Motion of a sphere parallel to plane walls in a Poiseuille flow. Application to field-flow fractionation and hydrodynamic chromatography

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The motion of a solid spherical particle entrained in a Poiseuille flow between parallel plane walls has various applications to separation methods, like field-flow fractionation and hydrodynamic chromatography. Various handy formulae are presented here to describe the particle motion, with these applications in mind. Based on the assumption of a low Reynolds number, the multipole expansion method coupled to a Cartesian representation is applied to provide accurate results for various friction factors in the motion of a solid spherical particle embedded in a viscous fluid between parallel planes. Accurate results for the velocity of a freely moving solid spherical particle are then obtained. These data are fitted so as to obtain handy formulae, providing e.g. the velocity of the freely moving sphere with a 1% error. For cases where the interaction with a single wall is sufficient, simpler fitting formulae are proposed, based on earlier results using the bispherical coordinates method. It appears that the formulae considering only the interaction with a nearest wall are applicable for a surprisingly wide range of particle positions and channel widths. As an example of application, it is shown how in hydrodynamic chromatography earlier models ignoring the particle-wall hydrodynamic interactions fail to predict the proper choice of channel width for a selective separation. The presented formulae may also be used for modeling the transport of macromolecular or colloidal objects in microfluidic systems.

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1. Introduction

Various processes involve the motion of suspended particles in a Poiseuille flow of a viscous fluid. This paper is concerned in particular with the field-flow fractionation (FFF) and the capillary hydrodynamic chromatography (CHDC) methods of separation (Giddings, 1978; Noel et al., 1978). In FFF, rigid particles of various sizes are entrained in a Poiseuille flow in a channel between two parallel walls. In the direction perpendicular to the walls, particles are submitted to a force field (e.g. gravity) and, depending on their size, either to Brownian forces or to lift forces due to the flow field, or to both. The particles move across the streamlines, until these various effects balance (or nearly balance), and the particles reach an equilibrium position across the channel. This equilibrium position (or mean position in the presence of Brownian motion) depends on the particle size and mass (or other field-sensitive property), and the time particles spend in the channel is determined by the local flow axial velocity. Thus a selection by the residence time results in separation of different particle species. Particles of the order of 0.05–20 μm in a channel of width 20–500 μm are standard.

The focus of the present paper is on the modeling of the retardation due to the hydrodynamic interactions with walls. Particles being small, the flow field Reynolds number is small enough for Stokes equations to apply around a particle. Moreover, the particles modeled here are assumed to be solid and spherical. The volume fraction of the suspension is assumed to be low enough compared with unity for hydrodynamic interactions between particles to be negligible. Thus, we will consider a single particle. The particle is assumed to be freely rotating (the external torque is zero). We are interested in the motion of the particle having reached a lateral equilibrium position between the parallel walls. Then the total force on the particle is zero and the fluid mechanics problem is that of a freely moving (and freely rotating) sphere in a Poiseuille flow between parallel walls.

In CHDC (Giddings, 1978; Noel et al., 1978), particles entrained in a Poiseuille flow in a cylindrical capillary tube are submitted to a strong Brownian motion and are uniformly distributed in the
tube cross-section. Here by extension, we consider CHDC in a Poiseuille flow between parallel walls. Particles are uniformly distributed across the gap between the walls. The only limitation is that they cannot penetrate the walls. Considering the minimum distance between a particle center and the wall, larger particles are, on average, centered closer to the channel mid plane where the fluid velocity is larger. They are thus moving faster and separation is achieved in this way.

In FFF, due to an applied transverse field, the particles are usually closer to an accumulation wall. In their study on particle migration in FFF channels, Williams et al. (1992) and Williams (1994) employed simple formulae for the translating velocity of a freely moving sphere by fitting (for small and large gaps between particle and wall) and interpolating by cubic splines (for intermediate distances) theoretical results of Goldman et al. (1967a,b) obtained for a linear shear flow. The two-wall effect was simplified by using a wall-superposition approximation in Williams (1994). The articles (Williams et al., 1992; Williams, 1994) therefore neglected the curvature of the flow velocity profile as well as the actual simultaneous effects of both walls, so that their expressions are valid only provided the ratio of the particle radius to the distance between walls is small.

There have been various theoretical works on the motion of a freely moving spherical particle in Poiseuille flow between parallel walls, on the basis of Stokes equations. Among them, we may quote Ho and Leal (1974) who used the method of reflexions, Staben et al. (2003) who used the boundary-integral technique, Jones (2004) who used an expansion in multipoles and Bhattacharya et al. (2005a,b, 2006) who applied a Cartesian representation method, based on multipole expansions.

The large body of theoretical works provides an extensive set of results with various levels of mathematical complexity depending on the provided accuracy, results being often given as tables or graphs. Now, for practical applications, it might be sufficient to have handy formulae with an accuracy that is appropriate for standard experimental practice. This is the purpose of the present paper. Based on earlier theoretical results detailed below, it will provide handy formulae for the translating velocity of a freely moving sphere in a Poiseuille flow between two parallel walls. From these formulae, it will then deduce the consequences of hydrodynamic interactions with walls for the separation techniques mentioned above.

The problem and notation will be presented first in Section 2. Formulae for a sphere interacting with one wall will be obtained from earlier results taking into account the linear shear flow (Chaoui and Feuillebois, 2003) and the quadratic contribution of the flow field (Pasol et al., 2006) calculated in bipolar coordinates. On this basis, fitting formulae will be derived, Section 3, for a sphere interacting with the nearest wall and using the wall-superposition approximation, that is superimposing contributions from both walls to the particle velocity in the sense of the first term of the method of reflexions (Ho and Leal, 1974).

Formulae taking into account the simultaneous contribution of two walls will be derived, Section 4, from precise results provided by the multipole expansion (Ekiel-Jeżyewska and Wajnryb, 2006) coupled to the Cartesian representation (Bhattacharya et al., 2005a,b). In the presence of planar walls, this method relies on expanding the flow in a wall-bounded system using two basis sets of Stokes flows. The spherical basis set of multipolar flows is used to describe the interaction of the fluid with the particles, and the Cartesian basis set is used to account for the presence of the walls. The key point in this procedure is the transformation relations between spherical and Cartesian basis sets of solutions of Stokes equations. The transformation formulae enable construction of Stokes-flow fields that satisfy appropriate boundary conditions both on the planar walls and on the spherical particle surfaces. Our results will be compared with Staben et al. (2003). Here, results for the friction coefficients will be fitted using these precise numerical data and accurate results for lubrication, that is for small gap between the sphere and wall(s). The particle velocity will then be obtained in terms of these fitted formulae for the friction coefficients. Simpler fitting formulae based on results for a single wall may be sufficient for some range of parameters. The validity of the one-wall and wall-superposition approximations will also be discussed by comparison with the more general formula for the particle interacting simultaneously with both walls.

On the basis of the derived formulae, we will then calculate the retention ratio R, a parameter for the characterization of retention in the FFF and CHDC methods of separation, Section 5. The general case of an inhomogeneous dilute suspension of spherical particles in Poiseuille flow will be presented as well as the particular case of a homogeneous suspension involved in CHDC.

2. A freely moving sphere in two-dimensional Poiseuille flow. Problem and notation

Consider (Fig. 1) a channel of width w between parallel walls. Let x and y be coordinates along the wall, and z be the coordinate across the channel (walls are at z=0 and w). A spherical particle with radius a is centered at a distance Z from the nearest wall (say that at z=0 without loss of generality).

The unperturbed fluid flow in the channel is a Poiseuille flow along x with parabolic velocity:

$$u = 6 \langle u \rangle \frac{Z}{w} \left(1 - \frac{Z}{w}\right),$$

where $\langle u \rangle$ denotes the average fluid velocity across the channel width, viz:

$$\langle u \rangle = \frac{1}{w} \int_{0}^{w} u \, dz.$$

The unperturbed flow (1) may be regarded as the sum of a linear shear flow and a quadratic flow:

$$u = k_3 z + k_q z^2,$$

with rate of shear

$$k_3 = 6 \frac{\langle u \rangle}{w},$$

and curvature of the velocity profile

$$k_q = -6 \frac{\langle u \rangle}{w^2}.$$

The spherical particle is freely moving in this Poiseuille flow. The velocity of its center, say U, is lower than that of this ambient flow because of the flow profile curvature (this is the Faxen term, as detailed below) and of hydrodynamic interactions with walls. Our
goal is to derive handy expressions for force and torque, respectively. The second subscript denotes the translation \((t)\), rotation \((r)\), linear shear flow \((s)\) and quadratic shear flow \((m)\). Here, the forces \(F\) and \(c\) are always positive. For a sphere far from any wall the limits \(c_1=U_0\) and \(c_2=0\) are for a sphere rotating with velocity \(\Omega_0\) along \(y\) in a fluid at rest; \(U_0=U_0(y)\) is the particle velocity normalizing by the channel width \(w\), in particular:

\[
\alpha = \frac{a}{w}, \quad s = \frac{Z}{w},
\]

with ranges of variations \(0 \leq \alpha \leq 0.5\) and \(0 \leq s \leq 1-\alpha\). Alternative useful variables are the dimensionless gaps \(1-\alpha\) between the particle surface and the nearest wall and \(\alpha\) between the particle surface and the other wall:

\[
\alpha_1 = \frac{Z_0 - \alpha \frac{s}{a}}{a}, \quad \alpha_2 = \frac{w - Z_0 - \alpha}{a} = \frac{1-s}{a} - 1.
\]

We define the dimensionless velocities:

\[
\hat{U}^t = \frac{U^t}{k_0 a}, \quad \hat{U}^q = \frac{U^q}{k_0 a^2}.
\]

The expressions for these dimensionless velocities were obtained in Goldman et al. (1967b), Chaoui and Feuillebois (2003) and Pasol et al. (2006), respectively, in term of dimensionless friction factors:

\[
\hat{U}^t = \frac{(S)_{yx} c_{xy} f_{xy} + (S)_{xy} f_{xy}}{c_{yx} f_{xy} + f_{xy}}, \quad \hat{U}^q = \frac{(S)_{yx} c_{xy} f_{xy} + (S)_{xy} f_{xy}}{c_{yx} f_{xy} - f_{xy}}.
\]

Here, the \(S\)'s and the \(c\)'s denote dimensionless friction factors for the forces \(F\) and torques \(C\), respectively. The superscripts denote translation \((t)\), rotation \((\tau)\), linear shear flow \((s)\) and quadratic shear flow \((q)\). The first subscript in \(f\) and \(c\) denotes the direction of the force and torque, respectively. The second subscript denotes the direction of the sphere motion or applied flow. Specifically:

- \(f_{xy}^t = -(f_{xy}^t/S \mu a \mu u_x)\), \(c_{xy}^t = (C_{xy}/8 \pi a^3 \mu u_x)\)
- for a sphere translating with a velocity \(U_x\) along \(x\) in a fluid at rest;
- \(f_{xy}^q = (f_{xy}^q/S \mu a^2 \mu \Omega_0)\), \(c_{xy}^q = -(C_{xy}/8 \pi a^3 \mu \Omega_0)\)
- for a sphere rotating with velocity \(\Omega_0\) along \(y\) in a fluid at rest;
- \(f_{xy}^s = (f_{xy}^s/S \mu a k_z \Omega_0)\), \(c_{xy}^s = (C_{xy}/4 \pi a^3 k_z \Omega_0)\)
- for a sphere held fixed in a linear shear flow \(k_z\) along \(x\); and
- \(f_{xy}^m = (f_{xy}^m/S \mu a k_z Z_0)\), \(c_{xy}^m = (C_{xy}/8 \pi a^3 k_z Z_0)\)
- for a sphere held fixed in a quadratic shear flow \(k_z^2\) along \(x\).

Here, \(\mu\) denotes the fluid dynamic viscosity. By their definition, all above friction coefficients are positive when there is only one wall at \(z=0\). However for two walls, only the ones with subscripts \(xx\) and \(yy\) are always positive. For a sphere far from any wall the friction coefficients tend to unity, except \(c_{xy}^t\) and \(f_{xy}^q\) which vanish.

The articles (Goldman et al., 1967b; Chaoui and Feuillebois, 2003; Pasol et al., 2006) are concerned with a particle interacting with a single wall. It is important to note that the formulas (10) were solely derived on the basis of the definitions of the friction factors and of the equations of motion for a freely moving sphere. Therefore, they are naturally valid for a particle interacting with two walls. The friction factors then depend on \(s\) and \(x\), whereas when interacting with a single wall they only depend on the normalized distance to the wall, that is \(Z/a = s/\alpha\).

It may also be remarked that from the Lorentz reciprocity theorem (see e.g. Happel and Brenner, 1973), the force friction factor for rotation is related to the torque friction factor for translation:

\[
f_{xy}^t = \frac{4}{3} c_{xy}^t.
\]

An alternative useful normalization for the velocities \(\hat{U}^t\) and \(\hat{U}^q\) is the velocity of a sphere embedded in unbounded flow, that is \(\lim_{x 
arrow \infty} \hat{U}^t \equiv \lim_{x 
arrow \infty} \hat{U}^q\), respectively. From Faxen formula (see e.g. Happel and Brenner, 1973), these limits are \(k_d Z\) for the linear shear flow and \([(1+(a/6)^2)U_0^2/k_d^2]z_c = k_d(Z^2+a^2/3)\) for the quadratic flow. Dimensionless velocities defined for each of these ambient flow fields by

\[
\hat{U}^t = \frac{U^t}{k_d Z} = \frac{Z}{s} \hat{U}^t, \quad \hat{U}^q = \frac{U^q}{k_d(Z^2+a^2/3)} = \frac{Z^2}{s^2+1/3} \hat{U}^q,
\]

then appear as "wall correction factors".

The decomposition (6) of the velocity of a freely moving sphere in the flow field (3) is rewritten with (4), (5), (12) as

\[
\frac{U}{b} c_{15} = s U^t - \left(1 + \frac{a^2}{3}ight) \hat{U}^q
\]

and with (10), (12) as

\[
U = c_{15} f_{xy}^t + f_{xy}^q c_{xy}^q.
\]

Here,

\[
f_{xy}^q = \frac{s^2}{3} - s^2 f_{xy}^q, \quad c_{xy}^q = \frac{2}{3} c_{xy}^q - 2 a c_{xy}^q
\]

denote, respectively, the friction factor for the force and torque on a fixed sphere in Poiseuille flow.

Our task is now to evaluate the various friction factors and then, from these results, to construct simple efficient formulae for these quantities and for the particle velocity. This will be done in the following sections.

3. The nearest wall and wall-superposition approximations

3.1. Velocity of a freely moving sphere interacting with the nearest wall

Let \(U^t\) be the sphere velocity assuming that it is only interacting with the wall at \(z=0\). The main results for a sphere in a parabolic shear flow will be recalled and a simple fitting formula for \(U^t\) will be derived therefrom.

Peculiar difficulties in the fitting arise when the dimensionless gap between the sphere and the nearest wall is small, \(e_1 \ll 1\), that is in the lubrication regime. Indeed, the friction coefficients for translation and rotation are singular in the limit \(e_1 \rightarrow 0\). The following limit expressions were derived in Goldman et al. (1967a) and O’Neill and Stewartson (1967):

\[
f_{xy}^t = -\frac{8}{15} \log e_1 + 0.9543 - \frac{64}{375} e_1 \log e_1 + O(e_1),
\]

\[
c_{xy}^t = -\frac{1}{10} \log e_1 - 0.1929 - \frac{43}{250} e_1 \log e_1 + O(e_1),
\]

\[
c_{xy}^q = -\frac{2}{5} \log e_1 + 0.3817 - \frac{66}{125} e_1 \log e_1 + O(e_1).
\]
The friction coefficients were calculated more recently in bispherical coordinates with a $10^{-16}$ accuracy for values of the gap between the sphere and the wall down to $2 \times 10^{-6}$ sphere radius: Chaoui and Feuillebois (2003) provided results for $f_{xy}^p, f_{zy}^p, f_{zx}^p/a$, and Pasol et al. (2006) for the $f_{xy}^p/a^2$ friction factors. From their results, Chaoui and Feuillebois (2003) also provided improved lubrication expressions like:

$$
c_{xy} = - \frac{1}{10} \log \epsilon_1 - 0.1929 - \frac{43}{250} \epsilon_1 \log \epsilon_1 + 0.1006 \epsilon_1 \\
-0.0369 \epsilon_1 \log \epsilon_1 + 0.09449 \epsilon_1^2.
$$

(19)

Accurate results for $U^c$ are obtained from these results for the friction coefficients, using (13) with (12), (10). For practical purposes, we now derive formulae fitting these accurate results in the whole range of distances. The idea is to base the fitting on lubrication expressions valid for small distances ($\epsilon_1 \ll 1$) and construct therefrom expressions that are valid for any distance. First, remark that for $\epsilon_1 \ll 1$ terms in $\log \epsilon_1$ appear in the numerators and denominators of Eqs. (10). Then $(U^c)^{-1} \sim \log \epsilon_1$. The difficulty is that terms in $\log \epsilon_1$ cannot be kept for the region $\epsilon_1 \gg 1$ since they become increasingly large. This increase is unphysical, unlike that for $\epsilon_1 \ll 1$. Then the idea is to use a function which behaves like $\log \epsilon_1$ for $\epsilon_1 \rightarrow 0$ and stays bounded for $\epsilon_1 \rightarrow \infty$. For that purpose, we replace in the lubrication expressions $\epsilon_1$ by the regularized variable:

$$
\epsilon_1 = \epsilon_1 + 1 + \frac{s/\bar{x} - 1}{s/\bar{x} - 1} = 1 - \frac{\bar{x}}{s}
$$

and use:

$$
\epsilon_1 = \log \left( 1 - \frac{\bar{x}}{s} \right).
$$

(20)

which vanishes for $\epsilon_1 \rightarrow \infty$. The first terms for the inverse of the velocity then are of the form $C \epsilon_1 + 1$, where $C$ is a constant. The remainder for non-small $\epsilon_1$ is simply fitted with a polynomial in $s/\bar{x}$ which vanishes at large distance from the wall, $s/\bar{x} \rightarrow \infty$. Results for the velocities in (12) are the following formulae which fit the results from the method of bispherical coordinates with a $10^{-3}$ precision (the largest error being for intermediate distances $\epsilon_1 \sim 0.1$):

$$
\tilde{U}^0 = \left[ -0.269 \epsilon_1 + 1 - 0.2710 \frac{\bar{x}}{s} - 0.1039 \left( \frac{s}{\bar{x}} \right)^2 \\
-0.08994 \left( \frac{s}{\bar{x}} \right)^3 + 0.08511 \left( \frac{s}{\bar{x}} \right)^4 - 0.1392 \left( \frac{s}{\bar{x}} \right)^5 \right]^{-1}
$$

(21a)

$$
\tilde{U}^{0'} = \left[ -0.292 \epsilon_1 + 1 - 0.2935 \frac{\bar{x}}{s} - 0.1269 \left( \frac{s}{\bar{x}} \right)^2 \\
+ 0.4860 \left( \frac{s}{\bar{x}} \right)^3 - 0.2763 \left( \frac{s}{\bar{x}} \right)^4 + 0.0025 \left( \frac{s}{\bar{x}} \right)^5 \right]^{-1}
$$

(21b)

The velocity of the freely moving particle is then obtained by (13) with (21). It is important to note for applications that the particle velocity is quite different from the local fluid velocity (3) calculated at the sphere center $z=0$. This may be emphasized by calculating their ratio, which we call the particle velocity correction factor. For instance in a linear shear flow, this ratio is precisely $\tilde{U}^{-1}$ from its definition (12a). Williams et al. (1992) gave approximations for this correction factor by fitting results from Goldman et al. (1967b). Far from the wall on the scale of the particle ($s/\bar{x} \gg 1$), they used the approximation from Goldman et al. (1967b):

$$
\tilde{U}^{-1} \simeq 1 - \frac{5}{16} \left( \frac{s}{\bar{x}} \right)^3.
$$

(22)

For the case when the gap between the particle and the wall is small ($\epsilon_1 = s/\bar{x} - 1 \ll 1$), that is in the lubrication regime, they corrected a formula of Goldman et al. (1967b) and proposed:

$$
\tilde{U}^{-1} \simeq \frac{1}{0.66 - 0.269 \log \left( \frac{s}{\bar{x}} \right)}.
$$

(23)

Our result for the particle velocity correction factor in a linear shear flow, Eq. (21a), is compared with the approximations (22) for large gap and (23) for small gap in Fig. 2. It is clear that, because of its construction from results in bispherical coordinates, the solid curve representing Eq. (21a) is practically the same as the cubic spline fit of results of Goldman et al. (1967a,b) by Williams (1994, Fig. 6).

The one-wall approximation will be used in the next subsection in the wall-supersposition approximation. The accuracy of the one-wall and wall-supersposition approximations will be discussed by comparison with the results for two walls in Section 4.4.

### 3.2. Ho & Leal’s supersposition of single walls at the level of particle velocity

Ho and Leal (1974) used the method of reflexions to calculate the translation velocity of a sphere that is small compared with the channel width, that is $s \ll 1$, and far from any wall, that is $s/\bar{x} \ll 1$ and $(1-s)/\bar{x} \ll 1$. They found that a good approximation is obtained by superimposing the influences of both walls, since the simultaneous influence of both walls produces smaller effects, of order $O(s^4)$. The sphere translational velocity $U$ is then written as

$$
U = U^0 + U^1 - U^0
$$

(24)

in which $U^0$ and $U^1$ are the velocities of the sphere when interacting only with the wall at $z=0$ and $w$, respectively ($U^0(a,w,Z)$ is given by (13) with (21) and $U^0(a,w,Z) = U(a,w,0-Z)$) and $U^0$ is the velocity of the particle in Poiseuille flow when ignoring any interaction with walls. This velocity is obtained by applying Faxen’s formula:

$$
U^0 = \left[ u + \frac{a^2}{6} \nabla^2 u \right]_{z=Z} = \left< u \right> [6s(1-s)-2x^2].
$$

(25)

This velocity is subtracted in Eq. (24) since the entrainment by the unperturbed flow is already included when calculating $U^0$ with the
wall in \( z=0 \). It should therefore not be counted twice when adding up the influence of the other wall in \( z=w \) with \( U'' \).

The range of validity of formula (24) will be found later in Section 4 which presents an expression for the sphere velocity taking both walls into account, thus valid for all \( z \)’s.

3.3. Superposition of single walls at the level of friction factors

In this approach, an approximation for each friction factor is obtained in the generic form:

\[
\bar{f} \approx f' + f'' - f^0
\]

(26)

by adding contributions from both walls (\( f' \) and \( f'' \)) and subtracting the common part, that is the friction factor in the absence of walls, \( f^0 \). Then these approximations are introduced in (10), (12), (13) to obtain an approximation for the particle velocity. This approach is classical (see Bhattacharya et al., 2005a and references therein). It was applied in Bhattacharya et al. (2005a) using the results for the friction factors from the multipole method. Independently, it was checked in two companion papers (Ekiel-Jezewska and Wajnryb, 2006; Pasol et al., 2006) that the results obtained for the friction factor from the multipole method (Ekiel-Jezewska and Wajnryb, 2006) agree with the results from the bispherical coordinates method (Pasol et al., 2006, see also Chaoui and Feuillebois, 2003) up to a high degree of accuracy. Thus, the wall-superposition approximation at the level of friction factors calculated from the bispherical coordinates method gives the same result as in Bhattacharya et al. (2005a).

A higher accuracy in the two-wall lubrication regime was obtained in Ekiel-Jezewska et al. (2008) by using the wall-superposition for friction factors and the results from multipoles. Their results as well as higher order terms in one-wall lubrication friction factors from the method of bispherical coordinates in Chaoui and Feuillebois (2003) will be used in Section 4.3 to construct fitting formulae for all \( s’ \)'s and \( s'' \)'s, based again on this idea of wall-superposition at the level of friction factors.

4. A freely moving sphere interacting with both walls in Poiseuille flow

When the particle size is of the same order as the channel width, the simultaneous importance of both walls should be taken into account. This is even more critical for applications where the particle size may happen to be nearly as large as the channel width (Moon and Giddings, 1996). The appropriate precise method used here to account for hydrodynamic interactions between a sphere and two walls is the multipole expansion method, corrected for lubrication. It is schematically presented in Section 4.1.

4.1. The multipole expansion method and the Cartesian representation

The dependence of the friction factors and the particle’s free-motion velocities on \( \epsilon_1 \) and \( \epsilon_2 \) has been determined numerically with the use of the multipole expansion (see Cichocki et al., 1994). In this method, the induced forces and the Green tensors are used to replace the Stokes problem (the equations and the boundary conditions at the particle surface) by boundary-integral equations for the unknown force density. These equations are then subsequently projected onto a complete set of multipole functions (Ekiel-Jezewska and Wajnryb, 2009). As the result, an infinite set of linear algebraic equations is obtained. To be solved for the force multipoles, the equations are truncated at a certain value of the truncation parameter \( L \).

In the theoretical algorithm, the lubrication correction is taken into account, which takes care of the hydrodynamic interactions between close solid surfaces in a relative motion. The point is to add the corresponding asymptotic lubrication expressions to the friction coefficients, and subtract their expansion, combining the coefficients with those resulting from the multipole expansion. These new coefficients, now free from the lubrication divergence, exhibit a fast convergence with the truncation parameter \( L \). As the result, the method is very precise for relatively low values of \( L \). Moreover, the accuracy is strictly controlled by analyzing the dependence of the results on a specific value of \( L \), including the extrapolation of the results to \( L \rightarrow \infty \).

In the present approach, the Green function takes into account that the fluid is bounded by and sticks to two parallel hard walls. Our numerical simulations were performed using the algorithm from Bhattacharya et al. (2005a,b, 2006), which combines spherical and Cartesian representations of the Stokes flow. The Cartesian representation is used to describe flow interaction with planar walls, and the spherical representation to describe flow interaction with the particle. The key element of the method is a set of transformation relations between the Cartesian and spherical representations. In this paper, we use a one-particle version of this code, for a particle in Poiseuille flow. The method has also been used to calculate hydrodynamic interactions in multiparticle systems.

4.2. Results from multipole expansion method taking into account particle interaction with walls

Results from the multipole expansion method have been obtained with the use of the available numerical code. The velocity \( U \) of the freely moving sphere normalized by the maximum velocity of the Poiseuille flow, \( u_m = \frac{1}{2} \langle u \rangle \), is plotted versus \( z \) and \( s \) in Fig. 3. Only half of the particle velocity profile (\( s \leq 0.5 \)) is represented by symmetry. For \( z \rightarrow 0 \), that is for a point particle, the plotted profile is simply one half of the Poiseuille flow parabola. From the geometrical restriction \( s \geq z \) on the particle center position, the relevant domain decreases for increasing \( z \). In the limit \( s = z = 0.5 \), the sphere touches both walls. It is observed that the particle velocity decreases sharply in the lubrication region, when the particle is interacting with one wall for \( (s-z)/z < 1 \). When \( z \) is close to 0.5 (and thus \( s \) also), there are lubrication regions on both walls and the decrease is even sharper.

Results for the normalized translational velocity \( U/u_m \) of the sphere in Poiseuille flow are provided in Table 1. For completeness, the results for the normalized rotational velocity \( \Omega w / u_m \) (where \( \Omega \) is the dimensional rotational velocity along the y-axis) are also presented. A comparison between the results of the multipole method and the results obtained by the boundary-integral technique in Staben et al. (2003) is available in Bhattacharya et al. (2006) (see their Tables I and II). More data are compared here in Table 1. The maximum difference of around 1.5% occurs for a particle close to a wall. The precision of the boundary-integral calculations decreases in this regime, whereas the multipole method retains its accuracy. On the other hand, the advantage of the boundary-integral method is that it provides the possibility to consider non-spherical particles, as in Staben et al. (2003).

4.3. Fitting formulae for the friction factors and particle velocity

The accurate results taking into account interactions with both walls (Section 4.2) are used in this subsection to derive fitting
of the lubrication terms of Poiseuille flow presented in Ekiel-Jezewska et al. (2008) for two walls proceeding in the same way as described in Section 3.1 for one wall, we then fitted the O(1) remainder terms by polynomials in $x$ and $s-1/2$. From the symmetry of $f_{xx}^0$, $f_{yy}$, and $f_{xy}$ with respect to the middle of the channel, we use even polynomials in $s$ and $s-1/2$, respectively, see Eqs. (8).

The way followed for the two-wall lubrication in Ekiel-Jezewska et al. (2008) is to superimpose the lubrication force or torque of each individual wall and adjust the constants in such a way as to account for two walls at the same time. The terms in log$c_i$ and $c_i \log e_i$ are added to terms in log$c_j$, constant and $c_j \log e_j$, where $e_i, e_j$ are the gaps between the sphere and wall 1, 2, respectively, see Eqs. (8).

A problem is that, outside the lubrication regime, $e_i$ and $e_j$ become large and the log terms then blow up. A possible way is, like in Section 3.1, to regularize by introducing the variable

$$\varepsilon_i = \frac{e_i}{1+e_i}, \quad i = 1, 2,$$

which has the limit of unity for $e_i \rightarrow \infty$ so that

$$L_i = \log \varepsilon_i, \quad i = 1, 2$$

vanishes for $e_i \rightarrow \infty$. We then substitute the $\varepsilon_i$’s with the $\varepsilon_i$’s in the lubrication formulae. Note also that the $c_i$’s may also be expressed as

$$\varepsilon_1 = 1 - \frac{x}{2}, \quad \varepsilon_2 = 1 - \frac{x}{2}.$$

In order to obtain a sufficient accuracy for $c_{xx}$ and $c_{yy}$, we found that it was more efficient to keep terms like $e_1 L_1$ rather than $\varepsilon_1 L_1$. Expanding the lubrication terms of $c_{xx}$ and $c_{yy}$ for $e_i \gg 1$, we found the appropriate constants multiplying $\varepsilon_1$ and $\varepsilon_2$ for these friction factors to be regular while retaining the correct symmetry or antisymmetry.

The results for the fitting formulae are

$$f_{xx}^0 = 0.954 - \frac{8}{15} (L_1 + L_2) - \frac{64}{375} (\varepsilon_1 L_1 + \varepsilon_2 L_2)$$

$$+ 0.0511 + 0.0190 \left( s - \frac{1}{2} \right) ^2 - 0.613 \left( s - \frac{1}{2} \right) ^4$$

$$+ z \left[ -0.961 - 4.75 \left( s - \frac{1}{2} \right) ^2 + 0.881 \left( s - \frac{1}{2} \right) ^4 \right]$$

$$+ z^2 \left[ 3.59 + 2.77 \left( s - \frac{1}{2} \right) ^2 + 84.4 \left( s - \frac{1}{2} \right) ^4 \right],$$

$$(29)$$

$$c_{xx}^0 = -\frac{1}{10} (L_1 - L_2) - \frac{43}{250} (\varepsilon_1 L_1 - \varepsilon_2 L_2) + 0.0209 (\varepsilon_1 - \varepsilon_2)$$

$$- 0.007 \left( s - \frac{1}{2} \right) + 0.037 \left( s - \frac{1}{2} \right) ^3 + z \left[ 0.097 \left( s - \frac{1}{2} \right) ^2 - 0.310 \left( s - \frac{1}{2} \right) ^4 \right]$$

$$+ z^2 \left[ 2.17 \left( s - \frac{1}{2} \right) ^2 - 7.21 \left( s - \frac{1}{2} \right) ^4 \right].$$

$$(30)$$

$$c_{yy}^0 = \frac{2}{5} (L_1 + L_2) - \frac{66}{125} (\varepsilon_1 L_1 + \varepsilon_2 L_2) + 0.1579 (\varepsilon_1 + \varepsilon_2 - 1)$$

$$- 0.206 + 0.0992 \left( s - \frac{1}{2} \right) ^2 + 0.323 \left( s - \frac{1}{2} \right) ^4$$

$$+ z \left[ -0.101 - 1.70 \left( s - \frac{1}{2} \right) ^2 + 16.83 \left( s - \frac{1}{2} \right) ^4 \right]$$

$$+ z^2 \left[ 0.307 + 15.4 \left( s - \frac{1}{2} \right) ^2 + 28.7 \left( s - \frac{1}{2} \right) ^4 \right].$$

$$(31)$$

The coefficient $f_{xy}^0$ is obtained from (11) and (30).

For the force and torque on a sphere held fixed in Poiseuille flow, there is no relative motion of surfaces, hence no shear in narrow gaps. As a consequence for the coefficients defined in (15) there is no lubrication term which may give a singularity for vanishing gap. So, fitting is easily done by polynomials in $x$ and $s-1/2$. Symmetry in...
than 1.3%, 1.9% and 1.7%, respectively. As for multipoles, the precision provided by (29), (31) and (32) is better.

The precision on the difference between the fitting formula (30) and the accurate result is less than 0.004. The precision is good, except near walls where \( c^p \) is small. For the reason just explained, the lack of local precision was found to be unimportant. The fitting formulae (30) and (33) for the torques were thus found to be sufficient in view of calculating the velocity.

The normalized velocity \( U/um \) is calculated from (14) using these fittings for the friction coefficients. Let \( U_{fit}/um \) be the fitted result. The precision of \( U_{fit}/um \) by comparison with the accurate result \( U/um \) obtained with the multipoles is less than 1% for the whole range of parameters. The largest error occurs for small \( \alpha \) and \( s \); this is shown in Fig. 4 which displays contours of the relative difference \( (U_{fit}−U)/U \). By comparison with the precision obtained for the friction factors, that on the velocity is a little better; indeed, it happens that some errors compensate.

The difference between the accurate and the fitted velocity normalized by the maximum Poiseuille velocity \( um \) is also useful (see Section 5). It is represented in Fig. 5. It is observed that the difference is less than 0.004 for the whole range of parameters.

The fitting formulae (29)–(33) for the friction factors may also be applicable to freely translating particles submitted to an external torque along \( y \), say \( C_{ry} \), like polar or polarizable particles in an electric field or magnetic particles in a magnetic field. In that case, the solution of the particle equations of motion gives the translation velocity \( U+Ur \), where \( U \) is given by Eq. (14) and

\[
U_r = \frac{C_{ry}}{8\pi s^2 \mu} \left( \frac{f_{ry}}{C_{ry} s^2 - f_{ry} c_{ry}} \right)
\]

The lack of precision on the torque \( c_{ry} \) does not affect this value since, as just said above, it has little influence on the denominator.
4.4. Ranges of validity of the nearest wall and wall-superposition approximations

Depending on the application and required accuracy in the domain of interest, simpler formulae based on a single wall (see Fig. 2) might be sufficient. The results for the velocity of a freely moving sphere in Poiseuille flow when using the nearest wall (Section 3.1) and wall-superposition (at the level of particle velocity, Section 3.2) approximations are here compared with the fitting formula obtained for two walls, \( U_{\text{fit}} = \frac{u_{m}}{C_0} U_0 \), Section 4.3.

The difference \( \left( \frac{U_{\text{fit}} - U_0}{C_0} \right) \frac{u_{m}}{U_0} \) is plotted in Fig. 6. Consider e.g. the 0.02 difference line: it is observed that the nearest wall approximation provides this accuracy for \( \alpha_0 = 0.18 \), that is when the sphere diameter is up to 36% of the channel width. On the other hand, it is quite surprising from Fig. 6 that the nearest wall approximation appears to be good for \( s = 0.5 \), that is when the sphere is closer to the middle of the channel. To study this two-wall effect in more detail, the limit case in which the sphere is located in the center of the channel (\( s = 0.5 \)) is considered in Fig. 7. Here, the relative difference \( \left( \frac{U - U_0}{U} \right) \) is plotted versus \( \alpha \). The function \( \alpha^5 \) is plotted as a dashed line for comparison. It is seen that \( \left( \frac{U - U_0}{U} \right) \) is practically of the order of \( \alpha^5 \) for small \( \alpha \). This is not in contradiction with, but yet surprisingly smaller than, the \( O(\alpha^4) \) expected from the method of reflexions as applied in Ho and Leal (1974). For \( \alpha \) in the range \([0.25, 0.96]\), the one-wall approximation is even better: it is 0.22% for \( \alpha = 0.4 \) and 0.44% for \( \alpha = 0.45 \). Note also that, by comparison, the Faxen approximation \( U^0 \) (viz no-wall approximation) gives for \( s = 0.5 \) a poorer relative accuracy \( \left( \frac{U^0 - U}{U} \right) \) of 5.5% for \( \alpha = 0.4 \) and 14% for...
Brownian motion can be characterized by a thermodynamic force $F$ here is assumed to be independent of the position of particles. We take two parallel plates in a Poiseuille flow. An external force field separation techniques. Section 4.3 are applied to calculate quantities relevant to particle

$$\langle c \rangle = \int_0^{1-x} c \, ds.$$  

(38)

Calculating this quantity we then derive:

$$\frac{c}{\langle c \rangle} = \frac{e^{-((x-2)/\lambda)}}{2 [1 - e^{-(1-2x)/\lambda}]}.$$  

(39)

In separation techniques, $\lambda$ is usually small, so that particles concentrate near the accumulation wall.

The retardation of particles in the channel is characterized by a parameter $R$ called the retardation factor or retention ratio:

$$R(x, \lambda) = \int_0^{1-x} \frac{c(s) U(s, x)}{\langle c \rangle \langle u \rangle} \, ds.$$  

(40)

Note that the integration in Eq. (40) is over the accessible part of the channel width, with the particles at least a radius away from either wall.

If particles are located far away from walls, a classical assumption in the modeling of separation methods is that they move with the velocity of the carrying fluid, $U(s, x) = U(s)$ (here, we neglect the pressure gradient term in the Frenkel formula (25)). The retention ratio may then be calculated, while keeping the steric exclusion zones expressed as the bounds in the integral, with the classical result (Giddings, 1978):

$$R(x, \lambda) = 6 (x - x^2) + 6 \lambda (1 - 2x) \left[ \coth \left( \frac{1 - 2x}{2\lambda} \right) - \frac{2\lambda}{1 - 2x} \right].$$  

(41)

For point particles ($x = 0$) in Poiseuille flow, the retention ratio is obtained from (41) by Giddings (1978):

$$R(0, \lambda) = \lim_{x \to 0} R(x, \lambda) = 6 \lambda \left[ \coth \left( \frac{1}{2\lambda} \right) - 2\lambda \right].$$  

(42)

For hydrodynamic chromatography, $\lambda \to \infty$ leads to

$$R_\infty = 1 + 2x - 2x^2.$$  

(43)

The expression (41) for the retention ratio is valid only when there are no hydrodynamic interactions between the walls and particles. Yet, this is not the case in separation techniques. These interactions are accounted for here by using the two-wall fitting formula presented in Section 4.3. The error in using the fitting formula instead of the accurate results for $U(s, x)$ leads to a difference:

$$\Delta R \leq \int_0^{1-x} \frac{c(s) \Delta U}{\langle c \rangle \langle u \rangle} \, ds$$

with (see Fig. 5):

$$\frac{\Delta U}{\langle u \rangle} = \frac{|U - U_{\text{ref}}|}{\langle u \rangle} = \frac{3 |U - U_{\text{ref}}|}{u_m} \leq 0.006$$

giving:

$$\Delta R \leq 0.006.$$  

(44)

Note that $U(s, x)/\langle u \rangle$ in (40) could be calculated using either the nearest wall or wall-superposition approximation. The formulae are simpler, at the expense of an added error, which can be evaluated with either Fig. 6 or Fig. 8, respectively.

The retention ratio $R$ may generally be calculated from (40) for an arbitrary distribution of particles interacting with both walls.
In this application, we will present the effect of the concentration distribution for Brownian particles (39) on the effective retardation of particles. But before presenting this effect in a general configuration, let us consider the particular case of CHDC. There is then no external force and the particle concentration is uniform across the channel. The retention ratio \( R_{eh} \) is simply:

\[
R_{eh}(x) = \frac{2}{1-2x} \int_{0}^{1/2} U(s,x) \frac{1}{4\pi s} ds
\]  

(45)

The integral represents the average of the velocity wall correction factor across half the channel width. \( R_{eh}(x) \) is represented in Fig. 9 (top solid line). The result is quite different from the one for point particles (43) (top dashed line in Fig. 9). The retention ratio for a uniform distribution (45) was also calculated numerically by Staben et al. (2003) (their Fig. 10). Our result (top solid line in Fig. 9) is very close with theirs for \( 2x < 0.7 \). In fact, the difference would be hardly visible in Fig. 9 (top solid line). In our calculation, integrals were evaluated using a five-point adaptative Newton–Cote technique, allowing a tolerance of 10^{-3}. Staben et al. (2003) found that the average velocity is at a maximum of \( R_{eh}^{\text{max}} = 1.18 \) when the distance between walls is such that \( 2x = 0.42 \). We obtain the maximum value of \( R_{eh}^{\text{max}} = 1.19 \) for \( 2x = 0.41 \). This maximum may be explained physically by remarking that increasingly large particles are shifted away from walls by steric effects and their velocity thus increases, but there is the concurrent retarding effect of hydrodynamic interactions. For large distance between walls \( x \ll 1 \), asymptotic formulae may be obtained. Staben et al. (2003) result for the average velocity is:

\[
R_{eh}^{\text{asymp}} = 1 + 2x - 7.71x^2 + O(x^3).
\]

The 2x term is due to purely steric effects. Fitting then the term at order \( O(x^2) \) on the basis of accurate results provided by the multipole method, the asymptotic result for the CHDC retention ratio is:

\[
R_{eh}^{\text{asymp}} = 1 + 2x - 7.71x^2 + O(x^3).
\]

(46)

Fitting our results in the whole range \( 2x \leq 1 \), we also propose the following ad-hoc formula that is valid with a relative error less than 4%:

\[
R_{\text{interp}}^{\text{ch}} = \left[ -4.437 - 4.543(1-x) + 54.47(1-x)^2 - 64.47(1-x)^3 \right. \\
+ 19.95(1-x)^4 \right]^{1/4}.
\]

(47)

For the more general case of a non-homogeneous Brownian suspension with the equilibrium concentration distribution (39), the retention ratio \( R(x,\lambda) \) taking into account the hydrodynamic interactions with walls is calculated from (40) for various values of \( \lambda \). The result is presented in Fig. 9 and compared with that of Giddings (1978) which ignores interactions with walls. It appears that the retention ratio taking wall effects into account has a maximum for a set of values of \( x \) and \( \lambda \). The values of \( x \) for which \( R(x,\lambda) \) is at a maximum, say \( x^{\text{max}}_\lambda \), is represented as a function of \( \lambda \) in Fig. 10. The retention ratio will then be less than the maximum value \( R^{\text{max}}(\lambda) = R(x^{\text{max}},\lambda) \) that is plotted versus \( \lambda \) in Fig. 11.

The selectivity of separation is better estimated in terms of the “selectivity parameter” \( \text{dlog}R/\text{dlog}x \). This parameter is plotted in Fig. 12 versus \( 2x \) for various values of \( \lambda \). A selective separation is obtained if \( \text{dlog}R/\text{dlog}x \) is as large as possible. Depending on \( \lambda \) and on the application, the best range of sizes for particle separation might be a subdomain of either \( x < x^{\text{max}} \) or \( x > x^{\text{max}} \).

Fig. 9. Comparison between values of the retention ratio, Eq. (41), calculated with only steric effects (Giddings, 1978) (dashed lines) and Eq. (40) with hydrodynamic interactions with walls (solid lines). Values of \( \lambda \) are, from bottom to top, 0.01, 0.05, 0.1, 0.2, 0.5. The thicker dashed and solid lines represent \( \lambda \to \infty \) which corresponds practically to hydrodynamic chromatography, Eqs. (43) and (45), respectively. The uncertainty when using the fitted formula for \( U \) is given by \( \Delta R \leq 0.006 \), Eq. (44).

Fig. 10. Values \( x^{\text{max}}_\lambda \) for which the retention ratio is at a maximum.
Fig. 12. Values of the selectivity parameter $d\log R/d\log z$ versus $2\xi$. Comparison between values calculated with only steric effects, Eq. (41) from Giddings (1978) (dashed lines) and with hydrodynamic interactions with walls, Eq. (40) (solid lines). Values of $\lambda$ are, from top to bottom, 0.01, 0.05, 0.1, 0.2, $\infty$ (the thicker solid and dashed lines represent $\lambda \rightarrow \infty$).

Consider for instance FFF in the Earth gravitational field. The transverse component, $F$, of the gravity force varies like $x^2$ so that $\lambda$ varies like $x^{-3}$ (except for a vertical configuration in which case $F=0$). But $\lambda$ is large anyway for Brownian particles; then we may consider the limit $\lambda \rightarrow \infty$ for simplification. This limit corresponds practically to CHDC. Fig. 12 shows that there is a region $2\xi \lesssim 0.5$ where $d\log R/d\log z$ is large. However, in practice, too narrow channels may be blocked by the largest sample particles and are thus avoided. The maximum value of normalized diameter $2\xi$ used in experiments was 0.47 (Moon and Giddings, 1996). We consider that the practical domain corresponds to $2\xi < 0.5$ and thus $d\log R/d\log z > 0$. Fig. 12 shows that a maximum selectivity of 0.038 is obtained for $2\xi = 0.18$. It is smaller than the maximum expected from the model which does not consider hydrodynamic effects, that is 0.091 for $2\xi = 0.45$. Nevertheless, Fig. 12 shows that a much larger selectivity would be obtained if separations could be implemented in the range $0.5 < 2\xi < 0.9$.

In FFF, the force field generally depends on particle size, which makes this separation method well suited for particle-size analysis. Hence, Fig. 12 for finite values of $\lambda$ represents the sole contribution of the steric effect to the selectivity taking into account the hydrodynamic wall–particle interactions, i.e. the selectivity that would be obtained by keeping the force field constant whatever the size. In order to exploit these results in a proper way, the variation of $F$ with $x$ (for instance for a lift force), and thus of $\lambda$ with $x$, should be taken into account. This is a topic for future projects.

6. Summary of results and conclusion

Accurate results for a freely moving spherical particle in Poiseuille flow between two parallel walls are provided on the basis of the multipole expansion (Ekiel-Jeżewska and Wajnryb, 2006) coupled to the Cartesian representation (Bhattacharya et al., 2005a,b). These results take into account the simultaneous contribution of both walls.

The main purpose of this paper is to provide handy formulae in view of applications. Fitting formulae are derived, based on these accurate results and on lubrication results for two walls (Ekiel-Jeżewska et al., 2008). Simpler fitting formulae with an accuracy which is appropriate to some domain in $\alpha$ and $s$ are also proposed.

1. The most precise formulae fitting the friction factors are given in Section 4.3. They are valid in the whole range of distances and sphere radius to channel width ratio. The particle velocity is then derived using (14). The precision of the fitting, as compared with the exact results from the multipole method, is better than 1% as displayed in Fig. 4.

2. If the domain of $(\alpha,s)$ is limited, it may be sufficient to use the superposition of two walls on the level of velocity. Then one should use the approximation (24) together with the result for a single wall (see next item) and (25) for no wall. The precision of this approximation may be estimated from Fig. 8: accepting for instance a relative difference of 0.02 with the accurate results from multipoles, the formula is valid in the $(\alpha,s)$ region located above the 0.02 curve when $\alpha > 0.25$ and valid for any $s \geq \alpha$ when $\alpha \leq 0.25$.

3. An even simpler formula based on the hydrodynamic interactions with the nearest wall is obtained by fitting accurate results from the method of bispherical coordinates (which are in excellent agreement with results from the multipole method for one wall, see Chaoui and Feuillebois, 2003; Pasol et al., 2006; Ekiel-Jeżewska and Wajnryb, 2006). The expression for the velocity is (13) with the fitting (20) and (21). The precision of this approximation may be estimated in this case with Fig. 6, with the procedure explained above for Fig. 8. Moreover, this nearest wall approximation is surprisingly good for a sphere close to the middle of the channel, as shown by Fig. 7. (Note that the no-wall approximation, viz the simple use of Faxen formula, is much poorer as shown at the end of Section 4.4.) Thus, the nearest wall approximation might be sufficient for many practical purposes.

These handy formulae may be used in various applications, for instance the CHDC and FFF methods of separation used in analytical chemistry.

For Brownian particles in CHDC, it is shown how earlier models of these techniques, which ignored hydrodynamic interactions with walls, fail to predict the proper choice of channel width for a selective separation. Indeed, it is observed in Fig. 12 that the selectivity parameter $d\log R(\alpha,s)/d\log z$ (based on the retention ratio $R(\alpha,s)$ for $\lambda \gg 1$) when considering hydrodynamic interactions is at maximum of 0.038 for a value of $2\alpha = 0.18$, which could not be found when ignoring hydrodynamic interactions. Although performing particle separation in the range $2\alpha \gtrsim 0.5$ can be risky because of particle blockage, the result that a large selectivity is obtained in this range might find practical applications for systems with a narrow particle-size distribution.

The curves in Figs. 9 and 12 are therefore fundamental for improving the selectivity of separation techniques. This is a topic for a future work in this field. The dispersion around the average could be studied in more detail following Brenner and Gaydos (1977) with the use of the present formulae for the particle velocity.

FFF techniques using other separation forces for non-Brownian particles could be studied along the same lines, taking proper account of the variation of the force (e.g. lift force) with the particle size.

Finally, the presented formulae may in general find applications in the transport of macromolecular or colloidal objects in microfluidic systems, in particular when the sizes of the objects become comparable to that of the channel.

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