary reason for this is the lack of detailed experimental information on the flow characteristics of two-phase systems. This is patently an area that warrants further attention, but one that is not without its challenges.

Although many good studies have been reported on liquid-phase distribution, and on axial and radial dispersion in the trickle-bed operation of packed towers (e.g., Farid and Gunn, 1979), no similar studies have been undertaken to investigate the gas-phase behaviour of these systems. This may be partly because at the low liquid rates usually employed in these studies, liquid-phase mixing is considered to be the more important of the two. However, the results of Linek et al. (1978) clearly demonstrate that under mass transfer conditions, interactions between the phases can be significant, and hence it would appear that the flow characteristics of the gas phase play a more important role in determining packed tower performance at these low liquid rates than has hitherto been realized. In a like manner, at the extremely high liquor rates used in the work of Hatton and Woodburn (1978), liquid-phase dispersion is assumed to be unimportant relative to that in the gas phase, although again it is clear from their results that conditions in both phases contribute to the overall dispersion of the gas phase.

NOTATION

c_m = weighted average solute concentration vector
c_m^* = Fourier transform of c_m
c_{mo} = initial value for c_m
c_i = solute concentration in phase i
d_p = nominal packing particle size
E_G = gas-phase axial dispersion coefficient
i = imaginary number $i = \sqrt{-1}$
$K_y a$ = volumetric overall mass transfer coefficient
k' = reaction rate constant
k = dimensionless reaction rate constant $k = k'/K_y a$
m = equilibrium constant ($c_y = mc_x$ at equilibrium)
$\mathbf{P}$ = matrix of Peclet numbers for overall axial dispersion (Part I, Eq. 22)
$\mathbf{P}_c$ = matrix of Peclet numbers in the constant coefficient axial dispersion model
$\mathbf{P}_\eta$ = matrix of Peclet numbers for radial dispersion
P_{ye} = effective y-phase Peclet number when phase interactions are ignored
Pe_G^o = experimental Peclet number determined in absence of mass transfer, $Pe_G = u_G d_p/E_g$
Pe_G^* = experimental Peclet number determined under mass transfer conditions, $Pe_G = u_G d_p/E_G$
$\mathbf{R}$ = matrix of effective rate constants in axial dispersion model (Part I, Eq. 20)
$\mathbf{R}_c$ = matrix of effective rate constants in the constant coefficient axial dispersion model
R = matrix of reaction rate constants for homogeneous reactions
Re_G = gas-phase Reynolds number $Re_G = \rho_G u_G d_p/\mu_g$
Re_L = liquid-phase Reynolds number $Re_L = \rho_L u_L d_p/\mu_L$
u_i = superficial velocity of phase i

V = matrix of effective velocities (Part I, Eq. 21)
V_c = matrix of effective velocities in the constant coefficient axial dispersion model
$V(\eta)$ = matrix of radially dependent velocities
V_{ye} = effective y-phase velocity when phase interactions are ignored

Greek Letters

ϵ = matrix of hold-up ratios
ζ = normalised axial coordinate
η = normalised radial coordinate
λ = Fourier transform variable
μ = viscosity
ρ = density
τ = dimensionless time
ω = matrix of weighting functions for concentration averaging

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Experimental and Mathematical Modeling of Three-Dimensional Natural Convection in an Enclosure

Three-dimensional natural convection patterns in an enclosure under conditions simulating flow in glass-making furnaces have been calculated using an approximate method in which solutions for the temperature and velocity fields in orthogonal two-dimensional planes are superposed. The computational method was tested by comparison with measurements of the temperatures and velocities in an enclosure heated and cooled from above and with a controlled heat loss from the sides. Good agreement was found between measurements and computations under conditions of interest in glass-making furnaces, wherein the motion in one of the two orthogonal planes is dominant.

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SCOPE

Glass is produced by feeding a solid mixture consisting primarily of sodium carbonate and silica onto the surface of a glass melt. The solids float over the melt near their point of introduction to the furnace and are heated from below by the melt and from above by radiation from the flames and refractory. The heat transfer to the bottom surface of the floating solids is controlled by the natural circulation pattern which is driven by radiation from the flame to that portion of the melt not covered by the feed solids. The flow in the glass melt is predominantly two-dimensional, laminar and Newtonian, with a Rayleigh number of 10^6 to 10^8, and a Prandtl number of 200 to 5,000.

Numerical methods for solving natural convection patterns are well developed (Catton and Edwards, 1967; Mallison and de Vahl Davis, 1973; Ozoe et al., 1974) and have been applied with considerable success for determining the dominant two-dimensional motion in glass melts (Noble et al., 1972). Interest in obtaining the complete three-dimensional field has been motivated by the importance of flows in the plane orthogonal to that of the dominant flow; the flow in the transverse plane is needed for applications such as determining residence-time

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