Dynamic simulation of one and two particles sedimenting in viscoelastic suspensions of FENE dumbbells

Housam Binous, Ronald J. Phillips

Department of Chemical Engineering and Materials Science, University of California at Davis, Davis, CA 95616, USA

Received 18 February 1998

Abstract

A new method is introduced for simulating the motion of particles in a viscoelastic fluid. The viscoelastic fluid is represented as a suspension of finite-extension-non-linear-elastic (FENE) dumbbells in Newtonian solvent. Instead of using an averaging technique to derive a continuum constitutive model, such as that of Chilcott and Rallison, we calculate directly the particle–particle and particle–bead interactions by using a modified version of the Stokesian dynamics method. This method is applied to a series of sedimentation problems, including the sedimentation of a single sphere, a non-spherical particle, and two spheres. It is found that the drag of a single sphere can be increased significantly by the presence of the dumbbells, and at high Deborah numbers (De) the sphere velocity becomes time-periodic. Non-spherical particles rotate as they fall such that their long axis is ultimately pointed in the direction of gravity. Two sedimenting spheres are in most cases attracted to each other, and they turn such that their line-of-centers is in the direction of gravity. The rotational velocity of the spheres in a side-by-side configuration is in the same direction as it would be in Newtonian fluid, in keeping with experimental observations found in the literature. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Viscoelastic; FENE; Dumbbell; Particle; Sphere

1. Introduction

It is now widely known that the motion of particles in non-Newtonian fluids differs dramatically from the corresponding behavior in Newtonian fluids. Because of the ubiquitous nature of suspensions in nature and industry, several methods for simulating the interactive motion of N spheres in Newtonian fluids have been developed, including Stokesian dynamics [1] and lattice-Boltzmann techniques [2]. In this paper, we present a Stokesian-dynamics-based approach for calculating the motion of spheres sedimenting in non-Newtonian fluids. The non-Newtonian character of the suspending fluid is incorporated by including thousands of bead-and-spring dumbbells in the simulation, and the bead–
sphere and sphere–sphere interactions are calculated by using relations from low Reynolds number (Re) hydrodynamics. In principle the method could be applied to $N$ spheres. However, to present the method and evaluate its merits, here we limit ourselves to one- and two-particle simulations, for which there is a relatively large amount of experimental and theoretical information available.

The connection between suspensions of bead-and-spring dumbbells and certain types of viscoelastic fluids is well-established. The Oldroyd-B or convected Jeffreys constitutive equation can be derived directly by applying methods of kinetic theory to a suspension of Hookean dumbbells [3,4]. However, Hookean dumbbells are known to exhibit a physical behavior in elongational flows, because the velocity field is such that they extend indefinitely and become infinitely long. In our simulations we therefore use Nonlinear-Finite-Extension-Elastic (FENE) dumbbells [3,5], which have a maximum extension length at which the connecting force becomes infinite. Like the continuum constitutive model of Chilcott and Rallison [6], our simulations are therefore primarily characterized by three dimensionless parameters: the dimensionless dumbbell concentration $c$; the dimensionless maximum extension length $L$; and the Deborah number (De), which is a ratio of the characteristic relaxation time of the fluid to a characteristic time for the flow. Precise definitions of these parameters are given below. Although the dumbbell contribution to the stress does shear-thin in our simulations, as it does in the FENE–P model [5], that contribution is always small compared to the solvent contribution. Our suspensions are therefore best thought of as Boger fluids in that they exhibit normal stresses and elasticity, but very little shear-thinning.

There have been numerous studies of the motion of particles sedimenting in various types of viscoelastic fluids [7]. For the sedimentation of a sphere at low Reynolds number in a Boger fluid, Walters and Tanner [8] have suggested that the elasticity has little effect or causes a slight drag reduction at low Deborah numbers, but causes a marked increase in drag as the De increases. This conclusion was reached by a compilation of experimental data. Recent experiments by Solomon and Muller [9] show a dramatic increase in the drag with increasing Deborah numbers, with the increase being as large as a factor of 3 for Deborah numbers greater than unity. This drag increase was affected by the solvent quality, with low-extensibility polymer solutions showing very small deviations from Newtonian behavior, and greater extensibility leading to the large increase in the drag. The numerical results of Chilcott and Rallison [6] and Harlen [10] also suggest the possibility of drag enhancement in Boger fluids. In particular, Chilcott and Rallison proposed a mechanism for drag enhancement that involves extended dumbbells in the wake of the sphere; we show below that this mechanism is very obvious in our simulations, and under most conditions leads to a drag increase relative to the Newtonian case. Significantly less work has been done to study sedimentation of non-spherical particles in viscoelastic fluids. However, in the absence of inertia it has been shown experimentally that axisymmetric particles tend to settle in the direction of their axis of rotation [11–13], and this tendency is consistent with what is predicted by the second-order fluid constitutive model [14].

An early study of the motion of two spheres in polymer solutions was that of Riddle et al. [15], who found that two spheres sedimenting in a tube move together or apart, depending on whether their initial separation is less or greater than a critical value. More recently, Joseph et al. [16] have shown that, during sedimentation at low Reynolds number, groups of spheres in polymer solutions tend to cluster together into rows oriented in the direction of motion. This behavior appeared in several fluids with varying degrees of shear-thinning, including some in which shear-thinning would be expected to be absent at the shear rates of interest. In contrast, Gheissary and van den Brule [17] show two-sphere sedimentation results in which the spheres fall as a vertical doublet if the fluid exhibits shear-thinning,
but which show the two spheres separating in a Boger fluid. For the latter, separating spheres, even if the two spheres are initially oriented vertically, after separating they assume a ‘tilt’ and lose their vertical orientation (see Fig. 7(c) in Gheissary and van den Brule’s paper). It is not yet clear what distinguishes the results of Gheissary and van den Brule from those of Joseph et al. [16], although Gheissary and van den Brule [17] suggest that elasticity drives particles apart, and hence by inference it must be other effects, such as shear-thinning, that lead to the particles forming vertical chains. Allen and Uhlherr [18] show photographs of suspensions sedimenting in shear-thinning, elastic polymer solutions, and show the presence of marked inhomogeneities in the suspension microstructure.

Relatively few theoretical and numerical calculations have been performed involving two or more particles. Brunn [19] showed that, in a second-order fluid, two sedimenting spheres will come together, rotate to a vertical orientation, and fall as a doublet. Phillips [20] used the ‘reciprocal theorem approach’ described by Leal [21] to extend the Stokesian dynamics method to low Deborah number, and showed that in multisphere simulations particles tend to cluster when sedimenting in a second-order fluid. This observation agrees qualitatively with the photographs reported by Allen and Uhlherr [18]. Feng et al. [22] used POLYFLOW to simulate the sedimentation of two circular particles in two dimensions in an Oldroyd-B fluid. Their results show the circular particles moving together if started close enough to one another, and maintaining an approximately constant separation otherwise.

It has been noted that viscoelastic instabilities can be found in multi-dimensional flows, such as flow past a cylinder or flow around a falling sphere [23]. In particular, Bisgaard [24] noted the development of a time-dependent flow in the wake behind a sphere sedimenting in a circular tube. The sphere was falling in a shear-thinning viscoelastic fluid with a Deborah number greater than 40, but at low Reynolds number. Similar experiments by Arigo et al. [25] did not show such an instability, although the latter experiments were performed with a different fluid. McKinley et al. [26] did find an instability in flow around a cylinder held fixed between flat plates. At a critical Deborah number, a three-dimensional secondary flow developed in the wake of the cylinder; at a higher Deborah number, the flow became time-periodic. The presence of secondary flows and transitions to time-periodic behavior have similarly been found in flow through abrupt contractions [27,28].

In this paper, we present a new approach for calculating the effect of non-Newtonian rheology on particle motion, where elastic effects are incorporated by directly calculating particle–dumbbell interactions, rather than by applying a finite-element or spectral numerical method to a set of non-linear differential equations. Before explaining the details of our calculations, we note some advantages and disadvantages of our method. The most obvious disadvantage is the need to account for the dumbbells independently, rather than in an averaged sense as is done in a continuum constitutive model. However, the constitutive model that results from the averaging process is invariably non-linear, resulting in non-linear governing equations, even in the absence of inertia. In contrast, under inertia-free or low Reynolds number conditions, the hydrodynamics of the individual particles and dumbbells before averaging are governed by the linear Stokes equations, greatly facilitating the calculation of hydrodynamic interactions, even for tens of thousands of dumbbells. Note, however, that the simulation proceeds by numerical integration of a large system of first-order, non-linear differential equations for the particle positions, and hence all the non-linear behavior of the continuum constitutive equation approach is retained. Furthermore, low Reynolds number interactions can be calculated without the need for a mesh such as is used in finite element calculations. In simulations where the particles are moving and the geometry changes with every time step, the lack of a need for a mesh is an important advantage.
In addition, the proposed simulation method is very flexible in that it is straightforward to incorporate non-Hookean springs of any type that is desired. In particular, it has been suggested that incorporating viscous damping in the springs may have advantages in some circumstances [29]. The two-bead dumbbells can also be altered to have a distribution of spring force constants, or to be multibead chains of beads connected by springs or rods (cf. [30]), yielding fluids with multiple relaxation times. Hence, with this method the prospect of performing multispher simulation in shear-thinning, viscoelastic fluids with multiple time constants is conceivable, provided current methods in parallel computing are implemented.

In the sections that follow we first describe the method we use to calculate hydrodynamic interactions between non-Brownian, sedimenting spheres and Brownian, bead-and-spring dumbbells. Then in Section 3, we report results from simulations which were performed with only the dumbbells present, in order to ‘measure’ the rheological properties of our suspensions; we compare our results with what is predicted for a Chilcott-Rallison fluid, and also with the simulation results of van den Brule [31]. Our results for a single sphere falling in a suspension of dumbbells are presented in Section 4, and we compare our calculated results for the drag on the sphere with theoretical and experimental results from the literature. We also describe an instability that develops in our simulations at Deborah numbers of about five, in which the sphere velocity develops an oscillatory component that shows some qualitative similarities with instabilities reported for other multi-dimensional flows with an elongational component [24,26]. Simulation of a non-spherical particle is described in Section 5, where a single particle consisting of two spheres ‘glued’ together is monitored as it sediments and assumes a preferred downward orientation due to the presence of the dumbbells. Two spheres moving freely in various configurations are discussed in Section 6, and it is shown that the two spheres generally move together as they sediment, rotating so that ultimately their line-of-centers is oriented in the direction of gravity. In Section 7, we discuss the interpretation of our results in terms of dumbbell–particle interactions in order to provide intuitive insight into the complex problem of particle motion in viscoelastic fluids, and Section 8 consists of some concluding remarks.

2. Method of simulation

Our simulations are performed by using an extension of the Stokesian dynamics method for calculating the motion of $N_s$ spheres in a Newtonian fluid at low Reynolds number [1,32]. In this method, sphere–sphere interactions are separated into two categories, those being so-called ‘far-field’ and ‘near-field’ interactions. The far-field interactions are calculated rigorously by using a multipole moment expansion to compute the fluid velocity disturbance caused by each sphere in terms of the forces and torques being applied to it. (Note that in this discussion we refer to the FT-method described in [32].) The effect of the fluid velocity disturbance caused by one sphere on the velocity of another sphere is evaluated by using Faxén’s formulae. The final result is the far-field, ‘grand mobility matrix’ that relates the forces and torques on $N_s$ suspended spheres to the translational and rotational velocities of those spheres.

The near-field interactions are lubrication interactions that arise when two spheres in relative motion come very close together. Such interactions are dominated by the two nearly touching spheres and are relatively unaffected by any other spheres; hence, they are accounted for by using a pairwise additivity approximation. This method of accounting for near-field interactions works well when spheres are in
relative motion, such as in sedimentation or shear flows. However, in porous media, where spheres are in fixed positions, the lubrication interactions play no role and the method is only as accurate as the far-field terms that are included [33].

In our current simulations, sphere–sphere interactions are computed by using the FT-method of Durlofsky et al. [32], with both far- and near-field interactions. However, we must also account for the presence of thousands of beads which comprise the bead-and-spring dumbbells. We take these beads to be freely rotating, and hence they are free of any applied torque. The applied force is determined by the spring length, by using the same expression used by Chilcott and Rallison [6]:

\[
F^b_j = \frac{KR}{1 - R^2/L^2} = \frac{3kT/L_{eq}^2 R}{1 - R^2/L^2}
\] (1)

In Eq. (1), \(F^b_j\) is the force on the \(j\)th bead, \(R\) is the vector from the \(j\)th bead to the bead with which it is connected, \(R\) is the magnitude of \(R\), \(k\) is Boltzmann’s constant and \(T\) is temperature. The parameter \(L\) is the length of the dumbbell at its maximum extension, and \(L_{eq}\) is the equilibrium length of the dumbbell in the absence of flow as \(L \to \infty\) (i.e. for Hookean dumbbells). For separations much smaller than \(L\), Hooke’s law for linear springs is recovered, with the spring constant \(K\) equal to \(3kT/L_{eq}^2\). Eq. (1) is a close approximation to the inverse Langevin force law for polymer deformation, which can be derived from molecular arguments [5].

Given this force, the far-field velocity disturbance \(u'\) at some position \(x\) caused by a bead at \(x_b\) is [32]

\[
u(x) = \frac{1}{8\pi \eta_b} \left( 1 + \frac{a_b^2}{6} \nabla^2 \right) J(x - x_b) \cdot F_b,
\] (2)

where \(\eta_b\) is the viscosity of the Newtonian suspending solvent and \(a_b\) is the bead radius. The Oseen tensor \(J\) is the Green’s function for Stokes flow, given by,

\[
J(x) = \frac{1}{r} (I - xx),
\] (3)

where \(r = (x \cdot x)^{1/2}\) is the magnitude of \(x\).

Combining the fluid velocity disturbance obtained from Eq. (2) with the relevant Faxén formula yields a \(3 \times 3\) mobility tensor. The Faxén formula that relates fluid disturbances to changes in a sphere’s translational velocity is

\[
U' = \left( 1 + \frac{a^2}{6} \nabla^2 \right) u',
\] (4)

where \(a_s\) is the sphere radius and \(U'\) is the change in the sphere velocity caused by the disturbance velocity \(u'\). Combination of Eq. (2) and Eq. (4) yields

\[
U' = \frac{1}{18 \pi \eta_b} \left[ \left( 1 + \frac{a_b^2}{6} \nabla^2 + \frac{a_s^2}{6} \nabla^2 \right) J \right] \cdot F_b,
\] (5)

where the term in square brackets is the \(3 \times 3\) portion of the grand mobility matrix that relates the force on a particular bead to the resulting change in a sphere’s translational velocity. We note that an identical
expression relates the force on the sphere to the resulting change in the bead’s translational velocity, as is expected since the mobility matrix is symmetric. To include the rotational velocities and torques on the spheres, the Faxén formula for rotational velocity is used,

$$\Omega' \frac{1}{2} \nabla \times \mathbf{u}'$$

where $$\mathbf{u}'$$ is again given by Eq. (2).

Because Stokes’ equations are linear, the contributions of all the particle interactions can be summed, leading to a matrix equation of the form:

$$
\begin{bmatrix}
U \\
\Omega \\
U_b
\end{bmatrix} = 
\begin{bmatrix}
M_{SS}^{UF} & M_{SS}^{UL} & M_{SB}^{UF} \\
M_{SF}^{UF} & M_{SF}^{UL} & M_{SF}^{SF} \\
M_{UF}^{BS} & M_{UL}^{BS} & M_{SB}^{BS}
\end{bmatrix} \cdot
\begin{bmatrix}
\mathbf{F} \\
\mathbf{L} \\
\mathbf{F}_b
\end{bmatrix}
$$

In Eq. (7), $$\mathbf{U}$$, $$\Omega$$, $$\mathbf{F}$$ and $$\mathbf{L}$$ are vectors of dimension $$3N_s$$ containing the translational and rotational velocities, forces and torques of the spheres, respectively. The vectors $$\mathbf{U}_b$$ and $$\mathbf{F}_b$$ have dimension $$3N_b$$ and contain the translational velocities and forces on the beads, respectively. The subscripts of the components of the grand mobility matrix refer to whether that term relates translational velocities to forces ($$\Omega_{UF}$$), rotational velocities to forces ($$\Omega_{SF}$$), etc.; the superscripts refer to whether the interaction is between two spheres (SS), two beads (bb), or a sphere and a bead (bS or Sb).

In calculating the bead velocities we modify Eq. (7) in two important ways. First, because there are so many more beads than spheres, it is convenient to neglect bead–bead interactions (i.e. the $$\Omega_{bb}^{bb}$$ matrix) in many simulations. As discussed below, we have evaluated the importance of these interactions by doing several test simulations in which bead–bead interactions are included within a specified radius of interaction. Bead–bead interactions rarely change the qualitative behavior in the sedimentation problems studied here, but generally they do affect the quantitative results for parameters such as the drag coefficient.

The second feature of the bead motion that is not included in Eq. (7) is the random or Brownian steps that they take each time step. Without such a Brownian diffusion term, there is nothing to prevent the spring force from collapsing the two beads of a dumbbell to a single point if the dumbbell is located in a stagnant region of fluid. The required random term is calculated by using the algorithm of Ermak and McCammon [34], which gives the change in position $$\Delta \mathbf{r}_b^i$$ of the $$i$$th bead in a time step $$\Delta t$$ as

$$
\Delta \mathbf{r}_b^i = \left[ \mathbf{u}^{\infty} (\mathbf{x}) + \frac{\mathbf{F}_b^i}{6\pi \eta_d a_b} + \sum_{j=1}^{N_b} \mathbf{M}_{bb}^{ij} \cdot \mathbf{F}_s^j + \sum_{j=1}^{N_b} \mathbf{M}_{UL}^{ij} \cdot \mathbf{L}_s^j + \sum_{j=1, j \neq i}^{N_b} \mathbf{M}_{UF}^{ij} \cdot \mathbf{F}_b^j \right] \Delta t + \mathbf{R}^i (\Delta t).
$$

The calculation of the random term $$\mathbf{R}^i (\Delta t)$$ is accomplished by using the method described by Ermak and McCammon [34], using a constant Stokes–Einstein diffusion coefficient $$D$$ given by

$$
D = \frac{kT}{6\pi \eta_d a_b}.
$$

Periodic boundary conditions would seem a natural choice for simulations of this type; they would have the desirable effect of producing an infinite suspension of FENE dumbbells with only a finite system of
one periodic unit cell. However, it is known from previous work [33,35,36] that, in three-dimensions, imposing periodic boundary conditions with the nearest-image convention results in grand mobility matrices that are not positive definite. A positive-definite mobility is required for the overall system to dissipate energy. One way to avoid this problem is to use the Ewald summation technique to sum the far-field interactions of all the periodically replicated spheres and beads in a convergent and relatively efficient manner [35,36]. Such an approach would necessarily result in a simulation of a periodic array of spheres in a suspension of dumbbells, since the spheres must be replicated along with the dumbbells.

Because we are currently more interested in examining finite groups of spheres rather than full suspensions, we have chosen not to use periodic boundary conditions and Ewald sums. Instead, we fill a finite volume surrounding the sedimenting spheres with dumbbells, and leave the region outside that finite volume a dumbbell-free, pure fluid. The volume is spherical in shape and has a radius from 10 to 20 sphere radii. All of the results presented here are unaffected by the finite nature of the dumbbell suspension. Our choice for the size of the finite region was in part motivated by the numerical calculations of flow around spheres and cylinders performed by Chilcott and Rallison [6]. In those calculations, as a boundary condition the fluid velocity was set equal to the bulk, imposed velocity at a distance of 10 radii from the cylinder or sphere being studied. Because the sedimenting spheres are subjected to a gravitational force and the dumbbells are not, if no precautionary steps are taken then in a short time the spheres will fall through the finite dumbbell region and find themselves in a pure Newtonian fluid. To prevent that from happening, at regular intervals dumbbells from the upper end (i.e. in the sphere wake) of the dumbbell region are removed and new dumbbells are introduced below the dumbbell region. The newly introduced dumbbells have a random orientation and the bead-to-bead separation is equal to the mean separation that would exist in a suspension of dumbbells in a quiescent fluid.

In the discussion of our simulation results given below, we characterize our dumbbell suspensions in terms of dimensionless groups suggested by Rallison and Hinch [37] and Chilcott and Rallison [6]. We define a dimensionless concentration \(c\) as

\[
c = n\pi a_b^3.
\]

where \(n\) is the number of dumbbells per unit volume. We also define a dimensionless Deborah number as a ratio of the relaxation time for the fluid to a characteristic flow time. For the sedimentation problems we study here, this yields

\[
De = \frac{\lambda U_T}{a_s},
\]

where \(U_T\) is the terminal sedimentation velocity of a sphere with radius \(a_s\) in a dumbbell suspension. The fluid time constant \(\lambda\) is given by,

\[
\lambda = \frac{6\pi \eta_s a_b}{4K},
\]

where \(K\) is the spring constant as given in Eq. (1). Finally, we define a dimensionless maximum spring extension \(\dot{L}\) as \(\dot{L} = L/a_s\). (Note that using \(a_s\) to scale \(L\) differs from some previous work, where the extensibility \(L\) and bead-to-bead vector \(\dot{R}\) are scaled such that \(\langle \dot{R} \dot{R} \rangle = I\) at equilibrium.) The root-
mean-square equilibrium bead separation $L_{\text{eq}}$ is related to the other parameters by

$$L_{\text{eq}}^2 = \frac{3kT}{K} = \frac{18\pi\eta_b a_s D}{K}.$$  

(13)

For our single-sphere sedimentation simulations, we have chosen the spring constant $K$ such that the equilibrium root-mean-square spring length $L_{\text{eq}}$ equals the sphere radius $a_s$ at a temperature of 25°C. For non-spherical particle and two-sphere simulations, $K$ is such that $L_{\text{eq}} = 0.77a_s$. The dimensionless maximum extension $\hat{L}$ is varied from 1.73 to 10. In all of our simulations, the bead size is one-tenth of the sphere size, $a_b = 0.1a_s$. The simulations were run in parallel on four HP 712/100 workstations by using Message-Passing Interface (MPI) software; a typical simulation with one or two spheres required about 8 h.

### 3. Rheological properties of dumbbell suspensions

We have performed dynamic simulations of sheared suspensions of FENE dumbbells in order to calculate directly the material functions of our fluids. These simulations do not include any suspended spheres, and are similar to those performed by van den Brule [31], except that we consider exclusively two-bead dumbbells. In reality these simulations correspond to transient experiments in which stress growth is monitored following the inception of a steady-shear flow. The objective is therefore both to examine the transient relaxation behavior and the steady, or long-time values of the material functions. Although we do not attempt to present an exhaustive survey of all the rheological features of dumbbell suspensions, these results serve the purpose of identifying the key features of our suspensions: they show very little shear-thinning, exhibit a positive first normal stress difference and a negligibly small second normal stress difference, and the elongational viscosity asymptotes to a finite value in the limit of increasing elongation rate. All of the results presented in this section were obtained by using a system of 20 000 non-interacting FENE dumbbells with a maximum extension length of $\hat{L} = 3.16$.

We first consider a suddenly imposed steady-shear flow with constant shear rate $\dot{\gamma}$ acting on a suspension of FENE dumbbells. To perform such simulations, the imposed velocity $u^\infty(x')$ in Eq. (8) is taken to be $u_x^\infty = \dot{\gamma}y$, $u_y^\infty = 0$ and $u_z^\infty = 0$, leading to a velocity gradient tensor given by

$$\Delta^\infty_u = \dot{\gamma}(t) \begin{bmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}.$$  

(14)

The polymer contribution to the stress $\tau_p$ is calculated by using Kramers’ expression [4,5]:

$$\tau_p = -n\langle RF_b \rangle + nkTI.$$  

(15)

The angular brackets in Eq. (15) denote an average over all dumbbells. Knowing the positions and forces of all the beads, Eq. (15) makes it a simple matter to calculate the polymer contribution to any component of the stress tensor.
The material functions of interest for a suddenly imposed shear flow are the viscosity $\eta^+(t, \dot{\gamma})$ given by

$$\eta^+(t, \dot{\gamma}) = \frac{\tau_{xy}}{\dot{\gamma}},$$  

(16)

the first normal stress coefficient $\psi_1^+(t, \dot{\gamma})$ given by

$$\psi_1^+(t, \dot{\gamma}) = \frac{\tau_{xx} - \tau_{yy}}{\dot{\gamma}^2},$$  

(17)

and the second normal stress coefficient $\psi_2^+(t, \dot{\gamma})$ given by

$$\psi_2^+(t, \dot{\gamma}) = \frac{\tau_{yy} - \tau_{zz}}{\dot{\gamma}^2}. $$  

(18)

The superscript $+$ in Eqs. (16)–(18) indicates that the constant shear rate $\dot{\gamma}$ is applied instantaneously for times $t > 0$. At long times, $t \gg \lambda$, the viscosity $\eta^+(t, \dot{\gamma})$ and normal stress coefficients $\psi_1^+(t, \dot{\gamma})$ and $\psi_2^+(t, \dot{\gamma})$ relax to their steady values, denoted by, $\eta(\dot{\gamma})$, $\psi_1(\dot{\gamma})$ and $\psi_2(\dot{\gamma})$, respectively. In the discussion below the transient functions are presented in dimensionless form, with the non-dimensionalization achieved as follows:

$$[\dot{\eta}^+] = \frac{\eta^+ - \eta_s}{nkT\lambda},$$  

(19)

$$\dot{\psi}_1^+ = \frac{\psi_1^+}{2nkT\lambda^2}$$  

(20)

and

$$\dot{\psi}_2^+ = \frac{\psi_2^+}{2nkT\lambda^2}.$$  

(21)

The dimensionless functions $\dot{\eta}(\dot{\gamma})$, $\dot{\psi}_1(\dot{\gamma})$ and $\dot{\psi}_2(\dot{\gamma})$ are normalized in an analogous fashion. The normalization quantities used in Eqs. (19)–(21) are suggested by the kinetic theory of dumbbell solutions [5].

In Fig. 1 we plot the transient shear viscosities $[\dot{\eta}^+]$ as a function of time for a range of dimensionless shear rates $\lambda \dot{\gamma}$. A very prominent overshoot is present before the steady-state values are reached. This overshoot increases as the imposed shear rate increases, as is often seen experimentally in polymer solutions. Similar behavior is seen in Fig. 2, where values of $\dot{\psi}_1^+$ are plotted vs. time. However, the overshoot for the first normal stress coefficients is less pronounced than that for the shear viscosities. Values of $\dot{\psi}_2^+$ are negligibly small at all shear rates studied. The steady-state values of the viscosity $\dot{\eta}(\dot{\gamma})$ and the first normal stress coefficient $\dot{\psi}_1(\dot{\gamma})$ are plotted vs. shear rate in Fig. 3. Both of these functions show shear-thinning behavior, as found in the FENE-P model [5]. We note, however, that the viscosity plotted only accounts for the polymer contribution to the stress, the solvent viscosity $\eta_s$ having been subtracted out as shown in Eq. (19). For virtually any liquid solvent, this polymer contribution is quite small, and hence the overall behavior of the solvent plus the polymer exhibits negligible shear-thinning and can be thought of as Boger fluid. The range of $\lambda \dot{\gamma}$ covered in Fig. 3 is corresponds to what one
expects to find in one of our simulations. These results are in good agreement with the calculations of van den Brule [31], who also showed that such simulations yield the correct asymptotic behavior in the limit $\lambda\dot{\gamma} \to \infty$.

Next we turn our attention to a suddenly imposed, steady elongational flow with elongation rate $\dot{\varepsilon}$. For this case, the imposed velocity $\mathbf{u}^\infty(\mathbf{x})$ in Eq. (8) is given by $u_x^\infty = -(\dot{\varepsilon}x)/2$, $u_y^\infty = -(\dot{\varepsilon}y)/2$ and
$u_z^\infty = \dot{\varepsilon} z$, yielding a velocity gradient tensor of the form

$$\nabla u^\infty = -\dot{\varepsilon}(t) \begin{bmatrix} \frac{1}{2} & 0 & 0 \\ 0 & \frac{1}{2} & 0 \\ 0 & 0 & -1 \end{bmatrix}.$$  \(22\)

The material function of interest is the elongational viscosity

$$\hat{\eta}^+(t, \dot{\varepsilon}) = \frac{\tau_{zz} - \tau_{xx}}{\dot{\varepsilon}},$$  \(23\)

with the dimensionless polymer contribution $\hat{\eta}^+$ given by

$$\hat{\eta}^+ = \frac{\tilde{\eta}^+ - 3\eta_s}{3nK\lambda}.$$  \(24\)

In Fig. 4 the transient elongational viscosity is plotted as a function of time. The overshoot that is observed for the shear viscosity and first normal stress coefficient is not apparent here, as is often the case experimentally [3], and the elongational viscosity has reached its steady value by a dimensionless time of approximately unity for $\lambda \dot{\varepsilon} = 5.8$. Values of the steady state elongational viscosity $\hat{\eta}$ are plotted vs. the elongation rate $\dot{\varepsilon}$ in Fig. 5 for dumbbells with a maximum extension $\hat{L}$ equal to 10. A steep rise in the elongational viscosity is evident at a dimensionless elongation rate of approximately unity.

Our simulation result for the limiting elongational viscosity as $\dot{\varepsilon} \to \infty$ at $\hat{L} = 10$ is 175. Our results for the zero-shear rate viscosity and first normal stress coefficient at $\hat{L} = 3.16$ are 0.84 and 0.78, respectively. These results compare favorably to the analytical expressions provided by Wiest and Tanner [38] evaluated at the same conditions, which yield a value of 200 for the elongational viscosity and 0.86 and 0.69 for the zero-shear rate viscosity and first normal stress coefficient, respectively. Finally, we have calculated the average end-to-end distance for our suspensions as a function of shear
rate, and these results are shown in Fig. 6. For the case of Hookean dumbbells for which the maximum extension is unbounded, \( \hat{L} \to \infty \), our results are in good agreement with the relationship given by Bird et al. [39]. As one would expect, for FENE dumbbells the extension is significantly reduced, particularly when the extension is comparable to \( \hat{L} \).

4. Sedimentation of a single sphere

We now consider the motion of a single sphere sedimenting through a suspension of FENE dumbbells. The simulation is performed by integration as described in Eq. (8), and is fully three-dimensional and captures time-dependent effects. Under conditions in which dumbbells become highly
extended, the random-step term in Eq. (8) can lead to numerical instabilities if the time step $\Delta t$ is too large. The reason for this is that, when they are very extended, small changes in the length of the dumbbells leads to very large changes in the spring force. To maintain numerical stability, we use a two-time step method: the motion of beads attached to elongated springs is calculated with an interval ten times smaller than that used for the sphere and the other beads.

The initial state of the sphere in a spherical volume of dumbbells is shown in Fig. 7; the spherical volume shown has a radius of 10. As the sphere moves downward, the dumbbells elongate, particularly

Fig. 6. Average end-to-end distance versus dimensionless shear rate for $L^2 = 10, 100$ and infinity.

Fig. 7. Initial condition for a particle settling in a suspension of dumbbells. The lines represent dumbbells.
in the wake region, as shown in Fig. 8(a–d). In Figs. 8(a) and (b), only dumbbells with lengths greater than 1.5 are shown, and in Figs. 8(c) and (d) only those with lengths greater than three are shown. For all the simulations shown in Fig. 8(a–d), the maximum extension length $\hat{L}$ is 10 and the dimensionless concentration $\hat{c}$ is 0.015. At a relatively low De of 1.82, only a slight wake is visible, whereas at higher Deborah numbers of 2.39 and 4.18 there is an unmistakable region of concentrated, elongated dumbbells just downstream of the falling sphere. The orientation and extension of dumbbells in this region is in agreement with the theoretical predictions of Chilcott and Rallison [6], and similar orientational effects have been seen in optical birefringence measurements for flow past a cylinder [40]. Interestingly, at Deborah numbers greater than five the most concentrated region of dumbbells is not directly behind the sphere, but is somewhat to one side. A possible explanation for this is provided below in our discussion of a transition to a time-dependent settling velocity. First, however, we discuss the steady behavior.

We consider both the short- and long-time motion of the sedimenting sphere. Note, however, that the inertia of both the fluid and the spheres is neglected in Eq. (7), and therefore overshoot phenomena such as described by Rajogopalan et al. [41] are not seen in our simulations. A typical plot of velocity vs. position is given in Fig. 9 for the conditions $L = 10$, $c = 0.006$ and $De = 4.0$. It can be seen that the
sphere reaches its terminal velocity after falling approximately 30 sphere radii, a result that is in reasonable agreement with the distance of five times the De, $5\text{De}$, that was determined by Chilcott and Rallison [6]. To obtain additional quantitative data, we define a drag coefficient $K$ according to

$$K = \frac{U_0}{U},$$

(25)

where $U_0$ is the Stokes settling speed in a fluid with the zero-shear viscosity of the dumbbell suspension and $U$ is the averaged, terminal velocity of the sphere in our simulations. Plots of $U/U_0$ as a function of time for De ranging from 0.82 to 5.96 are shown in Fig. 10 for $\hat{L} = 10$ and $c = 0.006$. The enhanced drag caused by the presence of the dumbbells is evident, and that effect grows as the Deborah number increases. Standard deviations in these and other simulations in which the instability was absent are approximately 6%. The oscillatory behavior at the highest De is discussed further below.

It is illuminating to consider plots of the steady drag coefficient $K$ as a function of De, as such data is readily compared with the previous work. In Fig. 11 $K$ is plotted vs. De at $c = 0.006$ for maximum extension lengths $\hat{L}$ of 1.73, 3 and 10. For a value of $\hat{L}$ equal to 10, there is a slight reduction in the drag at De less than 2, followed by an increase so abrupt that $K$ is only slightly below 1.5 at a De of 2.2. The qualitative shape of the curves at $\hat{L} = 10$ and $\hat{L} = 3$ in Fig. 11 is very close to that proposed by Walters and Tanner [8], as mentioned in the Section 1. The reason for the increase in the drag can be seen in Fig. 8. Extended dumbbells in the wake of the sphere slow it down, just as described by Chilcott and Rallison [6]. One can appreciate this effect qualitatively by considering the velocity perturbation caused by an extended dumbbell in a Newtonian fluid at low Reynolds number. This effect is depicted in Fig. 12, where one sees that the beads move the fluid along the axis of the dumbbell toward the midpoint, at which point the fluid turns and moves radially outward in a direction orthogonal to the axis. The concentration of beads in the region directly behind the sphere therefore causes a strong velocity disturbance that slows the sphere down as required by the Faxén formula given in Eq. (4).
The results for values of $\hat{L}=1.73$ and $3$ in Fig. 11 show the dramatic effect that the maximum extension length has on the drag. For $\hat{L}$ equal to $3$, the behavior is qualitatively similar to that at $\hat{L}$ equal to $10$, but the drag enhancement is reduced significantly. However, when $\hat{L}$ is as low as $1.73$, the drag is reduced slightly instead of being enhanced, and the deviation from Newtonian behavior is very slight. The observation that the drag coefficient is decreased slightly at lower extension lengths and increased at higher ones was made previously by Chilcott and Rallison [6], although their drag increases were more modest than those shown in Fig. 11. We note also that we performed simulations in which we varied the relaxation time $\lambda$ and sphere velocity while keeping the Deborah number in Eq. (11) fixed. Varying the dimensional parameters by a factor of five yielded essentially identical results so long as

---

**Fig. 10.** Sedimentation rate $U/U_0$ versus dimensionless time for $\hat{L}=10$, $c=0.006$ and $\text{De}=0.82$, 1.36, 2.34 and 5.96.

**Fig. 11.** Drag coefficient $K$ vs. Deborah number for $c=0.006$ and $\hat{L}^2=3$, 9 and 100.
the Deborah number was unchanged. This was not true of the dimensionless concentration: variations in \( n \) and \( a_b \) in Eq. (10) such that the parameter \( c \) is fixed can yield small variations in the results for the drag coefficient \( K \). As stated above, all results here pertain to a bead size that is one-tenth of a sphere radius, \( a_b = 0.1 \ a_s \).

The fact that the drag coefficient of a sphere falling through a Boger fluid is not uniquely determined by the Deborah number has also been noted in previous experimental work. In particular, Solomon and Muller [9] discuss this issue at length. They plot data from the literature [42,43] showing that the drag behavior for spheres in polyisobutylene/polybutene and polyacrylamide/corn syrup Boger fluids differ qualitatively, with the former showing an abrupt increase in the drag and the latter a decrease at a Deborah number of approximately unity. They also investigated the drag behavior of three polystyrene-based Boger fluids with different levels of solvent quality. They found the low extensibility fluid showed only small deviations from Newtonian behavior at all Deborah numbers tested. In contrast, fluids with greater extensibility exhibited a drag enhancement of more than 300% for \( \text{De} > 1 \). These findings are qualitatively similar to our results shown in Fig. 11.

We now turn our attention to the oscillations apparent in the plot of \( U/U_0 \) vs. time at a \( \text{De} = 5.96 \) in Fig. 10. What we believe to be the key to understanding this time-dependent behavior can be found in Fig. 8(d), where the greatest concentration of extended dumbbells is shown to be offset to one side rather than directly behind the falling sphere. From videos we have made of the simulation, it appears that as the sphere falls at higher Deborah numbers its position in a cloud of extended dumbbells becomes unstable. The sphere then gets ‘popped’ out into a region where the dumbbells are less extended, where it begins to fall faster, generates a new cloud of extended dumbbells, and so on. It is quite rare to find the sphere in the center of a dumbbell cloud in a simulation, and the relative positions of the sphere and the dense region change continually. A relatively high drag coefficient and this asymmetric positioning of the most dense region of dumbbells invariably accompany the time-dependent oscillations, and they can always be made to disappear by lowering \( \text{De} \) or \( c \). As discussed in more detail below, including bead–bead interactions in the simulations raises the minimum values of \( \text{De} \) and \( c \) at which the oscillations occur, but not the occurrence itself.

In Fig. 13(a) we show the power spectrum of the velocity data plotted in Fig. 10. These spectra were obtained by using a Kaiser window and performing a Fast Fourier Transform. The presence of a dominant peak is first apparent at a \( \text{De} = 5.35 \) and occurs at a frequency \( f_0 \) of approximately 16. At a Deborah number of 8.26 the peak is larger and is shifted to a higher frequency. The trend towards
higher frequencies continues as De is increased to 9.83, at which point harmonics also appear at frequencies $f_1 = 2f_0$ and $f_2 = 3f_0$. The presence of these oscillations may be related to the elastic wake instability that was noted by Bisgaard [24] in his studies of flow around a sphere falling in a tube. In Fig. 13(b), the frequency and squared amplitude (as measured by the standard deviation of the sphere velocity) of the oscillations are plotted vs. De. Both curves are similar to the behavior described by McKinley et al. [28] in their Figs. 9–12. In particular, the squared amplitude grows linearly with De, as expected for a supercritical Hopf bifurcation [28].
Before proceeding to discuss simulations involving non-spherical particles and more than one sphere, we note the potential importance of dumbbell–dumbbell interactions in our one-sphere simulations. As discussed in more detail in Section 7, the concentration of dumbbells used in our one-sphere simulations was sufficiently high that the dumbbell–dumbbell interactions did seem to have a significant effect in test simulations. Including the dumbbell–dumbbell interactions in the manner described in Section 7 generally reduced the drag coefficient to a magnitude close to the Newtonian value. However, higher drag coefficients of 2–3, and also the time-periodic behavior, could always be recovered by increasing the dumbbell concentration significantly beyond the values used here. At lower dumbbell concentrations where dumbbell–dumbbell interactions are not important, the drag coefficients were always within 10% of unity, as was found by Chilcott and Rallison [6]. In the non-spherical particle and two-sphere simulations described in Sections 5 and 6, we always used low dumbbell concentrations such that the drag coefficient of a single particle at moderate De would be approximately unity. Our test simulations in which dumbbell–dumbbell interactions were included consistently showed that they are negligible under those conditions.

5. Sedimentation of a non-spherical particle

To explore the effect of non-spherical shape on particle motion, we have done simulations with an elongated particle formed by two spheres ‘glued’ together. Shown in Fig. 14 is such a non-spherical particle that is formed by applying equal and opposite ‘connecting’ forces \( F_c \) to each of the two spheres. These forces ensure that the relative velocity of the spheres along their line of centers is zero. The particle’s inclination angle with respect to the vertical direction is denoted by \( \theta \) so that an angle \( \theta \) of 90° describes a horizontal particle. The trajectories of the particle and the beads are calculated by integrating the following three equations for the sphere and bead velocities:

\[
(U_1 - U_2) \cdot \mathbf{e} = 0
\]  

(26)

![Fig. 14. Non-spherical particle formed by ‘gluing’ two spheres together.](image)
The forces $F_c$ are computed at each time step by using Eqs. (26) and (27). The vector $e$ in Eq. (26) is a unit vector along the line-of-centers of the two spheres.

In Fig. 15(a)–(c), the change in orientation of a non-spherical particle is shown as it sediments starting from an initial position in which its long axis is horizontal, or perpendicular to the direction of gravity. The maximum extension length $L$ is 10, the dimensionless concentration $c$ is 0.003, and the Deborah number $D_e$ is approximately 3. Only those dumbbells that are extended beyond a dimensionless length of 1.5 are shown. Starting from the horizontal orientation shown in Fig. 15(a), the particle rotates as it falls as shown in Fig. 15(b), until finally its long axis is parallel to the direction of motion, as shown in Fig. 15(c). As noted in the Section 1, this tendency for elasticity to cause elongated particles to orient themselves with their long axis in the direction of the motion has been studied previously [12,14,19,44], and it appears that the current method of simulation captures the qualitative behavior very nicely.

It is instructive to consider more detailed comparisons, particularly with the experiments of Chiba et al. [12], who examined slender rods with aspect ratios of approximately 60 and 120 falling through various fluids. Although the aspect ratio in the two-sphere particle used in the simulations is only 2, the qualitative behavior appears to be quite similar. In Fig. 16 we plot the inclination angle $\theta$ as a function of time as the particle falls, starting from its initial horizontal orientation (i.e. with $\theta = 90^\circ$). The initial rotation is slow, followed by a rapid rotation toward vertical, and the process ends by a second period of slow rotation until the particle is completely aligned with the direction of motion. This behavior corresponds closely to that shown in Fig. 12(a) of Chiba et al.’s paper, where the inclination angle is similarly plotted vs. time, in this case for rods with aspect ratios of 60 falling in polyacrylamide solutions with concentrations of 0.02, 0.05 and 0.1 wt.%. Particularly for the more elastic 0.1 wt.% solution, the behavior is very similar to what we show in Fig. 16. We note that Chiba et al. [12] found that long bodies do not always rotate to a completely vertical configuration: for certain conditions, they reach a steady angle $\theta_s$. They found that the angle $\theta_s$ decreases to zero as the level of elasticity or shear-thinning increases, and as the velocity of the body decreases. Our simulations, which correspond to a fluid with only elastic characteristics and motion at very low Reynolds number, always yield rotation to a completely vertical orientation, which is consistent with Chiba et al.’s observations.

In Figs. 17 and 18 we show two further comparisons with Chiba et al.’s experiments. In Fig. 17, the vertical distance of fall $z$ is plotted as a function of the horizontal distance travelled $y$. The particle’s center of mass has a curved trajectory with an inflection point, just as is shown in Fig. 10 of Chiba et al.’s paper. In Fig. 18, the horizontal velocity of the particle $U_y$ is plotted vs. the inclination angle $\theta$. The horizontal velocity is approximately zero at the horizontal and vertical orientations corresponding to angles of $90^\circ$ and $0^\circ$, and reaches its maximum value at approximately $\theta = 45^\circ$. This behavior is close to that shown in Fig. 11(a) in Chiba et al.’s paper, although the maximum horizontal velocity in their experiments was reached at inclination angles somewhat less than $45^\circ$, depending on the concentration of polyacrylamide.
Fig. 15. Dynamic simulation of a non-spherical particle falling in a dumbbell suspension for De ≈ 3, \( L = 10 \), \( c = 0.003 \) and \( t/\lambda \) = (a) 1.2, (b) 48 and (c) 96.
6. Sedimentation of two spherical particles

The simplest sedimentation problem involving more than one particle is that of two spheres interacting while falling parallel or perpendicular to their line-of-centers. Under low Reynolds number conditions in a Newtonian fluid, two such spheres maintain their initial configuration as a consequence...
of the reversibility of Stokes’ equations. As discussed by Joseph et al. [16], inertia causes two spheres in a horizontal configuration to move apart, while for the vertical configuration it causes what they describe as a ‘drafting, kissing and tumbling’ motion. Here we consider two-sphere interactions in a viscoelastic fluid where elastic effects dominate inertial effects, leading to qualitatively different behavior from what is seen in Newtonian fluids.

Like Feng et al. [22], we have performed both dynamic and static simulations. The dynamic simulations are accomplished by direct application of Eqs. (7) and (8) as described above, and correspond to two freely sedimenting spherical particles. For the static simulations, the two-sphere configuration is held fixed by applying equal-and-opposite forces $F_c$ to the spheres, as shown in Fig. 19, thus ensuring that the sphere velocities remain equal. At each time step, the following equations are used to compute $F_c$, $U_1$ and $U_2$ in a static simulation:

$$U_1 = U_2$$

$$\begin{bmatrix} U_1 \\ U_2 \end{bmatrix} = M_{UF}^{SS} \cdot \begin{bmatrix} F_g + F_c \\ F_g - F_c \end{bmatrix} + M_{UF}^{sb} \cdot F_b$$  (30)

and

$$U_b = M_{UF}^{bb} \cdot F_b + M_{UF}^{SS} \cdot \begin{bmatrix} F_g + F_c \\ F_g - F_c \end{bmatrix}.$$  (31)

The static runs were performed in a volume with a dimensionless radius of 10, while the dynamic simulations were performed in a volume with a radius of 20 to avoid effects due to the finite size of the dumbbell-filled region. The static results are presented as $F_{c,z}/F_g$ and $F_{c,y}/F_g$ for the vertical and
horizontal configurations, respectively. Positive values of the forces indicate an attraction between the particles.

As discussed in the Section 1, there is not yet complete agreement about the roles of elasticity and shear-thinning in the two-sphere problem. Joseph et al. [16] report that two spheres close enough together will form a doublet that falls vertically in a viscoelastic fluid. They state that more than one property affects the aggregation process: a high elastic stress ratio $N_1/\lambda$ was found to be sufficient but not necessary for aggregation, and the presence of shear-thinning plus memory was likewise deemed to be sufficient but not necessary for spheres to come together. In contrast, Gheissary and van den Brule [17] suggest that elastic forces drive particles apart in end-to-end settling, because spheres settling vertically in their Boger fluids moved apart, whereas in shear-thinning fluids they were able to observe aggregation.

The limited number of theoretical studies that have been performed for the two-sphere problem suggest that elasticity does promote aggregation. These studies include calculations with particles in second-order fluids [19,20], in which it is found that two particles always attract each other, in both horizontal and vertical configurations, ultimately leading to a doublet that falls vertically. Feng et al. [22] performed dynamic and static simulations of circular particles (or cylinders) moving in two dimensions. For the vertical configuration, their static runs showed an attraction for small initial separations $s_0 < 7$, a repulsion for intermediate separations $7 < s_0 < 10$, and no interaction for large separations $s_0 > 10$. In their dynamic simulations, the cylinders were attracted to each other and fell as a doublet for $s_0 < 7$, showed a transient repulsion but then an attraction leading to doublet formation for $s_0 = 8$, and moved to a steady separation of 9.85 when the initial separation was $s_0 = 10$.

Joseph et al. [16] conducted an extensive experimental investigation of two spheres settling starting from a side-by-side configuration. Their results show a somewhat complicated dependence on the spheres’ initial separation $s_0$, leading them to define two distance parameters $\delta_c$ and $\delta_t$, where $\delta_t < \delta_c$. When the separation $s_0$ is large, $s_0 > \delta_c$, the two spheres do not interact. For intermediate separations, $\delta_t < s_0 < \delta_c$, the spheres interact and initially show an attraction, with the line-of-centers turning from horizontal. However, the spheres eventually separate or stop interacting, and do not chain. For small separations, $s_0 < \delta_t$, the falling spheres attract, with the line-of-centers turning as they interact such that they ultimately form a two-sphere doublet that falls vertically downward. The two-dimensional
dynamic simulations of Feng et al. [22] correctly reproduced the qualitative features of this doublet formation for circular particles moving in an Oldroyd-B fluid.

6.1. Two spheres sedimenting end-to-end

Results for our static simulations with the spheres oriented vertically are shown in Fig. 20. These simulations were performed at De \( \approx 3 \), \( \hat{L} = 10 \) and \( c = 0.003 \). The results indicate a weak repulsion for small separations, \( s_0 < 4 \), and a weak attraction for larger separations, where \( s_0 > 4 \). There is no evidence of any transition from attraction to repulsion at large separation, such as was observed by Riddle et al. [15]. However, it has been suggested that the separation seen by Riddle et al. may be connected with a negative wake, and negative wakes appear to be closely associated with shear-thinning [45], which is absent in our simulations.

Characteristic results from our dynamic simulations are shown in Figs. 21 and 22, and are consistent with the static results. These calculations correspond to initial separations of \( s_0 = 12 \) and \( s_0 = 2.1 \), respectively, under conditions where De \( \approx 3 \), \( \hat{L} = 10 \) and \( c = 0.003 \). The quantities plotted are \( \Delta x \), \( \Delta y \) and \( \Delta z \), or the separation distance between the spheres in the \( x \), \( y \) and \( z \) directions, respectively. From Fig. 21, one sees that the attraction is effective over large distances, bringing together spheres that are separated by as much as 12 radii. We performed many simulations of this type, with initial separations spanning the range from 2 to 20, and in general the results were quite similar. We found that the \( \Delta x \) and \( \Delta y \) values fluctuated around zero, while \( \Delta z \) fell gradually to a distance of approximately 4. In Fig. 22, the values of \( \Delta x \), \( \Delta y \) and \( \Delta z \) are plotted vs. time for an initial separation of 2.1. The particles move apart to a vertical separation of approximately 4, the same final vertical separation that is attained starting from large initial separations. Multiple simulations with initial separations between 2 and 20 all
consistently moved to this configuration, and in almost every case $\Delta x$ and $\Delta y$ were much smaller than $\Delta z$. Hence, our overall result can be summarized by saying that two spheres settling along their line-of-centers fall as a doublet with a small but persistent gap between them. For the conditions of the simulations reported here, if the initial separation was less than 4, then the particles separated slightly,
and if the initial gap was larger than 4, even as large as 20, then the particles moved together until the surface-to-surface gap between them was 2.

Although the fact that the spheres move together is partially in agreement with the experimental observations of Joseph et al. [16], the reason why the spheres move apart when their initial separation is very small is not clear. One possible explanation is that extended dumbbells are excluded from the region between the spheres at small separations, making it impossible to capture strong elastic stresses in the gap region without shorter dumbbells. This problem could potentially be alleviated by doing simulations with a distribution of dumbbell lengths; indeed, we have done simulations with dumbbells having maximum extension lengths of 3 (as opposed to 10), and found that the eventual, steady-state sphere–sphere separation was smaller for the shorter dumbbells. However, we also note recent experimental work by Bot et al. [46] showing that, in a polyacrylamide-based Boger fluid, two vertically oriented, sedimenting spheres will separate when starting close together and move together when starting far apart, in either case ending up in the same, stable arrangement. These observations are entirely consistent with our numerical results.

6.2. Two spheres sedimenting side-by-side

We now consider two spheres whose initial configuration is such that their line-of-centers is perpendicular to gravity. Results from our static simulations are shown in Fig. 23, where it can be seen that the two particles attract each other over the entire range of separations where they interact. These simulations were performed under the conditions $\text{De} \approx 3$, $\hat{L} = 10$ and $c = 0.003$. The attraction is quite strong for $s_0 < 4$, and its strength increases as the separation distance decreases.

![Fig. 23. Static results for the horizontal two-sphere configuration for $\text{De} \approx 3$, $\hat{L} = 10$, and $c = 0.003$.](image)
Results for dynamic simulations at the same values of \( \text{De}, \hat{L}, \) and \( c \), and for initial separations of 3, 8 and 12, are shown in Figs. 24–26. In Fig. 24, one sees that for \( s_0 = 3 \) the spheres attract and touch (i.e. \( \Delta y \) quickly falls to 2), and then the doublet rotates until the line-of-centers is approximately vertical.

![Fig. 24. Dynamic simulation of two spheres falling side-by-side for \( s_0 = 3 \), \( \text{De} \approx 3 \), \( \hat{L} = 10 \), and \( c = 0.003 \).](image1)

![Fig. 25. Dynamic simulation of two spheres falling side-by-side for \( s_0 = 8 \), \( \text{De} \approx 3 \), \( \hat{L} = 10 \), and \( c = 0.003 \).](image2)
The final vertical separation is 4, as expected from the results in Figs. 21 and 22. For the larger initial separation of $s_0 = 8$ shown in Fig. 25, the behavior is similar, except that the sphere attraction and the turning of the line-of-centers occurs simultaneously instead of sequentially. At this separation we have done simulations where, after turning, the spheres fall as a vertical doublet, but there have also been simulations where the spheres separate as the line-of-centers is turning toward vertical. Neither behavior occurs exclusively. Finally, our results at the large separation $s_0 = 12$ in Fig. 26 show negligible sphere–sphere attraction, and the initial configuration is retained except for some dispersion in the $\Delta z$ separation. All of these results from our dynamic simulations agree qualitatively with the two-dimensional results of Feng et al. [22].

Because our simulations include rotation–translation coupling, we can also calculate rotational velocities during the sedimentation process. In Fig. 27, it is shown that the spheres maintain the direction of rotation that is present when two spheres sediment side-by-side in a Newtonian fluid. The magnitude of the rotation gradually decreases as the spheres’ line-of-centers becomes vertical, and the spheres stop rotating at a time $t/\lambda \approx 80$. These features agree with the results of Joseph et al. [16] and Feng et al. [22]. The issue of rotation is of particular interest because it has been found experimentally that, for a particle settling near a wall, the direction of rotation in a viscoelastic fluid is opposite to what it is in a Newtonian fluid [47]. Becker et al. [48] show that the retarded motion expansion to $O(De^2)$ does not reproduce this change in rotation, nor does it show a particle–wall attraction, as is found experimentally. Similarly, Feng et al. [22] found that a particle falling near a single wall (or in a very wide channel) is repelled by the wall, not attracted to it. The latter calculation was performed with the Oldroyd-B constitutive equation. By extending the current method to include walls, we have reproduced both the ‘anomalous rotation’ and the particle–wall attraction [49]. Hence, our direct simulation method does appear to yield qualitatively correct results for the effect of
viscoelasticity on rotation as well as translation of sedimenting particles, both in pure fluids and near a single wall.

7. Discussion

7.1. Qualitative interpretation of the simulation results

In addition to being a useful method for calculating particle trajectories, the method of simulation developed here provides interesting qualitative insights into the reasons the particles behave the way they do. Such insight is obtained by considering how the particle motion affects the location, orientation and stretching of the dumbbells, and in turn how the dumbbells reflect back and influence the particle. In this way one can extrapolate one’s intuition from the relatively well-understood problems of low-Reynolds-number hydrodynamics to corresponding non-Newtonian problems. Such an extrapolation is by no means trivial, but it does at least provide a starting point for conceptualizing interparticle interactions in a new and potentially powerful way.

In the case of a single sphere, the dumbbells are nearly horizontal in the region immediately upstream of the sphere, and they are highly extended and oriented vertically in the downstream region. Similar orientational distributions were noted by Claey and Brady [50] in simulations of a sphere sedimenting in a suspension of force-free rods. As noted by Chilcott and Rallison [6], and as shown in Fig. 8, the downstream region extends many radii away from the sphere. The situation is represented schematically in Fig. 28. The main contribution to the drag increase experienced by the sphere comes...
from the vertical dumbbells in the wake region, which has been identified previously as consisting of ‘birefringent strands’ [10]. The arrows in Fig. 28 represent fluid being pulled upward by the dumbbells in the wake, and it is this pulling action that increases the drag on the sphere.

A schematic illustration similar to that in Fig. 28, but for a non-spherical particle, is shown in Fig. 29. Like in the Newtonian case, as the elongated particle falls it also moves horizontally (to the left when it is oriented as shown in Fig. 29). By dividing the space around the particle into Regions 1–3 as shown, and calculating the contributions of each region to the net turning couple on the body, we have
found that dumbbells in Regions 1 and 2 tend to turn the particle to its vertical orientation, while those in Region 3 have the opposite effect. Region 1 has the most important contribution, and it is also the region where the dumbbells are most elongated.

The turning couple responsible for the particle’s rotation is zero when the angle $\theta$ is 0 or 90°. When the particle is vertical (i.e. $\theta = 0$), the arrows representing how the fluid is affected by the dumbbells are parallel to the long axis of the particle, as shown in Fig. 30. Hence, the oriented and extended dumbbells downstream have negligible contributions to a turning couple. When the body is horizontal (i.e. $\theta = 90^\circ$), the downstream dumbbells contribute significant turning couples (cf. Fig. 31); however, the dumbbells just downstream of one of the spheres contributes a couple equal in magnitude and opposite in direction to the dumbbells downstream of the other sphere. Hence, the combined effect on the overall turning couple is negligible. These observations can be illustrated nicely by a static simulation like those of Section 6, where in this case we hold the non-spherical particle at a fixed orientation by application of a force $F_{c,n}$ in a direction normal to the line-of-centers to each of the two spheres (in opposite directions), cancelling any couple induced by the dumbbells. The results of such a simulation for which $c = 0.003$, $\hat{L} = 10$ and $De \approx 3$ are shown in Fig. 32, where negative values of $F_{c,n}$ indicate that there is a turning couple. As observed above in Fig. 14, the tendency to rotate is strongest at $\theta \approx 45^\circ$, and is absent at $\theta = 0$ and 90°.

For the case where two spheres are sedimenting freely and are not joined together to form a single particle, the region between the spheres plays a critical role. In Fig. 33, we have again divided the space around the spheres into three regions. Region 1, which is in the wake of the upper sphere, is the location of the most extended and oriented dumbbells. These dumbbells tend to cause the spheres to separate, because they slow the top sphere down more than the bottom sphere. The dumbbells in Region 2 between the particles tend to move fluid out of the gap between the spheres, hence drawing them together and opposing the separation induced by Region 1. The horizontal dumbbells in Region 3 are less extended than those in the other two regions. Region 3 causes the bottom sphere to slow down...
more than the top sphere, and therefore helps to bring the spheres together like Region 2. Overall, our simulations show that when the spheres are separated by a center-to-center distance greater than about 4, Regions 2 and 3 are strong enough to cause them to move together. As the distance between them gets smaller, Region 2 in particular has a weaker influence, and there is an approximate balance between the influences of the three regions at the separation of 4. As mentioned previously, the value of

Fig. 31. Non-spherical particle falling with its long-axis horizontal.

Fig. 32. $F_{c,n}$ vs. inclination angle for De ≈ 3, $\hat{L} = 10$, and $c = 0.003$. 
the final separation depends on the characteristics of the dumbbells, particularly the maximum extension length.

The contributions from these three regions makes the case of end-to-end settling significantly more complicated than side-by-side settling. The latter configuration is shown in Fig. 34, where we also
define Regions 1 and 2. In this side-by-side configuration, it is Region 1 that contributes nearly all of
the non-Newtonian interaction. The flow in the region between the two spheres orients and extends
the dumbbells, which are left in the wake region as the larger spheres sediment. The extended
dumbbells in Region 1 tend to pull fluid from between the spheres as shown, hence drawing the spheres
together.

7.2. Importance of dumbbell–dumbbell interactions

We have done several simulations in an attempt to assess the importance of dumbbell–dumbbell
interactions on our results. Because it is not practical to calculate the interactions between all the
dumbbells, these test simulations were performed by calculating interactions between dumbbells within
a specified distance of one another, that distance generally being \(2a_s\); furthermore, only dumbbells
extended beyond a specified length, usually \(2a_s\), were considered. In nearly all the cases, the results of
these simulations agreed qualitatively with results obtained in the complete absence of dumbbell–
dumbbell interactions. The one case where significant differences were observed is the one-sphere drag
coefficient calculations discussed above, in which it was found that the drag becomes significantly
closer to the Newtonian value when dumbbell–dumbbell interactions are included. However, even with
dumbbell–dumbbell interactions, it is still possible to obtain high drag coefficients and the transition to
time-periodic behavior if the dumbbell concentration is increased by a sufficient amount.

In addition to doing particle simulations with and without dumbbell–dumbbell interactions, we have
calculated the velocity profiles with and without dumbbell contributions and compared contour plots.
The reason such a comparison is relevant is that, without dumbbell–dumbbell interactions, each
dumbbell is responding to the velocity field contributed by the spheres as if no other dumbbells were
present. Hence, if the dumbbells collectively cause a significant modification of the overall velocity
field, then not accounting for dumbbell–dumbbell interactions results in dumbbells that are deforming
under the influence of an inappropriate velocity field. In all of the non-spherical particle and two-sphere
simulations, the velocity contours with and without dumbbell contributions look very similar. This
finding is consistent with finite element calculations in other geometries that show that velocity fields
in Boger fluids often exhibit relatively small deviations from Newtonian behavior [51]. In contrast, the
one-sphere simulations in which the drag coefficient differed significantly from unity did show
deviations from the Newtonian velocity profile, confirming the potential importance of dumbbell–
dumbbell interactions in that geometry. In the non-spherical particle and two-sphere simulations, the
conditions used were always such that the dumbbell concentration was low and dumbbell–dumbbell
interactions negligible.

Leal [21] defines a ‘weak’ viscoelastic effect as one which causes a small deviation from Newtonian
behavior at any instant in time, but leads to a significant accumulative change in particle orientation or
distribution. In contrast, ‘strong’ viscoelastic effects exhibit large deviation from Newtonian behavior
instantaneously. In the absence of dumbbell–dumbbell interactions, our simulations show that ‘weak’
effects (i.e. re-orientation of a non-spherical particle or redistribution of two sedimenting spheres) can
be calculated very successfully by the method proposed here. The drag coefficient calculations (i.e. a
‘strong’ effect) show remarkably good qualitative agreement with experimental results, in terms of the
predicted dependence of the drag coefficient on both the Deborah number and polymer extensibility,
but the possibility that neglecting dumbbell–dumbbell interactions affected those results cannot be
ruled out.
8. Conclusion

We have presented a new method for simulating the motion of particles in viscoelastic fluids. We have shown that the method predicts the correct qualitative behavior for several problems involving one and two spherical particles and one non-spherical particle. We find the drag on a single particle falling in a Boger fluid at moderate Deborah numbers to be enhanced significantly relative to what it would be in a Newtonian fluid with the same viscosity; in addition, the increase in drag is strongly dependent on the extensibility of the dumbbells, as has been shown experimentally. However, those findings must be interpreted in light of the possible effects of having neglected dumbbell–dumbbell interactions, as discussed above. We also find that a single non-spherical particle tends to rotate as it sediments so that its long axis lies in the direction of gravity, and that the two sedimenting spheres generally tend to move together and fall end-to-end rather than side-by-side. In addition to predicting the correct behavior in these test cases, the method allows a direct examination of what is happening in the flow to lead to such dramatic differences between particles sedimenting in Newtonian and non-Newtonian fluids. We have found the method to be similarly capable of predicting particle–wall interactions [49], providing comparable insight into those problems.

Rather than accounting for viscoelasticity in the fluid by using a non-Newtonian constitutive model in conjunction with the laws of continuum mechanics, in this method it is accounted for by the presence of many non-linear bead-and-spring dumbbells suspended in a Newtonian solvent. The calculation of interactions between spheres and thousands of dumbbells may at first seem computationally costly. However, the ability to calculate those interactions by using the linear relations of low-Reynolds-number hydrodynamics mitigates that cost significantly, making the method potentially very useful for N-particle simulations that are not feasible by other approaches. We would add that modifications such as using a distribution of spring force constants, yielding a fluid with a spectrum of relaxation times, are trivially easy to implement with this method. Hence, the prospect of calculating weak viscoelastic interactions in N-particle simulations in a fluid with a multiple relaxation times is feasible with this approach, even in the presence of one or two walls.

Acknowledgements

This work was supported by grant CTS-94-00737 from the National Science Foundation.

References


