31

Dispersion of solute in wall-bounded parallel shear flows

J.S. YU[1] and W.S. CHANG[2]

[1]*Department of Mechanical Engineering, West Virginia Institute of Technology, Montgomery, WV 25136, USA;*
[2]*Aero Propulsion and Power Laboratory, Wright Research and Development Center, Wright-Patterson AFB, Ohio 45433, USA*

Abstract. For laminar dispersion in wall-bounded straight channels, a rigorous method of solution to the basic convective diffusion equation is developed by using the Green's function. The method leads to a self-consistent and computationally useful procedure for the determination of the concentration distribution at arbitrary times. The dispersion approximation of Gill and Sankarasubramanian [5, 6] and the alternative approach of Smith [14, 15] for the transverse mean concentration are critically examined. It is shown that these latter methods are valid only under restricted conditions. For initial-value problems where the dispersion approximation applies, a purely algebraic method, distinctively different from the work of DeGance and Johns [10, 11] for the determination of the dispersion coefficients is developed.

The truncated dispersion approximation for the area-weighted transverse mean concentration of an inert solute due to prescribed initial conditions in flows without internal sources is analyzed in detail. It is shown that, by using the central moments of the mean concentration distribution, suitable criteria for the applicability of the second-order approximation can be reasonably established. Results pertaining to a concentrated initial distribution in Poiseuille pipe flow are presented as an example.

1. Introduction

The dispersal of a solute miscible in a slowly moving fluid is produced by the combined action of molecular diffusion and convective transport. The change of the solute concentration in the course of time is governed by the convective diffusion equation. With prescribed initial conditions of an inert solute in steady rectilinear flow through straight circular tubes, asymptotically effective solutions for the area-weighted transverse mean concentration $\bar{\theta}(\tau, X)$ have been formulated by Taylor [1], Aris [2], and Chatwin [3] at large times and by Lighthill [4] at small times. In the dispersion formulation developed by Gill and co-workers [5–9], it is maintained that in flows free of internal sources the evolution of $\bar{\theta}(\tau, X)$ due to a prescribed initial input is governed by the longitudinal dispersion equation,

$$\frac{\partial \bar{\theta}}{\partial \tau} = \sum_{j=0}^{\infty} K_j(\tau) \frac{\partial^j \bar{\theta}}{\partial X^j} .$$ (1.1)

Here, in nondimensional notations to be clarified later, τ is the time, X the axial distance, and the K_j's are the phenomenological dispersion coefficients. For an initial input belonging to the 'product' class, these authors have established the necessary equations for the determination of the K_j's and showed that (1.1) is exact and complete as a bulk-model equation for describing the dispersion of the solute. DeGance and Johns [10, 11], under the asssumption that (1.1) is indiscriminately valid for all initial conditions, developed a procedure for the sequential construction of these dispersion coefficients. Thus by employing a large enough number of terms, it appeared that (1.1), whenever applicable, may be used as

an effective approximation for the determination of $\bar{\theta}(\tau, X)$. With a different perspective within the context of source-free flow of an inert solute, however, Frankel and Brenner [12] most recently showed that no systematic improvement on the pointwise accuracy of $\bar{\theta}$ can be obtained from (1.1) by embodying the higher-order terms (higher than 1 to include diffusion) if the dispersion coefficients K_j, which for a given initial condition are inherently time-dependent, are replaced by their long-time ($\tau \to \infty$) asymptotic counterparts which are constants irrespective of the initial conditions.

On the basis of a method of solution to the convective diffusion equation by employing the Green's function [13, 14], this work presents the formulation of a self-consistent approximation by which a solution that converges in the mean can be computationally determined with prescribed accuracy. A critical examination made by virtue of this method shows that the dispersion formulation of (1.1) can be rigorously derived only if certain conditions are met by the initial concentration distribution and the prescribed source strength. Under such conditions, a purely algebraic procedure, in contrast to that of DeGance and Johns [10], is formulated for the systematic determination of the phenomenological coefficients. To test the effectiveness of the dispersion approximation of finite orders, the specific case of an inert solute in Poiseuille pipe flow with a transversely-uniform concentrated input is considered.

The usefulness of the dispersion formulation is essentially limited to its second-order approximation unless the coefficients K_j can be evaluated to arbitrary orders in practice. Aware of the fact that such an approximation has shortcomings irreconcilable with physical realities, Smith [15, 16] proposed a memory displacement representation for the determination of the area-weighted mean concentration. It is shown that Smith's approach can be precisely derived under the same conditions that the dispersion formulation is mathematically valid. The particular form of the new ansatz posed by Smith, however, is revealed to be the lowest-order nontrivial approximation of the exact solution obtained under additional restrictions.

2. Analysis by using the Green's function

The dispersion of a solute with prescribed distributions of source strength and initial concentration in a long straight channel of constant flow cross section is considered. The flow velocity, which is steady, laminar, and rectilinear in the axial direction, is assumed to depend only on the transverse coordinates. The solute may be inert or may undergo first-order irreversible chemical reactions in the fluid bulk and on the transverse flow boundaries. It is assumed that the solution is dilute so that effects on the flow velocity due to variations in solute concentration can be neglected. Under these conditions, the local concentration of the solute C is described by the basic convective diffusion equation

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2} + \nabla_A^2 C\right) - u\,\frac{\partial C}{\partial x} - k_A' C + q' \,,$$

$$\partial C/\partial N' + k_s' C = 0 \quad \text{on } S \,,$$

(2.1)

and an initial distribution of C which is assumed to be square-integrable. Here t is the time, x the axial distance, ∇_A^2 the transverse Laplace operator, u the local flow velocity, k_A' is the bulk reaction constant, k_s' is a constant used to account for surface reaction and wall

absorption, q', assumed to be square-integrable, is the local source strength, D the molecular diffusion coefficient, and $\partial/\partial N'$ denotes differentiation in the direction of the outward normal to the transverse flow boundary S.

Let C_0 be a reference solute concentration, u_0 a reference velocity, $\bar{u}$ the average flow velocity, L a characteristic length of the transverse flow area, and let

$$\theta = C/C_0 , \qquad \tau = Dt/L^2 , \qquad X = (x - \bar{u}t)/\mathrm{Pe}\, L ,$$

$$k_A = k_A' L^2/D , \qquad k_s = k_s' L/D , \qquad U = (u - \bar{u})/u_0 , \tag{2.2}$$

$$\nabla^2 = L^2 \nabla_A^2 , \qquad \partial/\partial N = L\, \partial/\partial N' , \qquad q = q' L^2/DC_0 ,$$

where $\mathrm{Pe} = Lu_0/D$ is the diffusion Peclet number. Then equation (2.1) can be written as

$$\frac{\partial \theta}{\partial \tau} = \frac{1}{\mathrm{Pe}^2} \frac{\partial^2 \theta}{\partial X^2} + (\nabla^2 - k_A)\theta - U \frac{\partial \theta}{\partial X} + q ,$$

$$\partial \theta/\partial N + k_s \theta = 0 \quad \text{on } S . \tag{2.3}$$

The eigenvalue problem for diffusion across the transverse flow area is

$$\nabla^2 \psi_n = -\lambda_n^2 \psi_n ,$$

$$\partial \psi_n/\partial N + k_s \psi_n = 0 \quad \text{on } S , \tag{2.4}$$

where ψ_n is the eigenfunction associated with the eigenvalue λ_n. Since the flow is laterally bounded, the sequence of values of λ_n ($n = 0, 1, \ldots$) is nonnegative, discrete and denumerably infinite, and the functions ψ_n form an infinite orthonormal set, that is,

$$\frac{L^2}{A} \int \int \psi_n \psi_m \, dy \, dz = \delta_{nm} , \tag{2.5}$$

where δ_{nm} is the Kronecker delta, A is the transverse flow area, (y, z) represents the nondimensionalized transverse coordinates in unit of the characteristic length L, i.e.,

$$\int \int dy \, dz = A/L^2 , \tag{2.6}$$

and the integration is over the entire transverse flow area.

The linearity of (2.3) means that its solution can be represented by the expansion

$$\theta = \sum_{n=0}^{\infty} \phi_n(\tau, X)\psi_n(y, z) . \tag{2.7}$$

Using (2.7) for θ in (2.3), multiplying by ψ_n (n fixed) and integrating over the transverse flow area by applying the orthogonal relation (2.5), we obtain

$$\frac{\partial \phi_n}{\partial \tau} = \frac{1}{\mathrm{Pe}^2} \frac{\partial^2 \phi_n}{\partial X^2} - \beta_n^2 \phi_n - \sum_{m=0}^{\infty} u_{nm} \frac{\partial \phi_m}{\partial X} + q_n , \quad \text{for } n = 0, 1, \ldots , \tag{2.8}$$

where

$$\beta_n^2 = \lambda_n^2 + k_A \, , \tag{2.9}$$

$$u_{nm} = \frac{L^2}{A} \int \int U \psi_n \psi_m \, \mathrm{d}y \, \mathrm{d}z \, , \tag{2.10}$$

$$q_n(\tau, X) = \frac{L^2}{A} \int \int q(X, y, z, \tau) \psi_n \, \mathrm{d}y \, \mathrm{d}z \, . \tag{2.11}$$

The initial conditions to be applied to the solutions are

$$\phi_n(0, X) = \frac{L^2}{A} \int \int \theta_0(X, y, z) \psi_n \, \mathrm{d}y \, \mathrm{d}z \, , \tag{2.12}$$

where $\theta_0(X, y, z)$ is the prescribed initial concentration distribution of the solute.

By using the variable transformation

$$X_n = X - u_{nn} \tau \, , \tag{2.13}$$

equation (2.8) is reduced to the form

$$\frac{\partial \phi_n}{\partial \tau} = \frac{1}{\mathrm{Pe}^2} \frac{\partial^2 \phi_n}{\partial X_n^2} - \beta_n^2 \phi_n - \sum_{m \neq n}^{\infty} u_{nm} \frac{\partial \phi_m}{\partial X} + q_n \, . \tag{2.14}$$

In terms of the Green's function created by an instantaneous point source located at X_s at time τ_s (see for example Morse and Feshbach [17]),

$$G(\tau, X_n | \tau_s, X_s) = \frac{\mathrm{Pe}}{2\sqrt{\pi(\tau - \tau_s)}} \exp\left[-\frac{\mathrm{Pe}^2 (X_n - X_s)^2}{4(\tau - \tau_s)} - \beta_n^2 (\tau - \tau_s) \right] , \tag{2.15}$$

the complete, but not directly useful, solution to equation (2.14) is

$$\phi_n(\tau, X) = \phi_{n0} + \rho_n - \sum_{m \neq n}^{\infty} u_{nm} \int_0^{\tau} \mathrm{d}\tau_s \int_{-\infty}^{\infty} \mathrm{d}X_s \frac{\partial \phi_m(\tau_s, X_s)}{\partial X_s} G(\tau, X_n | \tau_s, X_s) \, , \tag{2.16}$$

where

$$\phi_{n0}(\tau, X) = \int_{-\infty}^{\infty} \phi_n(0, X_s) G(\tau, X | 0, X_s) \, \mathrm{d}X_s$$

$$= \frac{\mathrm{Pe}}{2\sqrt{\pi\tau}} \, \mathrm{e}^{-\beta_n^2 \tau} \int_{-\infty}^{\infty} \phi_n(0, X_s) \exp\left[-\frac{\mathrm{Pe}^2 (X - X_s)^2}{4\tau} \right] \mathrm{d}X_s \tag{2.17}$$

is the contribution to ϕ_n arising from the initial distribution, and

$$\rho_n(\tau, X) = \int_0^{\tau} \mathrm{d}\tau_s \int_{-\infty}^{\infty} \mathrm{d}X_s q_n(\tau_s, X_s) G(\tau, X | \tau_s, X_s) \tag{2.18}$$

is that due to the distributed source. Now expand $\partial \phi_m(\tau_s, X_s)/\partial X_s$ in the source coordinates on the right-hand side of (2.16) about (τ, X_n) of the observation coordinates in a series in

powers of $(\tau_s - \tau)$ and $(X_s - X_n)$ and then perform the required integrations over time and space term by term. The resulting series can then be reverted back to the original coordinates (τ, X). The complicated algebraic maneuvers will not be presented. The final result is

$$\phi_n(\tau, X) = \phi_{n0}(\tau, X) + \rho_n(\tau, X)$$

$$- \sum_{m \neq n}^{\infty} \sum_{\mu=0}^{\infty} \sum_{\nu=0}^{[\mu/2]} \sum_{\gamma=0}^{\mu-2\nu} u_{nm} \Omega_n(\mu, \nu, \gamma; \tau) \frac{\partial^{2\nu+\gamma+1}}{\partial X^{2\nu+\gamma+1}} \left(\frac{\partial^{\mu-2\nu-\gamma} \phi_m}{\partial \tau^{\mu-2\nu-\gamma}} \right) \tag{2.19}$$

where $[\mu/2]$ designates the integral part of the argument and

$$\Omega_n(\mu, \nu, \gamma; \tau) = \frac{(-1)^\mu}{\text{Pe}^{2\nu}} \frac{(\mu-\nu)! u_{nn}^\gamma}{\gamma! \nu! (\mu-2\nu-\gamma)!} \beta_n^{-2(\mu-\nu+1)} \left[1 - e^{-\beta_n^2 \tau} \sum_{\sigma=0}^{\mu-\nu} \frac{(\beta_n^2 \tau)^\sigma}{\sigma!} \right]. \tag{2.20}$$

The expansion procedure used in obtaining (2.19) from (2.16) involves no *ad hoc* assumptions of any kind. These equations for $n = 0, 1, \ldots$, therefore, are mathematically exact for the description of the functions $\phi_n(\tau, X)$ if the latter are analytic in their domain of definition.

3. Computationally self-consistent approximation

The functions $q_n(\tau, X)$ given by (2.11) are the component coefficients in the series representation of the source strength with the transverse eigenfunctions as the basis, i.e.,

$$q(\tau, X, y, z) = \sum_{n=0}^{\infty} q_n(\tau, X) \psi_n(y, z). \tag{3.1}$$

We shall assume that the series of the right-hand side converges uniformly to $q(\tau, X, y, z)$. Thus, since the eigenfunctions ψ_n form an infinite orthonormal set, one has as an approximation with a sufficiently large $\mathcal{N}$ [18],

$$q_n(\tau, X) \simeq 0 \quad \text{for } n > \mathcal{N}, \tag{3.2}$$

and accordingly, from equation (2.8),

$$\frac{\partial \phi_n}{\partial \tau} = \frac{1}{\text{Pe}^2} \frac{\partial^2 \phi_n}{\partial X^2} - \beta_n^2 \phi_n - \sum_{m=0}^{\infty} u_{nm} \frac{\partial \phi_m}{\partial X} + q_n \quad \text{for } n = 0, 1, \ldots, \mathcal{N}, \tag{3.3a}$$

$$\frac{\partial \phi_n}{\partial \tau} = \frac{1}{\text{Pe}^2} \frac{\partial^2 \phi_n}{\partial X^2} - \beta_n^2 \phi_n - \sum_{m=0}^{\infty} u_{nm} \frac{\partial \phi_m}{\partial X} \quad \text{for } n > \mathcal{N}. \tag{3.3b}$$

The solutions of these equations are given by equation (2.19) with the use of, to the order of approximation $\mathcal{N}$ defined by condition (3.2), $\rho_n = q_n = 0$ for $n > \mathcal{N}$. These solutions, however, are not directly useful as before.

The following observations can be made on equation (2.19) for the purpose of obtaining solutions that are computationally useful. The term $\phi_{n0}(\tau, X)$ decays, at any given time τ, according to the factor $\exp(-\beta_n^2 \tau)$ indefinitely to zero as β_n^2 increases (see (2.17)) and the

functions $\Omega_n(\mu,\ \nu,\ \gamma;\ \tau)$ under the summations, where the exponential term decays in the same fashion (see (2.20)), are seen to become independent of time but inversely proportional to β_n^2 raised to progressively higher powers as the integer difference $\mu - \nu$ increases. Let β_n^2 be arranged in an increasing sequence so that $\beta_{n+1}^2 > \beta_n^2$. It is evident that, except at times when the source strength is instantaneously infinite, if

$$\beta_{\mathcal{N}}^2 \tau \gg 1\ ,\tag{3.4}$$

then the term ϕ_{n0} for $n > \mathcal{N}$ can be neglected and the small value of $\phi_n(\tau, X)$, to the order of $1/\beta_n^2$, may be closely approximated by retaining only the term containing $\Omega_n(0, 0, 0; \tau)$ in (2.19). We have therefore in this approximation

$$\phi_n(\tau, X) = -\frac{1}{\beta_n^2} \sum_{\substack{m \neq n}}^{\infty} u_{nm} \frac{\partial \phi_m}{\partial X} \quad \text{for } n > \mathcal{N}\ ,\tag{3.5}$$

where the condition $\rho_n = q_n = 0$ for $n > \mathcal{N}$ has been imposed. Use of (3.5) in itself for the terms with $m > \mathcal{N}$ leads to the approximate prescription, good to the level of the first-order axial derivatives,

$$\phi_n(\tau, X) = -\frac{1}{\beta_n^2} \sum_{m=0}^{\mathcal{N}} u_{nm} \frac{\partial \phi_m}{\partial X} \quad \text{for } n > \mathcal{N}\ .\tag{3.6}$$

With (3.6) substituted into (3.3a) for the terms with $m > \mathcal{N}$ under the summation, we obtain the system of simultaneous equations

$$\frac{\partial \phi_n}{\partial \tau} = -\beta_n^2 \phi_n - \sum_{m=0}^{\mathcal{N}} u_{nm} \frac{\partial \phi_m}{\partial X} + \sum_{m=0}^{\mathcal{N}} \left[\frac{1}{\text{Pe}^2} \delta_{nm} + k_{nm}^{(\mathcal{N})} \right] \frac{\partial^2 \phi_m}{\partial X^2} + q_n$$

$$\text{for } n = 0, 1, 2, \ldots, \mathcal{N}\ ,\tag{3.7}$$

where

$$k_{nm}^{(\mathcal{N})} = \sum_{l=\mathcal{N}+1}^{\infty} \frac{u_{nl} u_{lm}}{\beta_l^2}\ .\tag{3.8}$$

Equation (3.7), with the use of the initial conditions of (2.12), can be solved, if not analytically, at least computationally for the coefficient functions $\phi_n(\tau, X)$, $n \leqslant \mathcal{N}$, by the application of Fourier and Laplace transforms. The prescription procedure leading to (3.7) implies that, at any given time, after $\mathcal{N}$ becomes sufficiently large, the larger the value of $\mathcal{N}$, the better is the approximation and the more efficient is the series representation of (2.7).

4. Some fundamental relations

The dispersion formulation of equation (1.1) is derived under the assumption that the local concentration $\theta(\tau, X, y, z)$ can be represented by a linear combination of the axial derivatives $\partial^j \bar{\theta}(\tau, X)/\partial X^j$ ($j = 0, 1, \ldots$) [6]. DeGance and Johns [10] tacitly accepted this assump-

tion in their derivation of the pertinent dispersion coefficients. In this section, we shall attempt to establish the condition under which certain basic relations can be derived for the valid construction of the dispersion formulation.

In order to relate the present approach to the description of the dispersion formulation, it is necessary to eliminate the time derivatives of the ϕ-functions in (2.19). Differentiating (2.8) with respect to τ and switching the orders of differentiation between τ and X, one has

$$\frac{\partial^2 \phi_m}{\partial \tau^2} = \frac{1}{\mathrm{Pe}^2} \frac{\partial^2}{\partial X^2} \frac{\partial \phi_m}{\partial \tau} - \beta_m^2 \frac{\partial \phi_m}{\partial \tau} - \sum_{l=0}^{\infty} u_{ml} \frac{\partial}{\partial X} \frac{\partial \phi_l}{\partial \tau} + \frac{\partial q_m}{\partial \tau} .$$

Now use equation (2.8) again for the τ-derivatives on the right-hand side of the above equation, it is readily seen that the result is an expression involving the X-derivatives of the ϕ-functions only. By repeating this procedure, higher order τ-derivatives can similarly be obtained and we find in general that these can be written in the form

$$\frac{\partial^s \phi_m}{\partial \tau^s} = \sum_{l=0}^{\infty} \sum_{k=0}^{2s} T_{ml}^{(s,k)} \frac{\partial^k \phi_l}{\partial X^k}$$

$$+ (1 - \delta_{s0}) \left[\frac{\partial^{s-1} q_m}{\partial \tau^{s-1}} + (1 - \delta_{s1}) \sum_{l=0}^{\infty} \sum_{p=1}^{s-1} \sum_{k=0}^{2p} T_{ml}^{(p,k)} \frac{\partial^{k+s-p-1} q_l}{\partial X^k \partial \tau^{s-p-1}} \right] , \tag{4.1}$$

where the coefficients $T_{ml}^{(s,k)}$, beginning with

$$T_{ml}^{(0,0)} = \delta_{ml} , \qquad T_{ml}^{(0,k)} = 0 , \quad \text{for } k > 0 ,$$

$$T_{ml}^{(1,0)} = -\beta_l^2 \delta_{ml} , \qquad T_{ml}^{(1,1)} = -u_{ml} , \qquad T_{ml}^{(1,2)} = \frac{1}{\mathrm{Pe}^2} \delta_{ml} , \tag{4.2}$$

are determined, for $s \geq 2$, by using the following recurrence relations:

$$T_{ml}^{(s,0)} = \sum_{n=0}^{\infty} T_{mn}^{(s-1,0)} T_{nl}^{(1,0)} ,$$

$$T_{ml}^{(s,1)} = \sum_{n=0}^{\infty} T_{mn}^{(s-1,0)} T_{nl}^{(1,1)} + T_{mn}^{(s-1,1)} T_{nl}^{(1,0)} ,$$

$$T_{ml}^{(s,k)} = \sum_{n=0}^{\infty} \sum_{j=k-2}^{k} T_{mn}^{(s-1,j)} T_{nl}^{(1,k-j)} \quad \text{for } 2 \leq k \leq 2(s-1) , \tag{4.3}$$

$$T_{ml}^{(s,2s-1)} = \sum_{n=0}^{\infty} T_{mn}^{(s-1,2s-3)} T_{nl}^{(1,2)} + T_{mn}^{(s-1,2s-2)} T_{nl}^{(1,1)} ,$$

$$T_{ml}^{(s,2s)} = \sum_{n=0}^{\infty} T_{mn}^{(s-1,2s-2)} T_{nl}^{(1,2)} .$$

Employing (4.1) for the τ-derivatives in the exact solution (2.19), we find

$$\phi_n(\tau, X) = Z_n(\tau, X) - \sum_{l=0}^{\infty} \sum_{k=1}^{\infty} D_{nl}^{(k)}(\tau) \frac{\partial^k \phi_l}{\partial X^k} , \tag{4.4}$$

where

$$D_{nl}^{(k)}(\tau) = \sum_{m \neq n}^{\infty} \sum_{j=0}^{k-1} \sum_{\mu=k-1-[j/2]}^{\infty} \sum_{\nu=0}^{[(k-j-1)/2]} u_{nm} T_{ml}^{(\mu+j-k+1,j)} \Omega_n(\mu, \nu, k-2\nu-j-1; \tau) ,$$

(4.5)

$$\text{for } n, l \geq 0 \text{ and } k \geq 1 ,$$

otherwise $D_{nl}^{(k)} = 0$, and

$$Z_n(\tau, X) = \phi_{n0}(\tau, X) + \rho_n(\tau, X) + Q_n(\tau, X) .$$

(4.6)

Here $\phi_{n0}(\tau, X)$ and $\rho_n(\tau, X)$ are given by (2.17) and (2.18), respectively, and

$$Q_n(\tau, X) = -\sum_{m \neq n}^{\infty} \sum_{\mu=0}^{\infty} \sum_{\nu=0}^{[\mu/2]} \sum_{\gamma=0}^{\mu-2\nu} u_{nm} \Omega_n(\mu, \nu, \gamma; \tau) \Gamma_m(\tau, X) ,$$

(4.7a)

with

$$\Gamma_m(\tau, X) = (1 - \delta_{\mu-2\nu-\gamma,0}) \left[\frac{\partial^{\mu} q_m}{\partial X^{2\nu+\gamma+1} \partial \tau^{\mu-2\nu-\gamma-1}} + (1 - \delta_{\mu-2\nu-\gamma,1}) \right.$$

$$\left. \times \sum_{l=0}^{\infty} \sum_{s=1}^{\mu-2\nu-\gamma-1} \sum_{k=0}^{2s} T_{ml}^{(s,k)} \frac{\partial^{k+\mu-s} q_l}{\partial X^{k+2\nu+\gamma+1} \partial \tau^{\mu-2\nu-\gamma-1-s}} \right] .$$

(4.7b)

We proceed from here on in this paper to use $\phi_0(\tau, X)$ as a reference coefficient function for the purpose of developing the necessary relations that are adaptable to the description of the dispersion formulation. A compelling reason for this particular choice is that $\phi_0(\tau, X)$ is equal to the area-weighted transverse average concentration for the case of an inert solute (see Sec. 6(a)).

Introduce the Fourier transform of $Z_n(\tau, X)$,

$$V_n(\tau, \omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} Z_n(\tau, X) e^{i\omega X} dX ,$$

(4.8)

where $Z_n(\tau, X)$ is defined by (4.6), and the inversion formula,

$$Z_n(\tau, X) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} V_n(\tau, \omega) e^{-i\omega X} d\omega ,$$

(4.9)

and let

$$R_n(\tau, \omega) = V_n / V_0 .$$

(4.10)

If $R_n(\tau, \omega)$ exists, we can write $V_n = R_n V_0$ and, by virtue of the inversion formula, one has

$$Z_n(\tau, X) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} R_n(\tau, \omega) V_0(\tau, \omega) e^{-i\omega X} d\omega .$$

(4.11)

If the function R_n is analytic in the entire complex ω-plane, then it is valid to write

$$R_n(\tau, \omega) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m!} R_n^{(m)}(\tau)(-i\omega)^m ,$$

(4.12)

where

$$R_n^{(m)}(\tau) = (-i)^m \left. \frac{\partial^m R_n}{\partial \omega^m} \right|_{\omega = 0} . \tag{4.13}$$

It will be shown that the dispersion formulation can be rigorously derived under the condition of (4.12). Substituting (4.12) into (4.11) and performing the inversion, the result is

$$Z_n(\tau, X) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m!} R_n^{(m)}(\tau) \frac{\partial^m Z_0}{\partial X^m} , \tag{4.14}$$

where imposition has been made of the identity

$$\frac{\partial^m Z_0}{\partial X^m} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} (-i\omega)^m V_0(\tau, \omega) e^{-i\omega X} d\omega . \tag{4.15}$$

The physical significance of condition (4.12) perhaps can best be illustrated by considering dispersion in flows without sources. Setting $\rho_n = Q_n = 0$, then $Z_n(\tau, X) = \phi_{n0}(\tau, X)$ from (4.6) and equation (4.14) becomes

$$\phi_{n0}(\tau, X) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m!} R_n^{(m)}(\tau) \frac{\partial^m}{\partial X^m} \phi_{00}(\tau, X)$$

which must hold true for all time τ. At $\tau = 0$, equation (2.17) gives $\phi_{n0}(0, X) = \phi_n(0, X)$ which is given by equation (2.12) with the use of the specified initial condition. It is thus seen that, for this particular case, the restrictive condition of (4.12) is none other than to require the initial distribution of ϕ_n to be a linear combination of the X-derivatives of ϕ_0.

To proceed to derive the required relations, we use (4.14) in (4.4) to get

$$\phi_n(\tau, X) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m!} R_n^{(m)} \frac{\partial^m Z_0}{\partial X^m} - \sum_{l=0}^{\infty} \sum_{k=1}^{\infty} D_{nl}^{(k)}(\tau) \frac{\partial^k \phi_l}{\partial X^k} . \tag{4.16}$$

By differentiating (4.4) with $n = 0$, one has

$$\frac{\partial^m Z_0}{\partial X^m} = \frac{\partial^m \phi_0}{\partial X^m} + \sum_{l=0}^{\infty} \sum_{k=1}^{\infty} D_{nl}^{(k)}(\tau) \frac{\partial^{k+m} \phi_l}{\partial X^{k+m}} , \tag{4.17}$$

Employing (4.17) in (4.16) yields, after some simple manipulations,

$$\phi_n(\tau, X) = \sum_{k=0}^{\infty} S_{n0}^{(k)}(\tau) \frac{\partial^k \phi_0}{\partial X^k} + \sum_{l=1}^{\infty} \sum_{k=1}^{\infty} S_{nl}^{(k)}(\tau) \frac{\partial^k \phi_l}{\partial X^k} , \tag{4.18}$$

where

$$S_{nl}^{(k)}(\tau) = \frac{(-1)^k}{k!} R_n^{(k)} \delta_{l0} + (1 - \delta_{k0})\left(-D_{nl}^{(k)} + \sum_{m=0}^{k-1} \frac{(-1)^m}{m!} R_n^{(m)} D_{0l}^{(k-m)} \right) . \tag{4.19}$$

By proper truncation, equation (4.18) may be solved for ϕ_n in terms of the X-derivatives of ϕ_0. For the sake of keeping the notations simple, we shall, in what follows, use ∞ to mean that the number of terms included in the summation is large enough for achieving

convergence. We can show that the solution is

$$\phi_n(\tau, X) = \sum_{k=0}^{\infty} A_{nk}(\tau) \frac{\partial^k \phi_0}{\partial X^k} ,$$

(4.20)

where

$$A_{nk}(\tau) = S_{n0}^{(k)}(\tau) + (1 - \delta_{k0}) \sum_{l=1}^{\infty} \sum_{j=1}^{k} S_{nl}^{(j)}(\tau) A_{l,k-j}(\tau) .$$

(4.21)

It must be noted that the solution given by (4.20) is valid only if the function $R_n(\tau, \omega)$ defined by (4.10) is analytic in the complex ω-plane.

5. Derivation of the dispersion formulation

The local solute concentration as before is given by (2.7) and the weighted transverse mean concentration is defined as

$$\bar{\theta}(\tau, X) = \int \int w(y, z)\theta \, dy \, dz = \sum_{n=0}^{\infty} w_n \phi_n(\tau, X) ,$$

(5.1)

where the weight function $w(y, z)$ is normalized with respect to the transverse flow area and where on the extreme right-hand side,

$$w_n = \int \int w(y, z)\psi_n(y, z) \, dy \, dz .$$

(5.2)

If the condition expressed by equation (4.12) is fulfilled, then relation (4.20) holds true. Substitution of (4.20) for ϕ_n in equation (5.1) leads to

$$\bar{\theta}(\tau, X) = \sum_{k=0}^{\infty} h_k(\tau) \frac{\partial^k \phi_0}{\partial X^k} ,$$

(5.3)

where

$$h_k(\tau) = \sum_{n=0}^{\infty} w_n A_{nk}(\tau) .$$

(5.4)

Thus one has, by virtue of (5.3),

$$\frac{\partial^j \bar{\theta}}{\partial X^j} = \sum_{k=0}^{\infty} h_k(\tau) \frac{\partial^{k+j} \phi_0}{\partial X^{k+j}} \quad \text{for } j = 0, 1, 2, \ldots ,$$

(5.5)

from which one obtains, after some algebra, the basic relation, as it will be soon seen, required for the derivation of the dispersion approximation,

$$\phi_0 = \sum_{j=0}^{\infty} \mathcal{H}_j(\tau) \frac{\partial^j \bar{\theta}}{\partial X^j} ,$$

(5.6)

where, using determinant notation, the coefficients of the X-derivatives are given by

$$
\mathcal{K}_j = \frac{(-1)^j}{h_0^{j+1}}
\begin{vmatrix}
h_1 & h_2 & h_3 & \cdots\cdots\cdots & h_j \\
h_0 & h_1 & h_2 & \cdots\cdots\cdots & h_{j-1} \\
0 & h_0 & h_1 & \cdots\cdots\cdots & h_{j-2} \\
\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \\
0 & 0 & 0 & \cdots & h_0 & h_1 & h_2 \\
0 & 0 & 0 & \cdots & 0 & h_0 & h_1
\end{vmatrix}
\quad \text{for } j \geq 1 ,
\tag{5.7}
$$

with $\mathcal{K}_0 = 1/h_0$ and $\mathcal{K}_j = 0$ for $j < 0$.

Substituting (2.7) into (2.3) for θ in the transverse diffusion and axial convection terms, multiplying through by the weight function $w(y, z)$, and integrating the resulting equation over the flow cross-section, we have for the transverse mean concentration,

$$
\frac{\partial \bar{\theta}}{\partial \tau} = \frac{1}{\text{Pe}^2} \frac{\partial^2 \bar{\theta}}{\partial X^2} - \sum_{n=0}^{\infty} \beta_n^2 w_n \phi_n - \sum_{n=0}^{\infty} u_n \frac{\partial \phi_n}{\partial X} + \bar{q} ,
\tag{5.8}
$$

where β_n^2 is given by (2.9),

$$
u_n = \int\int wU\psi_n \, dy \, dz ,
\tag{5.9}
$$

and

$$
\bar{q}(\tau, X) = \int\int wq \, dy \, dz .
\tag{5.10}
$$

Now employ (4.20) for ϕ_n on the right-hand side of equation (5.8), then use (5.6) for the X-derivatives of ϕ_0 in the resulting equation. We find presently the equation governing the weighted transverse mean concentration,

$$
\frac{\partial \bar{\theta}}{\partial \tau} = \sum_{j=0}^{\infty} K_j(\tau) \frac{\partial^j \bar{\theta}}{\partial X^j} + \bar{q}(\tau, X) ,
\tag{5.11}
$$

where the dispersion coefficients

$$
K_j(\tau) = \frac{1}{\text{Pe}^2} \delta_{j2} - \sum_{k=0}^{j} \sum_{n=0}^{\infty} \mathcal{K}_{j-k}(\tau)[\beta_n^2 w_n A_{nk}(\tau) + u_n A_{n,k-1}(\tau)\delta_{k0}] .
\tag{5.12}
$$

Equation (5.11) is identical in form to equation (1.1) for purely initial-value problems ($\bar{q} = 0$). The present derivation shows, however, that this particular formulation is not indiscriminately mathematically equivalent to the original problem of equation (2.3) unless condition (4.12) is satisfied by the prescribed initial solute concentration and the distributed source strength.

6. Dispersion coefficients in flow due to an initial input belonging to the product class

The transport of a solute which is instantaneously injected into a flowing stream of a carrier fluid is a basic process in numerous applications. In many cases, the distribution of the initial

solute input can be approximated to have a form belonging to the product class. Gill and Sankarasubramanian [6] provided a method of determination of the area-weighted transverse mean concentration based on the scond-order dispersion approximation for the transport of an axisymmetric but radially partially uniform slug in laminar pipe flow under the condition of zero wall flux. Since instantaneous injection is equivalent to specifying the initial concentration distribution, the following discussion is made in the context of the latter by equating the source strength to zero.

If the initial concentration distribution of the solute is in the product class, i.e.,

$$\theta_0 = \theta(0, X, y, z) = f(X)g(y, z) ,$$ (6.1)

then use of equations (2.12) and (2.17) gives

$$\phi_{n0} = \frac{g_n \, \mathrm{Pe}}{2\sqrt{\pi\tau}} \, \mathrm{e}^{-\beta_n^2 \tau} \int_{-\infty}^{\infty} f(X_s) \exp\left[-\frac{\mathrm{Pe}^2(X - X_s)^2}{4\tau}\right] \mathrm{d}X_s ,$$ (6.2)

where

$$g_n = \frac{L^2}{A} \int \int g(y, z)\psi_n \, \mathrm{d}y \, \mathrm{d}z .$$ (6.3)

For purely initial-value problems, $q_n = Q_n = \rho_n = q = 0$, we have from (4.10) and (4.12),

$$R_n^{(0)} = R_n = \frac{V_n}{V_0} = \frac{\phi_{n0}}{\phi_{00}} = \frac{g_n}{g_0} \, \mathrm{e}^{-(\beta_n^2 - \beta_0^2)\tau} ,$$

$$R_n^{(m)} = 0 \quad \text{for } m > 0 .$$ (6.4)

Hence, from (4.19),

$$S_{nl}^{(k)} = R_n \delta_{k0}\delta_{l0} + (1 - \delta_{k0})(-D_{nl}^{(k)} + R_n D_{0l}^{(k)}) ,$$ (6.5)

where the function $D_{nl}^{(k)}$ is defined by (4.5).

(a) *Area-weighted transverse mean concentration of an inert solute*

For a chemically inert solute in the absence of sorption with the wall, one has $k_A = k_s = 0$ and $\beta_n^2 = \lambda_n^2$. The transverse eigenvalue problem is

$$\nabla^2 \psi_n = -\lambda_n^2 \psi_n ,$$

$$\partial \psi_n / \partial N = 0 \quad \text{on } S ,$$ (6.6)

which, in conjunction with the orthonormal relation of (2.5), gives for the lowest eigenmode, $\psi_0 = 1$ and $\lambda_0 = 0$. Since the concentration·is to be averaged over the flow cross-section, then

$$w(y, z) = L^2/A ,$$ (6.7)

and it follows from (5.2), (5.4), (5.7), and (5.9) that $w_n = \mathcal{H}_n = h_n = \delta_{n0}$ and $u_n = u_{0n}$. Use

of these results in (5.1) gives

$$\bar{\theta}(\tau, X) = \phi_0(\tau, X) \tag{6.8}$$

and from (5.12), noting that $\beta_0 = \lambda_0 = 0$, we have

$$K_0(\tau) = 0 \, ,$$

$$K_j(\tau) = \frac{1}{Pe^2} \, \delta_{j2} - \sum_{n=1}^{\infty} u_{0n} A_{n,j-1} \quad \text{for } j \geq 1 \, , \tag{6.9}$$

where A_{nj} is recursively determined from (4.21) and $u_0 = u_{00} = 0$ has been used.

The dispersion coefficient K_0 is identically zero in the course of time. We proceed to examine the asymptotic behaviors at small and at large times of the higher-order dispersion coefficients as prescribed by (6.9) where it is to be noted that $\beta_n^2 = \lambda_n^2 > 0$ because $n \geq 1$.

At small times as $\tau \to 0$, equation (6.4) gives $R_n \to g_n / g_0$, and equation (2.20) gives $\Omega_n(\mu, \nu, \gamma; \tau) = O(r^{\mu - \nu + 1})$. With the use of these asymptotic results, one has from (4.5) and (6.5) that as $\tau \to 0$,

$$S_{nl}^{(k)} \sim D_{nl}^{(k)} = \begin{cases} O(\tau^{[k/2]+1}) & \text{for even } k \, , \\ O(\tau^{k-[k/2]}) & \text{for odd } k \, . \end{cases}$$

We then obtain from (4.21) by finite induction,

$$A_{n0} = R_n \, ,$$

$$A_{nk} = O(\tau^{[k/2]+1}) \quad \text{for } k \geq 1 \, .$$

Substitution of these into (6.9) gives, as $\tau \to 0$,

$$K_1(\tau) \to - \sum_{n=1}^{\infty} u_{0n} g_n / g_0 \, ,$$

$$K_2(\tau) \to 1 / Pe^2 \, , \tag{6.10}$$

$$K_j(\tau) = O(\tau^{[(j-1)/2]+1}) \quad \text{for } j \geq 3 \, .$$

The last result shows that $K_j \to 0$ as $\tau \to 0$ for $j \geq 3$. Therefore at sufficiently small times the second-order dispersion approximation can be adequately used for describing the evolution of the area-weighted transverse mean concentration distribution.

At large times, the same asymptotic limiting procedure as above, but in the limit as $\tau \to \infty$, gives, for $n \geq 1$: $R_n = (g_n / g_0) e^{-\lambda_n^2 \tau} \to 0$, $\Omega_n(\mu, \nu, \gamma; \tau) = O(\tau^0)$, and $S_{nl}^{(k)} \to -D_{nl}^{(k)} = O(\tau^0)$. Hence we have from (4.21), $A_{n0} = R_n \to 0$ and $A_{nk} = O(\tau^0)$ for $k \geq 1$, and finally from (6.9) the asymptotic results as $\tau \to \infty$,

$$K_1(\tau) \to 0 \, ,$$

$$K_j(\tau) = \frac{1}{Pe^2} \, \delta_{j2} + O(\tau^0) \quad \text{for } j \geq 2 \, . \tag{6.11}$$

Thus, at sufficiently large times, the change of the area-weighted mean concentration is characterized by constant dispersion coefficients with the absence of convective transport $(K_1 \to 0)$ in a coordinate moving with the average flow velocity. At such times, the simplest approximation is then given by the second-order equation with $K_0 = K_1 = 0$ which was used by Taylor [1] in his analysis. The condition under which such an approximation becomes valid will be examined in a later section.

(b) *Inert solute with a transversely uniform initial concentration distribution*

We shall make use of the preceding results to obtain the explicit expressions of the dispersion coefficients for the determination of the area-weighted transverse mean concentration under the specific conditions that the solute is inert without wall sorption and that the initial input of the solute is uniform over the transverse area. Such an initial distribution falls in the product class as defined by (6.1) with $g(y, z) = 1$, and we have, from (6.3) and (6.4), $R_n = g_n = \delta_{n0}$. When this is used in (6.5) we get $S_{nl}^{(k)} = \delta_{n0}\delta_{k0}\delta_{l0} - (1 - \delta_{k0})D_{nl}^{(k)}$, and then (4.21) reduces to

$$A_{nk} = \delta_{n0} - (1 - \delta_{n0})(1 - \delta_{k0})\left(D_{n0}^{(k)} + \sum_{l=1}^{\infty}\sum_{j=1}^{k} D_{nl}^{(k)} A_{l,k-j}\right). \tag{6.12}$$

The explicit evaluation of A_{nk}, though using only elementary algebra involving multiple summations, turned out to be a tedious and extremely long task. $D_{nl}^{(k)}$ is defined by (4.5) which requires the use of (4.3) for $T_{ml}^{(s,k)}$ and (2.20) for $\Omega_n(\mu, \nu, \gamma; \tau)$. With $D_{nl}^{(k)}$ so determined and then substituted into (6.12), the resulting multiple summations are so complicated that, for $k \geq 2$, computer algebra had to be employed to carry out the necessary algebraic maneuvers. Presently we have persevered to formulate A_{nk} for $k \leq 4$ in this manner.

The dispersion coefficients are given by (6.9) in terms of A_{nk} because the solute is inert and no sorption with the wall occurs $(\beta_0 = \lambda_0 = 0, \beta_n = \lambda_n > 0$ for $n > 0)$. We record the explicit expressions of the first few as follows.

$$K_0 = K_1 = 0 \,,$$

$$K_2 = \frac{1}{\mathrm{Pe}^2} + \sum_{n=1}^{\infty} u_{0n}\lambda_n^{-2}(1 - \mathrm{e}^{-\lambda_n^2 \tau}) \,,$$

$$K_3 = -\sum_{n=1}^{\infty} \frac{u_{0n}u_{nn}}{\lambda_n^4}[1 - (1 + \lambda_n^2\tau)\mathrm{e}^{-\lambda_n^2\tau}]$$

$$-\sum_{n=1}^{\infty}\sum_{\substack{l=1 \\ l \neq n}}^{\infty} \frac{u_{0n}u_{0l}u_{nl}}{\lambda_n^2\lambda_l^2}\left(1 + \frac{\lambda_l^2}{\lambda_n^2 - \lambda_l^2}\mathrm{e}^{-\lambda_n^2\tau} + \frac{\lambda_n^2}{\lambda_l^2 - \lambda_n^2}\mathrm{e}^{-\lambda_l^2\tau}\right).$$

$$\tag{6.13}$$

Similar results have been obtained for K_4 and K_5 by using computer algebra but their expressions are too lengthy to be recorded here.

7. Dispersion approximation for the area-weighted mean concentration of an inert solute

We shall be concerned with the determination of the area-weighted transverse mean concentration distribution of an inert solute under the conditions that the solute is sorption

free with the wall, that the source strength is zero, and that the initial concentration distribution belongs to the product class. The results derived in Sec. 6(a) are to be used in the present discussion.

Truncation of equation (1.1) leads to the dispersion approximation

$$\frac{\partial \bar{\theta}_N}{\partial \tau} = \sum_{j=0}^{N} K_j(\tau) \frac{\partial^j \bar{\theta}_N}{\partial X^j} ,$$ (7.1)

where the number of terms N required for convergence of the approximate solution $\bar{\theta}_N(\tau, X)$ to the exact solution $\bar{\theta}(\tau, X)$, for flow with a given Peclet number, depends exclusively on time τ. Equation (6.10) shows that $N = 2$ as $\tau \to 0$ and (6.11) requires that $N \geqslant 2$ as $\tau \to \infty$. For finite time τ, (7.1) in principle could be used to yield a valid approximation by including an ever larger number of terms on the right-hand side until $\bar{\theta}_N$ absolutely converges. Although (6.9) shows that $K_j(\tau)$ in principle can be algebraically determined to all orders, their explicit evaluation, however, is a matter of every-increasing complexity as j increases. As a result, only a few low-order coefficients can be realistically determined and therefore equation (7.1) cannot in reality be employed in the sense of convergence-in-the-mean at arbitrary times. Because of its basic role in the application of the dispersion formulation, the conditions under which $\bar{\theta}_N$ for $N = 2$ can be used as an efficient representation of $\bar{\theta}$ will be examined. This is possible if a suitable number of dispersion coefficients of orders successively higher than $N = 2$ can be explicitly evaluated.

(a) *Central moments of the mean concentration distribution*

In order to see how well $\bar{\theta}_N$ of (7.1) serves to represent the exact solution $\bar{\theta}$, it is necessary, following Aris [2], to evaluate and compare their respective integral moments which can readily be obtained from (7.1) by applying the procedure used by Subramanian [19]. The nth central moment (hereafter moment) of the exact mean concentration distribution is defined as

$$\mu_n(\tau) = \int_{-\infty}^{\infty} X^n \bar{\theta} \, dX .$$ (7.2a)

and that of the Nth order approximate mean concentration distribution,

$$\mu_n^{(N)}(\tau) = \int_{-\infty}^{\infty} X^n \bar{\theta}_N \, dX ,$$ (7.2b)

for any non-negative integral value n. Multiplying (7.1) by X^n, integrating by parts over X by imposing the condition that $X^n \, \partial^j \bar{\theta}_N / \partial X^j \to 0$ as $X \to \infty$ for $j, n \geqslant 0$, it follows, by allowing $N \to \infty$, that the exact moments are governed by the equation

$$\frac{d\mu_n}{d\tau} = \sum_{j=0}^{n} (-1)^j \frac{n!}{(n-j)!} K_j \mu_{n-j} \quad \text{for } n \geqslant 0 ,$$ (7.3a)

with $\mu_{-1} = \mu_{-2} = 0$. Applying the same procedure to equation (7.1) for a fixed N, we find

$$\frac{d\mu_n^{(N)}}{d\tau} = \sum_{j=0}^{n} (-1)^j \frac{n!}{(n-j)!} K_j \mu_{n-j} \quad \text{for } n \leqslant N ,$$

$$\frac{d\mu_n^{(N)}}{d\tau} = \sum_{j=0}^{N} (-1)^j \frac{n!}{(n-j)!} K_j \mu_{n-j}^{(N)} \quad \text{for } n > N ,$$ (7.3b)

with $\mu_{-1}^{(N)} = \mu_{-2}^{(N)} = 0$. Given an initial concentration distribution, the same initial conditions are to be applied to the solutions of (7.3a) and (7.3b) which are identical in form for $n \leq N$. It therefore follows that $\mu_n(\tau) = \mu_n^{(N)}(\tau)$ for $n \leq N$. Thus the Nth order dispersion approximation gives precisely the exact moments up to the same order. We note in passing that this identity has been used by DeGance and Johns [10] as the definition of the Nth order approximation in formulating the necessary equations for the determination of the dispersion coefficients. The solutions of equations (7.3a) and (7.3b) are recorded here as follows:

$$\mu_n(\tau) = m_n + \sum_{j=0}^{n} (-1)^j \frac{n!}{(n-j)!} \int_0^\tau K_j(\eta)\mu_{n-j}(\eta)\,\mathrm{d}\eta \quad \text{for } n \geq 0, \tag{7.4a}$$

and

$$\mu_n^{(N)}(\tau) = \mu_n(\tau) \quad \text{for } 0 \leq n \leq N,$$

$$\mu_n^{(N)}(\tau) = m_n + \sum_{j=0}^{N} (-1)^j \frac{n!}{(n-j)!} \int_0^\tau K_j(\eta)\mu_{n-j}^{(N)}(\eta)\,\mathrm{d}\eta \quad \text{for } n > N, \tag{7.4b}$$

where

$$m_n = \mu_n(0) = \mu_n^{(N)}(0) \tag{7.5}$$

is the nth moment of the initial distribution. It follows from (7.4a) and (7.4b) that

$$\mu_n - \mu_n^{(N)} = \sum_{j=0}^{n-N-1} (-1)^j \frac{n!}{(n-j)!} \int_0^\tau K_j(\eta)(\mu_{n-j}(\eta) - \mu_{n-j}^{(N)}(\eta))\,\mathrm{d}\eta$$

$$+ \sum_{j=N+1}^{n} (-1)^j \frac{n!}{(n-j)!} \int_0^\tau K_j(\eta)\mu_{n-j}(\eta)\,\mathrm{d}\eta \quad \text{for } n > N. \tag{7.6}$$

Using the asymptotic results of (6.11), simple induction of (7.4a) yields

$$\mu_n(\tau) = O(\tau^{[n/2]}) \quad \text{for } n \geq 2 \text{ as } \tau \to \infty.$$

Now applying the induction procedure to (7.6), it can easily be shown that

$$\mu_n - \mu_n^{(N)} = O(\tau^{[(n-N+1)/2]}) \quad \text{for } n > N \text{ as } \tau \to \infty.$$

By employing the second moment μ_2, which is positive definite, as an effective measure of the departure of the approximate moment $\mu_n^{(N)}$ from the exact moment μ_n, there yields the large-time asymptotic behavior

$$\frac{\mu_n - \mu_n^{(N)}}{\mu_2^{n/2}} = O(\tau^{[(n-N+1)/2]-n/2}) \quad \text{for } n > N \text{ as } \tau \to \infty. \tag{7.7}$$

For small enough times, the moments under the integral signs in (7.6) may be replaced by their initial values. The small difference can be estimated by neglecting the contribution from the first summation term and one thus has

$$\mu_n - \mu_n^{(N)} = \sum_{j=N+1}^{n} (-1)^j \frac{n!}{(n-j)!} \, m_{n-j} \int_0^{\tau} K_j(\eta) \, d\eta \,,$$

which, after making use of the asymptotic results of (6.10), becomes

$$\frac{\mu_n - \mu_n^{(N)}}{\mu_2^{n/2}} = O(\tau^{[N/2]+2}) \quad \text{for } n > N \text{ as } \tau \to 0 \,, \tag{7.8}$$

where $\mu_2 = m_2 = O(\tau^0)$ has been implemented.

From equations (7.7) and (7.8), it is clear that for all $N \geq 2$ and $n > N$,

$$\frac{\mu_n - \mu_n^{(N)}}{\mu_2^{n/2}} \to 0 \quad \text{as } \tau \to 0 \text{ and as } \tau \to \infty \,. \tag{7.9}$$

It has been shown that $\mu_n^{(N)} = \mu_n$ for $n \leq N$. If $\bar{\theta}_N$ of the dispersion approximation with $N \geq 2$ is to be regarded as an efficient representation of the mean concentration $\bar{\theta}$, then acceptable quantitative agreement must exist between their respective moments having orders greater than N. Equation (7.9), using the standard deviation $(\sqrt{\mu_2})$ of the distribution as a basic measure, shows that this is certainly true at asymptotically small and large times. At finite times, however, such niceties cannot be expected to hold unaltered and the use of $\bar{\theta}_N$ to represent $\bar{\theta}$, therefore, must be carefully justified.

(b) *Criteria for application of the second-order dispersion approximation*

The approximation obtained by setting $N = 2$ in equation (7.1), first formulated by Gill [20] in that the author has showed that K_2 is time-dependent, clearly represents the simplest approach. A number of cases of theoretical and practical importance have been investigated on the basis of this model (e.g., see [9] and related references cited therein), even though its uniform applicability has been rightfully questioned (Booras and Krantz [21], Smith [15, 16]). The model is simple to use because the evaluation of K_1 and K_2 requires little or no effort. It is therefore of considerable interest to find out, for flow with specified Peclet numbers, the approximate time requirements for using its pertinent solution as an effective representation of the mean concentration distribution.

Expressed through its own Fourier transform, the exact mean concentration of the solute can be written in the form

$$\bar{\theta}(\tau, X) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega X - \omega^2 \mu_2/2} \left[e^{-(i\omega)^2 \mu_2/2} \int_{-\infty}^{\infty} e^{i\omega X'} \bar{\theta}(\tau, X') \, dX' \right] d\omega$$

$$= \frac{\mu_0}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega X - \omega^2 \mu_2/2} \left[1 + \sum_{n=1}^{\infty} \frac{(i\omega)^n}{n!} \mu_2^{n/2} c_n \right] d\omega \,, \tag{7.10}$$

where μ_0 is the total amount of solute and μ_2 is the variance of $\bar{\theta}$, and the set of quantities c_n satisfies the following identity in $a \equiv i\omega$,

$$1 + \sum_{n=1}^{\infty} \frac{a^n}{n!} \mu_2^{n/2} c_n = \frac{1}{\mu_0} e^{-a^2 \mu_2/2} \int_{-\infty}^{\infty} e^{aX} \bar{\theta} \, dX \,. \tag{7.11}$$

By expanding in the above equation the right-hand side about $a = 0$, it follows that

$$\mu_2^{n/2} c_n = \frac{1}{\mu_0} \lim_{a \to 0} \left[e^{-a^2 \mu_2/2} \int_{-\infty}^{\infty} e^{aX\bar{\theta}} \, dX \right]. \tag{7.12}$$

Since

$$\lim_{a \to 0} \frac{d^n}{da^n} e^{-a^2\mu_2/2} = (-1)^n \left(\frac{\mu_2}{2} \right)^{n/2} H_n(0),$$

$$\lim_{a \to 0} \frac{d^n}{da^n} \int_{-\infty}^{\infty} e^{aX\bar{\theta}} \, dX = \int_{-\infty}^{\infty} X^n \bar{\theta} \, dX = \mu_n,$$

equation (7.12) then gives, because $H_n(0) = 0$ for odd n,

$$c_n = \sum_{p=0}^{[n/2]} \frac{1}{2^p} \frac{n!}{p!(n-p)!} H_{2p}(0) \frac{\mu_{n-2p}}{\mu_2^{n/2-p}} \quad \text{for } n \geq 1, \tag{7.13}$$

where the Hermite polynomial

$$H_n(X) = (-1)^n e^{X^2} \frac{d^n}{dX^n} e^{-X^2}.$$

Carrying out the integration of (7.10), there follows

$$\bar{\theta} = \frac{\mu_0}{\sqrt{2\pi\mu_2}} \exp\left(\frac{-X^2}{2\mu_2} \right) \left[1 + \sum_{n=1}^{\infty} \frac{c_n}{n!} H_n\left(\frac{X}{\sqrt{2\mu_2}} \right) \right]. \tag{7.14}$$

The first three moments (μ_0, μ_1, μ_2) of the mean concentration distribution are all exactly given by the second-order dispersion approximation. It is obvious that we can write

$$\bar{\theta}_2 = \frac{\mu_0}{\sqrt{2\pi\mu_2}} \exp\left(\frac{-X^2}{2\mu_2} \right) \left[1 + \sum_{n=1}^{\infty} \frac{c_n^{(2)}}{n!} H_n\left(\frac{X}{\sqrt{2\mu_2}} \right) \right]. \tag{7.15}$$

Here

$$c_n^{(2)} = \sum_{p=0}^{[n/2]} \frac{1}{2^p} \frac{n!}{p!(n-p)!} H_{2p}(0) \frac{\mu_{n-2p}^{(2)}}{\mu_2^{n/2-p}}, \tag{7.16}$$

with $\mu_n^{(2)} = \mu_n$ for $n \leq 2$, and we thus have

$$c_n^{(2)} = c_n \quad \text{for } n \leq 2. \tag{7.17}$$

The difference between the exact and the approximate mean concentration can now be obtained by subtracting (7.15) from (7.14). Thus

$$\bar{\theta} - \bar{\theta}_2 = \frac{\mu_0}{\sqrt{2\pi\mu_2}} \exp\left(\frac{-X^2}{2\mu_2} \right) \sum_{n=3}^{\infty} \frac{c_n - c_n^{(2)}}{n!} H_n\left(\frac{X}{\sqrt{2\mu_2}} \right). \tag{7.18}$$

where the summation starts from $n = 3$ because of (7.17) and, from (7.13) and (7.16), one

has

$$c_n - c_n^{(2)} = \sum_{p=0}^{[n/2]} \frac{1}{2^p} \frac{n!}{p!(n-p)!} H_{2p}(0) \frac{\mu_{n-2p} - \mu_{n-2p}^{(2)}}{\mu_2^{n/2-p}} , \tag{7.19}$$

which clearly shows, using equation (7.9), that

$$c_n - c_n^{(2)} \to 0 \quad \text{as } \tau \to 0 \text{ and as } \tau \to \infty . \tag{7.20}$$

With the use of (7.18), we find that the following quantity which is a mean square relative error heavily weighted in favor of discrepancies at large distances from the origin,

$$\frac{\int_{-\infty}^{\infty} (\bar{\theta} - \bar{\theta}_2)^2 \exp\left(\frac{X^2}{2\mu_2}\right) dX}{\int_{-\infty}^{\infty} \bar{\theta}^2 \exp\left(\frac{X^2}{2\mu_2}\right) dX} = \frac{\sum_{n=3}^{\infty} \frac{2^n}{n!} (c_n - c_n^{(2)})^2}{1 + \sum_{n=3}^{\infty} \frac{2^n}{n!} c_n^2} \le \sum_{n=3}^{\infty} \frac{2^n}{n!} (c_n - c_n^{(2)})^2 , \tag{7.21}$$

obviously can be used as a suitable measure for the overall accuracy of $\bar{\theta}_2$ in its representation of $\bar{\theta}$. Since, as indicated by (7.20), that $c_n^{(2)} \to c_n$ asymptotically at both small and large times, it then follows that, among other possible stipulations, by demanding in equation (7.21),

$$|c_n(\tau) - c_n^{(2)}| \le \varepsilon \quad \text{for } n \ge 3 , \tag{7.22}$$

where ε, representing the tolerance for accepting $\bar{\theta}_2$ as an efficient representation of the mean concentration distribution, is a small number to be specified, the time limits for the applicability of the second-order dispersion approximation can be assessed in a systematic manner provided that $c_n(\tau)$ can be evaluated to arbitrary orders greater than 2.

In practice, however, only a few $c_n(\tau)$ of lower orders can be realistically computed. The above formulation therefore cannot be used in its full context. Chatwin [3] has essentially used a criterion equivalent to condition (7.22) with $n = 3$ as a measure for the accuracy of his analysis. A more detailed overall picture, perhaps, can better be obtained by including additional criteria with $n > 3$. A specific example is to follow in the next section.

8. Poiseuille pipe flow with a radially uniform initial concentration distribution

For the application of the results obtained in the preceding section, we consider here the determination of the area-weighted transverse mean concentration purely due to a transversely uniform initial input of a nonreactive solute ($k_A = k_s = 0$) in fully-developed laminar flow through a circular pipe. The dimensionless quantities are

$$X = \frac{x - u_0 t/2}{R \, \text{Pe}} , \qquad \rho = \frac{r}{R} , \qquad \tau = \frac{Dt}{R^2} , \qquad \text{Pe} = \frac{Ru_0}{D} ,$$

the weight function $w = 1/\pi$, the flow velocity $U = 1/2 - \rho^2$ and the initial solute input $\theta_0 = \theta(0, X, \rho) = f(X)$. Here x is the axial distance, r the radial distance, R the pipe radius,

u_0 the center line flow velocity, and $f(X)$ a prescribed square-integrable function. The required transverse eigenfunctions are

$$\psi_n = J_0(\lambda_n \rho)/J_0(\lambda_n) \quad \text{for } n = 0, 1, 2, \ldots, \tag{8.1}$$

with the corresponding eigenvalues given by $J_1(\lambda) = 0$; starting with $\lambda_0 = 0$, the eigenvalues are ordered in a sequence so that $\lambda_{n+1} > \lambda_n$.

(a) *Solution by self-consistent approximation*

The local concentration in this approximation is given by

$$\theta(\tau, X, \rho) = \sum_{n=0}^{\mathcal{N}} \phi_n(\tau, X) J_0(\lambda_n \rho)/J_0(\lambda_n) . \tag{8.2}$$

where $\mathcal{N}$ is sufficiently large so that the above solution can be considered as exact. The equations required for the determination of the functions $\phi_n(\tau, X)$ are given by equation (3.7) by setting $\beta_n = \lambda_n$ and $q_n = 0$, subject to the initial condition (2.13) which for the present case has the form

$$\phi_n(0, X) = \delta_{n0} f(X) . \tag{8.3}$$

Using the eigenfunctions (8.1) in (2.10), the factors u_{nm} are given by

$$u_{00} = 0 ,$$
$$u_{nn} = 1/6 \quad \text{for } n \neq 0 , \tag{8.4}$$
$$u_{nm} = -4(\lambda_n^2 + \lambda_m^2)/(\lambda_n^2 - \lambda_m^2)^2 \quad \text{for } m \neq n .$$

The solution of (3.7), subject to the initial condition of (8.3) can readily be obtained by applying the method of Fourier and Laplace transforms. We find the area-weighted mean concentration

$$\bar{\theta} = \phi_0 = \frac{1}{\sqrt{2\pi}} \sum_{j=0}^{\mathcal{N}} \int_{-\infty}^{\infty} F(\omega) \frac{B_j}{A_j'} \exp[a_j \tau + i(b_j \tau + \omega X)] \, d\omega , \tag{8.5}$$

where the number of terms $\mathcal{N}$ required for convergence can be realistically estimated [13],

$$F(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} f(X) e^{-i\omega X} \, dX \tag{8.6}$$

and $p = a_j + ib_j$ are the zeros of a determinant, call it $|A|$, having an element in the $(m+1)$th row and $(n+1)$th column given by

$$a_{mn} = \left(p + \lambda_m^2 + \frac{\omega^2}{Pe^2}\right)\delta_{mn} + \omega^2 k_{mn}^{(\mathcal{N})} + i\omega u_{mn} \quad \text{for } m, n = 0, 1, \ldots, \mathcal{N} , \tag{8.7}$$

$A_j' = d|A|/dp$, and B_j is the cofactor of the first element a_{00} of determinant $|A|$, both evaluated at $p = a_j + ib_j$.

(b) *Solution by dispersion approximation*

The results of Sec. 6(b) are applicable under the conditions specified at the beginning of this section. The dispersion coefficients can now be calculated by substituting (8.4) for u_{nm} in (6.13). The resulting expression of K_2 is exactly the same as that derived by Gill and Sankarasubramanian [5] and from the resulting expressions of K_3 and K_4, the steady-state ($\tau \to \infty$) values of Gill's [20] (A_k of author's equation (19)) are precisely reproduced. Noting that $K_0 = K_1 = 0$, one has thus in the dispersion approximation,

$$\frac{\partial \bar{\theta}_N}{\partial \tau} = \sum_{j=2}^{N} K_j(\tau) \frac{\partial^j \bar{\theta}_N}{\partial X^j} \quad \text{for } N = 2, 3, \ldots ,$$
(8.8)

subject to the initial condition

$$\bar{\theta}_N(0, X) = 2 \int_0^1 \rho \theta(0, X, \rho) \, d\rho = f(X) .$$
(8.9)

The complete solution is

$$\bar{\theta}_N(\tau, X) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} F(\omega) \exp\left[\sum_{j=2}^{N} (i\omega)^j \xi_j + i\omega X \right] d\omega \quad \text{for } N = 2, 3, \ldots ,$$
(8.10)

where $F(\omega)$ is given by (8.6) and

$$\xi_j(\tau) = \int_0^\tau K_j(\eta) \, d\eta ,$$
(8.11)

which, in this work, has been numerically evaluated for $j \leqslant 5$.

The dispersion coefficients K_2 through K_5 have been computed by employing the first 100 eigenvalues ($\lambda_n \leqslant 311.8$) so that little or no truncation error should occur in the process of performing the necessary numerical computations. The calculated results are plotted in Fig. 1 where it is seen that $K_2 - \text{Pe}^{-2}$ and K_5 are positive while K_3 and K_4 are negative, that these coefficients in general all increase monotonically in magnitude in the course of time (K_4 has a slight peak, not noticeable in Fig. 1, at $\tau \approx 0.6$), and that their steady-state values are reached practically at $\tau \approx 0.5$.

It is convenient to use a coordinate system in which $m_1 = 0$. Then one has from equations (7.4a) and (7.4b) that, because $K_0 = K_1 = 0$,

$$\mu_1^{(2)} = \mu_1 = 0 ,$$
$$\mu_2^{(2)} = \mu_2 = m_2 + 2\xi_2 .$$
(8.12)

Exact central moments of successively higher orders can be similarly evaluated by using (7.4a). In particular, we can numerically compute

$$\mu_3 = m_3 - 6\xi_3 ,$$
$$\mu_4 = m_4 + 12m_2\xi_2 + 12\xi_2^2 + 24\xi_4 ,$$
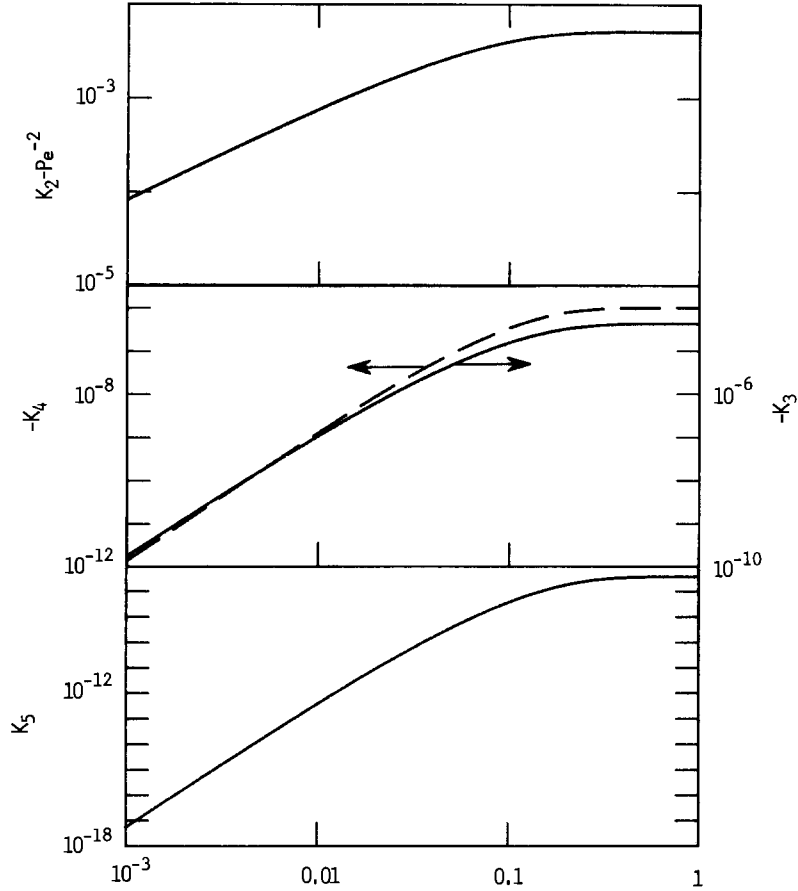$$\mu_5 = m_5 + 20m_3\xi_2 - 60m_2\xi_3 - 120\xi_2\xi_3 - 120\xi_5 .$$
(8.13)

Fig. 1. Dispersion coefficients for radially uniform initial input of an inert solute in Poiseuille pipe flow.

Correspondingly we find from (7.4b) the approximate moments,

$$\mu_3^{(2)} = m_3 ,$$

$$\mu_4^{(2)} = m_4 + 12m_2\xi_2 , \qquad (8.14)$$

$$\mu_5^{(2)} = m_5 + 20m_3\xi_2 .$$

Thus we have, by subtracting (8.14) from (8.13) and by using the result in (7.19), the effective criterion of (7.22) for using $\bar{\theta}_2(\tau, X)$ as an efficient representation,

$$|c_3 - c_3^{(2)}| = 1.5\sqrt{2}|\xi_3|/\mu_2^{1.5} \leq \varepsilon ,$$

$$|c_4 - c_4^{(2)}| = 6|\xi_4|/\mu_2^2 \leq \varepsilon , \qquad (8.15)$$

$$|c_5 - c_5^{(2)}| = 15\sqrt{2}|\xi_5|/\mu_2^{2.5} \leq \varepsilon ,$$

where μ_2 is given by (8.12).

(c) *An example*

We consider here an example with a radially uniform initial distribution of total mass unity concentrated at a section of the tube, that is, $\phi_0(0, X) = \theta_0 = f(X) = \delta(X)/\text{Pe}$. The central moments of the initial distribution are $m_0 = 1$ and $m_n = 0$ for $n \geq 1$. The solutions $\bar{\theta}$ of (8.5) by the self-consistent approximation and $\bar{\theta}_N$ $(2 \leq N \leq 5)$ of (8.10) by the dispersion approximation, with specified values of the Peclet number and the dimensionless time, can be computationally determined by using the method of fast Fourier transform [22].

Figure 2 shows a comparison of the computed solutions with $\text{Pe} = 100$ at $\tau = 0.1$. Numerical integration shows that $\bar{\theta}$ produces accurately the exact moments given by equation (8.13) and that all finite-order solutions of the dispersion approximation satisfy nearly precisely the moment requirements called for by (7.4b). $\bar{\theta}_2$ is Gaussian. $\bar{\theta}_3$ has a peak slightly downstream of the moving origin. The oscillatory character of $\bar{\theta}_4$ and $\bar{\theta}_5$ about zero, a few cycles upstream and a large number of cycles downstream extending far out to a large distance, is clearly devoid of physical meaning but strictly a mathematical fix due to the condition of square-integrability of the mean concentration distribution. It is clear that solutions of orders much higher than $N = 5$ must be pursued if the dispersion approximation is to be made effective.

Figure 3 shows the variations of $|c_n - c_n^{(2)}|$ given by (8.15) in the course of time for $\text{Pe} = 10$ and 100. As expected, these all tend to zero as $\tau \to 0$ and as $\tau \to \infty$. If the value of ε indicated in the figure is used as a criterion, we immediately see that the second-order approximation for $\text{Pe} = 10$ is efficient in the entire time domain and that, for $\text{Pe} = 100$, it is valid only in the regions outside of the range between the times indicated by the open circles in the graph. By specifying the value of ε, the quantitative relationship between τ and Pe for the applicability
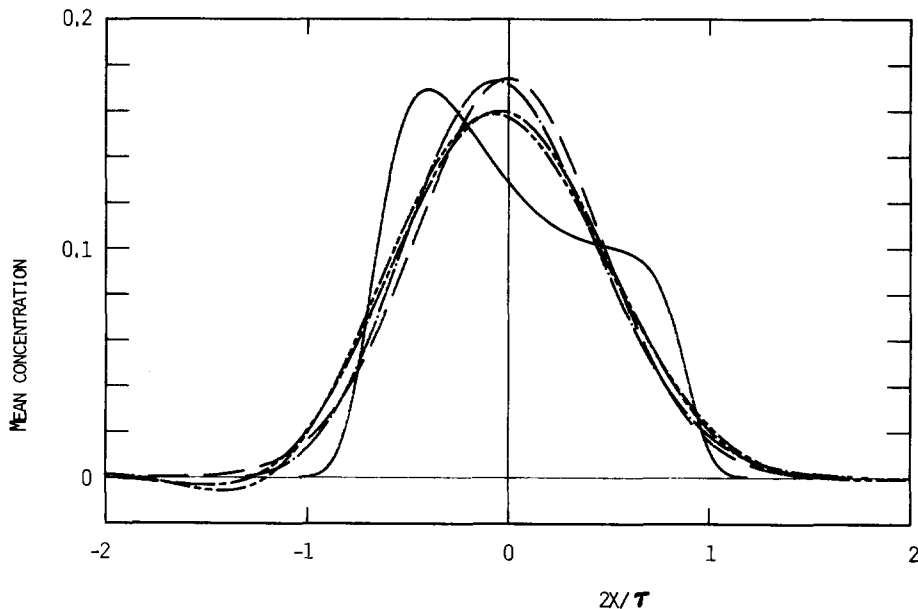

Fig. 2. Area-weighted transverse mean concentration profiles at $\tau = 0.1$ for $\text{Pe} = 100$. Exact (——); dispersion approximation: $N = 2$ (— —), $N = 3$ (—·—), $N = 4$ (—··—), $N = 5$ (—···—).
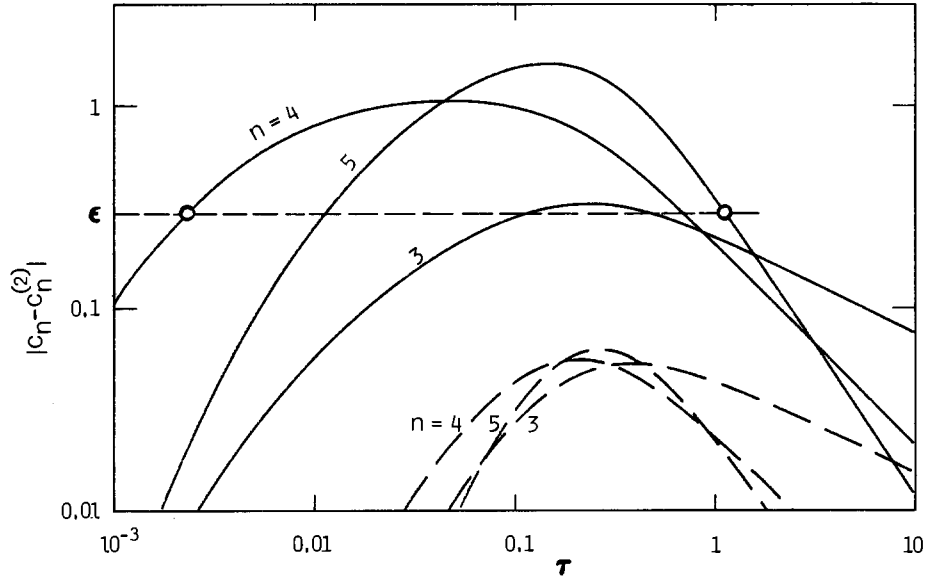
Fig. 3. Variations of $|c_n - c_n^{(2)}|$ of equation (8.15) with $m_2 = 0$ in the course of time for Pe = 10 (——) and Pe = 100 (——). With the use of ε as a criterion, the second-order dispersion approximation is not valid for Pe = 100 at times between the open circles, but it is valid in the entire time domain for Pe = 10.

of $\bar{\theta}_2$ in its representation of the solute concentration distribution therefore can be established by using the criteria of (8.15). It is clear that the smaller the value of ε, the better is the representation. Such relationships for $\varepsilon = 0.1, 0.15$ and 0.3 are shown in Fig. 4 where it is to be understood that $\bar{\theta}_2$ is valid in the open region lying to the left of each curve when
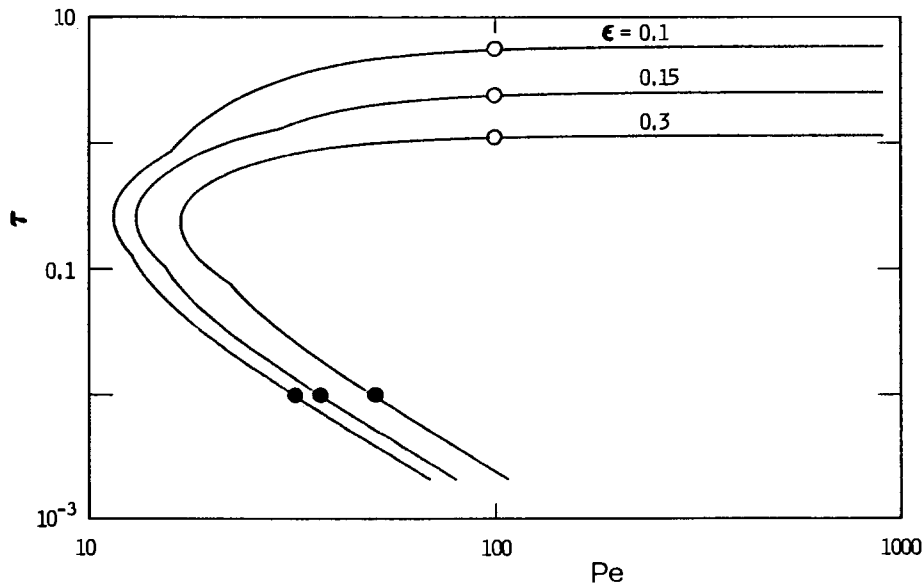

Fig. 4. Criteria for applicability of the second-order dispersion approximation obtained by using equation (8.15) with $m_2 = 0$ for $\varepsilon = 0.1, 0.15, 0.3$. $\bar{\theta}_2$ is valid in the open region lying to the left of each curve when the latter is viewed in the clockwise sense. Concentration profiles at times marked by open and closed circles are plotted in Figs 5 and 6 for visual comparison.

viewed in the clockwise sense. These results clearly demonstrate that at large enough times $\bar{\theta}_2$ is applicable independently of the Peclet number and that for flow with small enough Peclet numbers it is applicable at all times.

Figure 5 gives a comparison of $\bar{\theta}_2$ and $\bar{\theta}$ for Pe = 100 characterized by large times at $\tau = 1.1$ ($\varepsilon = 0.3$), 2.4 ($\varepsilon = 0.15$) and 5.6 ($\varepsilon = 0.1$). These are the conditions marked by open circles in Fig. 4. Also plotted here for comparison are the profiles of $\bar{\theta}_5$. It is seen that agreement between $\bar{\theta}_2$ and $\bar{\theta}$ is substantially and uniformly improved by reducing the value of ε, that the profiles of $\bar{\theta}$ are near Gaussian, and that $\bar{\theta}_5$ and $\bar{\theta}$ are essentially identical at all three times. This comparison apparently shows that $\bar{\theta}_5$ is uniformly a better representation than $\bar{\theta}_2$ at times large enough if the latter itself is a reasonable approximation.

At $\tau = 0.01$ for Pe = 32 ($\varepsilon = 0.1$), 50 ($\varepsilon = 0.15$) and 100 ($\varepsilon = 0.3$), which are the conditions marked with solid circles in Fig. 4, a comparison of $\bar{\theta}$, $\bar{\theta}_2$ and $\bar{\theta}_5$ is shown in Fig. 6. Here the effect of ε on the convergence of $\bar{\theta}_2$ to $\bar{\theta}$ is the same as before and $\bar{\theta}_5$ serves better than $\bar{\theta}_2$ in representing $\bar{\theta}$ for Pe = 32 and 50. But for Pe = 100 at a time $\tau = 0.01$, neither $\bar{\theta}_2$ nor $\bar{\theta}_5$ is satisfactory. This seems to indicate that at small times when $\bar{\theta}_2$ is not applicable, it is unlikely that $\bar{\theta}_5$ can be meaningfully effective (this is clearly also the case in Fig. 2).

From the comparisons shown in Figs 5 and 6, it appears, from a practical point of view, that $\varepsilon = 0.1$ is tolerably small for using $\bar{\theta}_2$ as a quantitatively efficient representation of the mean concentration distribution. The three individual criteria of (8.15) for $\varepsilon = 0.1$ are plotted separately in Fig. 7. The solid line is the same as that in Fig. 4 with $\varepsilon = 0.1$. Here it is seen that the large time requirements are exclusively determined by the third-order criterion, as has been done by Chatwin [3], but for $\tau < 1$ the 4th order criterion becomes dominant, while the 5th order criterion turns out primarily to accord a maximum Peclet number below which the second order dispersion approximation may be considered as tolerably valid at all times $\tau > 0$.
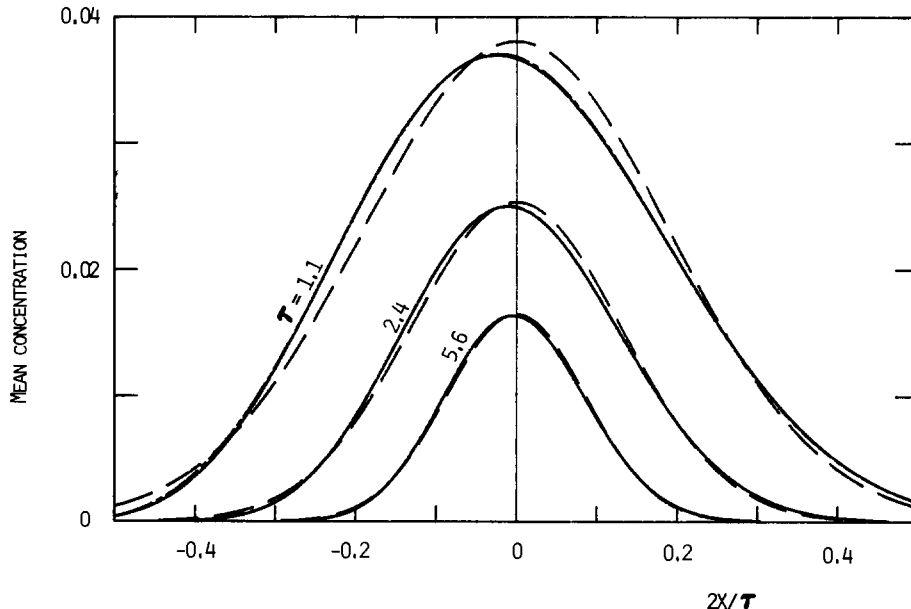

Fig. 5. Area-weighted transverse mean concentration profiles for Pe = 100 at τ = 1.1, 2.4, 5.6 (open circles in Fig. 4). Exact (———); dispersion approximation: N = 2 (— —), N = 5 (—·—).
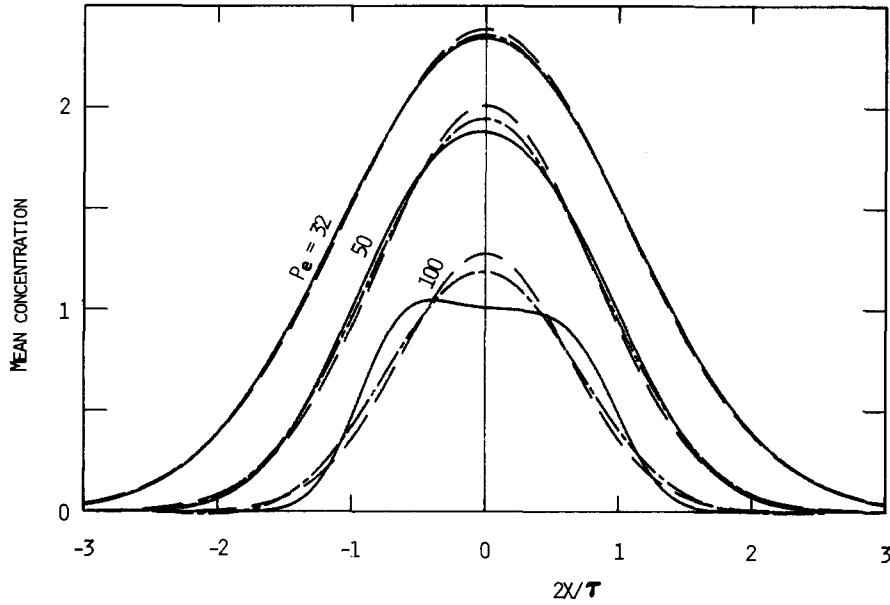
Fig. 6. Area-weighted transverse mean concentration profiles at $\tau = 0.01$ for Pe = 32, 50, 100 (solid circles in Fig. 4). Exact (———); dispersion approximation: $N = 2$ (— —), $N = 5$ (—·—).
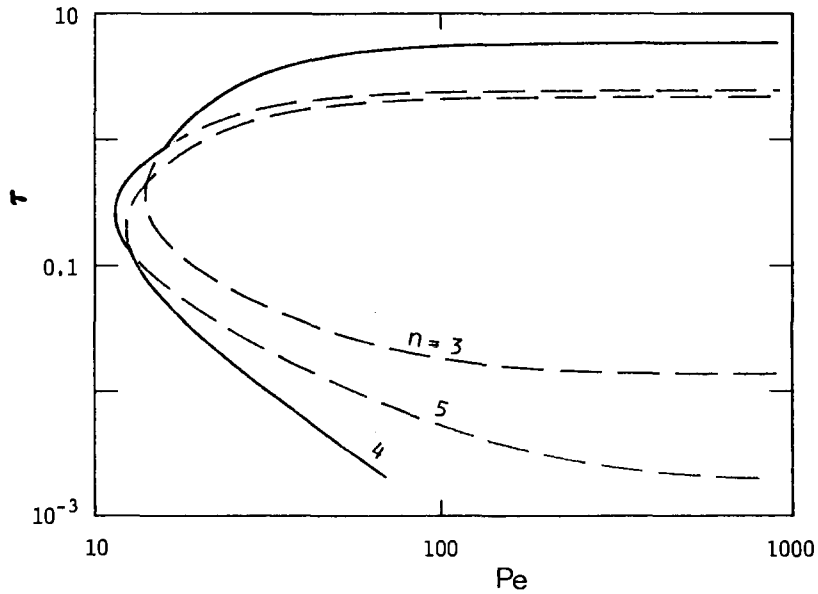

Fig. 7. Illustration of $|c_n - c_n^{(2)}| = \varepsilon$ given by equation (8.15) with $m_2 = 0$ for $\varepsilon = 0.1$. Combined criteria (———). At $\tau < 1$, the second-order dispersion approximation is valid for Peclet numbers determined by criteria of orders larger than 3. See legend of Fig. 4 for region of validity.

9. Relation to Smith's new ansatz for the area-weighted mean concentration

In conformity with Smith [15], the source strength and the initial concentration distribution are assumed to be independent of the transverse coordinates, the solute is considered to be inert, and sorption of solute is negligible. Thus the local concentration is governed by equation (2.3) with $k_A = k_s = 0$, and the transverse eigenvalue problem is prescribed by (2.4). The eigenfunctions form a complete orthonormal set as in (2.5). The lowest eigenvalue $\lambda_0 = 0$ with the corresponding eigenfunction $\psi_0 = 1$, thus the orthogonal relation,

$$\frac{L^2}{A} \int \int \psi_n(y, z) \, dy \, dz = \delta_{n0} \, . \tag{9.1}$$

The local concentration of the solute is given by (2.7) from which one evaluates the area-weighted mean concentration

$$\bar{\theta}(\tau, X) = \frac{L^2}{A} \int \int \theta \, dy \, dz = \frac{L^2}{A} \sum_{n=0}^{\infty} \phi_n(\tau, X) \int \int \psi_n \, dy \, dz = \phi_0(\tau, X) \, , \tag{9.2}$$

and hence obtains the initial condition $\bar{\theta}(0, X) = \phi_0(0, X) = \theta_0(X)$. Using (2.7) for the factor θ in the transverse diffusion and the axial convection terms, equation (2.3) becomes

$$\frac{\partial \theta}{\partial \tau} = \frac{1}{Pe^2} \frac{\partial^2 \theta}{\partial X^2} - \sum_{n=0}^{\infty} \lambda_n^2 \psi_n \phi_n - U \sum_{n=0}^{\infty} \psi_n \frac{\partial \phi_n}{\partial X} + q(\tau, X) \, . \tag{9.3}$$

Now multiplying the above equation by L^2/A and integrating over the transverse flow area, we find, for the transverse mean concentration,

$$\frac{\partial \bar{\theta}}{\partial \tau} = \frac{1}{Pe^2} \frac{\partial^2 \bar{\theta}}{\partial X^2} - \sum_{n=0}^{\infty} u_{0n} \frac{\partial \phi_n}{\partial X} + q(\tau, X) \, , \tag{9.4}$$

where u_{0n} is given by (2.10) and (9.1) has been used.

If the initial distribution and the source strength meet the requirement of (4.12), then (4.20) is valid and we have, with the substitution of ϕ_0 by $\bar{\theta}$ as is allowed by (9.2),

$$\phi_n(\tau, X) = \sum_{j=0}^{\infty} A_{nj}(\tau) \frac{\partial^j \bar{\theta}}{\partial X^j} \, . \tag{9.5}$$

Subtracting (9.4) from (9.3), making use of (9.5) for the terms involving ϕ_n, we find

$$\frac{\partial}{\partial \tau} (\theta - \bar{\theta}) - \frac{1}{Pe^2} \frac{\partial^2}{\partial X^2} (\theta - \bar{\theta}) = \sum_{j=0}^{\infty} M_j(y, z, \tau) \frac{\partial^j \bar{\theta}}{\partial X^j} \, , \tag{9.6}$$

where

$$M_j(y, z, \tau) = - \sum_{n=0}^{\infty} [A_{nj}(\lambda_n^2 \psi_n - u_{0n}) + (1 - \delta_{j0}) U \psi_n A_{n,j-1}] \, , \quad j = 0, 1, \ldots . \tag{9.7}$$

The solution of equation (9.6) is, taking into account the fact that $\bar{\theta} = \theta$ at $\tau = 0$,

$$
\theta - \bar{\theta} = \frac{\text{Pe}}{2\sqrt{\pi}} \sum_{j=0}^{\infty} \int_0^{\tau} d\tau_s \, M_j(y, z, \tau - \tau_s)
$$
$$
\times \int_{-\infty}^{\infty} dX_s \, \frac{\partial^j \bar{\theta}(X_s, \tau - \tau_s)}{\partial X_s^j} \frac{1}{\sqrt{\tau_s}} \exp\left[-\frac{\text{Pe}^2(X - X_s)^2}{4\tau_s} \right] . \tag{9.8}
$$

We proceed to show that the new ansatz proposed by Smith can be derived from equation (9.8) as a result of proper simplification. Expansion of $\bar{\theta}(X_s, \tau - \tau_s)$ about X gives

$$
\frac{\partial^j \bar{\theta}(X_s, \tau - \tau_s)}{\partial X_s^j} = \sum_{l=0}^{\infty} \frac{1}{l!} \frac{\partial^{j+l} \bar{\theta}(X, \tau - \tau_s)}{\partial X^{j+l}} (X_s - X)^l . \tag{9.9}
$$

Substituting (9.9) into (9.8), carrying out the integration over X_s term by term, we can show that

$$
\theta - \bar{\theta} = \sum_{j=0}^{\infty} \sum_{l=0}^{[j/2]} \int_0^{\tau} \frac{M_{j-2l}(y, z, \tau - \tau_s)}{l!} \left(\frac{\tau_s}{\text{Pe}} \right)^{2l} \frac{\partial^j \bar{\theta}(X, \tau - \tau_s)}{\partial X^j} d\tau_s . \tag{9.10}
$$

In terms of the memory displacement

$$
\tilde{X}(\tau_s) = \int_0^{\tau_s} \tilde{u}(r') \, d\tau' , \tag{9.11}
$$

where $\tilde{u}(\tau)$ is the velocity shift introduced by Smith, one has the series expansion

$$
\frac{\partial^j \bar{\theta}(X, \tau - \tau_s)}{\partial X^j} = \sum_{k=0}^{\infty} \frac{1}{k!} \frac{\partial^{j+k} \bar{\theta}(X - \tilde{X}(\tau_s), \tau - \tau_s)}{\partial X^{j+k}} \tilde{X}^k(\tau_s) . \tag{9.12}
$$

Use of the above expression in (9.10) gives

$$
\theta - \bar{\theta} = \sum_{j=0}^{\infty} \int_0^{\tau} L_j(y, z, \tau; \tau_s) \frac{\partial^j}{\partial X^j} \bar{\theta}(X - \tilde{X}(\tau_s), \tau - \tau_s) \, d\tau_s , \tag{9.13}
$$

with

$$
L_j(y, z, \tau; \tau_s) = \sum_{k=0}^{\infty} \sum_{l=0}^{[k/2]} \frac{1}{l!(j-k)!} M_{k-2l}(y, z, \tau - \tau_s) \tilde{X}^{j-k}(\tau_s) \left(\frac{\tau_s}{\text{Pe}} \right)^{2l} . \tag{9.14}
$$

Equation (9.13) is a complete representation relating the concentration variations across the transverse flow area to the X-derivatives of the area-weighted mean concentration in terms of the memory displacement $\tilde{X}(\tau_s)$. To obtain a series representation somewhat similar to the particular form proposed by Smith, it is necessary to further make use of the series expansion

$$
M_{k-2l}(y, z, \tau - \tau_s) = \sum_{k=0}^{\infty} \frac{1}{n!} \frac{\partial^n M_{k-2l}(y, z, \tau_s)}{\partial \tau_s^n} (\tau - 2\tau_s) , \tag{9.15}
$$

so that equation (9.13) can be written as

$$\theta - \bar{\theta} = \sum_{j=0}^{\infty} \sum_{n=0}^{\infty} \int_0^{\tau} l_{jn}(y, z, \tau_s) \frac{\partial^j}{\partial X^j} \bar{\theta}(X - \tilde{X}(\tau_s), \tau - \tau_s) \, d\tau_s , \tag{9.16}$$

where

$$l_{jn}(y, z, \tau_s) = \sum_{k=0}^{\infty} \sum_{l=0}^{[k/2]} \frac{1}{l! n! (j-k)!} \frac{\partial^n M_{k-2l}(y, z, \tau_s)}{\partial \tau_s^n} (\tau - 2\tau_s)^n \tilde{X}^{j-k}(\tau_s) \left(\frac{\tau_s}{\mathrm{Pe}}\right)^{2l}. \tag{9.17}$$

Truncation of equation (9.16) after $n = 0$ gives the first nontrivial approximation of the memory displacement formulation,

$$\theta - \bar{\theta} = \sum_{j=0}^{\infty} \int_0^{\tau} l_{j0}(y, z, \tau_s) \frac{\partial^j}{\partial X^j} \bar{\theta}(X - \tilde{X}(\tau_s), \tau - \tau_s) \, d\tau_s , \tag{9.18}$$

where

$$l_{j0}(y, z, \tau_s) = \sum_{k=0}^{j} \sum_{l=0}^{[k/2]} \frac{1}{l! (j-k)!} M_{k-2l}(y, z, \tau - 2\tau_s)(\tau - 2\tau_s)^n \tilde{X}^{j-k}(\tau_s) \left(\frac{\tau_s}{\mathrm{Pe}}\right)^{2l}. \tag{9.19}$$

For the lowest eigenmode, $\lambda_0 = u_{00} = 0$ and we have from (9.7),

$$M_0 = - \sum_{n=1}^{\infty} A_{n0}(\lambda_n^2 \psi_n - u_{0n}) = - \sum_{n=1}^{\infty} R_n(\lambda_n^2 \psi_n - u_{0n}) ,$$

where $R_n(\tau) = R_n^{(0)}(\tau)$ is defined by (4.13) and, to get to the second expression, equations (4.19) and (4.21) have been used. Thus if we confine ourselves to the special case (for example, the case of Sec. 6(b)) such that

$$R_n(\tau) = 0 \quad \text{for } n \geq 1 , \tag{9.20}$$

then $l_{00} = M_0 = 0$, and we have the result

$$\theta - \bar{\theta} = \sum_{j=1}^{\infty} \int_0^{\tau} l_{j0}(y, z, \tau_s) \frac{\partial^j}{\partial X^j} \bar{\theta}(X - \tilde{X}(\tau_s), \tau - \tau_s) \, d\tau_s , \tag{9.21}$$

which is identical to the new ansatz posed by Smith [15] (author's equation (1.5) with $l_j = l_{j0}$). Here the upper limit is τ instead of ∞ as used by Smith. But this does not change the nature of the equation because $\bar{\theta}$ under the integral sign is defined only for nonnegative time. The theoretical basis of Smith's new ansatz is therefore here established by recognizing that what it represents, valid under the simplifying condition of (9.20), is a truncated form of the full expansion given by equation (9.16). By retaining only the first term in equation (9.21), Smith introduced a delay-diffusion equation for the description of the area-weighted mean concentration distribution.

Figure 8 shows a comparison of the mean concentration distributions for the case of a radially uniform slug input of unit mass in fully developed laminar pipe flow obtained by Gill and Ananthakrishnan [23] through finite-difference integration, by Smith [15] from the delay-diffusion equation with a particular choice for the velocity shift $\tilde{u}$, and by the present
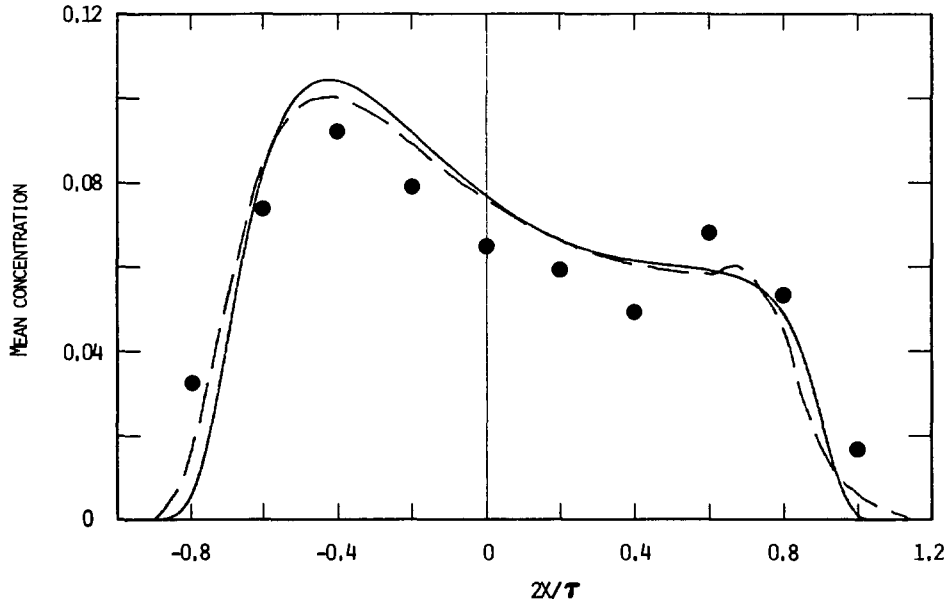
Fig. 8. Area-weighted transverse mean concentration profiles at $\tau = 0.1$ for Pe = 1000 in Poiseuille pipe flow with an initial slug input. Exact (——), finite-difference of Gill and Ananthakrishnan [23] (— —), and delay-diffusion of Smith [15] (●).

authors using the self-consistent formulation of Sec. 8(a). The self-consistent solution satisfies precisely the requirements $\mu_0 = 1$, $\mu_1 = 0$, and the higher-order integral moments given by (8.12) and (8.13). A numerical integration of the finite-difference solution shows that, although $\mu_0 = 1$ is satisfied, the magnitude of the integral of the first-moment distribution about the origin over the downstream region is nearly 7% larger than that over the upstream region. The requirement that $\mu_1 = 0$ is violated, showing that there are gross errors in the finite-difference solution. The double-peaked appearance of the distribution obviously is the result of its inherent inaccuracy. Though serving as a significant improvement over the Gaussian solution of the second-order dispersion approximation, the solution of Smith's evidently is far from being adequate for representing the mean concentration distribution at this stage ($\tau = 0.1$) of the dispersion process.

10. Concluding remarks

The dispersion formulation represented by equation (1.1) as originally advocated by Gill and Sankarasubramanian, or by equation (5.11) if there exists a distributed source strength, is not completely equivalent to the convective diffusion problem represented by equation (2.3) if the mathematical condition called for by (4.12) is not met.

Under the conditions that the dispersion formulation is valid, the pertinent coefficients in principle can be determined either by the method of Hermite moments of DeGance and Johns [10] or by the algebraic formulas developed in Sec. 5 of this paper. The use of the dispersion formulation for practical applications, however, is largely limited to the second-order approximation because the algebraic contents of the higher-order coefficients, even for the simplest cases, are highly complicated and extremely tedious to manage.

The case of an inert solute in source-free flows with an initial input belonging to the broad category of the product class has been examined in detail for the determination of the area-weighted mean concentration by means of the dispersion approximation of fixed orders. The present analysis shows that the second-order approximation is sufficiently efficient asymptotically both at small and at large times and its relative accuracy as a representation of the mean concentration distribution at arbitrary finite times can be systematically assessed if dispersion coefficients higher than the second order can be determined in succession. For an illustration of using the derived results, the case of an inert solute in Poiseuille piper flow with a radially uniform initial input is presented in detail.

The use of the self-consistent method in the form of equation (3.7) for practical calculations is a straightforward matter. For any desired accuracy, the convergent solution can be computationally determined by including a sufficiently large number of terms in these equations. The procedure of solution is especially simple with inert solute in flows free of sources for the determination of the area-weighted mean concentration because of the simplification that $\bar{\theta} = \phi_0$.

Like the dispersion formulation, the memory displacement approach suggested by Smith can be rigorously derived if the series expansion of equation (4.12) is valid. Under the condition that the distributions of the source strength and the initial concentration are transversely uniform, the difference $\theta - \bar{\theta}$ is exactly given by equation (9.8). The new ansatz proposed by Smith is shown to represent, subject to the additional restriction of equation (9.20), the lowest-order nontrivial approximation possible to the expansion of equation (9.16) which is a double-series representaion of the master solution given by equation (9.8).

References

1. G.I. Taylor, Dispersion of soluble matter in solvent flowing slowly through a tube, *Proc. Roy. Soc. Lond.* A219 (1953) 186–203.
2. R. Aris, On the dispersion of a solute in fluid flowing through a tube, *Proc. Roy. Soc. Lond.* A235 (1956) 67–77.
3. P.C. Chatwin, The approach to normality of the concentration distribution of a solute in a solvent flowing along a straight pipe, *J. Fluid Mech.* 43 (1970) 321–352.
4. M.J. Lighthill, Initial development of diffusion in Poiseuille flow, *J. Inst. Maths Applics* 2 (1966) 97–108.
5. W.N. Gill and R. Sankarasubramanian, Exact analysis of unsteady convective diffusion, *Proc. Roy. Soc. Lond.* A316 (1970) 341–350.
6. W.N. Gill and R. Sankarasubramanian, Dispersion of a non-uniform slug in time dependent flow, *Proc. Roy. Soc. Lond.* A322 (1971) 101–117.
7. R. Sankarasubramanian and W.N. Gill, Dispersion from a prescribed concentration distribution in time-variable flow, *Proc. Roy. Soc. Lond.* A329 (1972) 479–492.
8. R. Sankarasubramanian and W.N. Gill, Unsteady convective diffusion with interphase mass transfer, *Proc. Roy. Soc. Lond.* A333 (1973) 115–132.
9. M.R. Doshi, P.M. Daiya and W.N. Gill, Three dimensional laminar dispersion in open and closed rectangular conduits, *Chem. Eng. Sci.* 33 (1978) 795–804.
10. A.E. DeGance and L.E. Johns, The theory of dispersion of chemically active solute in a rectilinear flow field, *Appl. Sci. Res.* 34 (1978) 189–225.
11. A.E. DeGance and L.E. Johns, On the construction of dispersion approximations to the solution of the convective diffusion equation, *AIChE J.* 26 (1980) 411–419.
12. I. Frankel and H. Brenner, On the foundations of generalized Taylor dispersion theory, *J. Fluid Mech.* 204 (1989) 97–119.
13. J.S. Yu, Laminar dispersion for flow through round tubes, *ASME J. Appl. Mech.* 46 (1979) 750–756.
14. J.S. Yu, Dispersion in laminar flow through tubes by simultaneous diffusion and convection, *ASME J. Appl. Mech.* 48 (1981) 217–223.
15. R. Smith, A delay-diffusion description for contaminant dispersion, *J. Fluid Mech.* 105 (1981) 469–486.

16. R. Smith, Non-uniform discharge of contaminants in shear flows, *J. Fluid Mech.* 120 (1982) 79–89.
17. P.H. Morse and H. Feshbach, *Methods of Theoretical Physics* (Chapters 2 and 7), McGraw-Hill (1953).
18. H.S. Wilf, *Mathematics for the Physical Sciences*, Wiley (1962).
19. R.S. Subramanian, Unsteady convective diffusion in capillary chromatography, *J. Chromatogr.* 101 (1974) 253–270.
20. W.N. Gill, A note on the solution of transient dispersion problems, *Proc. Roy. Soc. Lond.* A298 (1967) 335–339.
21. G.S. Booras and W.B. Krantz, Dispersion in laminar flow of power-law fluids through straight tubes, *Ind. Eng. Chem. Fundam.* 15 (1976) 249–254.
22. E.O. Brigham, *The Fast Fourier Transform*, Prentice-Hall (1974).
23. W.N. Gill and V. Ananthakrishnan, Laminar dispersion in capillaries: part 4, the slug stimulus, *AIChE J.* 13 (1967) 801–807.