Dynamics of Self-Assembled Chaining in Magnetorheological Fluids

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The aggregation dynamics of paramagnetic spherical particles embedded in a viscous fluid is investigated via numerical simulations using a fully coupled three-dimensional model. Particles experience simultaneously Brownian motion, dipolar magnetic attraction, and multibody hydrodynamic interactions. When the dipole strength characterizing the ratio of magnetic attraction to random diffusion exceeds a critical value, particles join together forming supraparticle structures. As time evolves, particle/chain and chain/chain interactions lead to a continuous increase of the cluster size. The mean length of particle chains has a power-law dependence with respect to time, as predicted by the theory of diffusion-limited aggregation. Both the exponent and the characteristic time scale agree very well with the experimental results of Promislow et al. (J. Chem. Phys. 1995, 102, 5492).

1. Introduction

Fluids with tunable properties subject to electromagnetic external fields have different regimes. Ferrofluids are composed of small individual grains (size ~ 10 nm in stable Brownian suspensions) and exhibit strong hysteresis. When the field is removed, the particles keep their own irreversible magnetization (an overview on ferrofluids can be found in work by Rosenweig1,2 and on the magnetic properties of materials in work by Tabor3). In contrast, paramagnetic beads (size 1 µm or less) that can be embedded in fluids have nearly instantaneous reversible properties with minimal hysteresis (see for example the test on microspheres provided by Bang Laboratories4). These fluids behave usually as complex fluids when self-assembled structures are formed in the flow. Viscous fluids seeded either with colloidal or with noncolloidal particles are called electromechanical (ER) or magnetorheological (MR) fluids when macroscopic properties of the suspension can be modified under the influence of an external electric or magnetic field, respectively. When a constant field is applied, particles acquire an electric or magnetic dipole that tends to orient the particle mutual interactions. The relative motion of the particles leads to head-to-tail aggregation and chainlike structures are formed. In microfluidic applications, we can take advantage of the electrically or magnetically tunable property of the bulk viscosity. More importantly, ER and MR fluids can dynamically change their optical properties, anisotropy, mechanical rigidity, or electronic properties, often in a reversible way.

The applications are many, but the recent ones concern the development of DNA separation chips using self-assembled magnetic matrices.5 Also, optical properties related to chain orientation are easy to modify by an external field. Manipulating the chains appropriately could potentially lead to new novel applications such as micro-optical filters and gratings.6 More classical applications involve optimal control of dampers and hydraulic brakes, which can be achieved when an accurate model of the aggregation rate is known. In these applications, the concentration of magnetized particles is large and the fluid nearly solidifies when aggregation sets in.

In the past two decades, considerable interest in ER suspensions focused on theoretical predictions and experimental investigation of the aggregation rate that controls the apparent viscosity of the mixture (see Parthasarathy and Klingenberg8 or Gast and Zukoski9 for review articles). More recently, MR fluids have received considerably more attention as some practical problems associated with ER fluids are less pronounced in MR fluids. There are several complicating factors (e.g., surface charge, electrode polarization, etc.; see Promislow et al.10 for a brief discussion on limitations of ER fluids), which have limited the range of their application in microfluidics research today. However, the overall behavior of an MR suspension under the influence of a constant magnetic field is similar to that of an ER suspension under the influence of an electric field, and most of the concepts and fundamental ideas developed in ER research can be applied to MR suspensions.

When noncolloidal particles are dispersed in the suspension, attraction and aggregation in chains are expected for any magnitude of the magnetic dipole. Most applications deal with micrometer-sized or smaller particles. Therefore, aggregation can be hindered by Brownian diffusion as particles experience a random walk related to small-scale thermal fluctuations. A competition between dipolar attraction and diffusion that tends to homogenize the suspension leads to complex aggregation kinetics.

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When the external field is removed, reversible properties are encountered with paramagnetic beads. Losing immediately the magnetic dipole, particles involved in chain structures diffuse away in the bulk; the uniform initial suspension is then recovered.

The modeling of processes leading to chain formation is of fundamental significance for the determination of the aggregation kinetics. Theoretical models based on the Smoluchowski\textsuperscript{11} equation have been tested both in experiments and in simplified numerical simulations.\textsuperscript{12} Most experimental studies are closely related to measurements of the apparent viscosity of the mixture under specific conditions. However, Promislow et al.\textsuperscript{10} have investigated the influence of the mean cluster size. When the suspension is confined and the radius of the mean length of chains ($\langle S(t) \rangle$) is expected. The influence of particle concentration and applied field strength was systematically investigated, and good agreement with diffusion-limited predictions was achieved. In particular, Promislow et al.\textsuperscript{10} proposed a relevant characteristic time to scale the temporal evolution of the mean cluster size. When the suspension is confined by two flat plates perpendicular to the external field, the chain's length is limited and the radius of the aggregates depends on the experimental conditions.\textsuperscript{14} At high concentration, the chains tend to coarsen and the suspension loses the anisotropy.\textsuperscript{15}

In addition to the experimental work, numerical models have been proposed with different approximations of the exact physical mechanisms leading to simplified simulations. The complexity in MR fluids arises from the coupling of hydrodynamic and magnetic interactions during the Lagrangian motion of the particles. Klingenberg et al.\textsuperscript{16} and Haas\textsuperscript{17} provided the first simulations and used a dipolar representation of the magnetic interaction. The hydrodynamics, however, was crudely modeled by the Stokes drag law applying on each single particle separately, thus neglecting the multibody Stokesian interactions. They succeeded to reproduce the fibrous structures observed experimentally in two- and three-dimensional configurations (see Martin\textsuperscript{18} for simulations predicting the apparent viscosity of a sheared ER suspension). Bonnecaze and Brady\textsuperscript{19} reported a more detailed representation of hydrodynamic interactions, employing Stokesian dynamics for ER suspensions using a low-order multipole method for the multibody interaction. This technique has valuable properties when small-scale phenomena are investigated, but it is computationally more expensive when a large number of particles are involved, thus restricting the simulation to few particles. More recently, Ly et al.\textsuperscript{20} have adapted a fast multipole algorithm to deal with particle dynamics in MR fluids. However, the Stokes drag law was used again although the magnetic force was accurately represented by multipole expansions of the steady Maxwell equations. Ly et al.\textsuperscript{20} neglected the Brownian diffusion and their simulation was only in two-dimensional domains; however they obtained valuable results of the microstructure evolution.

In this paper, we present the first numerical model that takes into account all the important physical mechanisms encountered in a three-dimensional suspension of paramagnetic particles. Specifically, Stokesian hydrodynamic interactions, magnetic attraction, and Brownian diffusion are all modeled. On the basis of appropriate approximations we are able to obtain numerical results on the aggregation dynamics in MR fluids, which agree quantitatively with the experiments of Promislow et al.\textsuperscript{10} This paper is organized as follows. The next section will be devoted to a detailed presentation of physical issues for modeling. Then, results on the time evolution of the mean cluster size will be analyzed. Open issues and further developments of the model are discussed in the conclusion section.

2. Modeling of Particle Interactions

In the experiments of Promislow et al.,\textsuperscript{10} paramagnetic beads were uniformly sized with a mean radius $a$ of 0.3 μm; specifically, polystyrene beads were seeded with iron oxide (Fe$_3$O$_4$). As particles are embedded in aqueous solutions, the Reynolds and the Peclet numbers are quite small and thus the Stokes approximation for the flow motion is reasonable. The relative magnitude of sedimentation and Brownian diffusion defines the Peclet number $Pe = V_{sed}aD$, where $V_{sed}$ is the sedimentation velocity and $D$ the diffusion coefficient. In the experiments, $Pe = 0.01$ and thus buoyancy effects are not important.

2.1. Magnetic Attraction

Paramagnetic beads of radius $a$ placed in a uniform magnetic field acquire a dipole moment $\mathbf{m}$ aligned with the field given by

$$\mathbf{m} = \frac{4}{3} \pi a^3 \chi \frac{\mathbf{B}_0}{\mu_0}$$

where $\chi$ is the magnetic susceptibility, $\mu_0$ the magnetic permeability of a vacuum (nonmagnetic carrying fluid), and $\mathbf{B}_0$ is the external magnetic flux density. The uniform magnetic field produces no force on the particles; only the dipoles create a gradient of magnetic flux density $\mathbf{B}$ needed to generate a net force. For an isolated sphere, $\mathbf{B}$ is computed from

$$\mathbf{B} = \frac{\mu_0}{4\pi} \frac{3(\mathbf{m} \cdot \mathbf{r}) \mathbf{r} - \mathbf{m}}{r^5} + \mathbf{B}_0$$

where $\mathbf{r}$ is the vector originating from the center of the particle (see Figure 1); SI units are used in these equations as in the reference book of J. Jackson.\textsuperscript{21} The description of

\begin{figure}
\centering
\includegraphics[width=\textwidth]{Figure1.png}
\caption{Sketch and definition of geometric parameters: particle radius, $a$; $\mu_0$, magnetic permeability of the paramagnetic beads.}
\end{figure}

\begin{thebibliography}{1}
magnetic interactions in terms of simple magnetic dipoles captures the primary dynamics, even though a more detailed multipole representation may be warranted to resolve very localized variations in the magnetic field. Considering only pairwise interaction between particles, we obtain the following expression for the force acting on a magnetized particle

$$F_M = -\nabla U \quad \text{with} \quad U = -m \cdot B$$  \hspace{1cm} (3)$$

where $U$ is a dipolar potential. Dipoles will attract each other and aggregate head-to-tail. The effects of multibody and multipoles are secondary in the kinetics of attraction; see Lye et al.\(^\text{(20)}\) for a fast multipole algorithm dedicated to the determination of effective magnetic permeability of suspensions. Furst and Gast\(^\text{(22,23)}\) have demonstrated that mutual induction is important for the determination of the rupture threshold of chains in experiments investigating the microhydrology of MR fluids. Since our aim here is to examine the growth rate of supraparticle structures, we do not include these refinements. The attraction force $F_M$ is quite short range compared to the Stokes multibody interaction as it falls off as $r^{-3}$. At large separation the force will be below the level of Brownian agitation due to thermal fluctuations. As the particle disperses due to random walk or as longer chains form, the particles will move close enough and thus the magnetic force will become dominant.

### 2.2. Hydrodynamic Interaction

Hydrodynamic interactions in Stokes flows are particularly long range as the disturbance velocity decreases slowly like $r^{-1}$ in contrast to magnetic interaction forces. Multibody hydrodynamic interactions are important, and Stokes drag on a single sphere is a poor representation of these mechanisms.\(^\text{16,17,20}\) In the current work we have employed a low-order multipole representation of the disturbance induced by each particle. The forces acting on the spherical beads drive the flow via a Gaussian representation of the force monopole. This model is called the force coupling method (FCM) and has been extensively described in Maxey and Patel,\(^\text{(24)}\) Lomholt et al.,\(^\text{(25)}\) and Lomholt and Maxey.\(^\text{(26)}\) We will give only the basic features of this method adapted to the present configuration. The incompressible Stokes equations (eqs 4 and 5) are solved numerically by time marching. Specifically, we solve the equations in a three-dimensional domain

$$\nabla \cdot \mathbf{u} = 0$$  \hspace{1cm} (4)$$

$$0 = -\nabla p + \eta \nabla^2 \mathbf{u} + \mathbf{f}(x,t)$$  \hspace{1cm} (5)$$

where $\mathbf{f}(x,t)$ in eqs 5 and 6 denotes the force distribution experienced by the fluid (viscosity $\eta$) in response to the N particles centered at $Y_i(t)$. The term $\mathbf{F}_i$ involved in the forcing of the Stokes equations is related to the forces acting on the $i$th particle by eq 9. Also, $\Delta(x)$ in eq 7 is a spherical Gaussian envelope of finite width $\sigma$, i.e.

$$\Delta(x) = (2\pi\sigma^2)^{-3/2}e^{-(x^2/2\sigma^2)} \quad \text{with} \quad r = |x|$$  \hspace{1cm} (7)$$

The relation between $a$ and $\alpha (\alpha = a/(\sqrt{2}))$ has been derived analytically\(^\text{24}\) to give the exact Stokes velocity on a single sphere in an unbounded fluid. The velocity is determined by a convolution of the simultaneous disturbances of all the particles, i.e.

$$\mathbf{V}_i(t) = \int \int \mathbf{u}(x,t)\Delta(x - Y_i(t)) \, d^3x$$  \hspace{1cm} (8)$$

The force balance for a particle of mass $m_p$ is then

$$\dot{m}_p - m_p \frac{d\mathbf{V}_i}{dt} + \mathbf{F}_i = \mathbf{F}_{M,i} + \mathbf{F}_{B,i}$$  \hspace{1cm} (9)$$

The first term on the left-hand side is related to the excess of inertia of particles compared to the equivalent fluid mass $m_p$. It can be safely neglected, as particle and fluid density are typically close. $\mathbf{F}_{M,i}$ is the magnetic force acting on the particle due to all the other magnetized particles; pairwise interactions are summed over all the particles of the suspension. $\mathbf{F}_{B,i}$ is a random forcing that models thermal agitation. Notethat buoyancy has been neglected here since the Peclet number is small. The mean drift of the particles induced by buoyancy is very weak compared to thermally induced fluctuations. Therefore, the sedimentation can be neglected in the solution of the relative motion of the particles.

Various validations of FCM have been presented in distinct configurations; see Lomholt et al.\(^\text{(25)}\) and Lomholt and Maxey\(^\text{(26)}\) for a detailed experimental validation of the motion of two or three spherical particles. Sedimentation of a large number of particles has been successfully compared to experiments.\(^\text{(27)}\) Thus, FCM is a suitable model for microflows as demonstrated by Liu et al.\(^\text{(28)}\) Accuracy of the flow representation can easily be improved in the FCM model by using force dipole, lubrication model, and inertia effects, but the configurations we model here do not require these refinements because the volume fraction of the particles in our simulations is very small ($c < 0.03$).

### 2.3. Brownian Diffusion

Brownian motion and the effects of thermal fluctuations become an increasingly important feature for submicrometer sized particles. The dynamics of Brownian suspensions has been reviewed by Russell et al.\(^\text{(29)}\) and simulation results reported by Ladd et al.\(^\text{(30)}\) Phung et al.\(^\text{(31)}\) and Foss and Brady.\(^\text{(32)}\) The standard description is based on the Langevin model for Brownian motion, where the particles are subject to a random white-noise forcing from the thermal fluctuations. This yields the classic Stokes–Einstein result for the diffusivity $D$ of a single spherical particle that is independent of the mass of the particle, i.e.

$$D = k_B T / 6 \pi \eta a$$  \hspace{1cm} (10)$$

where $k_B$ denotes Boltzmann’s constant and $T$ the absolute temperature. In a dilute system the particles can be considered in isolation. However, there are long-range hydrodynamic interactions between particles in a suspens-
sion under low Reynolds number conditions. These modify the mobility of a system of particles and hence the diffusivities in response to thermal fluctuations. The resistance tensor is determined by the instantaneous configuration and must be recalculated as the particles move. The repeated computations of the resistance or mobility tensors can become lengthy as the system of particles becomes larger. Therefore, we keep a simple representation of the Brownian motion using a fluctuating force $F_{Bi}$ with zero mean and without correlation in time nor with the other particles. Specifically, we have

$$F_{Bi} = \xi \left( 12 \pi \eta a kT / dt \right)^{1/2}$$  \hspace{1cm} (11)

where $\xi$ is the time step selected for the numerical integration of the Lagrangian tracking of the particles and $\xi$ is a random vector with a Gaussian distribution. Such a simple representation may limit the accuracy of predicting thermally induced diffusion, especially at later times. The fluctuating forces may be correlated when the separation distance between the particles is small. Therefore, our model will be a reasonable approximation when the concentration of the suspension is low. Also, when chains are formed, thermal agitation of the chains may be poorly represented by uncorrelated forcing of the particles. In the current study, we focus primarily on the initial aggregation kinetics and these issues should be secondary.

Depending on the ratio $\lambda$ of the thermally induced agitation and the attraction potential $U$, particles can be evenly dispersed throughout the suspension or form supraparticle structures such as chains. This ratio is given by

$$\lambda = \lambda_{\text{max}} = \frac{9\pi a^3}{kT} \beta_o$$  \hspace{1cm} (12)

The onset of aggregation is expected to be in the range $\lambda > 1$, i.e., when attraction overcomes the Brownian random walk.

Due to magnetic attraction, particle clusters will be formed. As our model does not provide a real physical interface between the interior of the particles and the carrying fluid, we have to prevent overlapping of spheres by a repulsion barrier. It is activated only when particles are closer than a selected cutoff length, $R_{\text{ref}}$, of the order of 20% of the radius $a$. Then, particles are kept apart from each other by the addition of a repulsive velocity $V_{ij}$ given by

$$V_{ij} = -\frac{V_{\text{ref}}}{2a} \left[ \frac{R_{\text{ref}}^2 - r^2}{R_{\text{ref}}^2 - 4a^2} \right] x_{ij}$$  \hspace{1cm} (13)

where $x_{ij}$ is a vector connecting the centers of particles $i$ and $j$ ($r = |x_{ij}|$). Calibration of the relative motion of two or three spheres provides the determination of the velocity $V_{\text{ref}}$. We have checked systematically the stability of long chains, and more rigorous comparisons of different repulsion models have been presented in Dance et al.33

No other particle interactions are included in our model. In experiments, a surfactant layer prevents aggregation due to van der Waals forces and aqueous solution at neutral pH induces very weak electrostatic repulsion.

The numerical solution of the fully coupled fluid/particles suspension was obtained via direct simulations of the fluid flow using fast Fourier transforms in a periodic box. A second-order Adams–Bashforth scheme was used for the Lagrangian tracking of the particles.

3. Dynamics of Particle Chaining

Initially, the particles were seeded in a periodic box of width $L/a = 48$. The resolution was $128^3$ grid nodes for the solution of the fluid flow equations. Under the influence of hydrodynamic and magnetic forces, the particles tend to aggregate in linear clusters. An averaging of 10 initial random seedings was performed in order to evaluate the temporal growth of the mean cluster size. We used a simple and efficient algorithm of cluster detection proposed by Sevick et al.34 From the connectivity matrix, all the pairs of touching spheres were detected. We can determine the indirectly connecting pairs to provide a cluster size distribution $n(s)$, where $n(s)$ denotes the number of clusters of sizes in the suspension at the sampled time. Then, the mean cluster length $\langle S(t) \rangle$ is defined by

$$\langle S(t) \rangle = \frac{\sum_s s^2 n(s)}{\sum_s s n(s)}$$  \hspace{1cm} (14)

This quantity has been normalized with the particle diameter and plotted in all figures. The mean cluster size was evaluated in terms of particle number involved in the cluster.

We investigated different configurations corresponding to a particle volume fraction $c$ ranging from 0.3% to 3% and nondimensional dipole strength $\lambda$ from 1 to 104. This concentration is in the range of the experiments of Promislow et al.10 and thus comparisons are possible. Figure 2 shows typical initial conditions with 80 paramagnetic beads seeded at random positions in a cubic domain. Figure 3 shows a typical result: aggregation of particles is clear and chains of various lengths are formed but some particles are still isolated. As time evolves, these


particles will join together and form longer chains. Chain/chain interaction leads also to long structure formation. Most of the chains are linear as head-to-tail aggregation of magnetic dipoles is energetically preferable. As pointed out by Mohebi et al., and also observed in the experiments of Fermigier and Gast, lateral merging of chains is possible leading to thick clusters of particles. Such lateral merging is important for the resistance to deformation and threshold of rupture of long chains. It is directly connected to the yield stress of the suspension and has a strong impact on technological applications. In Figure 3, chain defects are observable; e.g., observe one chain divided into two branches that connect again. Careful experiments of microrheology using dual-trap optical tweezers have highlighted clearly the impact of annealing defects on the mechanical properties of chains.

Von Smoluchowski's theory provides a solid basis for predicting aggregation rates in very dilute solutions. It states that the rate of change in the number of clusters containing n particles is connected to the coagulation kernel that represents the rate of coalescence of two smaller clusters. In diffusion-limited aggregation, the numerical simulations based on the motion of oriented particles on periodic square lattices support the scaling evolution \( S(t) \sim t^z \). The exponent \( z \) is related to the diffusion coefficient of a cluster of length \( s \). Under the assumption of long chains, it is commonly accepted that the diffusion coefficient scales like \( s^{-4} \) which in turn leads to the relation \( S(t) \sim t^{1.5} \). Even though this behavior has never been strictly observed in experiments, it provides a valuable point of reference.

Having obtained detailed temporal information from the simulations, we now want to compare distinct time evolutions of the mean length of chains; the determination of a characteristic time scale is a critical issue. We can expect a collapse of all the data if the time scale is properly defined, i.e., accounting for the effect of the dipole strength \( \lambda \) and the particle volume fraction \( c \). A naive proposition of doublet formation is based on pure diffusion of particles in a very dilute suspension. Particles diffuse as long as collision does not occur. When two particles are touching, a doublet is formed. The characteristic time \( t_b \) proposed in Russel et al., in this case is inversely proportional to \( D \) and \( c \) as it corresponds to the flux of particles toward an isolated test particle, i.e.,

\[
t_b = \frac{1}{6 D c} \frac{a^2}{\langle S(t) \rangle}
\]

(15)

Promislov et al., have introduced a significant improvement in the scaling. The identification of mechanisms leading to chain formation is of fundamental significance for the timescale determination. In particular, the motion of a paramagnetic particle can be split into two main regimes. When particles are widely separated, the attraction force that decays quickly \( (r^{-3}) \) is not strong enough and particles diffuse randomly. When the separation between two beads or between a particle and a chain is smaller than a critical value \( r_c \), determined by \( \langle S(t) \rangle \sim 0 \), the attraction force becomes dominant and aggregation occurs. Therefore, the flux of particles captured by a single sphere has to be augmented. The capture volume related to the surface \( |U| = k_B T \) allows the determination of a new timescale based on the envelope of the anisotropic dipolar potential. The modified time scale \( t_{b*} \) given by

\[
t_{b*} = \frac{a^2}{24((1/3)^{1/2} - (1/3)^{3/2}) D c l}
\]

(16)

It is important to note that this characteristic time is inversely proportional to \( D c \) as before but also to the dipole strength \( \lambda \). The constant prefactor is derived based on the capture volume that corresponds to the combined conditions of attraction, i.e., \( U < 0 \) and \( r < r_c \).

Initially, the mean cluster size is equal to one particle diameter as all the particles are seeded randomly throughout the domain respecting the nonoverlapping condition. Figure 4 shows results for very dilute suspension \( c = 0.003 \) that experiences a constant magnetic field characterized by a dipole strength \( \lambda \) ranging from 1 to 10^4. We notice that in the case \( \lambda = 1 \), no chains are forming and the particles always diffuse randomly. The magnetic attraction is not strong enough to join the particles together as Brownian forcing dominates the behavior of the
Promislow et al. observed an opposite trend. Exponents are slightly increased, i.e., as a relevant time scale for a different concentration.

The collapse of all the temporal evolutions $S(t)$ clearly points out that $t_0^*$ is adequate to scale the data for different $\lambda$. The power law scaling $S(t) \sim t^{z}$ inferred from the simulations agrees with the theoretical predictions as well as the experiments on the aggregation kinetics. The exponent $z$ ranges from 0.55 to 0.70 as $\lambda$ increases from 10 to $10^4$. Promislow et al. observed an opposite trend when comparing experiments performed with $\lambda = 8.6$ and $\lambda = 34$ in a very dilute system. In our simulations, increasing the concentration to $c = 0.009$ (see Figure 5) does not change the main features of the aggregation kinetics. For $\lambda \gg 1$, the aggregation follows a power-law evolution after an initial transient of order $t_0^*$. The collapse of the data is very good and confirms the validity of $t_0^*$ as a relevant time scale for a different concentration. Exponents are slightly increased, i.e., $z = 0.65, 0.77$, and 0.88 for $\lambda$ equal to $10, 100$, and $10^4$, respectively.

Finally, we checked the validity of $t_0^*$ as a scaling time with another simulation at $c = 0.027$. We plot the evolutions related to the same value of the dipole strength $\lambda = 100$ and various concentrations $c$ (Figure 6). It is evident that the collapse still occurs even for this higher value of the concentration. Comparing our results with Promislow et al., we see that $z$ is increasing with $c$ although experiments demonstrated the opposite trend in more dilute configurations. However, the range of variation of the exponent value agrees well with their results ($z$ ranges between 0.48 and 0.75 on all their configurations). We note that in the experiments the chains were up to five times longer than those in the simulations and the asymptotic value of $z$ appears to depend on this. The mechanism proposed by Promislow et al. to explain the reduction of the exponent in the scaling law is related to thermal diffusion of long chains. When chains are formed, the lateral and longitudinal diffusion coefficients are different. During the aggregation process the hindered lateral diffusion of the chains leads to a reduction of head-to-tail attraction of the particles, followed by slower aggregation kinetics. In our simulations, we have simplified the interactions between the particles. The magnetic attraction due to the dipole strength is evaluated by direct summation of pair interaction. Also, the dipole strength is independent of spatial configuration of the particles because we neglected mutual interactions for the background magnetic field. During the thermal agitation of the particles aggregated in chains, the Brownian forcing is strongly correlated and we use a simple model of forcing for single particles. The mechanism of lateral/longitudinal diffusion of the chains may be poorly represented in the numerical model. In conclusion, we can say that discrepancies observed in the more concentrated suspensions are related to a lower accuracy of chain–chain interactions, which may be crucial when the concentration increases, and possibly the relatively short length of the chains in the simulations. The aggregation of chains can lead to the formation of columns as experimentally observed by Martin and Odinek.

4. Conclusion

We have presented both qualitative and quantitative results in order to identify the mechanisms by which particles aggregate in chainlike structures in MR fluids. The fully coupled three-dimensional numerical model we developed is an efficient tool to obtain statistics and comparison with experiments and theoretical predictions of aggregation kinetics. Both characteristic time scale and power-law exponents were successfully compared to experimental values even though trends in the latter seem to be opposite. The number of particles in the simulation is still limited to a few hundred because the cost of the solution for the flow equations (hydrodynamic interactions) increases rapidly with the width of the domain. When long chains are formed, the periodic boundary conditions lead to a blockage effect on the aggregation of the particles. The domain width we selected is adequate to observe aggregation for at least 1 decade of non-dimensional time units.

Future work will be devoted to MR fluids in complex geometries for electroosmotic flows; see Patankar and

Figure 5. Evolution of the mean cluster size in terms of particle diameter: +, $\lambda = 1$; $\Delta$, $\lambda = 10$; $\Box$, $\lambda = 100$; $*$, $\lambda = 10^4$; dashed line, $\sim t^{0.8}$. Concentration $c = 0.009$.

Figure 6. Evolution of the mean cluster size in terms of particle diameter. $\bigcirc$, $c = 0.003$; $\Diamond$, $c = 0.009$; $\triangleleft$, $c = 0.027$. Dipole strength $\lambda = 100$.  


Hu. In more recent experiments, Hayes et al. obtained some exciting new results on the dynamics of MR fluids in microchannels of different cross sections; both pressure-driven and electroosmotic flows were investigated. A more detailed modeling is required for microscale simulations, where mechanical properties of chains are important. The particles that we consider in this work are larger than about 100 nm. In this size range we simulated particulate microflows as a continuum. The robustness of continuum calculations in this context has been demonstrated previously in Israelachvili and Vergeles et al. for spheres approaching a plane wall. More recently, a systematic molecular dynamics (MD) study was undertaken by Drazer et al. for a colloidal spherical particle through a nanotube containing a partially wetting fluid.

For a generalized Lennard-Jones liquid, it was demonstrated that the MD simulations are in good agreement with the continuum simulations of Bungay and Brenner despite the large thermal fluctuations present in the system. This is true even for very small size particles of the order of 2 nm, i.e., much smaller than the ones we consider here.

Simulation of MR particulate microflows could play an important role in the development and active control of dynamically reconfigurable self-assembled structures. The possibility to target and precisely control the electro-optical as well as the mechanical properties of microstructures in a dynamic way using external fields will open new horizons in microfluidics research and will suggest new protocols in nanofabrication.

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