Numerical Solution for the Axial and Radial Diffusions of Solute Tracer in Laminar Pipe Fiow

Numerical Solution for the Axial and Radial Diffusions
of Solute Tracer in Laminar Pipe Flow

Kenji KUBO *, Toshiro ARATANI **, Akira MISHIMA * and Takeo YANO **

(Received September 1, 1978)

Abstract

Taylor (1953, 1954a) analysed the problem of the effective dispersion of soluble tracer injected into a small-bore tube through which a solvent is flowing. The correlation $E=R^2u^2/48D$ is shown to be valid for the condition $4L/R>>D/uR>>\sqrt{48}$. Aris (1956, 1959) extended the correlation so as to include the axial diffusion effect and obtained the correlation $E=D+R^2u^2/48D$ for the validity condition excluding $uR/D>>\sqrt{48}$. Lighthill (1966) found an exact analytical solution for the radial diffusion and convection. A numerical method was developed here incorporating both the axial and radial diffusion effects with convection, for the unsteady state dispersion of the soluble tracer substance in fully-developed pipe flow. Finite-differenced with respect to space variables, the question gave rise to a set of ordinary differential equations. Unsteady state solution with respect to concentration change could be obtained by numerical integration of these ordinary differential equations. From the unsteady state solution when the tracer is continuously introduced into the fluid flowing at average velocity u at an upstream cross-section of the pipe, step response was computed, as the average concentration change in the cross-section at distance L from the tracer input cross-section. The effective dispersion co-efficient was numerically derived from the step response curve by moment integration, and non-dimensionalized so as to give the effective Pe_d group, $Pe_d=2uR/E$. The Pe_d is correlated as a function of $Re\times Sc=2uR/D$, in comparison with the Taylor's and Aris' correlations, with which our numerical experiment shows close agreement even for higher value of the slender ratio (R/L). (R: radius of pipe, L: the length with which the response is concerned) General Pe_d vs. $Re\times Sc$ correlations are given as charts, that are valid even for higher value condition of slender ratio, utilizing the slender ratio, R/L, as a parameter.

1. Introduction

Dispersion of the fluid flowing through a tube or a duct is of considerable practical importance, since the dispersion greatly affects the performance of chemical equipments that have the tubular flow channel. The dynamics of tubular reactors and heat exchangers are directly influenced by the dispersion. The yield of the tubular reactor is as well shifted by the fluid mixing characteristics in the tube. From these technical requirements, the dispersion in a pipe has been described by an effective dispersion coefficient, on introducing average flow rate and concentration through the cross-section of the pipe. The dispersion effect in a pipe is caused essentially by the axial and radial molecular diffusions of soluble tracer accompanied with the

---

*   Department of Industrial chemistry
** Department of Chemical Engineering, University of Osaka Prefecture

pattern of flow velocity. The correlating equation between the molecular diffusion and the effective dispersion, if obtained, will be useful in the engineering field. In order to obtain this correlation, the governing equation for diffusion and convection must be solved, and experiments must be devised to check the accuracy of the solution or the correlating equation. These requirements motivated many investigators to study the dispersion in the pipe flow analytically and experimentally, initiated by Taylor (1953).

In a series of papers, Taylor (1953, 1954a, b) has treated analytically and experimentally the problem of the effective dispersion of soluble tracer in a fluid flowing through a tube of circular cross-section. For the laminar flow region, the concentration of tracer, c, as a function of distance along the pipe, x, of radial distance from the axis of the pipe, r, and of time, t, is described by the partial differential equation (1).

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial r^2} + \frac{1}{r} \cdot \frac{\partial c}{\partial r} \right) - 2u\left(1 - \frac{r^2}{R^2} \right) \frac{\partial c}{\partial x} , \qquad (1)$$

where u is the average linear velocity, R is the radius of the pipe and D is the molecular diffusion coefficient. As the axial diffusion term, $D \cdot \partial^2 c/\partial x^2$ is able to be neglected for a small-bore tube of long size, Taylor discussed the dispersion by convection together with an ingenious approximation technique for the case where radial diffusion was significant, and further found the correlation between effective (or virtural) dispersion coefficient, E, and molecular diffusion coefficient, D, as $E=R^2 u^2/48D$. Taylor also obtained the conditions $4L/R \gg D/uR \gg \sqrt{48}$, in which the correlation is valid, where L is the length of the pipe over which appreciable changes in concentration occur. Aris (1956, 1959) and Bournia, Coull & Houghton (1961) considered these validity conditions more closely. Aris extended the analysis of Taylor to include non-cylindrical pipes and the effects of axial diffusion. In particular, for cylindrical pipes, he obtained an approximate correlation which indicates that the effective dispersion coefficient is given by the sum of molecular diffusivity and the dispersion coefficient owing to velocity effect, i.e., $E=D+R^2 u^2/48D$ and that validity condition $D/uR > \sqrt{48}$ is not required. Bournia et al. performed displacements with gases flowing in cylindrical pipes and obtained good agreement with Taylor's correlation. Data from this investigation did not agree so well with the theoretical results of Aris in the low velocity range. Bailey and Gegarty (1962) obtained numerical solution of equation (1) by abbreviating as did Taylor, the axial diffusion term, $D \cdot \partial^2 c/\partial x^2$. Its solution was able to explain both Taylor's correlation and their experimental results for a dilute solution of potassium permanganate in water flowing through a capillary tube. Lighthill (1966), after abbreviating the axial diffusion term in equation (1), found a fully exact solution of the resulting equation by an elegant mathematical manipulation. Anathakrishnan, Gill and Barduhn (1965, 1966) obtained finite difference solutions of equation (1) for both large and small values of D/uR. Gill (1967) and Chatwin (1970) carried out numerical calculation of equation (1) more precisely. Evans and Kenney (1965) experimentally measured the dispersion coefficient of a pulse of ethylene injected into nitrogen flowing in a tube. Their data were in good agreement with the theory for large values of dimensionless time, $T=Dt/R^2$. Bruce Hunt (1977) investigated the combination of analytical reasoning and experimental observation for diffusion in fully-developed laminar pipe flow. His results indicated that diffusion in laminar flow depended very much upon the D/uR.

In this paper, numerical experiments were carried out for the case when all the terms in equation (1) are taken into consideration in order to obtain a more general correlation between the effective dispersion coefficient and the molecular diffusivity than the correlations of Taylor and Aris. The numerical method utilized in this work for the numerical experiment was an extended algorithm that had been proposed by us (1969) to solve a non-linear partial differen-

tial equation, governing a one dimensional dispersion model with non-linear chemical reaction rate process. As the average concentration change, computed from the solution of equation (1), at a cross-section of the pipe, the step response curves when a tracer injected into the fully-developed laminar pipe flow with axial and radial molecular diffusions were calculated numerically. The variances of the residence time distribution from the mean residence time were calculated via these step response curves by moment integration method in which integration of the response curve by adding various time weight is made. The effective dispersion coefficient, E, is evaluated by setting the variance equal to E/uR, which is the variance of one dimensional dispersion model. And the dependence of $Re_p \times Sc$ ($=2uR/D$) on the effective dispersion coefficient, E was discussed.

2. Numerical Method

For fully-developed laminar pipe flow, the diffusion-convection equation is given by equation (1). The boundary conditions proposed by Danckwerts (1953), are as follows:

$$t = 0 \quad x > 0 \quad c = 0,$$

$$t > 0 \quad x = 0 \quad D\left(\frac{\partial c}{\partial x}\right)_{x=0} = u(c_{x=0} - c^{in}),$$

$$x = L \quad \left(\frac{\partial c}{\partial x}\right)_{x=L} = 0, \tag{2}$$

$$r = 0 \quad \left(\frac{\partial c}{\partial x}\right)_{r=0} = 0,$$

$$r = R \quad \left(\frac{\partial c}{\partial x}\right)_{r=R} = 0.$$

In dimensionless coordinates equation (1) can be written as follows:

$$\frac{\partial C}{\partial T} = \frac{1}{Pe_x} \cdot \frac{\partial^2 C}{\partial X^2} + \frac{1}{Pe_r}\left(\frac{\partial^2 C}{\partial Y^2} + \frac{1}{Y} \cdot \frac{\partial C}{\partial Y}\right) - 2(1 - Y^2)\frac{\partial C}{\partial X}, \tag{3}$$

with the conditions

$$T = 0 \quad X > 0 \quad C = 0,$$

$$T > 0 \quad X = 0 \quad \left(\frac{\partial C}{\partial X}\right)_{X=0} = Pe_x(C_{x=0} - 1)$$

$$X = 1 \quad \left(\frac{\partial C}{\partial X}\right)_{X=1} = 0, \tag{4}$$

$$Y = 0 \quad \left(\frac{\partial C}{\partial Y}\right)_{Y=0} = 0,$$

$$Y = 1 \quad \left(\frac{\partial C}{\partial Y}\right)_{Y=1} = 0,$$

where $C = c/c^{in}$, $T = ut/L$, $X = x/L$, $Y = r/R$, $Pe_x = uL/D$ and $Pe_r = uR^2/DL = Pe_x \times (R/L)^2$.

Equation (1) is finite-differenced with respect to dimensionless space variables X and Y by second order finite-difference approximation formulae. The fluid in the pipe is divided into N parts in the axial direction and into K parts in the radial direction as shown in Fig. 1. Step input of concentration change is imposed at the input plane, and the output signal (step response) is detected as the concentration change at the output plane. The difference formulae for the axial direction are

Kenji KUBO, Toshiro ARATANI, Akira MISHIMA and Takeo YANO
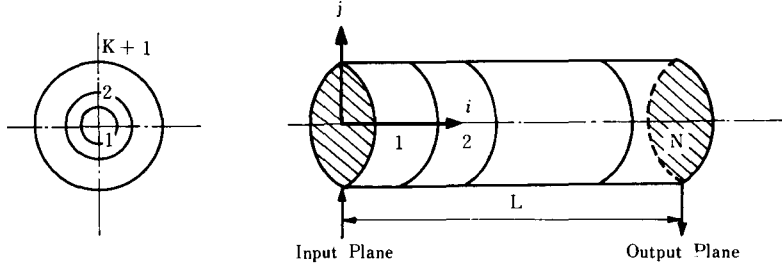
Fig. 1 Finite differenced cells with respect to axial and radial directions (i, j).

$$\frac{\partial^2 C_{i,j}}{\partial X^2} = N^2 (C_{i+1,j} + C_{i-1,j} - 2 C_{i,j}) , \tag{5}$$

$$\frac{\partial C_{i,j}}{\partial X} = \frac{N}{2} (C_{i+1,j} - C_{i-1,j}) , \tag{6}$$

where i and j represent sequential numbers of order for the axial and radial directions, respectively. The formulae in the cases of $i=1$ (at inlet) and $i=N$ (at outlet) are given by us (1969a);

$$i = 1 \quad \frac{\partial C_{i,j}}{\partial T} = \frac{N^2}{4 P e_x} (C_{3,j} - C_{1,j}) - \frac{N}{2} (C_{2,j} - C_{0,j}) , \tag{7}$$

$$i = N \quad \frac{\partial C_{N,j}}{\partial T} = \frac{N^2}{4 P e_x} [C_{N-1,j} - C_{N-3,j} - 4 (C_{N,j} - C_{N-2,j})] - \frac{N}{2} (C_{N-2,j} - 4 C_{N-1,j} + 3 C_{N,j}) . \tag{8}$$

The difference formulae for the radial direction are written as follows;

$$\frac{\partial^2 C_{i,j}}{\partial Y^2} = K^2 (C_{i,j+1} + C_{i,j-1} - 2 C_{i,j}) , \tag{9}$$

$$\frac{\partial C_{i,j}}{\partial Y} = \frac{K}{2} (C_{i,j+1} - C_{i,j-1}) . \tag{10}$$

The formulae in the cases of $j=1$ (at center of the pipe) and $j=K+1$ (at wall of the pipe) are as follows;

$$j = 1 \quad \frac{\partial C_{i,j}}{\partial T} = \frac{2K^2}{P e_r} (C_{i,2} - C_{i,1}) , \tag{11}$$

$$j = K + 1$$

$$\frac{\partial C_{i,j}}{\partial T} = \frac{K^2}{P e_r} (C_{i,K} - C_{i,K+1}) + \frac{K}{P e_r} (C_{i,K+1} - C_{i,K}) . \tag{12}$$

By the difference formulae (5)-(12), equation (3) is represented in various conditions as follows;

$$i = 1 , \ j = 1$$

$$\frac{dC_{i,j}}{dT} = \frac{2K^2}{P e_r} (C_{1,2} - C_{1,1}) + \frac{N^2}{4 P e_x} (C_{3,1} - C_{1,1}) - N \left[1 - \left(\frac{j-1}{K} \right)^2 \right] (C_{2,1} - 1.0) , \tag{13}$$

$i = 1, \; j = 2, \cdots, K$

$$\frac{dC_{i,j}}{dT} = \frac{K^2}{Pe_r}(C_{1,j+1} + C_{1,j-1} - 2C_{1,j}) + \frac{K^2}{2(j-1)Pe_r}(C_{1,j+1} - C_{1,j-1})$$

$$+ \frac{N^2}{4Pe_x}(C_{3,j} - C_{1,j}) - N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{2,j} - 1.0), \qquad (14)$$

$i = 1, \; j = K + 1$

$$\frac{dC_{i,j}}{dT} = \frac{K^2}{Pe_r}(C_{1,j-1} - C_{1,j}) + \frac{K}{Pe_r}(C_{1,j} - C_{1,j-1}) + \frac{N^2}{4Pe_x}(C_{3,j} - C_{1,j})$$

$$- N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{2,j} - 1.0), \qquad (15)$$

$i = 2, \cdots, N-1, \; j = 1$

$$\frac{dC_{i,j}}{dT} = \frac{2K^2}{Pe_r}(C_{i,2} - C_{i,1}) + \frac{N^2}{Pe_x}(C_{i+1,1} + C_{i-1,1} - 2C_{i,1})$$

$$- N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{i+1,1} - C_{i-1,1}), \qquad (16)$$

$i = 2, \cdots, N-1, \; j = 2, \cdots, K$

$$\frac{dC_{i,j}}{dT} = \frac{K^2}{Pe_r}(C_{i,j+1} + C_{i,j-1} - 2C_{i,j}) + \frac{K^2}{2(j-1)Pe_r}(C_{i,j+1} - C_{i,j-1})$$

$$+ \frac{N^2}{Pe_x}(C_{i+1,j} + C_{i-1,j} - 2C_{i,j}) - N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{i+1,j} - C_{i-1,j}),$$

$$\qquad (17)$$

$i = 2, \cdots, N-1, \; j = K + 1$

$$\frac{dC_{i,j}}{dT} = \frac{K^2}{Pe_r}(C_{i,j-1} - C_{i,j}) + \frac{K}{Pe_r}(C_{i,j} - C_{i,j-1})$$

$$+ \frac{N^2}{Pe_x}(C_{i+1,j} + C_{i-1,j} - 2C_{i,j}) - N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{i+1,j} - C_{i-1,j}), \; (18)$$

$i = N, \; j = 1$

$$\frac{dC_{i,j}}{dT} = \frac{2K^2}{Pe_r}(C_{i,2} - C_{i,1}) + \frac{N^2}{4Pe_x}[C_{i-1,1} - C_{i-3,1} - 4(C_{i,1} - C_{i-2,1})]$$

$$- N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{i-2,1} - 4C_{i-1,1} + 3C_{i,1}), \qquad (19)$$

$i = N, \; j = 2, \cdots, K$

$$\frac{dC_{i,j}}{dT} = \frac{K^2}{Pe_r}(C_{i,j+1} + C_{i,j-1} - 2C_{i,j}) + \frac{K^2}{2(j-1)Pe_r}(C_{i,j+1} - C_{i,j-1})$$

$$+ \frac{N^2}{4Pe_x}[C_{i-1,j} - C_{i-3,i} - 4(C_{i,j} - C_{i-2,j})]$$

$$- N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{i-2,j} - 4C_{i-1,j} + 3C_{i,j}), \qquad (20)$$

$i = N, \; j = K + 1$

$$\frac{dC_{i,j}}{dT} = \frac{K^2}{Pe_r}(C_{i,j-1} - C_{i,j}) + \frac{K}{Pe_r}(C_{i,j} - C_{i,j-1})$$

$$+ \frac{N^2}{4Pe_x}[C_{i-1,j} - C_{i-3,j} - 4(C_{i,j} - C_{i-2,j})]$$

$$- N\left[1 - \left(\frac{j-1}{K}\right)^2\right](C_{i-2,j} - 4C_{i-1,j} + 3C_{i,j}). \tag{21}$$

The Kutta-Merson process, summarized by Lapidus and Seinfeld (1971), was employed for integration of a set of these $N \times (K+1)$ ordinary differential equations, because automatic time increment setting gives this process good stability. The step response is expressed as the change in mean concentration through the cross-section at the outlet of the pipe. From the numerical solution of equation (1), the mean concentration is computed at $X=1$ by the integration of equation (22);

$$(\bar{C})_{X=1} = \frac{1}{\pi R^2}\int_0^R (C)_{X=L} \times 2\pi r \; dr = 2\int_0^1 (C)_{X=1} \times Y \; dY. \tag{22}$$

The mean, $\bar{T}$ and variance, $\overline{T^2}$ of residence time distribution are computed from the step response by the moment integration method, which is developed by Danckwerts (1953) and Van der Lann (1958) as follows;

$$\bar{T} = \int_0^\infty (\bar{C})_{X=1} \times T \; dT, \tag{23}$$

$$\overline{T^2} = \int_0^\infty (\bar{C})_{X=1} \times T^2 \; dT / \int_0^\infty (\bar{C})_{X=1} \times T \; dT. \tag{24}$$

3. Results and Discussion

Figure 2 shows the influence of Pe_r $(=(uR/D)(R/L))$ on the shape of the computed step responses, while Pe_x $(=(uR/D)(L/R))$ is fixed at $2^{12}=4096$. The shape of a step response curve is affected by the Pe_r group. The maximum difference on the step response curves for $Pe_r=1$ and $Pe_r=512$ is found to be 35%. When Pe_r decreases, the response becomes similar to that of piston flow. Inversely, increase in Pe_r gives rise to the long tailing on the step response curve.

The numerical integration error, brought about by the method used to compute the step response curve from equation (1) through equations (13)~(22), was examined by the computations with two different numbers of division with respect to space variables. The test indicates that the maximum error on the step responses for $N=10$, $K=10$ and $N=20$, $K=20$ is less than 0.05%. In this algorithm, the computing time to obtain a step response curve is about 10 minutes on a tiny general purpose electronic computer (Memory size = 48K bytes) for which the Gibbson-Mix value is 50μs.

To reveal the influence of Pe_x on the step response, the step responses for several values of Pe_x are shown in Table 1, while the value of Pe_r is held at 4.0. The maximum difference in the step response for $Pe_x=64.0$ and $Pe_x=8.59\times10^9$ $(\cong2^{33})$ is 3%. The influence of Pe_x on the step response is less than that of Pe_r. Therefore, it is valid to neglect the term of $D \cdot \partial^2 C/\partial X^2$ in
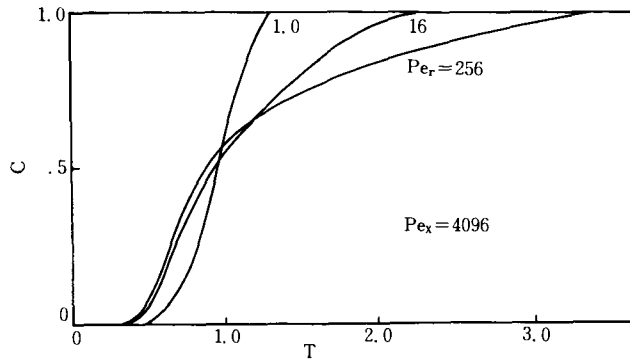
Fig. 2   Step response (concentration change with time) detected at output palne when step input is imposed
on the input plane.

Table 1   Concentration change with time detected at output plane when step input is imposed on the input
plane.

T	C (T)			
	$Pe_x=64$	$Pe_x=256$	$Pe_x=4096$	$Pe_x=8.59 \times 10^9$
0.000	0.0000000	0.0000000	0.0000000	0.0000000
0.320	0.0017484	0.0008601	0.0006581	0.0006458
0.640	0.1706966	0.1502197	0.1434003	0.1429375
0.960	0.5326071	0.5210361	0.5174938	0.5172684
1.280	0.8074933	0.8150576	0.8180043	0.8182153
1.600	0.9461374	0.9648916	0.9725180	0.9730641

Pe_r = 4.0 fixed

equation (1) in order to obtain the approximate solution of equation (1). However, when uL/D becomes small, i.e., when a fluid of relatively large molecular diffusivity is flowing very slowly, the influence of Pe_x on the step response curve can not be neglected.

When the effects of radial and axial diffusions and convection on the dynamics of chemical process equipments involving rate processes, it is not easy to use the simulation model which is expressed by equation (1). Thus,: the one-dimensional dispersion model is widely employed instead of the simulation model expressed by equation (1). By utilizing the moment integration method, the variance of resistence time distribution with the step response of equation (1) was calculated. On the other hand, the variance of the one-dimensional dispersion model is expressed by equation (25). By equating these two variances, the effective dispersion coefficient, E, is estimated as follows, and the effective Peclet group ($Pe_d=2uR/E$) can be computed by E.

$$\bar{T}^2 = \frac{2}{Pe} = \frac{2E}{uL} \qquad (25)$$

$$E = \frac{uL}{2} \times \bar{T}^2 \qquad \text{(This } \overline{T^2} \text{ is from equation (24))} \qquad (26)$$

In figure 3, Pe_d $(=uD/E=2uR/E)$ is plotted against the dimensionless group, $Re \times Sc$ $(=2uR/D)$. The Reynolds number, Re is based on the internal dimameter of the pipe. In addition, the approximate correlations of Taylor (1953) and Aris (1956) are shown in figure 3. These correlations hold on the condition of $4L/R > uR/D = Re \times Sc/2$. The condition of $4L/R = uR/D$ is shown by the dots in figure 3. Under the condition of $4L/R > uR/D$, that is, the left side of the dots in figure 3, the numerical solution shows good agreement with the Aris' and Taylor's correlations. The relative magnitude of axial and radial diffusion terms is thought to be estimated approximately as the ratio of the coefficients, $1/Pe_x$ and $1/Pe_r$. The ratio becomes Pe_r/Pe_x, or $(L/R)^2$. The slender ratio R/L of the pipe, i.e. the ratio of the radius of the pipe to the length of the section tested for its dynamics, will influence the correlation. Our numerical results can be applied for the Pe_d vs. $Re \times Sc$ correlation even with the slender condition by the calculated correlations as is shown in figure 3. Less slender condition will be realized in pipe flow in chemical equipments, while flow in a pipe line approximates the limiting slender condition, and so the axial diffusion term can be eliminated.
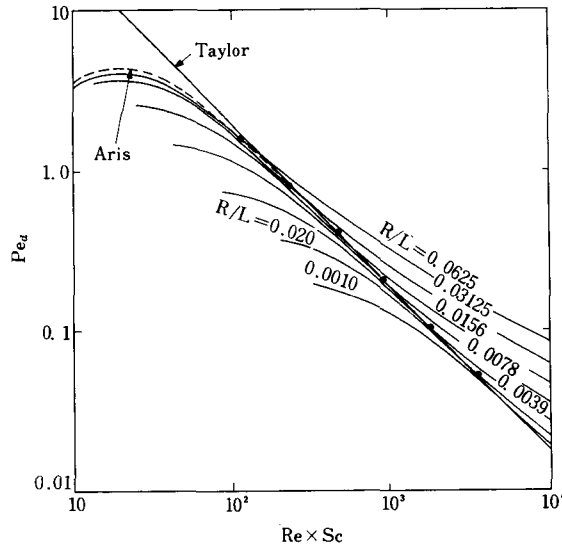

Fig. 3　Pe_d vs. $Re \times Sc$ correlation, when E in Pe_d is computed by moment integration method.

The other method of estimation for an effective dispersion coefficient from the step response, E, is as well derived as the inclination of step response curve at T=1.0, or the peak concentration value of impulse response at T=1.0. Impulse response of the dispersion model is written by equation (27) as derived by us (1968) and summarized by Wen and Fan (1975),

$$\bar{C}_I(T) = \frac{\sqrt{Pe}}{2\sqrt{\pi T}} e^{-\frac{Pe}{4T}(T-1)^2} \tag{27}$$

The slope, m of the step response curve at T=1.0 is derived as equation (28) from equation (27), since time derivative (slope) of step response leads to impulse response,

$$m = \left(\frac{d\bar{C}_s(T)}{dT} \right)_{T=1.0} = (\bar{C}_I(T))_{T=1.0} = \frac{\sqrt{Pe}}{2\sqrt{\pi}} \tag{28}$$
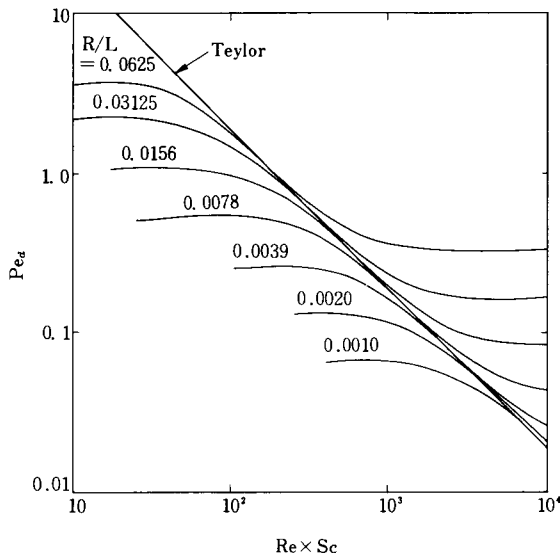
Fig. 4 Pe_d vs. $Re \times Sc$ correlation, when E in Pe_d is computed by slope method.

Then, effective Pe $(=uL/E)$ becomes equal to $4m^2 \pi$, and the effective dispersion coefficient, E is estimated as $uL/4m^2\pi$. Pe_d $(=Pe \times d/L)$ which is obtained by this method, is plotted against dimensionless groups, $Re \times Sc$ $(=2uR/D)$ in figure 4. In this slope method, the results of calculation show good agreement with that of the moment method (figure 3) under the condition of $4L/R > uR/D$. But, when $4L/R < uR/D$, the correlations by the two methods give different results. The Pe_d vs. $Re \times Sc$ correlation has been found, as seen in figures 3 and 4, to be affected considerably by the slender ratio, R/L, which is directly related to the ratio of the coefficients of the radial and axial diffusion terms in equation (1) or (3). Even for such slender case as $R/L = 0.0078$ in figure 3, the correlation deviates from Taylor's for $Re \times Sc > 5 \times 10^2$. By comparing figure 3 (moment integration method) with figure 4 (slope method), the correlation does not seem to be affected by method for the highly slender case, i.e. $R/L < 0.001$, with our numerical experiment range of $Re \times Sc$. In the experiments, the input-output plane length, L, must be as much as 1,000 times the diameter, R, to avoid deviation from the Taylor's correlation, when $Re \times Sc$ in greater than 10^4. For relatively high $Re \times Sc$, the method to evaluate the dispersion coefficient from the experimental step response should be considered, and the dispersion coefficient obtained must be corrected by the factor as the difference in the correlations (figures 3 and 4) produced in this work, according to slender ratio and the evaluating method of the effective dispersion coefficient, used in the respective experimental work.

Reference

Ananthakrishnan V., W. N. Gill and Allen J. Barduhn, A.I.Ch.E. Journal, 11, No. 6, 1063 (1965)

Ananthakrishnan V. and W. N. Gill, A.I.Ch.E. Journal, 12, No.5, 906 (1966)

Aratani T. and T. Yano, Bull. of Univ. of Osaka Pref., Series A. 18, No.2, 415 (1969a)

Aratani T. and T. Yano, Report of the Computer Center Univ. of Tokyo, 2, Nos. 1–2, 127 (1969b)

Aris R., Proc. Royal Soc. (London), A235, 66 (1956)

Aris R., ibid, A252, 538 (1959)

Bailey H. R. and W. B. Gogarty, ibid, **A269**, 352 (1962)

Bournia A., J. Coull and G. Houghton, ibid, **A261**, 227 (1961)

Bruce Hunt, Int. J. Heat Mass Transfer, **20**, 393 (1977)

Chatwin P. C., J. Fluid Mech., **43**, Part 2, 321 (1970)

Chia-Junk Hsu, A.I.Ch.E. Journal, **11**, No.5, 938 (1965)

Danckwerts P. V., Chem. Engg. Sci., **2**, 1 (1953)

Evans E. V. and C. N. Kenney, Proc. Royal Soc. (London), **A284**, 540 (1965)

Eugene H. Wissler, Chem. Engg. Sci., **24**, 527 (1969)

Gill W. N., Proc. Royal Soc. (London), **A298**, 335 (1967)

Lapidus L. and T.H. Seinfeld, Numerical solution of Ordinary Differential Equations, Academic Press, (1971)

Lighthill M. J., J. Inst. Mathe. Applic., **2**, 97 (1966)

Nunge, R. J., Lin, T. -S. and W. N. Gill, J. Fluid Mech. **51**, 363 (1972)

Taylor G., Proc. Royal Soc. (London), **A219**, 186 (1953)

Taylor G., ibid., **A223**, 446 (1954a)

Taylor G., ibid., **A225**, 473 (1954b)

Van der Laan, E. Th., Chem. Engg. Sci., **7**, 187 (1958)

Wen C. Y. and L. T. Fan, Models for Flow Systems and Chemical Reactors, Marcel Dekker, (1975)

Yano T. and T. Aratani, Seigyo Kogaku, **12**, No.1, 19 (1968)