Mixing and Contacting Models for Fluidized Beds

JOHN P. MIREUR and KENNETH B. BISCHOFF

University of Texas, Austin, Texas

Parameters of the two-phase model for mixing in fluidized beds were evaluated from a large amount of available literature data. The dense phase dispersion coefficients and the cross flow, or interphase transfer, coefficients, based on the consistent model calculations, were correlated with operating conditions. A strong effect of bed length-to-diameter ratio was found for the dense phase mixing but not for the cross flow. On the basis of the given kinetics information with the mathematical model parameter correlations, some chemical reactor conversions from the literature were predicted by a priori calculations.

Although much is known about the behavior of fluidized beds (*6, 14, 39*), it is still not possible to predict accurately their performance, for example, as a chemical reactor, from knowledge of only nominal operating variables. In other words, given the flow rates, particle properties, bed geometry, etc., the conversion to be expected can only be found through the use of many empirical factors. In fact, the usual procedure of operating a pilot plant and then scaling up is not even completely possible. The main problem lies in the extremely complicated hydrodynamics in the fluidized state. This is particularly true for gas-solid fluidization where it is found that large bubbles form in the bed in addition to the existing solid-fluid motion.

The origin and certain properties of fully developed bubbles have been worked out to a considerable degree by solving certain parts of the equations of motion for the system. A comprehensive discussion of much of this work, along with the extensive investigations of Rowe, is provided in the monograph of Davidson and Harrison (*3*). For our purposes, the points of interest are that given a single bubble diameter, the flow patterns of gas and solid and some aspects of mass transfer from the bubble can be mathematically predicted. Also, many qualitative aspects that are useful in formulating proper models are provided.

MODELS

The first attempts in the literature to apply a model to account for the nonideal (ideal being either plug flow or perfect mixing) flow patterns were to use the axial dispersion model for the whole bed. However, it was found to be very difficult to correlate the dispersion coefficients and not much more was done in this direction. The reason for the failure was that the presence of large bubbles precluded the use of any model whose basis lies in a diffusion type of mechanism, that is, a large number of small, random mixing steps.

Other types of chemical engineering operations have the same features, such as gas absorption or extraction. The classical type of model successfully used in these cases visualizes the system as containing two continuous phases with interphase transfer occurring. Thus, a logical step would be to apply these concepts to the case of a fluidized bed containing bubbles, leading to the general two-phase model shown in Figure 1. It should be noted that a dispersion type of model is to be used here only in the dense phase, not for the entire bed. Since the dense phase mixing is caused primarily by the bubble motion, this is still not entirely correct, and it is found from solids mixing studies that the experimental curves do not have quite the proper shape as found from the dispersion model, but the errors caused by this would probably not be too great in applications, as will be seen later.

This type of model was first proposed by Toomey, Shen, and Johnstone (*32, 35*) and subsequently used by Mathis and Watson (*19*), Lewis et al. (*16*), May (*20*), Lanneau (*13*), Gomezplata and Shuster (*9*), van Deemter (*36*), and many recent investigators. No one has yet used the complete model because of the large number of parameters, and many special cases have been defined, as discussed by Levenspiel and Bischoff (*15*). It seems that with our present state of knowledge, several of the parameters can be specified from the above-mentioned hydrodynamic studies, leaving a small number to be determined from mixing tests. Thus, if the *bubble phase* is defined to have no solids, $m = 0$. A certain amount of solids do

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John P. Mireur is with Celanese Chemical Company, Corpus Christi, Texas. Kenneth B. Bischoff is with the University of Maryland, College Park.

"rain" through the bubbles, but this would probably be very difficult to specify or measure.

The flow split and volume split are determined from the supposition that conditions in the *dense phase* at any flow rate must be the same as at minimum fluidization conditions. This seems reasonable from qualitative analysis of the hydrodynamic conditions that must exist for the fluidized state to exist at all. The theoretical analyses (*3*) as well as direct measurements by *x*-ray and other means (*27* to *29*, *38*) also substantiate this. Thus the flow to and volume of the dense phase will be taken to be those at minimum fluidization with all excess flow and bed expansion accounted for by the bubble phase.

Finally, under commercial operating conditions of interest where a large multiple of the minimum fluidizing velocity is used, the bubble velocity is much greater than the dense phase gas velocity. Therefore, the bubble gas has little time for backmixing, which leads to the assumption of plug flow in the bubble phase. This same approximation has been used by several earlier investigators of the two-phase model. With all these stipulations, there are two model parameters remaining that must be determined by mixing experiments: the cross flow or interphase transfer and the dense phase dispersion coefficient.

Mathematical models even more closely connected to the bubble information have also been proposed by Orcutt et al. (*25*), Davidson and Harrison (*3*), and Rowe (*30*). It would seem that these would be the most desirable since they are based on existing theoretical information, and this is undoubtedly true for those situations where definite single bubbles exist. Even here there remains the problem that bubble diameters cannot yet be accurately predicted and must be found empirically. Thus, as stated by Davidson and Harrison (*3*), there is at present no real reason to prefer one model over the other. However, it will probably not be too long before means are available for predicting single bubble sizes from first principles, in which case the direct bubble models would be the best, where applicable. Most commercial fluidized beds are operated under conditions where the gas flow rate is many times (5-10-20) the minimum. Here, the bed is a churning mass with bubbles of many sizes colliding, etc., and it is difficult to see how a direct bubble model could be used. In other words, there would always be a certain number of empirical parameters regarding bubble size distributions (even if the flow patterns could still be mathematically predicted) and so the above conclusion of not really being able to decide on one model over the other would probably be true.

DETERMINATION OF PARAMETERS

Based on the above discussion, the two-phase model was used in this study. After utilizing the hydrodynamic information fixing the dense phase at minimum fluidization conditions (*20*, *32*), there were still two parameters to be found from mixing tests. The most common test is the impulse response or residence time distribution experiment (*15*). In principle, both parameters could be found from various curve fitting methods comparing the model solutions with the experimental data. However, in practice, the precision is not good enough to determine the parameters uniquely. Therefore it is usually conceded that two types of experiments, essentially providing two independent "equations," are required to solve for the two parameters.

Several methods of doing this have been proposed. May (*20*) measured solids mixing and assumed that the dense phase gas and solids dispersion coefficients were the same. What were essentially residence time distribution experiments were then used to obtain the cross flow. Van Deemter (*36*) proposed using so-called backmixing experiments (steady tracer flow at downstream location) in conjunction with residence time distributions. This method would not require the assumption of equal solids and dense phase gas dispersion. Another possibility is to use backmixing plus chemical reaction data. One problem with utilizing existing data is that very few investigators have obtained two types of data on the same apparatus; thus, van Deemter (*36*) only presented results from a few runs of Gilliland and Mason (*7*, *8*). The present study used a method to circumvent partially this problem, which will be discussed below in detail.

MATHEMATICAL EQUATIONS

The velocities in the two phases are related to the (measured) superficial total velocity by

$$u = f_d\, v_d + f_b\, v_b \tag{1}$$

If the cross flow is expressed in the usual interphase mass transfer terms, mass balances on each phase (per unit total bed volume) are

$$f_b \frac{\partial C_b}{\partial t} + v_b f_b \frac{\partial C_b}{\partial z} = k(C_d - C_b) \tag{2}$$

$$f_d \frac{\partial C_d}{\partial t} + v_d f_d \frac{\partial C_d}{\partial z} = f_d D_d \frac{\partial^2 C_d}{\partial z^2} + k(C_b - C_d) - f_d k_r C_d \tag{3}$$

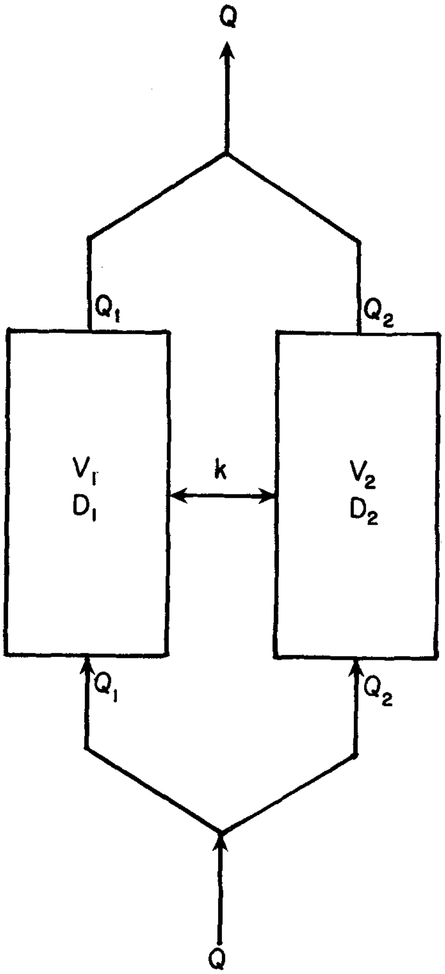

Fig. 1. The two-phase model for fluidized beds.

Van Deemter (*36*) presented solutions to the steady state version of Equations (2) and (3) with the backmixing boundary conditions. He found that for tracer concentrations negligible at the bottom (upstream) of the bed and for flow rates a few multiples of the minimum, the solutions reduced to the following form:

$$\frac{C_d}{C_o} = K_d \exp\left[\frac{\beta - 1}{2}\,\frac{kz}{u}\right] \tag{4}$$

$$\frac{C_b}{C_o} = K_b \exp\left[\frac{\beta - 1}{2}\,\frac{kz}{u}\right] \tag{5}$$

where

$$\beta^2 = 1 + 4\frac{u^2}{f_d D_d k}$$

$$= 1 + 4\frac{n_D}{n_k} \tag{6}$$

and

$$n_k = kL/u \tag{7}$$

$$n_D = uL/f_d D_d \tag{8}$$

The two integration constants K_d and K_b were different for each phase and also contained a constant representing the fraction of tracer injected into one phase. Now, in experiments where one injects and/or measures tracer within the fluidized bed, it is not possible to know which phase was being sampled, which leads to extreme difficulties in data interpretation. Note, however, from Equations (4) and (5), that the slopes of a semilog plot of tracer concentration vs. length are identical in both phases, so that the detailed sampling information is not required if one uses only the slope S of a semilog plot:

$$LS \equiv n_B = \frac{n_k}{2}(\beta - 1) \tag{9}$$

The slope, of course, has both parameters k and D_d in it and must be used in conjunction with residence time or other information.

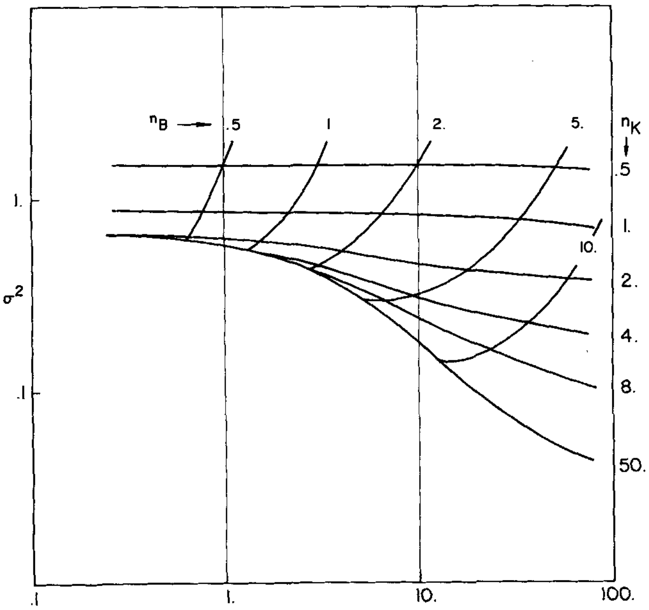

Fig. 2. Model parameters for $p = 0.40$.

Van Deemter (*36*) also found the variance of the residence time distribution using the methods of van der Laan (*37*). For the same approximations, the result was

$$\sigma^2 = \frac{2}{n_k}(1-p)^2 + \frac{2}{n_D} + \frac{2\beta}{n_D^2}\left\{\frac{\left\{\exp\left[\frac{1}{2}n_k(\beta-1)\right]-1\right\}\left\{\exp\left[-\frac{1}{2}n_k(\beta+1)\right]-1\right\}}{\left\{\exp\left[\frac{1}{2}n_k(\beta-1)\right]\right\} - \left\{\exp\left[-\frac{1}{2}n_k(\beta+1)\right]\right\}}\right\} \tag{10}$$

$$= \Psi\,(n_k, n_D) \tag{10a}$$

This, then, defined another relation between an experiment and the parameters n_k and n_D.

Since both Equations (9) and (10) are not explicitly solvable for n_k and n_D, it is convenient to utilize a graph such as Figure 2 when actually determining the parameters. Other interesting features of the model, however, can also be seen clearly from Figure 2. Over much of the region the functional dependence of the parameters is not the same for both types of experimental information. In other words, n_D (dense phase mixing) is most strongly a function of n_B (backmixing slope) and, similarly, n_k (cross flow) is primarily dependent on σ^2 (variance of residence time curve). In physical terms, backmixing is caused mostly by dense phase mixing and residence time distribution spreading by cross flow.

With a first-order chemical reaction, May (*20*) presented a solution, with finite bed boundary conditions, which in our terminology is

$$X = \left(\frac{u_{mf}}{u}\right) X_d + \left(1 - \frac{u_{mf}}{u}\right) X_b \tag{11}$$

where

$$X_d = 1 - \frac{1}{P}\sum_{i=1}^{3} M_i e^{\alpha_i}$$

$$X_b = 1 - \frac{a}{P}\sum_{i=1}^{3}\frac{M_i}{\alpha_i + a}e^{\alpha_i}$$

The α_i are the roots of the cubic equation

$$\alpha^3 + (a - b)\alpha^2 - (ab + ad + gd)\alpha - adg = 0$$

where

$$a = \frac{n_k}{1 - u_{mf}/u} \qquad b = n_D\left(\frac{u_{mf}}{u}\right)$$

$$d = n_D\left[1 - \frac{u_{mf}}{u}\right] \qquad g = \frac{n_R}{1 - u_{mf}/u}$$

and

$$n_R = f_d k_r L/u \tag{12}$$

$$M_1 = (\alpha_2 - b)\alpha_3 e^{\alpha_3} - (\alpha_3 - b)\alpha_2 e^{\alpha_2} + \frac{ab}{a + \alpha_2}\alpha_3 e^{\alpha_3} - \frac{ab}{a + \alpha_3}\alpha_2 e^{\alpha_2}$$

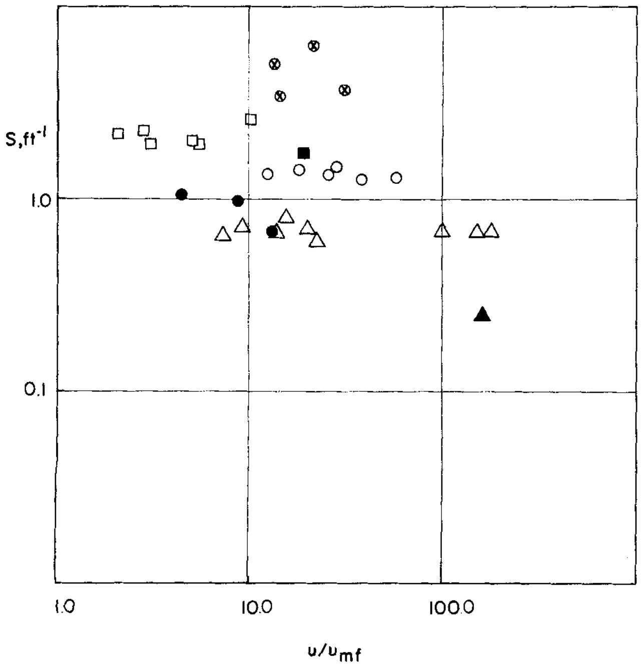

Fig. 3. Mixing slope data: ● (24), ○, ⊗ (17), ■ (34), △ (7), ▲ (26), □ (23).

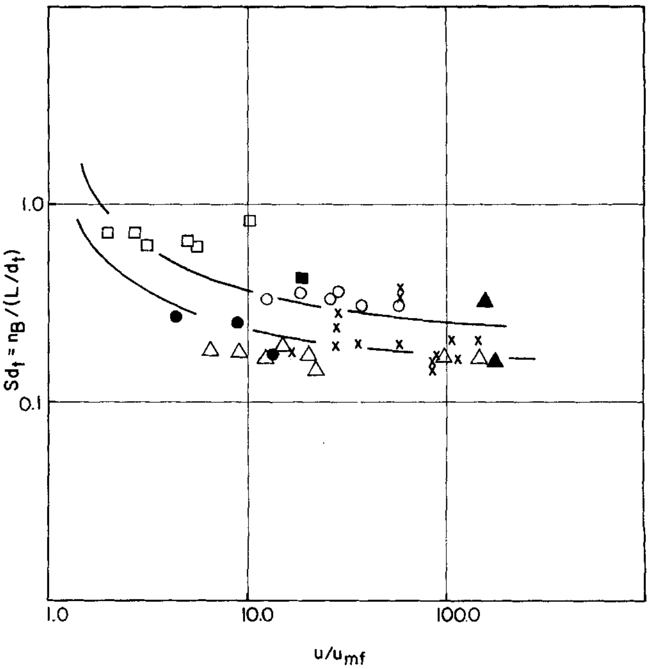

Fig. 4. Mixing slope correlation: ● (24), ○ (17), ■ (34), △, X (7), ▲ (26), □ (23).

$$M_2 = (\alpha_3 - b)\alpha_1 e^{\alpha_1} - (\alpha_1 - b)\alpha_3 e^{\alpha_3} + \frac{ab}{a+\alpha_3}\alpha_1 e^{\alpha_1} - \frac{ab}{a+\alpha_1}\alpha_3 e^{\alpha_3}$$

$$M_3 = (\alpha_1 - b)\alpha_2 e^{\alpha_2} - (\alpha_2 - b)\alpha_1 e^{\alpha_1} + \frac{ab}{a+\alpha_1}\alpha_2 e^{\alpha_2} - \frac{ab}{a+\alpha_2}\alpha_2 e^{\alpha_2}$$

$$P = \alpha_1 e^{\alpha_1}\left[\frac{a}{a+\alpha_2}(\alpha_3 - b) - \frac{a}{a+\alpha_3}(\alpha_2 - b)\right] + \alpha_2 e^{\alpha_2}\left[\frac{a}{a+\alpha_3}(\alpha_1 - b) - \frac{a}{a+\alpha_1}(\alpha_3 - b)\right] + \alpha_3 e^{\alpha_3}\left[\frac{a}{a+\alpha_1}(\alpha_2 - b) - \frac{a}{a+\alpha_2}(\alpha_1 - b)\right]$$

It is seen from the above that the conversion is also a function of both n_D and n_k and again would need to be used in conjunction with, for example, a backmixing experiment to obtain the parameters. Graphs similar to Figure 2 can be used to aid in the calculations. It is found that the conversion is most strongly a function of the cross flow, as also indicated by May (20), Orcutt et al. (25), Muchi (22), and Kobayashi and Arai (12).

RESULTS

As mentioned earlier, very few investigators have performed two types of experiments on the same system. Thus we believed that if the large amount of existing residence time and reaction data were to be utilized, some means must be used to estimate the backmixing information that presumably would have been obtained if the experiments had been done originally. Therefore, the available backmixing data were plotted to see if a correlation was possible. The slopes of the experimental data of several investigators were plotted against the ratio of actual to minimum velocity u/u_{mf} as shown in Figure 3. The use of u/u_{mf} as the independent variable seems to be preferable to something like Reynolds number, even though they both contain the same type of information: flow rate, solid properties, fluid properties. This is based on the bubble mechanics studies wherein it is found that the different flow regimes are dependent on u/u_{mf} (Rowe has also pointed this out). Obviously, as $u/u_{mf} \rightarrow 1.0$, the dependent variables should approach their fixed-bed values, which is a further aid in correlations. Figure 3 shows a large amount of scatter, some of which is undoubtedly partly caused by lack of a "scale factor" in the dimensional slope variable S. Various length parameters were tried and it was found that the bed diameter d_t gave the best correlation, as shown in Figure 4. The limit of (Sd_t) as $u/u_{mf} \rightarrow 1.0$ is very large, as found from fixed-bed data, and the indicated lines could thus be estimated.

It should be mentioned that only data for cracking catalyst type solids have been used in Figure 4, since it was felt that the additional complications from grossly different solid properties would be too difficult to account for at the present time. A large amount of glass bead data from Gilliland and Mason (7, 8) and Schügerl were also found to follow the same curve, only displaced upward (less mixing); further work would be required to define completely these trends. Thus Figure 4 was used to estimate the presumed slope to add to residence time and/or reaction data of the various investigators.

The use of graphs like Figure 2, then, permitted the determination of n_D and n_k as discussed above. The results for n_D are shown in Figure 5, where a wide range of bed length-to-diameter ratios, flow rates, etc., were used. It was found that the L/d_t ratio was quite significant, as shown in Figure 6. A replot of this correlation is shown in Figure 7, where the low u/u_{mf} region is again extrapolated to fixed-bed values. The correlation is not too rigorous, of course, since the points for it were obtained by "generating" slope data from Figure 3. Since n_D depends mostly on backmixing results, n_B, this generated information would have its strongest effect here. As has been stated, however, the desired end result, namely, the prediction of conversion for chemical reactions, does not depend very strongly on n_D, so Figure 7 may be sufficient.

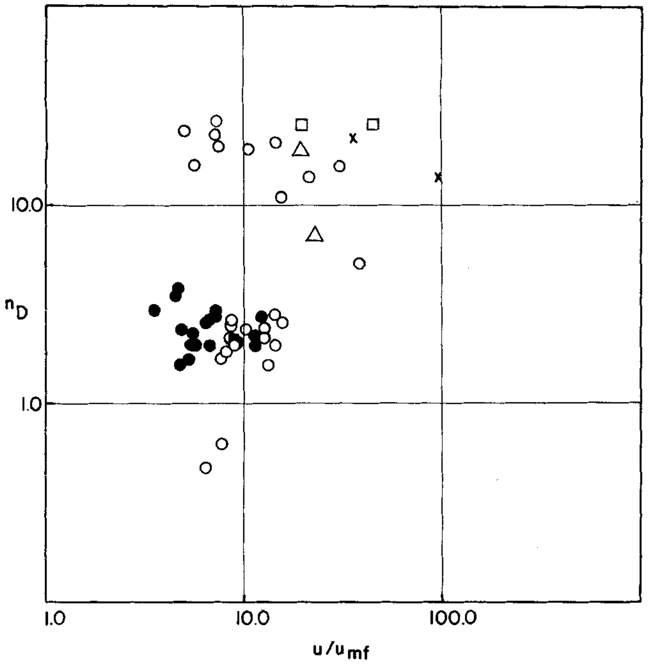

Fig. 5. Dense phase dispersion coefficient data: X (5), ⊗ (20), △ (8), □ (7), ○ (10), ● (2), ■ (11).

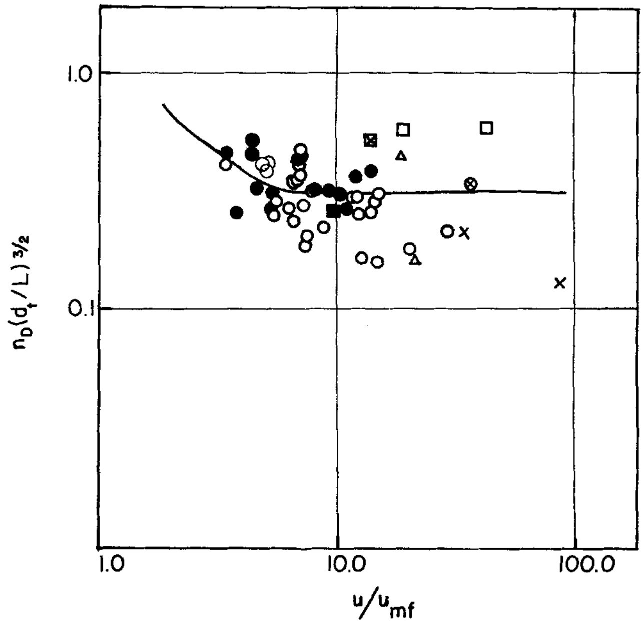

Fig. 7. Dense phase dispersion coefficient correlation: same data as in Figure 5 plus ☒ (1).

An interesting point from Figure 7 is the L/d_t dependence of dense phase mixing. The correlation states that the mixing is smaller for large L/d_t ratios, which is qualitatively correct. Since the data cover an absolute diameter range from about 1 in. to 5 ft., the correlation does have some utility. Also, it was found that the solid mixing point of May (*20*) essentially coincided with the gas mixing value, and the solid mixing data of Bart (*1*) agreed reasonably well with the other data. This lends some validity to May's assumption concerning equality of dense phase gas and solid mixing. A recent survey of solids mixing data by Mori and Nakamura (*21*) may be of use in further studies to clarify this point.

Finally, Figure 5 or 7 shows that for beds with $L/d_t \simeq 1$ commonly used commercially, the value of n_D is about 0.3. Now the parameter n_D is the same as the Peclet number for the dense phase, and it is known (*15*) that this range of values indicates almost perfect mixing. Thus for some applications, a useful simplification might be to assume a perfectly mixed dense phase.

The values of n_k are shown in Figure 8. All the obvious geometrical ratios such as L/d_t, d_t/d_p, etc., were tried in an effort to improve the correlation, but none seemed to help. Thus, the plain plot of n_k vs. u/u_{mf} was retained. As indicated in the figure, two main groupings of data appeared: that from residence time data and that from chemical reaction data.

The reaction data points were determined from an arbitrary portion of the existing literature. The newer, and thus possibly more reliable, data were saved for the final

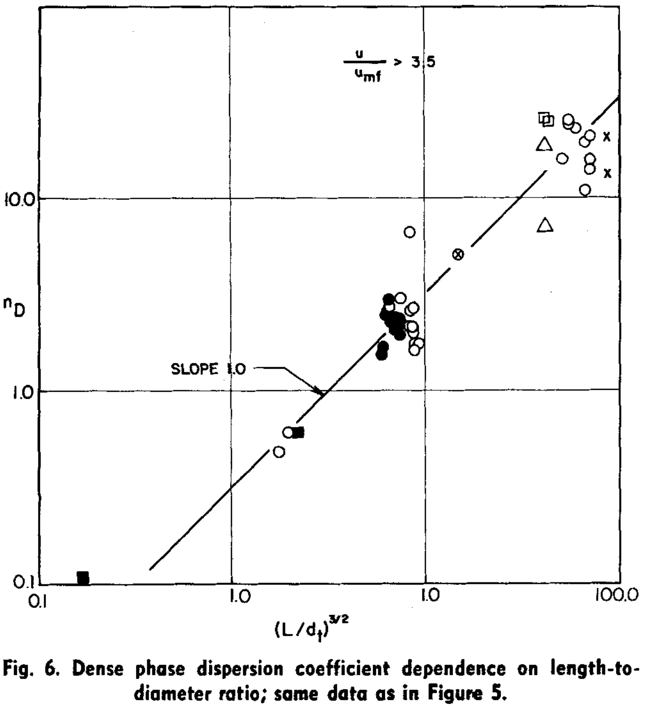

Fig. 6. Dense phase dispersion coefficient dependence on length-to-diameter ratio; same data as in Figure 5.

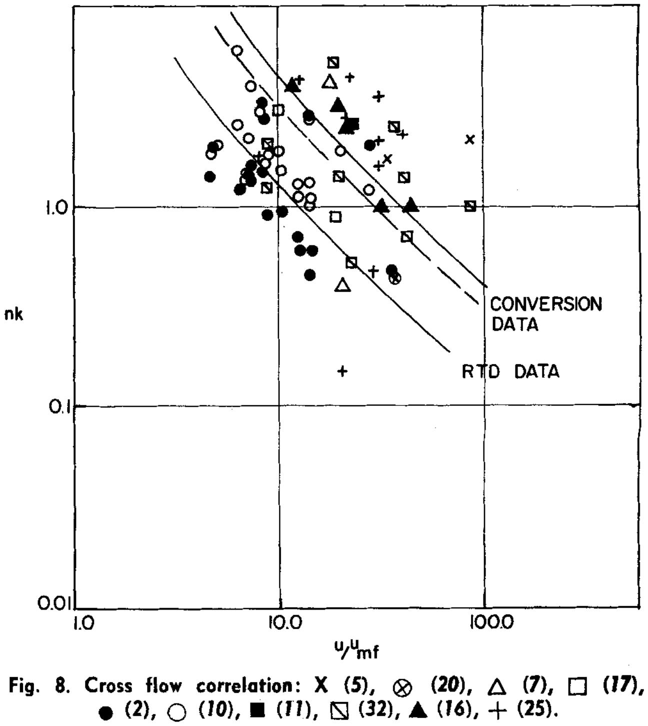

Fig. 8. Cross flow correlation: X (5), ⊗ (20), △ (7), □ (17), ● (2), ○ (10), ■ (11), ⧄ (32), ▲ (16), + (25).

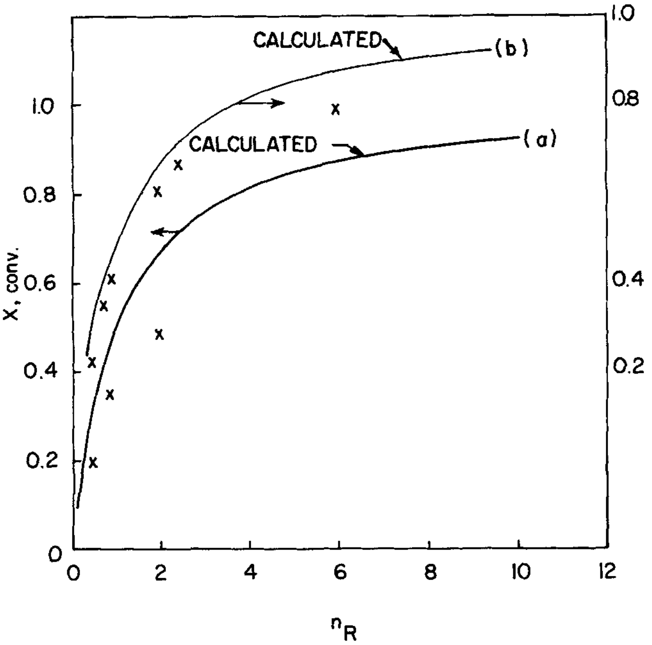

Fig. 9. Comparison of model predictions with data of reference 11. (a) $d_t = 0.89$ in.; $L = 1.7$ in.; $d_p = 107$ μ; $u = 0.11$ ft./sec.; $u/u_{mf} = 10$. (b) $d_t = 6$ in.; $L = 10$ in.; $d_p = 107$ μ; $u = 0.11$ ft./sec.; $u/u_{mf} = 10$.

part of the work—comparison of a priori model predictions with actual results. Thus only the older reaction data were used at this stage. It should be mentioned here that it was found that the model was incapable of representing some of the Schügerl et al. (*2, 10*) and the Massimilla and Johnstone (*18*) data in that the experimental combinations of slope and variance were such that no values of n_k and n_D could satisfy the equations. These were usually at low values of u/u_{mf} which are not of too much interest commercially.

The split in the data was unexpected but, upon reflection, there may be a good reason for it. The residence time data are usually obtained with a tracer that is *not* adsorbed on the catalyst, such as helium. This is done to get information on the between particles mixing, not clouded by adsorption complications. However, the reactant *is* adsorbed in order to have a chemical reaction and so is acting like a different type of tracer. Surprisingly, very little seems to have been done on the problem of comparing adsorbable and nonadsorbable tracers in the same system. Stemerding (*33*) gave a few points with carbon dioxide that seemed to indicate a difference, but firm conclusions could not be drawn. More recently, Echigoya et al. (*4*) showed that there were significant differences in the tracer output curves for adsorbable vs. nonadsorbable tracers and also that more accurate conversion predictions could be obtained by using the adsorbable tracer residence time distribution. General methods of treating this are not available yet, however, and more work in this area is definitely needed. The ultimate goal would be, of course, to take purely physical tracer data, adsorption data, and chemical reaction data and put them together to predict reactor performance.

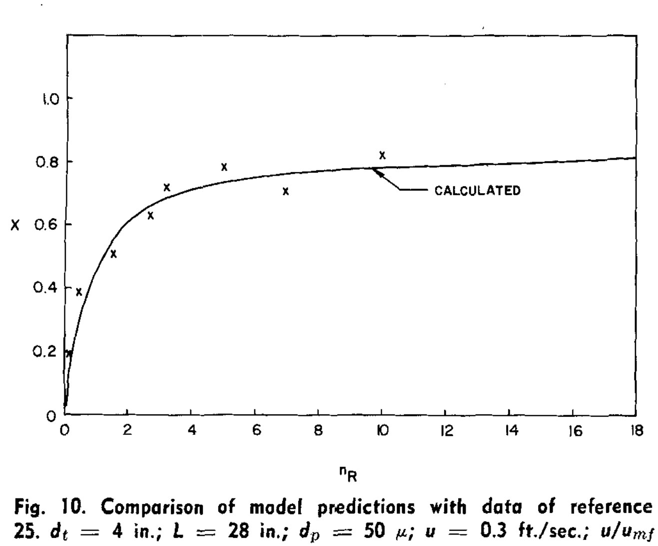

Fig. 10. Comparison of model predictions with data of reference 25. $d_t = 4$ in.; $L = 28$ in.; $d_p = 50$ μ; $u = 0.3$ ft./sec.; $u/u_{mf} = 21.5$.

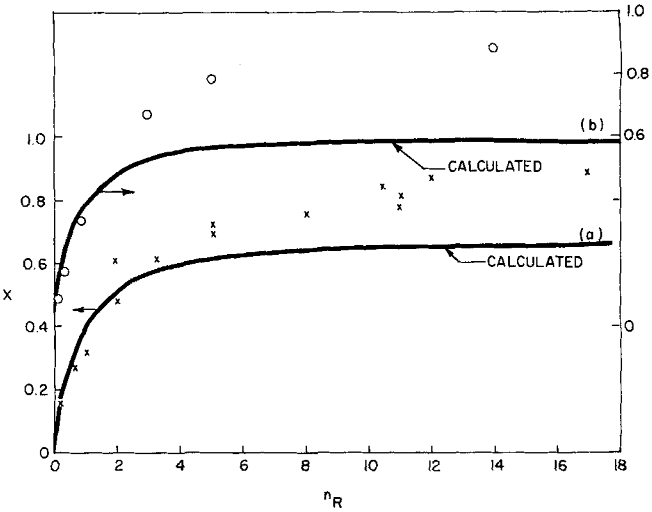

Fig. 11. Comparison of model predictions with data of reference 25. (a) $d_t = 6$ in.; $L = 24$ in.; $d_p = 50$ μ; $u = 0.46$ ft./sec.; $u/u_{mf} = 33$. (b) $d_t = 4$ in.; $L = 28$ in.; $d_p = 50$ μ; $u = 0.48$ ft./sec.; $u/u_{mf} = 43.6$.

TEST OF MODEL PREDICTIONS

The final test of any model is its success in predicting reactor performance. This was done by using the established parameter correlations along with an investigator's operating conditions and chemical kinetics data to attempt to make an a priori prediction of his results. Knowing velocities, particle and fluid properties, and bed geometry, we used Figures 7 and 8 to obtain n_D and n_k. In the absence of a definite conclusion regarding adsorption effects, the interpolated line in Figure 8, which was arbitrarily biased somewhat toward the reaction data, was used provisionally as the correlation. With the rate data, one can calculate n_R (the nominal space time) and then use all these values in Equation (11) to predict the conversion. Figures 9 through 12 show some results.

Figure 9 compares both very small and large fluidized bed conversion data of Ishii and Osberg (*11*) with the model predictions, with fair agreement. Orcutt et al. (*25*) gave three sets of data, as shown in Figures 10 and 11. The model predictions agreed perfectly with one set, fairly with another, and poorly with the third. The reason for these differences is not known. Finally, the data of Heidel et al. (*10*) are given in Figure 12 where fairly good agreement was obtained.

Since a rather large range of conditions is represented by Figures 9 to 12, it is felt that the model and correlations do a reasonable job of predicting reactor performance. Many of the model predictions are conservative, as can be seen from the graphs, but this may have been caused by the interpolation line used in Figure 8 rather than by the reaction line. Until methods are developed to

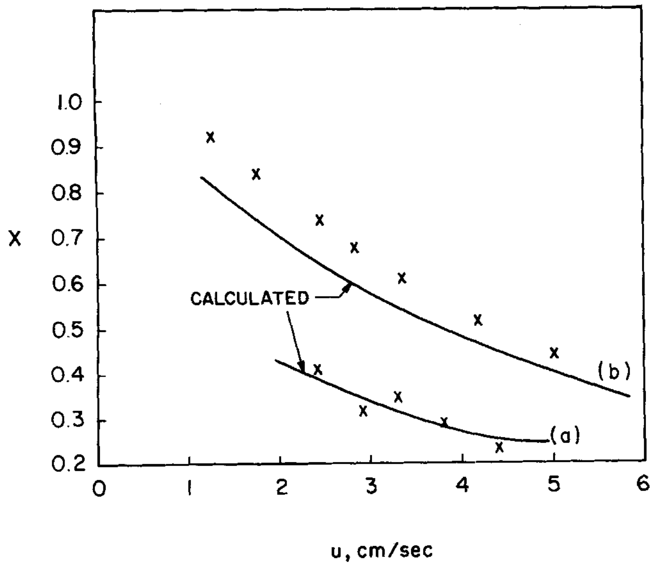

Fig. 12. Comparison of model predictions with data of reference 10. (a) $d_t = 10$ cm.; $L_{mf} \simeq 14.5$ cm.; $d_p = 75\ \mu$; $u_{mf} = 0.837$ cm./sec.; $k_r = 0.11$ sec.$^{-1}$. (b) $d_t = 7.5$ cm.; $L_{mf} \simeq 24$ cm.; $d_p = 50\ \mu$; $u_{mf} = 0.377$ cm/sec.; $k_r = 0.14$ sec.$^{-1}$.

account specifically for adsorption, however, this cannot be definitely known. Since a fluidized bed is such a complicated system, more work is definitely required, but some results, especially operating parameter studies, can now be obtained to some extent with the available information.

ACKNOWLEDGMENT

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NOTATION

C = concentration, moles/cu.ft.
d_p = particle size, ft.
d_t = tube size, ft.
D = diffusivity or diffusion coefficient, sq.ft./sec.
f = volume fraction of gas in phase
k = transfer coefficient, sec.$^{-1}$
k_r = first-order rate constant, sec.$^{-1}$
L = bed height, ft.
m = solids fraction in bubble phase, dimensionless
M_1, M_2, M_3 = constants in equation for conversion
n_B = LS, number of backmixing units, dimensionless
n_D = uL/f_dD_d, number of eddy diffusion units, dimensionless
n_k = kL/u, number of transfer units, dimensionless
n_R = f_dk_rL/u, reaction parameter (space time), dimensionless
p = $f_b/(f_b + f_d)$, dimensionless
P = constant in equation for conversion
Q = flow rate, cu.ft./sec.
S = mixing slope, ft.$^{-1}$
t = time, sec.
u = superficial velocity, ft./sec.
v = actual velocity in phase, ft./sec.
V = volume, cu.ft.
X = fraction conversion
z = position in bed, ft.

Greek Letters

α_1, α_2, α_3 = roots of equation for conversion
ϵ = bed voidage, dimensionless
σ^2 = variance of the residence time distribution, dimensionless

Subscripts

b = bubble phase
d = dense phase
e = bed exit conditions
k = interchange
mf = minimum fluidization
s = solids
t = total bed

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