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Lagrangian approach to time-dependent laminar dispersion in rectangular conduits. Part 1. Two-dimensional flows

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Time-dependent mean velocities and dispersion coefficients are evaluated for a general two-dimensional laminar flow. A Lagrangian method is adopted by which a Brownian particle is traced in an artificially restructured velocity field. Asymptotic expressions for short, medium and long periods of time are obtained for Couette flow, plane Poiseuille flow and open-channel flow over an inclined flat surface. A new formula is suggested by which the Taylor dispersion coefficient can be evaluated from purely kinematical considerations. Within an error of less than one percent, over the entire time domain and for various flow fields, a very simple analytical expression is derived for the time-dependent dispersion coefficient

$$\tilde{D}(\tau) = D + D^{\mathrm{T}}\left(1 - \frac{1-\mathrm{e}^{-a\tau}}{a\tau}\right),$$

where D is the molecular diffusion coefficient, D^{T} denotes the Taylor dispersion coefficient, τ stands for the non-dimensional time $\pi^2 Dt/Y^2$, Y is the distance between walls and $a = (N+1)^2$ is an integer which is determined by the number of symmetry planes N that the flow field possesses. For Couette and open-channel flow there are no planes of symmetry and $a = 1$; for Poiseuille flow there is one plane of symmetry and $a = 4$.

1. Introduction

The present work is concerned with the transport of a Brownian particle by a non-uniform flow.

The diffusion of a small sphere in a Poiseuillian velocity field was first studied by Taylor (1953) and Aris (1956), who found that the total dispersion is the result of two contributions: the molecular diffusion, and the so-called Taylor dispersion, which is produced by the coupling between the Brownian motion and the non-uniform velocity field. Doshi, Daiya & Gill (1978) extended Taylor's solution to include open and closed rectangular conduits.

Recently, Brenner (1980, 1982) has studied the Taylor dispersion phenomenon under very general conditions, viewing the solute transport process as occurring in an abstract multidimensional phase space.

The analysis performed in the present article adopts Brenner's point of view. In particular, the transport process is described from the perspective of a single

Brownian solute particle, thus eliminating the irrelevant question of initial solute concentration.

However, unlike Taylor and Brenner, who adopted a kinematic (Eulerian) approach,† we characterize the stochastic trajectory of a Brownian particle through a dynamical (Lagrangian) viewpoint. This point of view was also shared by Van den Broeck (1982), Dewey & Sullivan (1979, 1982) and others. However, the latter employed the known solution for the conditional probability distribution of particle position in a bounded region (Chandrasekhar 1943 and Budak, Samarski & Tikhonov 1964) to derive the time-dependent dispersion coefficient. In other words, tacitly, the solution of Fokker–Plank's equation for the probability distribution function in a bounded region was prerequisite to any progress in the analysis. In our treatment a purely Lagrangian approach is adopted to evaluate the first and second moments of the particle position and the explicit solution of the conditional probability distribution in a bounded region is not required.

Since kinematic and dynamical techniques provide alternatives but equivalent methods to describe the Brownian motion, they will lead to the same results, namely to the same values for the mean global particle velocity and the total dispersivity coefficient.

Finally, the general time-dependent velocity and dispersion of particles in a fluid flowing in a two-dimensional duct are discussed. Asymptotic expressions are obtained for three particular cases: Couette flow, Poiseuille flow, and an open flow over an inclined flat plate. Three-dimensional problems are discussed in a subsequent paper, Part 2, where the flows in rectangular closed and open ducts are addressed as particular cases.

2. Statement of the problem

A rigid particle is immersed in a fluid flowing in the x-direction, which is stationary in time and uniform along x.

Assume that the condition of local equilibrium is fulfilled, namely (i) a so-called averaging timescale t_a can be defined, which is much shorter than the macroscopic timescale and much longer than the correlation time of the Brownian velocity. (ii) when time is spanned through the t_a timescale the particle is in equilibrium with the surrounding fluid, i.e. it does not dissipate macroscopically. According to (ii) no external force besides the hydrodynamic interaction acts on the particle; moreover the condition (i) states that in the t_a timescale, variations in the flow field are not perceived.

Thus during a span of time of order t_a the particle rotates randomly innumerable times, without having any preferential orientation. That means that in the t_a timescale the particle behaves as if it were isotropic,‡ with drag coefficient Z.

Let us consider the motion of one particle. Its random position is described by the coordinates x, y and z of any of its points, where x, y and z form an orthogonal coordinate system.

Since in the t_a timescale the inertial force is negligible, the Langevin equation in the x-direction reads

$$Z[\dot{x}(t)-V(y(t),z(t))]=f(t), \tag{1}$$

† In a recent article, Dill & Brenner (1983) applied a mixed kinematic–dynamical method, but they concluded that the kinematic approach offers the most direct route of solution.

‡ For this reason, in the kinematic theory of gases, the probability function of the gas molecules in their phase space is independent of orientation of the molecules (see Lifschitz & Pitaevski 1981). This, however, is incorrect for particles very close to the boundaries.

where $V(y, z)$ is the velocity of the incident flow, while the Brownian force $f(t)$ satisfies the following statistical conditions at local equilibrium (see, for example, Van Kampen 1981; Mauri & Haber 1983):

$$\langle f(t) \rangle = 0, \tag{2a}$$

$$\langle f(t) f(t+\tau) \rangle = 2KTZ\,\delta(\tau), \tag{2b}$$

$$\langle f(t)\, V(y, z) \rangle = 0. \tag{2c}$$

Here K is Boltzmann's constant and T is the absolute temperature.

The y- and z-coordinates of the particle's position satisfy Langevin equations similar to (1) with $V = 0$. This results in a free diffusion process in the y- and z-directions.

3. Two-dimensional Taylor dispersion – general approach

Let us first consider the case of a two-dimensional velocity field $V(y)$ with $0 \leqslant y \leqslant Y$. This case includes most (but not all) of the ingredients of three-dimensional cases and is easier to analyse.

Substitution of (2) into (1) yields

$$\langle \dot{x}(t) \rangle = \langle V(y) \rangle = \langle V(y_0 + \Delta y) \rangle, \tag{3}$$

where y_0 is the known position at time $t_0 = 0$ of the particle and Δy is the displacement of the particle after time $\Delta t = t - t_0$. The foregoing mean velocity can be evaluated by

$$\langle V(y_0 + \Delta y) \rangle = \int_{-y_0}^{Y-y_0} V(y_0 + \Delta y)\, P(y_0 + \Delta y;\, \Delta t \,|\, y_0)\, \mathrm{d}(\Delta y), \tag{4}$$

where $P(y_0 + \Delta y;\, \Delta t\,|\, y_0)$ is the probability density of locating a particle at position $y_0 + \Delta y$ after a time period Δt given that the initial position of the particle is y_0.

Similarly, from (1) and ($2b, c$), one obtains for the velocity autocorrelation

$$\langle \dot{x}(t_2)\, \dot{x}(t_1) \rangle = 2D\delta(\Delta t) + \langle V(y_2)\, V(y_1) \rangle, \tag{5}$$

where $\Delta t = |t_2 - t_1|$, $D = KT/Z$ is the molecular diffusivity, while

$$\langle V(y_2)\, V(y_1) \rangle = \int_0^Y \int_0^Y V(y_2)\, V(y_1)\, P(y_2;\, t_2, y_1;\, t_1 \,|\, y_0)\, \mathrm{d}y_1\, \mathrm{d}y_2. \tag{6}$$

Here $P(y_1;\, t_1, y_2;\, t_2\,|\, y_0)$ is the probability density of locating the particle at position y_1 after a time period t_1 and at position y_2 after a time period t_2 given the particle was initially positioned at y_0. If $t_2 > t_1 > 0$ and the process is Markoffian,

$$\begin{aligned} P(y_2;\, t_2, y_1;\, t_1 \,|\, y_0) &= P(y_2;\, t_2 \,|\, y_1;\, t_1)\, P(y_1;\, t_1 \,|\, y_0) \\ &= P(y_1 + \Delta y_2;\, \Delta t_2 \,|\, y_1)\, P(y_0 + \Delta y_1;\, \Delta t_1 \,|\, y_0), \end{aligned}$$

where $$\Delta y_2 = y_2 - y_1, \quad \Delta y_1 = y_1 - y_0, \quad \Delta t_2 = t_2 - t_1.$$

Hence,

$$\begin{aligned} \langle V(y_2)\, V(y_1) \rangle = \int_{-y_0}^{Y-y_0} & V(y_0 + \Delta y_1)\, P(y_0 + \Delta y_1;\, \Delta t_1 \,|\, y_0)\, \mathrm{d}(\Delta y_1) \\ & \times \int_{-y_0-\Delta y_1}^{Y-y_0-\Delta y_1} V(y_1 + \Delta y_2)\, P(y_1 + \Delta y_2;\, \Delta t_2 \,|\, y_1)\, \mathrm{d}(\Delta y_2) \end{aligned} \tag{7}$$

for $t_2 > t_1$. Similar expressions can be obtained for the case where $t_2 < t_1$. Similarly, higher moments of the velocity profile can be obtained utilizing the relation between probability densities for Markoffian processes, namely,

$$P(y_n;t_n, y_{n-1};t_{n-1}\dots y_1;t_1\,|\,y_0) \\ = P(y_n;t_n\,|\,y_{n-1};t_{n-1})\,P(y_{n-1};t_{n-1}\,|\,y_{n-2};t_{n-2})\dots P(y_1;t_1\,|\,y_0),$$

where $t_n > t_{n-1} > \dots > t_1 > t_0$. Consequently only a single two-point conditional probability distribution is required to evaluate (4) and (6). If, however, the mean velocity or higher velocity moments are sought for a large number of particles placed initially at various y_0 positions, the initial distribution $P(y_0)$ is also needed. Generally, this distribution is *a priori* given and we shall thereafter assume it to be $1/Y$ (particles uniformly distributed at the initial position). In this case the ensemble mean velocity and the velocity autocorrelation are (denoted by double angular brackets):

$$\langle\langle V(y)\rangle\rangle = \frac{1}{Y}\int_0^Y \mathrm{d}y_0 \int_{-y_0}^{Y-y_0} V(y_0+\Delta y)\,P(y_0+\Delta y;\Delta t\,|\,y_0)\,\mathrm{d}(\Delta y), \tag{8}$$

$$\langle\langle V(y_1)\,V(y_2)\rangle\rangle = \frac{1}{Y}\int_0^Y \mathrm{d}y_0 \int_{-y_0}^{Y-y_0} V(y_0+\Delta y_1)\,P(y_0+\Delta y_1;\Delta t_1\,|\,y_0)\,\mathrm{d}(\Delta y_1) \\ \times \int_{-y_0-\Delta y_1}^{Y-y_0-\Delta y_1} V(y_1+\Delta y_2)\,P(y_1+\Delta y_2;\Delta t_2\,|\,y_1)\,\mathrm{d}(\Delta y_2). \tag{9}$$

The foregoing integrations rely heavily on the yet unknown and complex two-point conditional probability. For an unbounded velocity field, a simple Gaussian distribution could be applied,

$$P(y+\Delta y;\Delta t\,|\,y) = (4\pi D\Delta t)^{-\frac{1}{2}}\exp[-(\Delta y)^2/4D\Delta t], \tag{10}$$

since no external velocity components exist in the y-direction and the molecular diffusion mechanism prevails.

In our case, however, the particle is bounded by the conduit walls. Consequently, the Gaussian formula is inadequate and boundary conditions at the walls must be specified. A possible condition is that the particle bounces off the walls and no deposition occurs. In this case the probability of a particle reaching a position $y_1 = y_0+\Delta y$ after a time period Δt equals the sum of the probabilities of a particle reaching positions y_1, y_2, y_3, etc. *in an unbounded field* (see figure 1). These locations are multiple mirror images of y_1 with respect to the $y = 0$ and $y = Y$ planes.† Hence, integrating the velocity profile (or any other function of y defined in $0 < y < Y$) *over all possible positions of a bounded y-domain is identical to integrating over an infinite y-domain with a velocity field periodically extended to infinity such that its semi-period is* Y *and* $y = 0$ (*or* $y = Y$) *is a symmetry plane.*

Thus, instead of deriving the complex probability distribution for the bounded domain and then integrating over a finite domain, the Gaussian distribution can be used if we simply expand the velocity profile by a cosine Fourier series

$$V(y) = \sum_{n=0}^{\infty} V_n \cos(\gamma_n y) \tag{11}$$

† This notion was devised by Chandresekhar (1943) and Oppenheim & Mazur (1964) to derive the conditional probability distribution for a particle in a *bounded* quiescent fluid. Here we employ the same idea not for the probability distribution but rather to the velocity field.

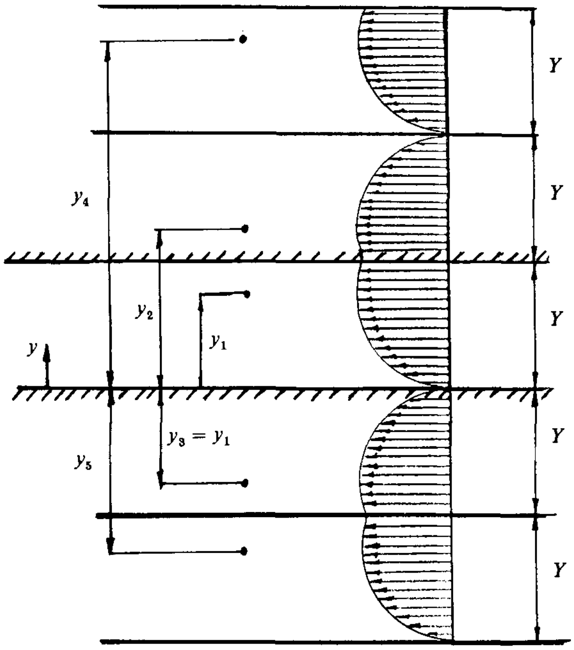

FIGURE 1. The extended velocity distribution.

and integrate over an infinite y-domain. Here

$$\gamma_n = \frac{n\pi}{Y}, \tag{12}$$

and the zero harmonic is

$$V_0 = \bar{V} = \int_0^Y V(y)\,\mathrm{d}y, \tag{13}$$

where $\bar{V}$ is the mean velocity of the fluid and

$$V_n = \frac{1}{Y}\int_0^Y V(y)\cos(\gamma_n y)\,\mathrm{d}y \quad (n = 1, 2, 3, \ldots) \tag{14}$$

are the amplitudes of the higher harmonics.

3.1. *The mean axial velocity and displacement of a particle*

Based on the analysis of the previous section, the mean velocity of a particle initially at y_0 is

$$\begin{aligned}\langle V(y)\rangle &= \int_{-y_0}^{Y-y_0} V(y_0+\Delta y)\,P(y_0+\Delta y;\,\Delta t\,|\,y_0)\,\mathrm{d}(\Delta y)\\ &= (4\pi D\Delta t)^{-\frac{1}{2}}\sum_{n=0}^{\infty} V_n\int_{-\infty}^{\infty}\cos\gamma_n(y_0+\Delta y)\exp\left[-(\Delta y)^2/4D\Delta t\right]\mathrm{d}(\Delta y)\\ &= \sum_{n=0}^{\infty} V_n\exp\left[-\gamma_n^2 D\Delta t\right]\cos(\gamma_n y_0) = \bar{V} + \sum_{n=1}^{\infty} V_n\exp\left[-\gamma_n^2 D\Delta t\right]\cos(\gamma_n y_0),\end{aligned} \tag{15}$$

where a formula in Gradshteyn & Ryzhik (1965, p. 410) was used to carry out the integration. For long time periods this result coincides with that observed by

Griffiths (1911) and Taylor (1953). Since when $\Delta t \to \infty$ the sum in (15) dies out, the mean velocity of a particle is independent of its initial position y_0 and is identical to the mean velocity of the fluid. Consequently, the ensemble mean velocity $\langle\langle V(y)\rangle\rangle$ would be unaffected by the initial probablity distribution after a long time period. If a uniform initial distribution is assumed, the ensemble average is

$$\langle\langle V(y)\rangle\rangle = \bar{V} \tag{16}$$

for all times. It is quite remarkable to notice that for this particular case only the ensemble mean yields a constant at all times. A pertinent timescale in the foregoing case is $Y^2/\pi^2 D$, approximately the time a particle will take to reach the walls by molecular diffusion.

It is useful to define a non-dimensional time variable based on this timescale,

$$\tau = \frac{\pi^2 t D}{Y^2}, \tag{17}$$

by which $\tau > 1$ and $\tau < 1$ pertain to 'long' time periods or 'short' time periods respectively. The mean distance travelled by a particle along x can easily be calculated by the following integration:

$$\begin{aligned}\langle x(t)\rangle &= \left\langle \int_0^t \dot{x}(t)\,\mathrm{d}t \right\rangle = \int_0^t \langle \dot{x}(t)\rangle\,\mathrm{d}t = \int_0^t \langle V(y)\rangle\,\mathrm{d}t \\ &= \bar{V}t + \frac{Y^2}{\pi^2 D}\sum_{n=1}^{\infty}\frac{V_n}{n^2}(1-\mathrm{e}^{-n^2\tau})\cos(\gamma_n y_0).\end{aligned} \tag{18}$$

The ensemble average for an initial uniform probability distribution is clearly

$$\langle\langle x(t)\rangle\rangle = \bar{V}t \tag{19}$$

for all times. Nonetheless, non-uniform initial distributions introduce a constant error. For example, if we decide to trace a small blob of particles located initially at $y_0 = \frac{1}{2}Y$ only even harmonics would survive in (18) and

$$\langle x(t)\rangle = \bar{V}t + \frac{Y^2}{\pi^2 D}\sum_{n=2,4}^{\infty}(-1)^{n/2}\frac{V_n}{n^2}(1-\mathrm{e}^{-n^2\tau}). \tag{20}$$

3.1.1. *Asymptotic short- and long-time solutions*

An asymptotic solution for the mean velocity and displacement can now be derived from the exact expressions of the previous section.

For short times it can easily be shown that

$$\langle \dot{x}(t)\rangle = V_0 + \sum_{n=1}^{\infty} V_n \cos(\gamma_n y_0) = V(y_0),$$

namely, the velocity of a particle initially positioned at y_0 coincides with the external velocity before lateral diffusion takes place. Correspondingly the displacement is

$$\langle x(t)\rangle = tV(y_0).$$

For long times, however, the mean displacement of a small blob of particles positioned initially at $y_0 = \frac{1}{2}Y$ is

$$\begin{aligned}\langle x(t)\rangle_{y_0=\frac{1}{2}Y} &= \bar{V}t+\frac{Y^2}{\pi^2 D}\sum_{n=2,4}^{\infty}\frac{(-1)^{n/2}V_n}{n^2}\\ &= \bar{V}t\left[1+\frac{1}{\tau}\sum_{n=2,4}^{\infty}\frac{(-1)^{n/2}}{n^2}\left(\frac{V_n}{\bar{V}}\right)\right],\end{aligned} \tag{21}$$

which deviates from the simple $\bar{V}t$ expression by a term decreasing linearly with time. Hence introducing a small centrally located Brownian blob and measuring the total displacement of the particles introduces a time-dependent error that can be estimated from (21).

To elucidate this result we consider three simple examples: (i) Couette flow, (ii) plane Poiseuille flow, (iii) open-channel flow over an inclined flat plate.

(i) Couette flow

$$\left.\begin{aligned} V(y) &= 2\bar{V}y/Y,\\ V_n &= \begin{cases} -\dfrac{8\bar{V}}{\pi^2 n^2} & n\text{-odd},\\ 0 & n\text{-even}.\end{cases}\end{aligned}\right\} \tag{22}$$

(ii) Plane Poiseuille flow

$$\left.\begin{aligned} V(y) &= 6\bar{V}\frac{y}{Y}\left(1-\frac{y}{Y}\right),\\ V_n &= \begin{cases} -\dfrac{24\bar{V}}{\pi^2 n^2} & n\text{-even},\\ 0 & n\text{-odd}.\end{cases}\end{aligned}\right\} \tag{23}$$

(iii) Open-channel flow over an inclined plate

$$\left.\begin{aligned} V(y) &= 3\bar{V}\frac{y}{Y}\left(1-\frac{y}{2Y}\right),\\ V_n &= -\frac{6\bar{V}}{n^2\pi^2}.\end{aligned}\right\} \tag{24}$$

For Couette flow, no deviation from the mean value $\bar{V}t$ occurs given that the initial location of the blob coincides with the location of the mean external velocity. However, a small deviation from the mean value $\bar{V}t$ exists for the second and the third cases, where the last is four times smaller. In figure 2(*a–c*), equation (18) is evaluated for various y_0 locations and velocity profiles. Generally, the flatter the velocity profile the smaller the deviation from $\bar{V}t$ that is expected (as can be verified from figure 2*a*). Figure 2(*a*) depicts the displacement for long times. Figure 2(*b*) describes explicitly the time dependence of $\langle x(t)\rangle$ for Poiseuille flow. Figure 2(*c*) indicates that the temporal variation of $\langle x(t)\rangle/\bar{V}t-1$ is proportional to $(1-e^{-4\tau})/\tau$ for $0<\tau<\infty$ with only a small error introduced at the walls and the midsection of the flow. Generally, for not too badly shaped velocity profiles ($V_n = O(n^{-q})$ $q \geqslant 2$), one can show from (18) that the temporal variation of the displacement can be represented by the expression

$$\frac{\langle x(t)\rangle}{\bar{V}t}-1 \sim \frac{1-e^{-a\tau}}{\tau}, \tag{25}$$

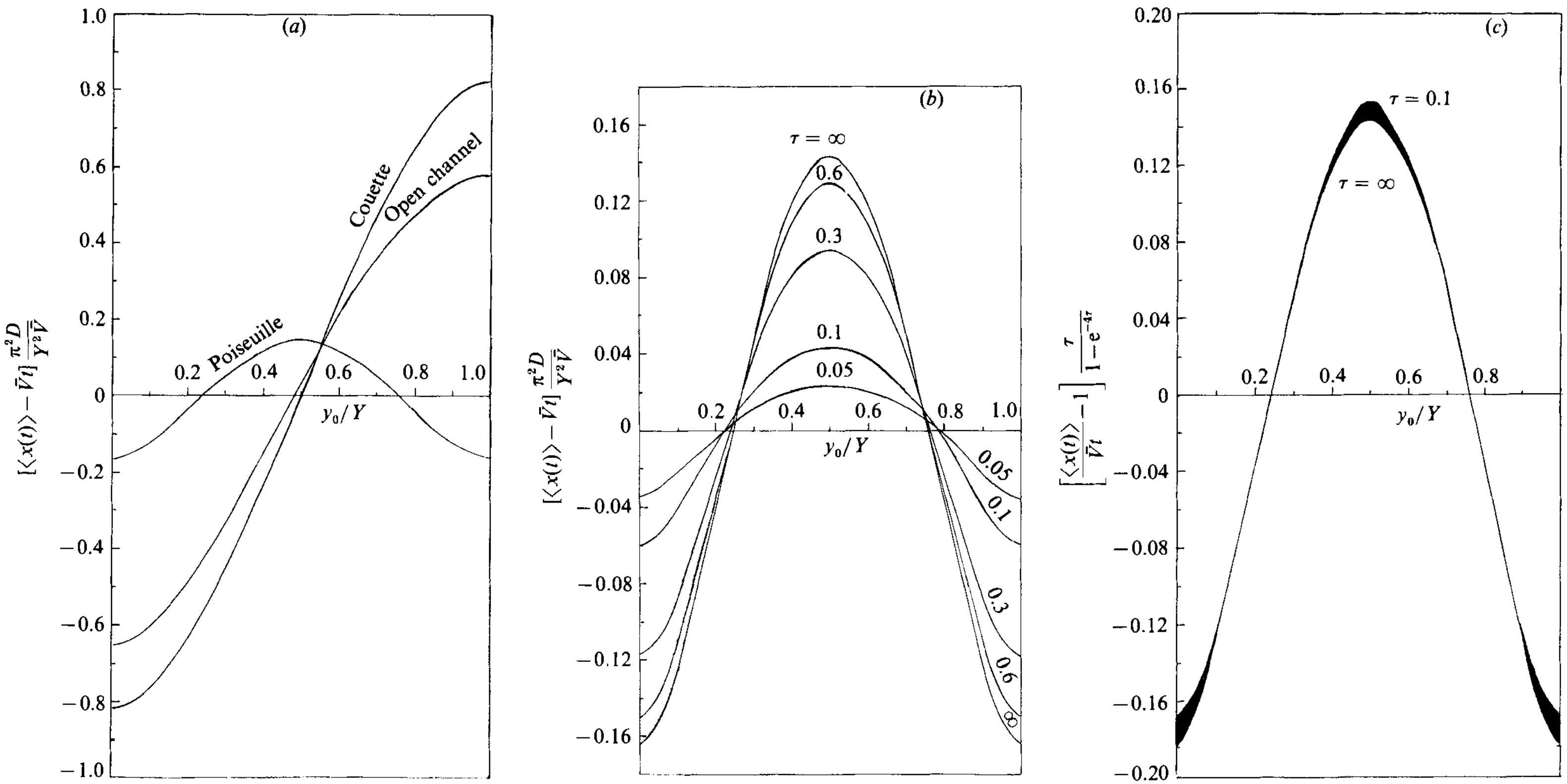

FIGURE 2. (*a*) The effect of the initial location of a particle for medium and long time periods. (*b*) The effect of the initial location on the mean displacement of a particle moving in Poiseuille flow for various times. (*c*) The generalized dependence of the mean displacement on the initial position of a particle moving in Poiseuille flow.

which is in good agreement with the exact solution. The factor a is discussed in a broader sense in §3.4 where it is used to describe the temporal variation of the dispersion coefficient. Note that the expression (25) does not depend on the initial position y_0 of the particle.

3.2. *The velocity autocorrelation*

Before calculating the dispersion coefficient, the integral in (7) must be evaluated. The arguments we used in the previous section are still valid and the integral

$$\int_{-y_0-\Delta y_1}^{Y-y_0\Delta y_1} V(y_1+\Delta y_2)\,P(y_1+\Delta y_2\,;\,\Delta t_2\,|\,y_1)\,\mathrm{d}(\Delta y_2)$$

can be replaced by an unbounded integral with a cosine Fourier expansion of the velocity field and a Gaussian probability distribution to yield

$$V_0+\sum_{n=1}^{\infty} V_n\,\mathrm{e}^{-n^2(\tau_2-\tau_1)}\cos(\gamma_n y_1).$$

Hence

$$\begin{aligned}\langle V(y_1)\,V(y_2)\rangle = {} & V_0\int_{-y_0}^{Y-y_0} V(y_0+\Delta y_1)\,P(y_0+\Delta y_1\,;\,\Delta t_1\,|\,y_0)\,\mathrm{d}(\Delta y_1)\\ & +\sum_{n=1}^{\infty} V_n\,\mathrm{e}^{-n^2(\tau_2-\tau_1)}\int_{-y_0}^{Y-y_0}\cos\gamma_n(y_0+\Delta y_1)\,V(y_0+\Delta y_1)\\ & \times P(y_0+\Delta y_1\,;\,\Delta t_1\,|\,y_0)\,\mathrm{d}(\Delta y_1)\end{aligned}\tag{26}$$

for $t_2 > t_1$. The first integral is similar to (4); thus

$$V_0\int_{-y_0}^{Y-y_0} V(y_0+\Delta y_1)\,P(y_0+\Delta y_1\,;\,\Delta t_1\,|\,y_0)\,\mathrm{d}(\Delta y_1) = V_0^2+V_0\sum_{n=1}^{\infty} V_n\,\mathrm{e}^{-n^2\tau_1}\cos(\gamma_n y_0).$$

The second integral can be evaluated as before, only this time $\cos(\gamma_n y)\,V(y)$ must periodically and symmetrically be extended to infinity, with the same period Y and symmetry with respect to $y = 0$ or $y = Y$. It can easily be shown that a product of any two periodic functions satisfying the foregoing conditions results in a periodic function of the same family. Since

$$\cos(\gamma_n y)\quad\text{and}\quad\sum_{m=0}^{\infty} V_m\cos(\gamma_m y)$$

possess these properties the second integral yields

$$\begin{aligned}&(4\pi D\,\Delta t_1)^{-\frac{1}{2}}\sum_{n=1}^{\infty}\sum_{m=1}^{\infty} V_n V_m\,\mathrm{e}^{-n^2(\tau_2-\tau_1)}\int_{-\infty}^{\infty}\cos\gamma_n(y_0+\Delta y_1)\cos\gamma_m(y_0+\Delta y_1)\\ &\quad\times\exp\left[-\frac{\Delta y_1^2}{D\Delta t_1}\right]\mathrm{d}(\Delta y_1)+V_0\sum_{n=1}^{\infty} V_n\,\mathrm{e}^{-n^2\tau_2}\cos(\gamma_n y_0)\\ &\qquad = V_0\sum_{n=1}^{\infty} V_n\,\mathrm{e}^{-n^2\tau_2}\cos(\gamma_n y_0)+\frac{1}{2}\sum_{m=1}^{\infty}\sum_{n=1}^{\infty} V_n V_m\,\mathrm{e}^{-n^2(\tau_2-\tau_1)}\\ &\qquad\qquad\times\{\mathrm{e}^{-(n+m)^2\tau_1}\cos(\gamma_{n+m} y_0)+\mathrm{e}^{-(n-m)^2\tau_1}\cos(\gamma_{n-m} y_0)\}\end{aligned}$$

(see Gradshteyn & Ryzhik 1965, p. 480.) Hence for $\tau_2 > \tau_1 > 0$ we have

$$\langle \dot{x}(\tau_2)\,\dot{x}(\tau_1)\rangle = 2D\delta(\tau_2-\tau_1)+V_0^2+V_0\sum_{n=1}^{\infty} V_n \cos(\gamma_n y_0)\,(\mathrm{e}^{-n^2\tau_1}+\mathrm{e}^{-n^2\tau_2})$$
$$+\frac{1}{2}\sum_{n=1}^{\infty}\sum_{m=1}^{\infty} V_n V_m\, \mathrm{e}^{-n^2(\tau_2-\tau_1)}\,[\mathrm{e}^{-(n-m)^2\tau_1}\cos(\gamma_{n-m}y_0)+\mathrm{e}^{-(n+m)^2\tau_1}\cos(\gamma_{n+m}y_0)], \quad (27)$$

and a similar expression for $\tau_1 > \tau_2 > 0$ (indices 1 and 2 should be interchanged). This result is exact and determines the time-dependent autocorrelation for the velocity of a Brownian particle initially located at y_0. The ensemble average for an initially *uniform* distribution is a drastic simplification of (27), namely

$$\langle (\dot{x}(\tau_2)\,\dot{x}(\tau_1)\rangle = 2D\delta(\tau_2-\tau_1)+V_0^2+\frac{1}{2}\sum_{n=1}^{\infty} V_n^2\, \mathrm{e}^{-n^2|\tau_2-\tau_1|} \quad (28)$$

for all $\tau_2, \tau_1 > 0$.

This important result is recovered from (27) for *any* initial probability distribution if $\tau_1 \to \infty$ and $\tau_2-\tau_1$ is finite. It could also be recovered if to start with we had assigned for the probability distribution $P(y_0+\Delta y_1;\,\Delta t_1\,|\,y_0)$ the value $1/Y$ in (9). Physically it means that after a long time t, the particle samples evenly all locations and its initial position y_0 is of no consequence.

3.3. *The dispersion coefficient*

The dispersion coefficient can be obtained directly from the velocity autocorrelation by utilizing the following time integration:

$$\langle x^2(t)\rangle = \left\langle \int_0^t \dot{x}(t_1)\,\mathrm{d}t \int_0^t \dot{x}(t_2)\,\mathrm{d}t_2 \right\rangle = \int_0^t\int_0^t \langle \dot{x}(t_1)\,\dot{x}(t_2)\rangle\,\mathrm{d}t_1\,\mathrm{d}t_2. \quad (29)$$

If we define a time-dependent dispersion coefficient as follows:

$$D^{\mathrm{s}}(t) = \frac{1}{2t}\,[\langle x^2(t)\rangle - \langle x(t)\rangle^2], \quad (30)$$

only (29) needs to be evaluated. (We preferred this form since it is easier to compare with experiment.) By the same token an ensemble dispersion coefficient can be defined as

$$\tilde{D}(t) = \frac{1}{2t}\,[\langle\!\langle x^2(t)\rangle\!\rangle - \langle\!\langle x(t)\rangle\!\rangle^2]. \quad (31)$$

The definition (30) is useful if a small blob is positioned initially at a certain location y_0 and sampled after a relatively short time period. However, both $D^{\mathrm{s}}(t)$ and $\tilde{D}(t)$ are expected to possess an identical form for long time periods.

Introducing (27) into (29) we obtain

$$\langle x^2(t)\rangle = 2Dt+\bar{V}^2t^2+\frac{2\bar{V}^2Y^4}{\pi^4D^2}\left\{\sum_{n=1}^{\infty}\frac{V_n}{\bar{V}}\cos(\gamma_n y_0)\,\frac{\tau}{n^2}\,(1-\mathrm{e}^{-n^2\tau})\right.$$
$$+\frac{1}{2}\sum_{\substack{n=1\\ m\neq n}}^{\infty}\sum_{\substack{m=1\\ m\neq 2n}}^{\infty}\frac{V_n}{\bar{V}}\frac{V_m}{\bar{V}}\cos(\gamma_{n-m}y_0)\,\frac{1}{m(2n-m)}\left[\frac{1-\mathrm{e}^{-(n-m)^2\tau}}{(m-n)^2}-\frac{1-\mathrm{e}^{-n^2\tau}}{n^2}\right]$$
$$+\frac{1}{2}\sum_{n=1}^{\infty}\frac{V_nV_{2n}}{\bar{V}^2}\cos(\gamma_n y_0)\left[\frac{1}{n^4}\,(1-\mathrm{e}^{-n^2\tau})-\frac{\tau\mathrm{e}^{-n^2\tau}}{n^2}\right]$$

$$-\frac{1}{2}\sum_{n=1}^{\infty}\sum_{n=1}^{\infty}\frac{V_n V_m}{\bar{V}^2}\cos(\gamma_{n+m}\,y_0)\frac{1}{m(2n+m)}\left[\frac{1-e^{-(n+m)^2\tau}}{(m+n)^2}-\frac{1-e^{-n^2\tau}}{n^2}\right]$$

$$+\frac{1}{2}\sum_{n=1}^{\infty}\left(\frac{V_n}{\bar{V}}\right)^2\left[\frac{\tau}{n^2}-\frac{1}{n^4}(1-e^{-n^2\tau})\right]\Bigg\}. \tag{32}$$

Hence

$$D^{s}=\frac{\langle x^2(t)\rangle-\langle x(t)\rangle^2}{2t}=D+\frac{\bar{V}^2Y^2}{\pi^2 D}$$

$$\times\left(\frac{1}{2}\sum_{n=1}^{\infty}\left(\frac{V_n}{\bar{V}}\right)^2\left[\frac{1}{n^2}-\frac{1}{n^4\tau}(1-e^{-n^2\tau})\right]-\frac{1}{2}\sum_{n=1}^{\infty}\frac{V_n V_{2n}}{\bar{V}^2}\frac{e^{-n^2\tau}}{n^2}\cos(\gamma_n\,y_0)\right.$$

$$+\frac{1}{2\tau}\sum_{\substack{n=1\\ m\neq n}}^{\infty}\sum_{\substack{m=1\\ m\neq 2n}}^{\infty}\left(\frac{V_n V_m}{\bar{V}^2}\right)\cos(\gamma_{n-m}\,y_0)\frac{1}{m(2n-m)}\left[\frac{1-e^{-(m-n)^2\tau}}{(m-n)^2}-\frac{1-e^{-n^2\tau}}{n^2}\right]$$

$$+\sum_{n=1}^{\infty}\frac{V_n V_{2n}}{\bar{V}^2}\cos(\gamma_n\,y_0)\left[\frac{1-e^{-n^2\tau}}{n^4}\right]$$

$$-\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}\frac{V_n V_m}{\bar{V}^2}\cos(\gamma_{n+m}\,y_0)\frac{1}{m(2n+m)}\left[\frac{1-e^{(n+m)^2\tau}}{(m+n)^2}-\frac{1-e^{-n^2\tau}}{n^2}\right]$$

$$\left.-\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}\frac{V_n V_m}{\bar{V}^2}\frac{1}{n^2m^2}(1-e^{-n^2\tau})(1-e^{-m^2\tau})\cos(\gamma_n\,y_0)\cos(\gamma_m\,y_0)\right). \tag{33}$$

The ensemble dispersion coefficient for an initially uniform probability distribution is a much simpler expression since all terms including $\cos(\gamma_n\,y_0)$ vanish:

$$\tilde{D}(\tau)=D+\frac{\bar{V}^2Y^2}{2\pi^2D}\sum_{n=1}^{\infty}\left(\frac{V_n}{\bar{V}}\right)^2\frac{1}{n^2}\left[1-\frac{1}{n^2\tau}(1-e^{-n^2\tau})\right]. \tag{34}$$

3.4. *Asymptotic short- and long-time dispersion coefficients and the approximate analytic expression*

The analytical expressions for D^{s} and $\tilde{D}$ can now be examined so that simplified asymptotic expressions can be derived for short and long time periods. If we retain terms inversely proportional to τ and neglect all other exponentially small terms, the medium-time dispersion coefficient is

$$D^{s}=D+\frac{\bar{V}^2Y^2}{2\pi^2D}\left\{\sum_{n=1}^{\infty}\left(\frac{V_n}{\bar{V}}\right)^2\frac{1}{n^2}-\frac{1}{\tau}\left[\sum_{n=1}^{\infty}\left(\frac{V_n}{\bar{V}}\right)^2\frac{1}{n^4}\right.\right.$$

$$\left.\left.-\sum_{n=1}^{\infty}\sum_{\substack{m=1\\ m\neq n}}^{\infty}\frac{V_n V_m}{\bar{V}^2}\frac{\cos(\gamma_{n-m}\,y_0)}{mn(m-n)^2}+\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}\frac{V_n V_m}{\bar{V}^2}\frac{\cos(\gamma_{n+m}\,y_0)}{mn(m+n)^2}\right]\right\}$$

$$+(\text{exponentially small terms in }\tau), \tag{35}$$

and

$$\tilde{D}=D-\frac{\bar{V}^2Y^2}{2\pi^2D}\sum_{n=1}^{\infty}\left(\frac{V_n}{\bar{V}}\right)^2\frac{1}{n^2}\left[1-\frac{1}{\tau n^2}\right]+(\text{exponentially small terms in }\tau). \tag{36}$$

For very large τ, Taylor's solution is recovered as a particular case:

$$\tilde{D}=D^{s}\overset{\text{def}}{=}\tilde{D}+D^{\mathrm{T}}=D+\frac{\bar{V}^2Y^2}{2\pi^2D}\sum_{n=1}^{\infty}\left(\frac{V_n}{\bar{V}}\right)^2\frac{1}{n^2} \tag{37}$$

and the initial position y_0 does not affect the results. However, for medium time periods y_0 enters the solution and its effect is depicted in figure 3(a, b) for Couette, plane Poiseuille, and open-channel flow over an inclined plate.

The Taylor dispersion coefficient for the foregoing cases is:

Couette flow
$$D^{\mathrm{T}} = \frac{\bar{V}^2 Y^2}{2\pi^2 D} \sum_{n=1,3}^{\infty} \frac{64}{\pi^4} \frac{1}{n^6} = \frac{1}{30} \frac{\bar{V}^2 Y^2}{D}; \tag{38a}$$

Poiseuille flow
$$D^{\mathrm{T}} = \frac{\bar{V}^2 Y^2}{2\pi^2 D} \sum_{n=2,4}^{\infty} \frac{576}{\pi^4} \frac{1}{n^6} = \frac{1}{210} \frac{\bar{V}^2 Y^2}{D}; \tag{38b}$$

open-channel flow
$$D^{\mathrm{T}} = \frac{\bar{V}^2 Y^2}{2\pi^2 D} \sum_{n=1,2}^{\infty} \frac{36}{\pi^4} \frac{1}{n^6} = \frac{2}{105} \frac{\bar{V}^2 Y^2}{D}. \tag{38c}$$

The first two results are identical to those obtained by Van den Broeck (1982). The Taylor dispersion coefficient for the last case is four times larger than the second case and the largest dispersion coefficient is associated with the Couette flow, which is the 'steepest' case.

It is interesting to note that the Taylor dispersion coefficient (37) can be obtained directly from the velocity distribution via the following simple formula:

$$D^{\mathrm{T}} = \frac{Y^2}{D} \int_0^Y v'^2(y)\, \mathrm{d}y, \tag{39a}$$

where
$$v'(y) = \frac{1}{Y} \int_0^y (V(y) - \bar{V})\, \mathrm{d}y. \tag{39b}$$

The proof is easy, using the cosine Fourier expansion for $V(y)$. This result substantiates the notion that a 'flat' velocity profile yields a small dispersion coefficient. However, not the fluctuations themselves but their *local* mean v' is of importance. The dispersion coefficient can be viewed as the product of the mean square of v' (used as a characteristic velocity) and the characteristic time Y^2/D it takes a particle to sample all lateral positions by molecular diffusion.

For very short times $\tau \ll 1$, (34) assumes the form

$$\begin{aligned} \tilde{D}(t) &= D + \tfrac{1}{2} t \sum_{n=1,2}^{\infty} V_n^2 + O(t^2) \\ &= D + t \left\{ \frac{1}{Y} \int_0^Y (V(y) - \bar{V})^2\, \mathrm{d}y \right\} + O(t^2), \end{aligned} \tag{40}$$

which indicates a linear growth with time due to convection only. The dispersion coefficient can be viewed as a product of the mean square of the velocity fluctuations by the elapsed time.

The following particular cases are addressed:

for Couette flow
$$\tilde{D} = D + t \frac{32}{\pi^4} \bar{V}^2 \sum_{n=1,3}^{\infty} \frac{1}{n^4} = D + \frac{t\bar{V}^2}{3}; \tag{41a}$$

for plane Poiseuille flow
$$\tilde{D} = D + t \frac{288}{\pi^4} \bar{V}^2 \sum_{n=2,4}^{\infty} \frac{1}{n^4} = D + \frac{t\bar{V}^2}{5}; \tag{41b}$$

for open-channel flow over an inclined plate
$$\tilde{D} = D + t \frac{18}{\pi^4} \bar{V}^2 \sum_{n=1,2}^{\infty} \frac{1}{n^4} = D + \frac{t\bar{V}^2}{5}. \tag{41c}$$

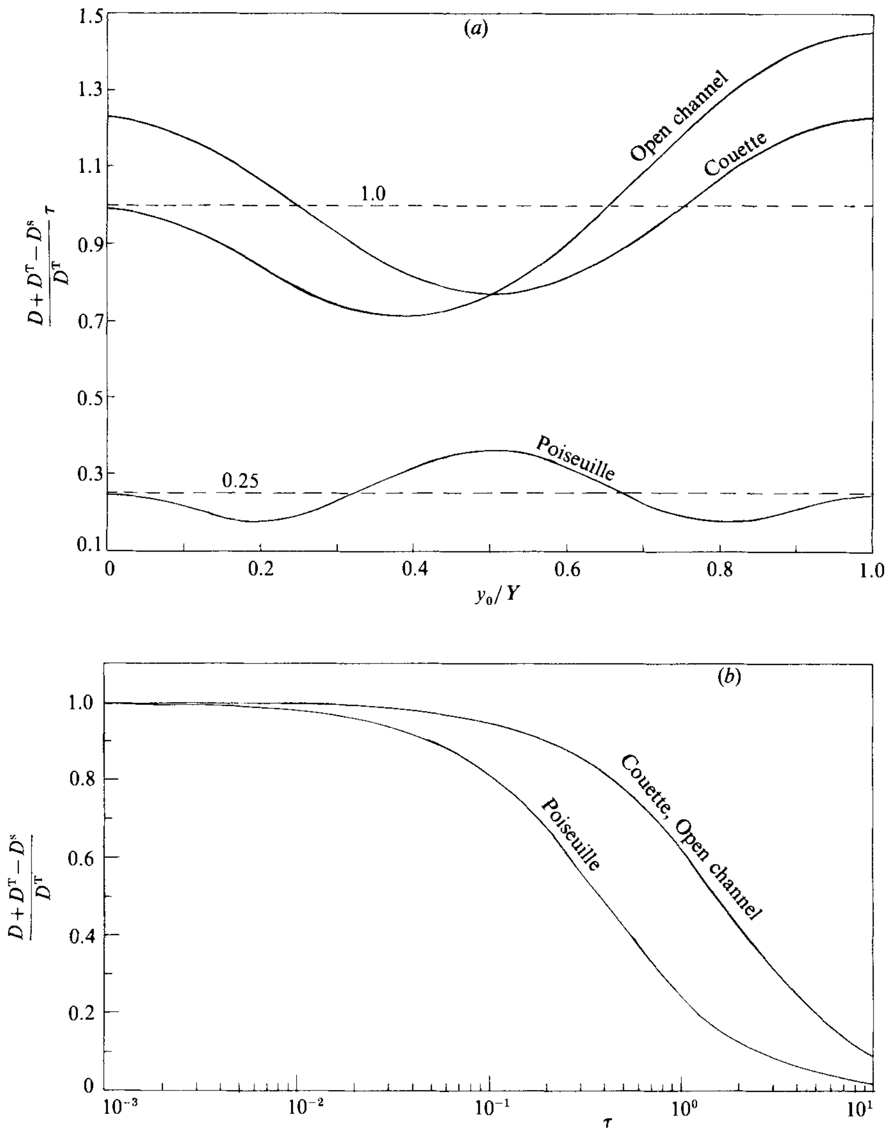

FIGURE 3. (*a*) The effect of the initial location of a particle on the dispersion coefficient for medium time periods. (*b*) The temporal variation of the dispersion coefficient for an initially uniform distribution of particles.

It can be shown from (34) that the time-dependent dispersion coefficient $\tilde{D}(\tau)$ assumes a very simple form according to the following single parameter formula:

$$\tilde{D}(\tau) = D + D^{\mathrm{T}}\left(1 - \frac{1-\mathrm{e}^{-a\tau}}{a\tau}\right), \tag{42}$$

where

	Couette	Poiseuille	Open
a	1	4	1
error	0.2 %	1 %	0.4 %

with an error of less than one percent over the entire time domain ($0 < \tau < \infty$).

The factor a can be evaluated quite simply by the following observation for a not too irregular velocity profile. If the velocity profile has no planes of symmetry within $0 < y < Y$, then $a = 1$; if it has N planes of symmetry within $0 < y < Y$ (e.g. Poiseuille flow possesses one plane of symmetry) then $a = (N+1)^2$. In other words, if $V_1 \neq 0$ then $a = 1$. If $V_n = 0$ for all $n = 1, 2, \ldots, N$ and $V_{N+1} \neq 0$ then $a = (N+1)^2$.

4. Conclusions

The Lagrangian approach, by which a single particle is traced, proved to be a very useful tool to obtain velocity correlations of any order. First- and second-order moments were analysed in detail and a solution is provided for a general two-dimensional, bounded velocity profile. Extending the velocity profile periodically made is possible to use the simple Gaussian, two-point, probability distribution. It also made it possible to decouple the kinematic problem from the stochastic one. Consequently, all that was needed was to expand the velocity profile by a cosine Fourier series. The fundamental solution for the mean velocity and dispersion coefficient irrespective of the initial distribution of particles was obtained. The introduction of the initial distribution amounts to a simple integration over all possible initial locations weighted by an *a priori* given initial probability distribution.

Taylor's dispersion coefficient is obtained as a particular case for very long times. It can be derived utilizing a very simple formula (39) which elucidates the physical meaning of the dispersion coefficient (the product of the mean square of the local mean of velocity fluctuations by the time it takes a particle to sample all transverse locations by molecular diffusion). For medium time periods, however, the dispersion coefficient is no longer a constant and an additional term is introduced which decays linearly with time. For short times, the molecular diffusion is insignificant and the results can be explained assuming convection only. Generally, the dispersion coefficient increases monotonically from its value for short times to its value according to Taylor's formula. For small and medium time periods, the initial location of the particle is important and slightly different results will be obtained for the mean velocity and dispersion coefficient. These differences will die out for long time periods and Taylor's result will be recovered.

A detailed analysis for three velocity profiles is carried out. It indicates that a 'flatter' profile results in a smaller dispersion coefficient. Particular attention is given to two initial cases: the uniform initial distribution of particles *vis-à-vis* the local Dirac-delta-function distribution. For the first case, most results simplify substantially: the dispersion coefficient assumes a very simple generalized analytic form (42) which deviates from the exact solution (34) by less than one percent for all times. The second case makes it possible to calculate ensemble mean velocities and diffusion coefficients for various initial probability distributions by means of a simple integration.

The parameter a defined in (42) can be used to predict the temporal variations of both the displacement and the dispersion coefficient over the entire time domain with an excellent accuracy. The derivation of a is purely kinematical and depends only on the number of symmetry planes that the flow field possesses. For N symmetry planes, $a = (N+1)^2$ for not too irregular velocity profiles (namely, $V_n = O(n^{-q})$ $q \geqslant 2$).

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