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TAYLOR DIFFUSION IN LAMINAR FLOW IN AN ECCENTRIC ANNULUS

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Abstract—Taylor diffusion in laminar flow along a straight pipe, the cross-section of which is an eccentric annulus, is studied analytically by an exact method which in principle is valid for all values of time. An expression is obtained for the apparent diffusion coefficient K_2 and is evaluated numerically for a wide range of values of the eccentricity ϕ and radius ratio ρ.

The apparent diffusion coefficient is inherently time dependent. However, because of the complexity of the equation that describes laminar convective diffusion in an eccentric annulus, the numerical results are limited to asymptotic values of K_2, that is, to dimensionless times τ large enough for K_2 to be effectively a constant independent of time. To estimate the value τ_m, above which τ is sufficient for this, an approximation which is excellent at large values of ρ, and moderately good for small values of ρ, is employed for small eccentricities. The results of this calculation are correlated by the formula

$$\tau_m \approx 31\, K_2^{0.93}.$$

It is found that eccentricity has an enormous effect on the asymptotic value of K_2. For example, with $\rho = 1.5$, the value of K_2 for $\phi = 0.5$ is approximately 250 times that for a concentric annulus. This remarkable result suggests that eccentricities in the interstices of packed beds may contribute significantly to the scatter of the experimental values of the apparent diffusion coefficient reported previously.

NOMENCLATURE

a, tube radius;

A_n, constant given by equation (7b);

A_{1n}, constant given by equation (32b);

A_{2n}, constant given by equation (32c);

B_n, constant given by equation (7b);

c, half the distance between poles in the bipolar coordinate system;

C, point concentration of solute;

C_m, mean concentration over a cross section;

C_0, initial concentration of solute in slug;

d_n, expansion coefficients defined by equation (40);

D, binary diffusion coefficient;

e, distance between the centers of the two circles in an eccentric annulus;

E, constant given by equation (7b);

f_k, functions defined by equations (13), (19a)–(19c), (20) and (21);

f_{s_1}, f_{t_1}, steady and transient parts respectively of f_1 as defined in equation (29);

F, constant given by equation (7b);

i, $\sqrt{-1}$;

$K_{i,\, i=1,2,\ldots}$, dimensionless coefficients occurring in the generalized dispersion model, equation (16);

K_2', dimensionless dispersion coefficient based on outer radius r_2 as reference length; defined by equation (34);

N_{Pe} Péclét number: $2r_2v_0/D$;

N_{Pe}', dimensionless group analogous to Péclét number: cv_0/D;

O, origin;

P, pressure;

r_1, radius of inner circle of annulus;

r_2, radius of outer circle of annulus;

t, time;

U, dimensionless velocity in z direction defined by equation (7a);

U_0, dimensionless average velocity defined by equation (8);
v_z, velocity of fluid in z direction;
v_0, average velocity in z direction;
v_{max}, maximum velocity in z direction;
V, dimensionless velocity in z direction defined on average velocity as basis by equation (9a);
V_{max}, dimensionless maximum velocity: v_{max}/v_0;
x, y, cartesian transverse coordinates as defined in Fig. 1;
X, dimensionless axial distance defined in equation (9d);
X_s, dimensionless slug length Dz_s/c^2v_0;
X_1, dimensionless axial distance measured from a plane moving with the average velocity of flow, $X_1 = X - \tau$;
Y_n, eigenfunctions defined by equations (39) and (39a);
z, axial coordinate;
z_s, slug length.

Greek letters

α, η coordinate for the inner circle in the eccentric annulus in bipolar system; defined by equation (6b);
β, η coordinate for the outer circle in the eccentric annulus in bipolar system; defined by equation (6c);
γ, inner radius/outer radius as defined in equation (5b); also used as dummy variable in integrations;
ζ, coordinate defined by equation (27);
η, bipolar coordinate; related to cartesian coordinates by equation (4c);
θ, dimensionless concentration defined in equation (9b);
θ_m, dimensionless mean concentration defined by equation (14);
κ, Taylor dispersion coefficient;
$\kappa_{i,\, i=2,3}$, coefficients occurring in a generalized dispersion model;
λ_n, eigenvalues of Sturm–Liouville problem defined byequations (39) and (39a);
μ, viscosity of fluid;
ξ, bipolar coordinate related to cartesian coordinates by equation (4d);
ρ, outer radius/inner radius, $\rho = (1/\gamma)$;
τ, dimensionless time defined in equation (9c);
ϕ, eccentricity as defined by equation (5a);
ψ, constant defined by equation (32a).

G. I. TAYLOR [13] first published a mathematical analysis of unsteady convective diffusion in a straight capillary. He considered a tube initially filled with fluid A into which a slug of arbitrary length of fluid B is introduced and the two fluids flow along the direction of the tube axis and mix with each other. Taylor showed that after a certain amount of time has elapsed, the mean concentration of solute B, which is miscible with and dispersed in fluid A, behaves as though it were diffusing with respect to a a plane moving at the mean speed of flow but with an apparent diffusion, or dispersion coefficient, κ for which he deduced the explicit expression,

$$\kappa = \frac{a^2 v_0^2}{48D}.$$

Aris [1] used an integral approach to generalize Taylor's results to include the effect of axial molecular diffusion which adds linearly to give

$$\kappa = D + \frac{a^2 v_0^2}{48D}.$$

Aris also showed how dispersion in a more general geometry could be analyzed by using the method of moments. These results can be used in conjunction with the one dimensional dispersion model

$$\frac{\partial C_m}{\partial t} + v_0 \frac{\partial C_m}{\partial z} = \kappa \frac{\partial^2 C_m}{\partial z^2}$$

to describe the mean concentration of B as a function of time and axial position.

Carrier [2] and Lighthill [6] also have analyzed the unsteady convective diffusion problem. Carrier obtained a solution for the case involving a periodic concentration input to the system and Lighthill found a solution valid for short periods of time for a step change input in concentration.

Gill [3] generalized Taylor's work by proposing a series expansion about the mean concentration to describe the local concentration distribution. This approach also led to an expression for the dispersion coefficient which reduced to Taylor's result in the limit of large Péclét numbers and to Aris' result for small Péclét numbers where axial molecular diffusion is significant.

It was a natural step to extend the concept of a dispersion model to various other flow geometries. Philip [10] analyzed dispersion in a parallel plate duct to obtain an expression for the dispersion coefficient. Recently Nunge *et al.* [8] have analyzed dispersion in curved tubes and channels and Gill *et al.* [5] solved the problem for a concentric annulus and Jeffrey–Hamel flows using Taylor-type dispersion models. Nunge and Gill [9] discussed the applicability of the results of the analyses of simple geometries to modelling the dispersion process in porous media.

Gill and Sankarasubramanian [4] have shown recently that the series expansion mentioned previously provides an exact solution of the unsteady laminar convective diffusion problem for flow in a circular tube and that the apparent diffusion coefficient varies in a predictable way with time. A generalized dispersion model for the area average concentration C_m which is given by

$$\frac{\partial C_m}{\partial t} + v_0 \frac{\partial C_m}{\partial z} = \sum_{i=2}^{\infty} \kappa_i(t) \frac{\partial i C_m}{\partial z^i}$$

evolves as a natural consequence of their analysis. Obviously, this model contains a set of apparent diffusion coefficients κ_i, the principal component of which is κ_2 and this coefficient is identical to κ in Taylor's analysis. It is the purpose of the present work to extend this technique to the analysis of Taylor diffusion in an eccentric annulus. An expression for the dimensionless form of κ_2, K_2, which appears as one of the eigenvalues in the problem, will be developed and the influence of the radius ratio and eccentricity parameter on the asymptotic value of K_2 will be determined. The results show that this dispersion coefficient is surprisingly sensitive to the degree of eccentricity of the system. This may have significant practical implications for transport processes in porous media.

ANALYSIS

For the eccentric annulus, shown in Fig. 1, the boundary conditions cannot be expressed in cylindrical coordinates at a constant value of one independent space variable. This difficulty can be overcome by using a bipolar coordinate system which is discussed in detail by Moon and Spencer [7], and has been used in previous analyses of transport processes in eccentric annuli.

For fully developed steady laminar flow in the z direction of a fluid with constant physical properties and no transverse velocity components, the convective diffusion equation is

$$\frac{\partial C}{\partial t} + v_z \frac{\partial C}{\partial z} = D\left[\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2}\right]. \quad (1)$$

Consider the dispersion of a slug which is initially z_s units in length and of uniform concentration C_0. For this, the boundary and

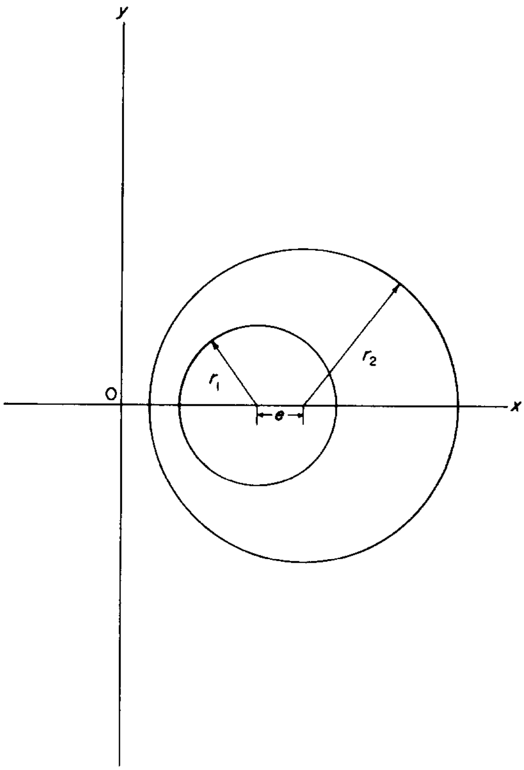

FIG. 1. Eccentric annulus geometry.

initial conditions may be written as

$$\left.\begin{aligned} &C(0,z,x,y) = C_0, \quad |z| \leqslant \tfrac{1}{2}z_s \\ &C(0,z,x,y) = 0, \quad |z| > \tfrac{1}{2}z_s \\ &C(t,\infty,x,y) = 0 \\ &\left.\frac{\partial C}{\partial n}\right|_{\text{inner wall}} = \left.\frac{\partial C}{\partial n}\right|_{\text{outer wall}} = 0 \end{aligned}\right\} \quad (2)$$

where n represents the direction normal to the walls. Also, by symmetry,

$$\left.\frac{\partial C}{\partial y}\right|_{y=0} = 0. \quad (2a)$$

The bipolar coordinates (ξ, η) are defined by

$$x + iy = i\,c\cot\left(\frac{\xi + i\eta}{2}\right) \quad (3)$$

from which one may derive the following relationships:

$$x = \frac{c\sinh\eta}{\cosh\eta - \cos\xi} \quad (4a)$$

$$y = \frac{c\sin\xi}{\cosh\eta - \cos\xi} \quad (4b)$$

$$e^{2\eta} = \frac{y^2 + (x+c)^2}{y^2 + (x-c)^2} \quad (4c)$$

$$\tan\xi = \frac{2yc}{x^2 + y^2 - c^2} \quad (4d)$$

$$y^2 + (x - c\coth\eta)^2 = \frac{c^2}{\sinh^2\eta}. \quad (4e)$$

Equation (4e) shows that lines of constant η are circles in the xy plane with center $(c \coth \eta, 0)$ and radius $(c/\sinh \eta)$. So, in the (ξ, η) system, the inner circle may be represented by a line $\eta = \alpha$ and the outer circle by a line $\eta = \beta$.

Defining

$$\phi = \frac{e}{r_2 - r_1} \quad (5a)$$

$$\gamma = \frac{r_1}{r_2} \quad (5b)$$

the following results are established immediately:

$$c = r_1 \sinh\alpha = r_2 \sinh\beta \quad (6a)$$

$$\cosh\alpha = \frac{1}{\gamma}\frac{\gamma(1+\phi^2) + (1-\phi^2)}{2\phi} \quad (6b)$$

$$\cosh\beta = \frac{\gamma(1-\phi^2) + (1+\phi^2)}{2\phi}. \quad (6c)$$

Using the above transformations, Snyder and Goldstein [12] obtained the laminar flow steady state velocity distribution in dimensionless form as

$$U = F + E\eta - \frac{\coth\eta}{2} + \sum_{n=1}^{\infty}\{A_n e^{n\eta} + (B_n - \coth\eta)e^{-n\eta}\}\cos n\xi \quad (7)$$

where

$$U = \frac{v_z}{\left[\frac{-c^2}{\mu}\frac{dP}{dz}\right]} \tag{7a}$$

and F, E, A_n, B_n are given as:

$$F = \frac{\alpha \coth \beta - \beta \coth \alpha}{2(\alpha - \beta)};$$

$$E = \frac{\coth \alpha - \coth \beta}{2(\alpha - \beta)}$$

$$A_n = \frac{\coth \alpha - \coth \beta}{e^{2n\alpha} - e^{2n\beta}};$$

$$B_n = \frac{e^{2n\alpha} \coth \beta - e^{2n\beta} \coth \alpha}{e^{2n\alpha} - e^{2n\beta}} \tag{7b}$$

The element of area dA in the (ξ, η) system is given by

$$dxdy = dA = \frac{c^2}{(\cosh \eta - \cos \xi)^2} d\xi d\eta$$

and the average dimensionless velocity is given by

$$U_0 = \frac{\int_\alpha^\beta \int_0^\pi \frac{c^2 U \, d\xi d\eta}{(\cosh \eta - \cos \xi)^2}}{\int_\alpha^\beta \int_0^\pi \frac{c^2 d\xi d\eta}{(\cosh \eta - \cos \xi)^2}}$$

$$= \frac{2}{\pi} \frac{1}{[\mathrm{cosech}^2 \alpha - \mathrm{cosech}^2 \beta]} \int_\alpha^\beta \int_0^\pi \frac{U \, d\xi \, d\eta}{(\cosh \eta - \cos \xi)^2} \tag{8}$$

where the integration with respect to ξ is performed in the interval $(0, \pi)$ instead of $(0, 2\pi)$ because of symmetry. New dimensionless variables may be defined as:

$$V = \frac{v_z}{v_0} = \frac{U}{U_0} \tag{9a}$$

$$\theta = \frac{C}{C_0} \tag{9b}$$

$$\tau = \frac{Dt}{c^2} \tag{9c}$$

$$X = \frac{Dz}{c^2 v_0} \tag{9d}$$

so that equation (1), and the boundary conditions (2), may be transformed to the bipolar system and represented in dimensionless form as:

$$\frac{\partial \theta}{\partial \tau} + V(\xi, \eta)\frac{\partial \theta}{\partial X} = (\cosh \eta - \cos \xi)^2 \times \left\{\frac{\partial^2 \theta}{\partial \xi^2} + \frac{\partial^2 \theta}{\partial \eta^2}\right\} + \frac{1}{N_{Pe}'^2}\frac{\partial^2 \theta}{\partial X^2} \tag{10}$$

with

$$\left.\begin{aligned}
&\theta(0, X, \xi, \eta) = 1, \quad |X| \leqslant \tfrac{1}{2} X_s \\
&\theta(0, X, \xi, \eta) = 0, \quad |X| > \tfrac{1}{2} X_s \\
&\theta(\tau, \infty, \xi, \eta) = 0 \\
&\left.\frac{\partial \theta}{\partial \eta}\right|_{\eta=\alpha} = \left.\frac{\partial \theta}{\partial \eta}\right|_{\eta=\beta} = 0 \\
&\left.\frac{\partial \theta}{\partial \xi}\right|_{\xi=0} = \left.\frac{\partial \theta}{\partial \xi}\right|_{\xi=\pi} = 0
\end{aligned}\right\} \tag{11}$$

where

$$N_{Pe}' = \frac{cv_0}{D} = (\tfrac{1}{2}\sinh \beta) N_{Pe} \tag{12a}$$

and N_{Pe} is the Péclét number defined as

$$N_{Pe} = \frac{2r_2 v_0}{D}. \tag{12b}$$

Now, let

$$\theta = \theta_m(\tau, X) + \sum_{k=1}^{\infty} f_k(\tau, \xi, \eta)\frac{\partial^k \theta_m}{\partial X^k} \tag{13}$$

where the dimensionless mean concentration θ_m is,

$$\theta_m(\tau, X) = \frac{\displaystyle\int_\alpha^\beta\int_0^\pi \frac{\theta c^2 d\xi d\eta}{(\cosh\eta - \cos\xi)^2}}{\displaystyle\int_\alpha^\beta\int_0^\pi \frac{c^2 d\xi d\eta}{(\cosh\eta - \cos\xi)^2}}$$

$$= \frac{2}{\pi}\frac{1}{[\mathrm{cosech}^2\alpha - \mathrm{cosech}^2\beta]}\int_\alpha^\beta\int_0^\pi \frac{\theta d\xi d\eta}{(\cosh\eta - \cos\xi)^2}. \tag{14}$$

When equation (13) is substituted into equation (10), the result is

$$\frac{\partial\theta_m}{\partial\tau} + \sum_{k=1}^{\infty}\left\{\frac{\partial f_k}{\partial\tau}\frac{\partial^k\theta_m}{\partial X^k} + f_k\frac{\partial^{k+1}\theta_m}{\partial\tau\partial X^k}\right\} + V\left\{\frac{\partial\theta_m}{\partial X} + \sum_{k=1}^{\infty} f_k\frac{\partial^{k+1}\theta_m}{\partial X^{k+1}}\right\}$$

$$= (\cosh\eta - \cos\xi)^2\left\{\sum_{k=1}^{\infty}\left(\frac{\partial^2 f_k}{\partial\xi^2} + \frac{\partial^2 f_k}{\partial\eta^2}\right)\frac{\partial^k\theta_m}{\partial X^k}\right\} + \frac{1}{N_{Pe}'^2}\left\{\frac{\partial^2\theta_m}{\partial X^2} + \sum_{k=1}^{\infty} f_k\frac{\partial^{k+2}\theta_m}{\partial X^{k+2}}\right\}. \tag{15}$$

To develop an exact dispersion model, equation (10) is multiplied throughout by the elemental area

$$\frac{c^2 d\xi d\eta}{(\cosh\eta - \cos\xi)^2}$$

and integrated over the cross section of the eccentric annulus. If one then introduces equation (13) into the result, the following generalized dispersion model is obtained:

$$\frac{\partial\theta_m}{\partial\tau} = \sum_{i=1}^{\infty} K_i(\tau)\frac{\partial^i\theta_m}{\partial X^i}. \tag{16}$$

By differentiating this with respect to X, k times, we obtain

$$\frac{\partial^{k+1}\theta_m}{\partial\tau\partial X^k} = \sum_{i=1}^{\infty} K_i(\tau)\frac{\partial^{i+k}\theta_m}{\partial X^{i+k}}. \tag{17}$$

It should be emphasized that the dispersion model, given by equation (16), is a direct consequence of integrating the convective diffusion equation (10) and does not involve any arbitrary assumptions.

Equations (16) and (17) are substituted into equation (15) to give, after some rearrangement,

$$\left\{\frac{\partial f_1}{\partial\tau} - (\cosh\eta - \cos\xi)^2\left(\frac{\partial^2 f_1}{\partial\xi^2} + \frac{\partial^2 f_1}{\partial\eta^2}\right) + V + K_1\right\}\frac{\partial\theta_m}{\partial X}$$

$$+ \left\{\frac{\partial f_2}{\partial\tau} - (\cosh\eta - \cos\xi)^2\left(\frac{\partial^2 f_2}{\partial\xi^2} + \frac{\partial^2 f_2}{\partial\eta^2}\right) + Vf_1 + K_1 f_1 + K_2 - \frac{1}{N_{Pe}'^2}\right\}\frac{\partial^2\theta_m}{\partial X^2}$$

$$+ \sum_{k=1}^{\infty}\left\{\frac{\partial f_{k+2}}{\partial\tau} - (\cosh\eta - \cos\xi)^2\left(\frac{\partial^2 f_{k+2}}{\partial\xi^2} + \frac{\partial^2 f_{k+2}}{\partial\eta^2}\right)\right.$$

$$\left. + Vf_{k+1} + \sum_{i=1}^{k+2} K_i f_{k+2-i} - \frac{1}{N_{Pe}'^2} f_k\right\}\frac{\partial^k\theta_m}{\partial X^k} = 0 \tag{18}$$

with the understanding that $f_0 = 1$. If equation (18) is satisfied by setting the coefficients of $(\partial^k \theta_m / \partial X^k)$ to zero for $k = 1, 2, 3 \ldots$ the following system of equations is generated:

$$\frac{\partial f_1}{\partial \tau} = (\cosh \eta - \cos \xi)^2 \left\{ \frac{\partial^2 f_1}{\partial \xi^2} + \frac{\partial^2 f_1}{\partial \eta^2} \right\} - V(\xi, \eta) - K_1(\tau) \quad (19a)$$

$$\frac{\partial f_2}{\partial \tau} = (\cosh \eta - \cos \xi)^2 \left\{ \frac{\partial^2 f_2}{\partial \xi^2} + \frac{\partial^2 f_2}{\partial \eta^2} \right\} - V(\xi, \eta) f_1 + \frac{1}{N_{Pe}'^2} - K_1(\tau) f_1 - K_2(\tau) \quad (19b)$$

and

$$\frac{\partial f_{k+2}}{\partial \tau} = (\cosh \eta - \cos \xi)^2 \left\{ \frac{\partial^2 f_{k+2}}{\partial \xi^2} + \frac{\partial^2 f_{k+2}}{\partial \eta^2} \right\} - V(\xi, \eta) f_{k+1} + \frac{1}{N_{Pe}'^2} f_k - \sum_{i=1}^{k+2} K_i f_{k+2-i}, \mathrm{k} = 1, 2, 3 \ldots \quad (19c)$$

From the boundary conditions (11), the $f_k(\tau, \xi, \eta)$ must satisfy the following conditions:

$$f_k(0, \xi, \eta) = 0 \quad (20)$$

$$\left.\begin{aligned} \left.\frac{\partial f_k}{\partial \eta}\right|_{\eta=\alpha} &= \left.\frac{\partial f_k}{\partial \eta}\right|_{\eta=\beta} = 0 \\ \left.\frac{\partial f_k}{\partial \xi}\right|_{\xi=0} &= \left.\frac{\partial f_k}{\partial \xi}\right|_{\xi=\pi} = 0. \end{aligned}\right\} \quad (21)$$

In addition, the definition of θ_m requires that

$$\int_\alpha^\beta \int_0^\pi \frac{f_k(\tau, \xi, \eta) \mathrm{d}\xi \mathrm{d}\eta}{(\cosh \eta - \cos \xi)^2} = 0. \quad (22)$$

If we multiply equation (19a) throughout by $\dfrac{1}{(\cosh \eta - \cos \xi)^2}$, integrate with respect to ξ from $\xi = 0$ to $\xi = \pi$ and with respect to η from $\eta = \alpha$ to $\eta = \beta$, invoke the conditions represented by equations (21) and (22), we get

$$\int_\alpha^\beta \int_0^\pi \frac{V(\xi, \eta) + K_1(\tau)}{(\cosh \eta - \cos \xi)^2} \mathrm{d}\xi \mathrm{d}\eta = 0. \quad (23)$$

By using equations (8) and (9a) in conjunction with equation (23) one can show that

$$K_1 = -1. \quad (24a)$$

If a similar procedure is applied to equations (19b) and (19c) we get the following expressions for the eigenvalues K_2 and K_{k+2}, $k = 1, 2, \ldots$ as:

$$K_2(\tau) = \frac{1}{N_{Pe}'^2} - \frac{2}{\pi} \frac{1}{[\mathrm{cosech}^2 \alpha - \mathrm{cosech}^2 \beta]} \int_\alpha^\beta \int_0^\pi \frac{V f_1}{(\cosh \eta - \cos \xi)^2} \mathrm{d}\xi \mathrm{d}\eta \quad (24b)$$

and

$$K_{k+2}(\tau) = -\frac{2}{\pi} \frac{1}{[\mathrm{cosech}^2 \alpha - \mathrm{cosech}^2 \beta]} \int_\alpha^\beta \int_0^\pi \frac{V f_{k+1}}{(\cosh \eta - \cos \xi)^2} \mathrm{d}\xi \mathrm{d}\eta \quad (24c)$$

$$k = 1, 2, 3 \ldots$$

Since it is difficult to solve for f_2, or the higher order functions, and since it has been shown by Gill and Sankarasubramanian [4] that truncating the series in equation (16) after the first two terms causes a negligible amount of error for the case of a straight tube, we will adopt the same procedure here. The truncated form of equation (16) after substituting for K_1 from equation (24a) is:

$$\frac{\partial \theta_m}{\partial \tau} + \frac{\partial \theta_m}{\partial X} = K_2(\tau) \frac{\partial^2 \theta_m}{\partial X^2}. \quad (25)$$

Except for the time dependence of K_2, which makes it valid for small τ, equation (25) is the equation Taylor used in his original work. This is to be solved with the boundary conditions

$$\left.\begin{aligned}\theta_m(0, X) &= 1, \quad |X| \leqslant \tfrac{1}{2}X_s \\ \theta_m(0, X) &= 0. \quad |X| > \tfrac{1}{2}X_s \\ \text{and} \\ \theta_m(\tau, \infty) &= 0.\end{aligned}\right\} \quad (26)$$

If one defines

$$\zeta = \int_0^\tau K_2(\gamma)\mathrm{d}\gamma \quad (27)$$

the well-known solution to equations (25) and (26) may be written as

$$\theta_m = \frac{1}{2}\left[\operatorname{erf}\left\{\frac{\frac{1}{2}X_s + X_1}{2\sqrt{\zeta}}\right\} + \operatorname{erf}\left\{\frac{\frac{1}{2}X_s - X_1}{2\sqrt{\zeta}}\right\}\right] \quad (28)$$

where

$$X_1 = X - \tau. \quad (28a)$$

Equation (24b) shows that K_2 depends on f_1, but solving for the complete f_1 function analytically is a difficult task and so we will solve only for the steady-state part, f_{s_1}, of this function. At a later stage, an approximate theory will be given which enables one to determine the magnitude of τ necessary for $f_1 \approx f_{s_1}$, for small eccentricities, and we will thus determine the region of applicability of the present analysis. Under these conditions, the dispersion coefficient K_2 has reached its asymptotic value and the constant dispersion coefficient model is valid.

The f_1 function may be separated into steady and transient parts, such that

$$f_1 = f_{s_1}(\xi, \eta) + f_{t_1}(\tau, \xi, \eta). \quad (29)$$

The equation for f_{s_1} is then, upon using the value of K_1 from equation (24a),

$$\frac{\partial^2 f_{s_1}}{\partial \xi^2} + \frac{\partial^2 f_{s_1}}{\partial \eta^2} = \frac{V - 1}{(\cosh \eta - \cos \xi)^2} \quad (30)$$

The appropriate boundary conditions are

$$\left.\begin{aligned}\left.\frac{\partial f_{s_1}}{\partial \eta}\right|_{\eta=\alpha} &= \left.\frac{\partial f_{s_1}}{\partial \eta}\right|_{\eta=\beta} = 0 \\ \left.\frac{\partial f_{s_1}}{\partial \xi}\right|_{\xi=0} &= \left.\frac{\partial f_{s_1}}{\partial \xi}\right|_{\xi=\pi} = 0\end{aligned}\right\} \quad (31)$$

Sankarasubramanian [11] has solved equation (30), along with equation (31) by using a cosine transform technique in the ξ coordinate. The solution is

$$f_{s_1} = \frac{1}{\pi}\left[\psi + \int_\alpha^\eta (\eta - \gamma)\int_0^\pi \frac{V(\xi', \gamma) - 1}{(\cosh \gamma - \cos \xi')^2}\,\mathrm{d}\xi'\mathrm{d}\gamma\right.$$
$$+ \frac{2}{\pi}\sum_{n=1}^{\infty}\left\{\left[A_{1n} + \frac{1}{2n}\int_\alpha^\eta \mathrm{e}^{-n\gamma}\right.\right.$$
$$\left.\times \int_0^\pi \frac{[V(\xi', \gamma) - 1]\cos n\xi'\mathrm{d}\xi'}{(\cosh \gamma - \cos \xi')^2}\mathrm{d}\gamma\right]\mathrm{e}^{n\eta}$$
$$+\left(A_{2n} - \frac{1}{2n}\int_\alpha^\eta \mathrm{e}^{n\gamma}\right.$$
$$\left.\left.\times \int_0^\pi \frac{[V(\xi', \gamma) - 1]\cos n\xi'\mathrm{d}\xi'}{(\cosh \gamma - \cos \xi')^2}\mathrm{d}\gamma\right)\mathrm{e}^{-n\eta}\right\}\cos n\xi \quad (32)$$

where

$$\psi = \frac{1}{2(\alpha - \beta)}\int_\alpha^\beta\int_0^\pi \{\gamma^2 - 2\gamma\beta - \xi'^2\}$$
$$\frac{V(\xi', \gamma) - 1}{(\cosh \gamma - \cos \xi')^2}\mathrm{d}\xi'\mathrm{d}\gamma \quad (32a)$$

$$A_{1n} = \frac{1}{2n(\mathrm{e}^{2n\alpha} - \mathrm{e}^{2n\beta})}\left[\mathrm{e}^{2n\beta}\int_\alpha^\beta \mathrm{e}^{-n\gamma}\right.$$
$$\times \int_0^\pi \frac{[V(\xi', \gamma) - 1]\cos n\xi'}{(\cosh \gamma - \cos \xi')^2}\mathrm{d}\xi'\mathrm{d}\gamma$$

$$+\int_{\alpha}^{\beta} e^{n\gamma}\int_{0}^{\pi}\frac{[V(\xi',\gamma)-1]\cos n\xi'}{(\cosh\gamma-\cos\xi')^2}\,d\xi'd\gamma\Bigg] \quad (32b)$$

and

$$A_{2n} = e^{2n\alpha}A_{1n}. \quad (32c)$$

$f_{s_1}(\xi, \eta)$, as given in equation (32), can be used in equation (24b), to determine asymptotic values of the dimensionless dispersion coefficient K_2.

DISCUSSION OF RESULTS

K_2 was evaluated numerically for ϕ ranging from 0·005 to 0·9 and ρ ranging from 1·25 to 100. In these computations, the contribution to K_2 due to axial molecular diffusion, represented by $(1/N_{Pe}'^2)$, has been omitted since this contribution can be added directly to the values presented.

K_2 was evaluated from equation (24b) by using double precision and Simpson's rule for the numerical integrations. The number of significant figures was determined by using different step lengths at the extreme values of the parameters. The accuracy of the dimensionless velocity and the f_{s_1} function was estimated by varying the number of terms used in the series for these functions.

At the higher eccentricities, both the series for the f_{s_1} function and that for the dimensionless velocity converge very slowly. Also, a larger number of steps is required to obtain sufficient accuracy in the numerical integrations. This requires prohibitively large amounts of computer time, and so only a limited number of results were obtained for $\phi = 0{\cdot}9$. At this value of eccentricity this problem is very severe and the results obtained are accurate only to within 5 per cent, but this is satisfactory to indicate trends reliably.

The dimensionless dispersion coefficient K_2 is based on c as reference length,

$$K_2 = \frac{\kappa D}{c^2 v_0^2}. \quad (33)$$

Because its physical significance is much clearer for presentation and comparison purposes, the outer radius, r_2 is chosen as the reference length and a new dimensionless dispersion coefficient K_2' is defined as

$$K_2' = \frac{\kappa D}{r_2^2 v_0^2} = K_2 \sinh^2\beta. \quad (34)$$

The axial diffusion contribution which has been omitted, correspondingly becomes

$$\frac{1}{N_{Pe}'^2}\sinh^2\beta = \frac{4}{N_{Pe}^2}.$$

The calculated values of K_2' have been tabulated in [11] and are plotted against ρ with ϕ as the parameter in Fig. 2. It is seen that eccentricity can have an enormous effect on the dispersion process. The values of K_2' for the

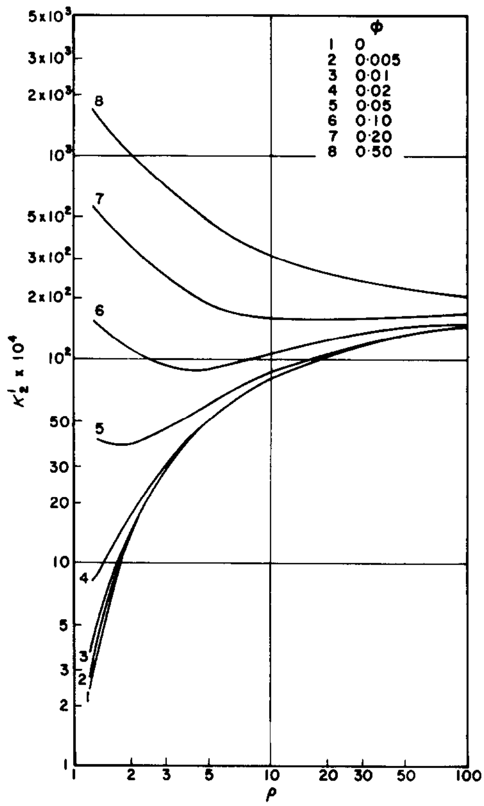

FIG. 2. Behavior of dispersion coefficient, $K_2' \times 10^4$, as a function of radius ratio, ρ, and eccentricity, ϕ.

concentric annulus from the results of Gill *et al.* [5] also are plotted in the same figure.

$\phi = 0$ cannot be used for calculations in bipolar co-ordinates since this is a singular point for the transformation given in equation (3). However, the results obtained for $\phi = 0{\cdot}005$ by the present approach are in excellent agreement with those for $\phi = 0$ given in [5] for all radius ratios investigated.

eccentric annulus with $\phi = 0{\cdot}1$ and $0{\cdot}5$ respectively; $\rho = 5$ in all cases. The eccentric annulus profiles are given at a section at $\xi = 0$ and $\xi = \pi$, that is, on the X axis.

Note that the velocity profiles become more asymmetric as the eccentricity increases. Differences in the velocity at different angular locations, and over the cross section at any one angular location become more pronounced

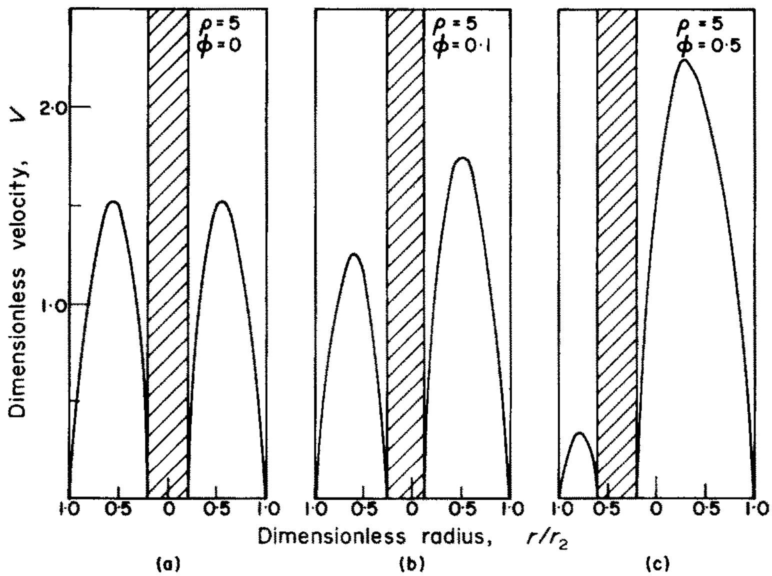

FIG. 3. Comparison of velocity profiles for increasing eccentricity for a given radius ratio, $\rho = 5$ at sections at $\xi = 0$ and $\xi = \pi$.

Although the complexities of the flow pattern in an eccentric annulus make an exact interpretation of the results represented in Fig. 2 extremely difficult, an attempt is made here to explain qualitatively, the behavior of the dispersion coefficient.

The first characteristic of interest is the increase in the value of the dispersion coefficient with increasing eccentricity for any radius ratio. This is explained by considering the velocity profiles shown in Fig. 3. Figure 3a gives the dimensionless velocity distribution for a concentric annulus and Figs. 3b and c for the with increasing eccentricity and cause an increase in the value of the dispersion coefficient. However, as the velocity differences become greater with increasing eccentricity, transverse diffusion starts playing a more important role in suppressing the extent of axial dispersion and above $\phi = 0{\cdot}5$, the dispersion coefficient shows a definite decrease with increasing eccentricity as confirmed by the limited results available at $\phi = 0{\cdot}9$. It is seen also that the relative increase in the dispersion coefficient with increasing eccentricity is far greater for small ρ than for large ρ. The reason is as follows: for a fixed value of the outer radius r_2, the radius of the inner circle and hence its size get smaller with increasing ρ. A displacement of this inner circle from a concentric position when it is smaller has less effect on the velocity field and therefore on the dispersion process. In fact, when $\rho = 100$, the inner circle is but a spot inside the outer circle and there is practically no effect on the dispersion coefficient as ϕ increases from 0 to 0·1 as observed in Fig. 2.

Another characteristic which should be noted in Fig. 2 is the minimum exhibited in the plots for $\phi = 0{\cdot}05$, 0·1 and 0·2. Such a minimum may very well exist for other eccentricities also, but it was not observed in the range of radius ratios investigated. Note also that this minimum shifts toward larger values of ρ as ϕ increases.

In an effort to explain this behavior, the dimensionless velocity, $V_{max} = (v_{max}/v_0)$, was computed for the same range of the parameters ϕ and ρ for which K_2' has been calculated. The plot of V_{max} vs. ρ with eccentricity ϕ as parameter is presented as Fig. 4. It is seen that the behavior of K_2', and V_{max} vs. ρ is similar for each

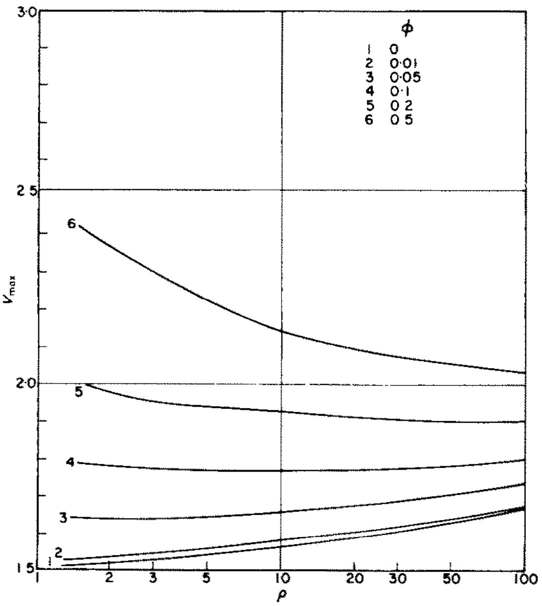

FIG. 4. Behavior of dimensionless maximum velocity, V_{max}, as a function of radius ratio, ρ, and eccentricity, ϕ.

value of eccentricity and in each case the curves exhibit minima at about the same value of ρ. Since the extent of axial dispersion, as pointed out previously, depends strongly on velocity differences over the cross section and since the ratio of maximum velocity to average velocity is an index of such differences, the dispersion coefficient behaves in sympathy with this ratio.

A qualitative perspective of the magnitude of the effect of eccentricity on the dispersion process can be obtained from Fig. 5 in which eccentric annuli are drawn to scale and the respective values of K_2' for each annulus is shown. Most dramatic, perhaps, is the 25-fold increase in the dispersion coefficient which occurs when ϕ is changed from 0 to 0·1 at $\rho = 1{\cdot}5$ and the eccentricity is barely perceptible by eye.

An important objective of studies of dispersion in relatively simple geometries is to suggest effects which may be significant in very complicated systems such as packed beds which occur in commercial applications. It seems relatively safe to speculate, on the basis of the very large effects observed in the present work, that eccentricities in the interstitial flow passages created by the arrangement of the particles in packed systems may contribute significantly to the scatter of the data which have appeared in the literature.

APPROXIMATION FOR SMALL ECCENTRICITIES

A deeper understanding of dispersion in eccentric annuli can be obtained if the mathematical aspects of the problem are reduced by considering an important asymptotic case. In the limiting case as $\phi \to 0$,

$$\frac{\partial C}{\partial \xi} \to \frac{\partial C}{\partial \theta} \text{ and } \frac{\partial^2 C}{\partial \xi^2} \to \frac{\partial^2 C}{\partial \theta^2}$$

where θ is the angular coordinate in a cylindrical coordinate system with its origin at the common center of the two circles. For a concentric annulus, due to angular symmetry,

$$\frac{\partial C}{\partial \theta} = \frac{\partial^2 C}{\partial \theta^2} \equiv 0$$

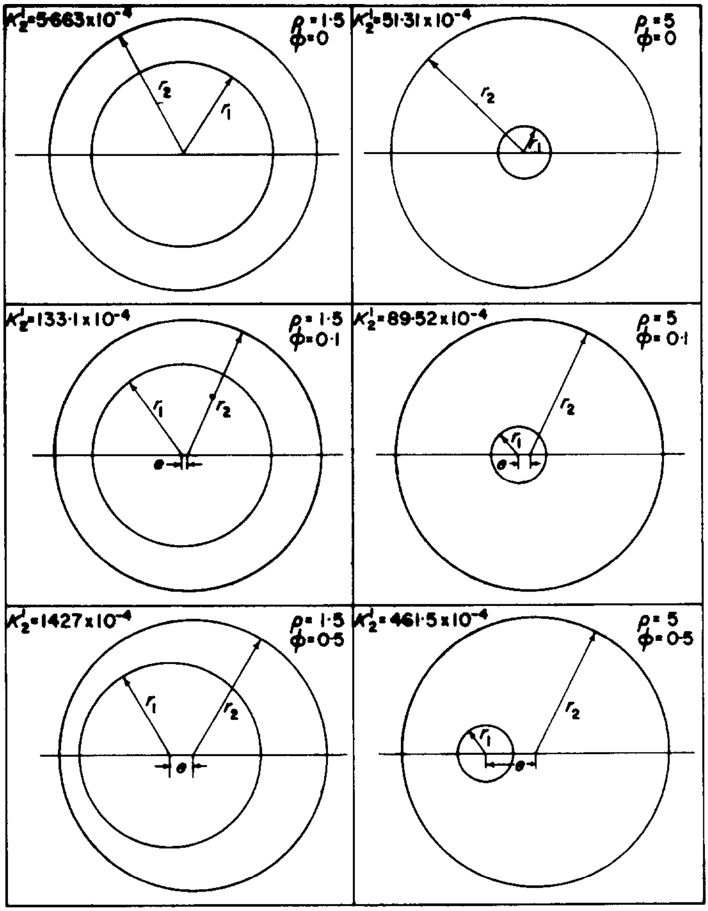

FIG. 5. Scale drawings illustrating the effect of radius ratio, ρ, and eccentricity, ϕ, on dispersion coefficient, K_2'.

and for the eccentric annulus with small eccentricities, the ξ dependence may be neglected. Accordingly, the ξ dependent part of the f_{s_1} function is small and may be neglected in using equation (24b) to calculate K_2. This approximation was used to calculate K_2' for small eccentricities in the region $\phi = 0{\cdot}005$–$0{\cdot}1$; it was found, as expected, that the approximate values approach the exact value as $\phi \to 0$. It was also found that the approximation is increasingly better for larger ρ. Table 1 gives some sample values of K_2', expressed as a percentage of the exact value, which were calculated using the small ϕ approximation. This approximation will now be used to establish

Table 1. Dispersion coefficient K_2' calculated using approximation for small eccentricities expressed as a percentage of the exact value

ϕ \ ρ	1·25	5·0	100·0
0·005	82·92	99·8	99·98
0·02	23·3	96·88	99·67
0·1	1·24	55·54	92·48

criteria for the dimensionless time required for the steady state dispersion coefficient to be valid. In the calculation of K_2, only the steady state part, f_{s_1}, of the f_1 function has been used and therefore the results apply only for large values of time. Consequently it is desirable to estimate the magnitude of τ required for the steady state function to be dominant so that K_2 becomes independent of time. To do this we shall employ the small eccentricity approximation to solve for f_{t_1}

MINIMUM TIME REQUIRED FOR THE DISPERSION COEFFICIENT TO BECOME CONSTANT

The equation for the transient part, f_{t_1}, of the f_t function is:

$$\frac{\partial f_{t_1}}{\partial \tau} = (\cosh \eta - \cos \xi)^2 \left[\frac{\partial^2 f_{t_1}}{\partial \xi^2} + \frac{\partial^2 f_{t_1}}{\partial \eta^2}\right]. \quad (35)$$

This is a very difficult equation to solve exactly and so we resort to approximations. For the case of small eccentricities, both cosh α and cosh β are very large compared to unity [see equations (6b) and (6c)] which is the upper bound on $|\cos \xi|$ and so, $\cos \xi$ may be neglected compared to $\cosh \eta$. In addition, $(\partial^2 f_{t_1}/\partial \xi^2)$ may be dropped for reasons discussed in the last section and so we may write, for small eccentricities.

$$\frac{\partial f_{t_1}}{\partial \tau} = \cosh^2\eta \frac{\partial^2 f_{t_1}}{\partial \eta^2} \quad (36)$$

with the conditions,

$$\left.\begin{aligned} \left.\frac{\partial t_{t_1}}{\partial \eta}\right|_{\eta=\alpha} = \left.\frac{\partial f_{t_1}}{\partial \eta}\right|_{\eta=\beta} = 0 \\ f_{t_1}(0, \eta) = -f_{s_1}(\eta) \end{aligned}\right\} \quad (37)$$

where the ξ dependent part of f_{s_1} also has been neglected. With these approximations, the solution for f_{t_1} can be written as

$$f_{t_1} = \sum_0^\infty d_n e^{-\lambda_n \tau} Y_n(\eta) \quad (38)$$

where Y_n satisfies

$$Y_n'' + \lambda_n \operatorname{sech}^2\eta \, Y_n = 0 \quad (39)$$

and

$$Y_n'(\alpha) = Y_n'(\beta) = 0. \quad (39a)$$

Equations (39) and (39a) constitute a Sturm–Liouville system which will give rise to the eigenvalues λ_n, and eigenfunctions Y_n. The expansion coefficients d_n will be given by

$$d_n = -\frac{\int_\alpha^\beta f_{s_1}(\eta) Y_n(\eta) \operatorname{sech}^2\eta \, d\eta}{\int_\alpha^\beta Y_n^2(\eta) \operatorname{sech}^2\eta \, d\eta} \quad (40)$$

The approximate form for f_{s_1} which neglects ξ dependence obeys

$$\int_\alpha^\beta f_{s_1}(\eta) \operatorname{sech}^2\eta \, d\eta = 0 \quad (41)$$

which is also the result one would obtain from equation (22) for small eccentricity.

For the system of equations (39) and (39a), $\lambda_0 = 0$ and $Y_0 = 1$ and therefore from equations (40) and (41) $d_0 = 0$. Consequently, for large values of dimensionless time, we may truncate the series for f_{t_1} and obtain

$$f_{t_1} = d_1 e^{-\lambda_1 \tau} Y_1(\eta). \quad (42)$$

This approximation was used to compare the relative magnitudes of $|f_{t_1}|$ and $|f_{s_1}|$ averaged over the interval (α, β) to determine the value of dimensionless time τ when the transient becomes negligible compared to the steady state solution. The value of τ so obtained is the minimum dimensionless time, τ_m, required for the dispersion coefficient to assume a constant value independent of τ. The results obtained by solving equations (39) and (39a) can be correlated with reasonable accuracy by the following approximate relation

$$\tau_m \approx 31 \, K_2^{0.93}. \quad (43)$$

Since eccentricity may increase K_2 by several orders of magnitude, it also increases markedly the residence time required for a constant coefficient dispersion model to apply.

CONCLUSIONS

As a result of the present study, the following observations may be made about the behavior of the dispersion coefficient.

1. The steady state dispersion coefficient, which is the asymptotic value of K_2 for large time, increases with increasing eccentricity ϕ and reaches a maximum at a value of ϕ above 0·5: then it decreases with further increase in ϕ. Such behavior is exhibited for all radius ratios investigated. However, the dispersion coefficient is much more sensitive to eccentricity for small values of radius ratio ρ; that is, the sensitivity is greater when the inner circle of the annulus is relatively large. The flow rate in an eccentric annulus for a given pressure gradient exhibits similar behavior, but it is not as dramatic as it is for the dispersion coefficient.

2. For values of ϕ between 0·05 and 0·2, the dispersion coefficient, when plotted against radius ratio ρ, at first decreases with increasing ρ, reaches a minimum at a certain value of ρ, and then starts increasing as ρ is increased further. The location of this minimum shifts in the direction of increasing ρ as eccentricity increases. Such a minimum may very well exist for $\phi < 0.05$ and $\phi > 0.2$ but it is not observed in the range of radius ratios investigated here. Interestingly, it turns out that the above behaviour is in sympathy with that of the ratio (v_{max}/v_0) with ϕ and ρ.

3. For small eccentricities, some approximations may be made which are excellent for large ρ and enable one to estimate the minimum dimensionless time τ_m above which K_2 is effectively constant.

4. The extremely large effect of eccentricity in annuli on the magnitude of the dispersion coefficient suggest that eccentricities in the interstitial flow passages created by the arrangement of particles in packed beds may contribute significantly to the scatter of the data that have been reported in the literature.

ACKNOWLEDGEMENT

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DIFFUSION DE TAYLOR AU SEIN D'UN ÉCOULEMENT LAMINAIRE DANS UN ESPACE ANNULAIRE EXCENTRIQUE

Résumé—Grâce à une méthode exacte, qui en principe est valable pour toutes les valeurs du temps, on étudie de façon analytique la diffusion de Taylor dans un écoulement laminaire le long d'un tube rectiligne dont la section droite est un anneau excentrique. on obtient une expression du coefficient K_2 de diffusion apparente et on l'évalue numériquement pour un large domaine de l'excentricité ϕ et du rapport des rayons ρ.

Le coefficient de diffusion apparente est dépendant du temps. Cependant, à cause de la complexité de l'équation décrivant la diffusion convective laminaire dans un anneau excentrique, les résultats numériques sont limités à des valeurs asymptotiques de K_2 correspondant à des temps adimensionnels τ suffisamment grands pour que K_2 puisse être effectivement une constante indépendante du temps. Afin d'estimer la valeur τ_m au dessus de laquelle τ correspond à cette condition on emploie, pour des petites excentricités, une approximation qui est excellente pour des grandes valeurs de ρ et modérément bonne pour des petites valeurs de ρ. Les résultats de ce calcul sont reliés par la formule:

$$\tau_m \simeq 31\, K_2^{0,93}.$$

On montre que l'excentricité a un effet considérable sur la valeur asymptotique de K_2. Par exemple, avec $\rho = 1{,}5$ et $\phi = 0{,}5$, la valeur de K_2 vaut approximativement 250 fois celle relative à un anneau concentrique. Ce résultats remarquable suggère que des excentricités dans les grappes peuvent contribuer de façon significatrice à la dispersion des valeurs expérimentales du coefficient de diffusion apparentes rapportées auparavant.

TAYLOR-DIFFUSION IN EINEM EXZENTRISCHEN RINGSPALT BEI LAMINARER STRÖMUNG

Zusammenfassung—Es wird die Taylor-Diffusion bei laminarer Strömung entlang eines geraden Rohres mit exzentrischem Ringspaltquerschnitt analytisch mit Hilfe einer exakten Methode untersucht, die im Prinzip für alle Zeitpunkte gültig ist. Man erhält eine Formel für den scheinbaren Diffusionskoeffizienten K_2, die für einen weiten Bereich der Exzentrizität ϕ und des Radienverhältnisses φ numerisch ausgewertet wird.

Der scheinbare Diffusionskoeffizient ist an sich zeitabhängig, es werden jedoch die numerischen Ergebnisse wegen der Komplexität der den Fall laminarer, konvektiver Diffusion in einem exzentrischen Ringquerschnitt beschreibenden Gleichungen auf asymptotische Werte für K_2 beschränkt, d.h. auf dimensionslose Zeiten τ, die so gross sind, dass K_2 effektiv eine von der Zeit unabhängige Konstante wird. Zur Abschätzung des Wertes τ_m, oberhalb dessen τ dieser Beziehung genügt, wird für kleine Exzentrizitäten eine Näherungsbeziehung verwendet, die für grosse Werte von φ mässig gut geeïgnet ist. Die Ergebnisse dieser Berechnung lassen sich durch die Formel

$$\tau_m \approx 31\ K_2^{0.93}$$

korrelieren. Es zeigte sich, dass die Exzentrizität einen enormen Einfluss auf den asymptotischen Wert von K_2 hat. Zum Beispiel ist für $\varphi = 1{,}5$ und $\phi = 0{,}5$ der Wert von K_2 ungefähr 250 mal grösser als der für einen konzentrischen Ringspalt. Dieses bemerkenswerte Ergebnis lässt vermuten, dass Exentrizitäten in den Zwischenräumen von Festbetten ganz beträchtlich zur Streuung der kürzlich mitgeteilten experimentellen Werte für den scheinbaren Diffusionskoeffizienten beitragen können.

ДИФФУЗИЯ ТЕЙЛОРА В ЛАМИНАРНОМ ПОТОКЕ ЧЕРЕЗ ЭКСЦЕНТРИЧЕСКИЙ КОЛЬЦЕВОЙ ЗАЗОР

Аннотация—Исследовалась аналитически с помощью точного метода, который в принципе является точным для всех значений времени, диффузия Тейлора в ламинарном потоке через прямую трубу, поперечным сечением которой является эксцентрический кольцевой зазор. Получено выражение для эффективного коэффициента диффузии K_2 и рассчитано численно для широкого диапазона значений эксцентричности ϕ и отношения радиуса ρ.

Эффективному коэффициенту диффузии присуща зависимость от времени. Однако, из-за сложности уравнения, описывающего ламинарную конвективную диффузию в эксцентрическом кольцевом зазоре, численные результаты ограничены до асимптотических значений K_2, т.е.до безразмерных времен τ, достаточно больших для того чтобы K_2 было постоянной величиной, не зависящей от времени. Для того чтобы оценить значение τ_m, выше которого τ является достаточной для этого величиной, использовано приближение для небольших эксцентричностей, которое оказывается отличным для больших значений ρ и довольно хорошим для малых значений ρ. Результаты этого расчета описаны формулой

$$\tau_m \approx 31 K_2^{0,93}.$$

Найдено, что эксцентричность оказывает громадное влияние на асимптотическое значение K_2. Например, при $\rho = 1{,}5$ значение K_2 для $\phi = 0{,}5$ оказывается в 250 раз больше его значения для кольцевого зазора. Этот замечательный результат предполагает, что эксцентричности в пустотах плотных слоев могут оказывать значительное влияние на экспериментальные значения эффективного коэффициента диффузии, рассмотренного ранее.