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Microstructure of flow-driven suspension of hardspheres in cylindrical confinement: a dynamical density functional theory and Monte Carlo study

Hsiu-Yu Yu^{†,‡,⊥}, Zahera Jabeen^{¶,⊥}, David M. Eckmann^{§,||}, Portonovo S. Ayyaswamy[¶], and Ravi Radhakrishnan^{‡,||}

[†]Department of Chemical Engineering, National Taiwan University, Taipei 10617, Taiwan

[‡]Department of Chemical and Biomolecular Engineering, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

[¶]Department of Mechanical Engineering and Applied Mechanics, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

[§]Department of Anesthesiology and Critical Care, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

^{||}Department of Bioengineering, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

Abstract

We have studied the microstructure of a flow-driven hardsphere suspension inside a cylinder using dynamical density functional theory and Monte Carlo simulations. In order to be representative of various physical conditions that may prevail in experiments, we investigate the problem using both the grand canonical (μVT) ensemble and the canonical (NVT) ensemble. In both ensembles, the hydrodynamic effect on the suspension mediated by the presence of the confining wall is implemented in a mean-field fashion by incorporating the thermodynamic work done by the inertial lift force on the particle given the average flow field. The predicted particle distribution in the μVT ensemble displays strong structural ordering at increasing flow rates due to the correspondingly higher particle concentrations inside the cylinder. In the NVT ensemble, for dilute suspensions we observe a peak in the distribution of density at a location similar to that of the Segré-Silberberg annulus, while for dense suspensions the competing effects of the inertial lift and the hardsphere interaction lead to the formation of several annuli.

1 Introduction

Particulate suspensions in cylindrical pipe flows have evinced great interest due to their relevance in a wide range of applications such as in oil, food, construction, pharmaceutical industry^{1–3} and in biomedical research related to cell sorting and blood rheology.⁴ The

Correspondence to: Ravi Radhakrishnan.

[⊥]These authors contributed equally

rheology of flow-driven suspensions depends on the properties of the particulates such as shape, size, and deformability, the Reynolds number of the flow^{5,6} and the nature of confinement.⁶⁻⁹ Also, in the presence of particulates, fluid flow has been shown to transition from a shear-thinning to a shear-thickening behavior as the volume fraction is increased.¹⁰ Particulates are also known to induce an early onset of turbulence¹¹ as well as cause jamming with denser suspensions.¹² Both hydrodynamic and thermodynamic interactions play important roles in influencing the rheological behaviors of particulate suspensions, especially when they are in confined flow geometries. To understand the microrheology in confined flows of particulate suspensions, the microstructure of such systems need to be investigated. Here, we develop a predictive framework for investigating the structural characteristics of a hardsphere suspension flow in cylindrical confinement through the use of thermodynamic potentials.

Segré and Silberberg¹³ demonstrated the ‘tubular pinch’ effect where neutrally buoyant spheres carried by a flow inside a cylinder tend to accumulate at a radial distance r nearly equal to two-thirds of the cylinder radius R from the axis, $r_S \sim 0.6R$. This effect was attributed to a lateral force due to inertia of the moving fluid acting on the spheres which moves them across the streamlines in the flow. Theoretical studies of lateral migration started with Rubinow-Keller¹⁴ and Saffman¹⁵ who showed that the direction of such a lateral force is proportional to the slip velocity of the sphere, and the lift force is directed towards the centerline when the sphere lags the flow velocity while it is directed away from the centerline when the sphere leads the flow velocity. Cox and Brenner¹⁶ calculated the lift force and torque acting on a sphere in a Poiseuille flow using the method of matched asymptotic expansions with the Reynolds number Re and the ratio of radius of the sphere to cylinder radius a/R as expansion parameters, but the solutions were expressed in the form of complicated unsolved integrals. Ho and Leal¹⁷ calculated the lift force acting on spheres between two parallel walls using the method of reflections and showed that both the shear flow acting on the sphere and the associated wall-corrections give rise to the lift-force. Cox and Hsu¹⁸ evaluated the expressions given by Cox and Brenner¹⁶ for a sphere near a single plane wall in a parabolic flow and showed the existence of an equilibrium location that matched the position of the Segré-Silberberg annulus. Schonberg and Hineh¹⁹ studied the inertial migration of a sphere confined between two planar walls and showed that the equilibrium position moves towards the wall as the Reynolds number Re is increased. Matas *et al*²⁰ carried out experiments at particle Reynolds number $Re \sim 67 - 2400$, and showed that the Segré -Silberberg annulus moves towards the wall at higher Reynolds numbers, and is accompanied by the appearance of a secondary inner annulus in addition to the Segré-Silberberg annulus, which dominates over the outer annulus as the Reynolds number is further increased. The presence of particles in a suspension also changes the velocity profile of the suspension, with blunted velocity profiles with characteristics similar to a Casson flow²¹ observed for particle volume fractions greater than 0.20, while parabolic profiles are obtained at low volume fractions.^{22,23}

Numerical studies have explored the behavior of suspension flows under confinement. For example, Nott and Brady²⁴ have studied the inertial migration of a suspension of spheres in a cylindrical confinement using Stokesian Dynamics. They have defined a ‘suspension temperature’ which highlighted the local variations in the suspension rheology. Shao and

Sun²⁵ numerically evaluated the effect of a pressure-driven flow on a suspension of hardspheres, and were able to extract migration velocity profiles and locations of the annuli. Yang and Hu²⁶ studied the inertial migration of a single sphere in a cylinder flow ($Re = 1 - 100$) using the arbitrary Lagrangian-Eulerian(ALE) finite element method and investigated the correlations with the lift-force and equilibrium locations. While these studies solve the full hydrodynamic equations, they have been restricted to limited time-scales and particle numbers due to the huge computational overhead involved. An attractive alternate method for solving this problem would be to invoke a mean-field particle distribution theory such as the framework of the classical density functional theory.

Classical density-functional theory (DFT) is a field theoretic approach that relates the equilibrium system free energy to spatially-varied densities.²⁷ Through an appropriate definition of the grand potential functional, DFT has been shown to be capable of predicting the equilibrium thermodynamics of simple fluids in the bulk,²⁸ in the vicinity of a wall,^{29,30} in a slit or a cylindrical tube,³¹⁻³⁴ or in a spherical pore.^{32,35,36} In the presence of transient diffusion or applied flows, the nonequilibrium effects may be incorporated in the DFT formalism and this generalized procedure is referred to as the dynamical density functional theory (DDFT),³⁷⁻⁴⁰ which has been successful in predicting the flow-driven particle correlations with weak or no hydrodynamic interactions.⁴¹⁻⁴⁴

In this paper, we study the steady state density distribution of a suspension of hardspheres confined in a cylinder due to flow-induced inertial migration using DDFT. In order to account for the fluid inertia in the framework, we apply rigorous ensemble averaging to relate the single-particle migration velocity to the flow-perturbed average particle distribution through the consideration of an additional work term in the total free energy functional. Other DDFT studies in the past have similarly investigated the microstructure of colloidal fluid flows in the limit of zero Reynolds number confined between boundaries by including the distortion of the particle pair correlation due to the flow in the limit of zero Reynolds number.^{41,44} Our current effort is an attempt to bridge the prevailing physics of the system over various fluid hydrodynamic time scales with its long-time configuration by employing appropriate free energy descriptions. We also put forward a complimentary approach to predict the spatially varying densities under flow conditions using Monte Carlo (MC) simulations. Methods based on DDFT and on MC involve distinct sets of assumptions, yet agree extremely well with each other. We discuss comparisons between them as further validations of our study.

In the following, flow profiles through the cylinder are assumed to be parabolic for dilute suspensions and a blunted parabolic profile with denser suspensions. We explore the behavior of the suspension in the grand canonical ensemble as well as in the canonical ensemble. When the equilibrium state is controlled by the chemical potential, the grand canonical ensemble is invoked; when the physical system requires that the number of particulates be held constant, the canonical ensemble is used. In both ensembles, the effect of flow is included implicitly via an energy cost introduced by the mean-field hydrodynamic interactions of the sphere. We show that the inertial migration of particles may introduce strong spatial correlations between particles. In addition to comparing the results between

DDFT and MC, we also compare our findings, where available, with those of previous experiments reported in the literature on flowing suspensions under confinement.

The paper is organized as follows. In section 2, we describe our model. In section 3, we elucidate the DDFT formalism and give a formal expression for the density distribution of hardspheres both under fixed chemical potential and fixed particle number conditions. In section 4, we detail the method of canonical and grand canonical MC simulations. In section 5, we present the results obtained using DDFT and MC and discuss the effect of flow on the structural properties of the hardsphere suspension. We finally conclude with a discussion of our results and ramifications of our study for more complex properties of the particulates.

2 Formulation of the problem

The system of interest consists of a suspension of N hardspheres each of diameter σ (radius a) confined in a cylinder of finite length H and radius R in a Newtonian solvent of viscosity η_0 (See Fig 1(a)). The axis of the cylinder is chosen to be along the x -direction, and the radial direction is in the $y-z$ axis. We define $r = (y^2 + z^2)^{1/2}$ as the radial distance from the center of the cylinder, with the axis of the cylinder being at $r = 0$ and position of the wall at $r = R$. We also define $h = R - r$ as the distance from the wall of the cylinder. The number density of the hardspheres inside the cylinder is $\rho = N/\pi R^2 H$ (in reduced units, $\rho\sigma^3$) and the corresponding volume fraction is $\phi = (\pi/6)\rho\sigma^3$. This suspension is subjected to a pressure-driven flow along the axis of the cylinder with a centerline velocity v_{max} . In the case of dilute suspensions, we assume the flow through the cylinder to be Poiseuille, with the velocity along the radial direction given by $v(r) = v_{max}(1 - r^2/R^2)$. For suspensions at moderate to high concentrations, the average flow field would appear to have a blunted velocity profile due to significant hydrodynamic interactions between the hardspheres as well as between the hardspheres and the confining wall. As will be demonstrated in the DDFT, these effects may be incorporated in the particle conservation equation in a mean-field manner by considering the disturbance velocity of a ‘chosen’ particle driven by such a blunted flow field. On this basis, we shall assume a Casson-like velocity profile with more concentrated systems. For mathematical simplicity, we approximate the Casson profile by a flat profile in the center of the cylinder where $v(r) = v'_{max} = v_{max}[1 - (R - L)^2/R^2]$ when $0 < r < R - L$ and a parabolic profile near the wall with $v(r) = v'_{max}(1 - r'^2/L^2)$ in the region $R - L < r < R$, where $r' = r - (R - L)$ (See Fig. 1(b)). For a given hardsphere of diameter σ , the Reynolds number of the sphere is then defined by $Re = \sigma v_{max}/2\nu$, where $\nu = \eta_0/\rho$ is the kinematic viscosity and ρ is the density of the fluid. The Péclet number is defined by $Pe = \sigma v_{max}/2D_0$ with the diffusion coefficient $D_0 = 2k_B T/6\pi\eta_0\sigma$.

Although the problem may in principle be generalized to any length scale, here, we choose the diameter of the hardspheres to be $\sigma = 5 \mu\text{m}$. When the micron-sized spheres are in water and are subjected to flows with velocity in the range of $0 - 5 \text{ cm/sec}$, the Reynolds number Re is in the range $\sim 0 - 0.125$ and the product of the Reynolds number and Péclet number $RePe$ varies from $0 - 1.8 \times 10^5$. Unless otherwise specified, the radius of the cylinder is chosen to be $R = 4\sigma$. The volume fractions studied lie in the range $\phi \in [0.05, 0.3]$. Since the

sphere diameters are fixed, a variation of the product RePe would correspond to a change in the centerline flow velocity $v_{\max}$ or equivalently the fluid flow rate.

3 Dynamical density functional theory

In this section, we derive the DDFT for a flowing hardsphere suspension in a cylinder. We start with the general theory following Marconi and Tarazona³⁷ and Archer and Evans,³⁸ but explicitly account for the effect of the flow. We then separately consider two different scenarios, one with a fixed chemical potential and the other with a fixed number of particles in the cylinder.

For a system of N particles, the time evolution of the probability density function of particles, $P^{(N)}(\mathbf{r}^N, t)$, is described by the Smoluchowski equation written as

$$\frac{\partial P^{(N)}}{\partial t} + \sum_{i=1}^N \nabla \cdot \mathbf{j}_i = 0, \quad (1)$$

where the flux $\mathbf{j}_i$ due to the forces acting on each particle i and the applied convective flow is defined by

$$\mathbf{j}_i = P^{(N)} \left[\mathbf{v}_i + \sum_{j=1}^N \frac{\mathbf{D}_{ij}}{k_B T} \cdot \mathbf{F}_j \right] \quad (2)$$

where,

$$\mathbf{F}_j = -\nabla_j U_N - k_B T \nabla_j \ln P^{(N)}, \quad (3)$$

k_B is the Boltzmann constant, and T the temperature. Here, $\mathbf{v}_i$ is the velocity of particle i and $\mathbf{D}_{ij}$ is the total diffusivity tensor describing the mobility of particle i due to the force exerted on particle j , $\mathbf{F}_j$ which consists of the contributions from the total potential energies U_N (including interparticle potentials and external potentials) and the Brownian interaction arising from the gradients in the probability density. If we arbitrarily pick one particular particle and integrate out the other $N-1$ particles' degrees of freedom, for indistinguishable particles, Eq. 1 becomes

$$\frac{\partial \rho(\mathbf{r}_1, t)}{\partial t} + N \int_V \cdots \int_V (\nabla \cdot \mathbf{j}_1) d\mathbf{r}_2 \cdots d\mathbf{r}_N + N(N-1) \int_V \cdots \int_V \int_S (\mathbf{j}_2 \cdot \hat{\mathbf{n}}_2) d\mathbf{r}_2 d\mathbf{r}_3 \cdots d\mathbf{r}_N = 0, \quad (4)$$

where the one-body density of the chosen particle is defined by

$$\rho(\mathbf{r}_1, t) = N \int_V \cdots \int_V P^{(N)}(\mathbf{r}^N, t) d\mathbf{r}_2 \cdots d\mathbf{r}_N \quad (5)$$

Here, we have invoked the divergence theorem on the 3rd term of Eq. 4 with $\hat{\mathbf{n}}_2$ being the outward-pointing surface normal vector on the system boundary. V and S are the system volume and boundary area, respectively. Since the particles within the entire system must be conserved, the 3rd term is zero. If we neglect the hydrodynamic interactions between individual particles, the total diffusivity tensor can be simplified as $\mathbf{D}_{ij} \approx \mathbf{D}_s \delta_{ij}$, where $\mathbf{D}_s$ is the self-diffusivity tensor for a particle and δ_{ij} is the Kronecker delta. Substituting Eq. 3 for the chosen particle 1 into Eq. 4 then yields the conservation equation of the one-body density,

$$\frac{\partial \rho(\mathbf{r}_1, t)}{\partial t} + \nabla \cdot \rho(\mathbf{r}_1, t) \langle \mathbf{v}_1 \rangle_1 = \nabla \cdot \left[\frac{\langle \mathbf{D}_s \rangle_1}{k_B T} \cdot \rho(\mathbf{r}_1, t) \nabla \langle U_N \rangle_1 \right] + \nabla \cdot [\langle \mathbf{D}_s \rangle_1 \cdot \rho(\mathbf{r}_1, t) \nabla \ln \rho(\mathbf{r}_1, t)], \quad (6)$$

where $\langle \rangle_1$ denotes a conditional average with the position of one particle held fixed. For a given quantity $A(\mathbf{r}^N)$, one may define $\langle A \rangle_1(\mathbf{r}_1) = \int_V \cdots \int_V P^{(N-1)}(\mathbf{r}^{N-1} | \mathbf{r}_1) A(\mathbf{r}^N) d\mathbf{r}_2 \cdots d\mathbf{r}_N$ with $P^{(N-1)}(\mathbf{r}^{N-1} | \mathbf{r}_1)$ being the conditional probability density function of finding $N-1$ particles given that the chosen particle 1 is fixed at $\mathbf{r}_1$.

In the presence of external potentials, the total potential energy may be expressed as the sum of the external and the interparticle potentials, $U_N = U_{\text{ext}} + U_{N,\text{inter}}$. If the external potential acts independently on each particle with a well-defined one-body distribution, we may express the effective total potential averaged over nonequilibrium configurations of particles 2 to N given that the particle 1 is fixed, by $\langle U_N \rangle_1 = U_{\text{ext}} + \langle U_{N,\text{inter}} \rangle_1$, where $\langle U_{N,\text{inter}} \rangle_1$ denotes the effective one-body potential due to the many-body interactions. For a system in equilibrium, this effective one-body potential in the fluid is related to the gradient of the one-body direct correlation function, $c^{(1)}(\mathbf{r}_1)$ defined by the variation of the excess

Helmholtz free energy functional, $k_B T c^{(1)}(\mathbf{r}_1) = - \frac{\delta \mathcal{F}^{\text{ex}}[\rho(\mathbf{r}_1)]}{\delta \rho(\mathbf{r}_1)}$.⁴⁵ If we assume that the equilibrium correlations between the particles may be applied to nonequilibrium situations, or more specifically, if the sum rule for many-body interactions holds true in nonequilibrium,^{37,38,41} then we arrive at

$$\langle U_{N,\text{inter}} \rangle_1 = - k_B T \nabla c^{(1)}(\mathbf{r}_1, t) = \frac{\delta \mathcal{F}^{\text{ex}}[\rho(\mathbf{r}_1, t)]}{\delta \rho(\mathbf{r}_1, t)}. \quad (7)$$

Given this relation, combining terms on the right hand side of Eq. 4 and replacing $\mathbf{r}_1$ by $\mathbf{r}$ leads to the mean-field DDFT equation in the presence of the applied flow and external potential,

$$\frac{\partial \rho(\mathbf{r}, t)}{\partial t} + \nabla \cdot \rho(\mathbf{r}, t) \langle \mathbf{v}_1 \rangle_1 = \nabla \cdot \frac{\langle \mathbf{D}_s \rangle_1}{k_B T} \cdot \rho(\mathbf{r}, t) \nabla \frac{\delta \mathcal{F}[\rho(\mathbf{r}, t)]}{\delta \rho(\mathbf{r}, t)}, \quad (8)$$

where $\mathcal{F}$ is the Helmholtz free energy defined as a functional of the one-body particle density,

$$\mathcal{F}[\rho(\mathbf{r}, t)] = \mathcal{F}^{\text{id}}[\rho(\mathbf{r}, t)] + \mathcal{F}^{\text{ex}}[\rho(\mathbf{r}, t)] + \int_V U_{\text{ext}}(\mathbf{r}, t) \rho(\mathbf{r}, t) d\mathbf{r} \quad (9)$$

with the ideal gas free energy given by $\mathcal{F}^{\text{id}} = k_B T \int_V \rho(\mathbf{r}, t) [\ln(\rho(\mathbf{r}, t) \Lambda^3) - 1] d\mathbf{r}$ and Λ is the thermal de Broglie wavelength of the particle. In the limit of $t \rightarrow \infty$, the left hand side of Eq. 8 is set to zero and we obtain a quasi-steady DDFT equation for $\rho(\mathbf{r})$.

In the mean-field formulation, we may write $\langle \mathbf{v}_1 \rangle_1 = \langle \mathbf{v}_1^0 \rangle_1 + \langle \mathbf{v}_1' \rangle_1$; where $\langle \mathbf{v}_1^0 \rangle_1$ is the conditional average equilibrium flow velocity at $\mathbf{r}$ (the center of particle 1), and $\langle \mathbf{v}_1' \rangle_1$ is the conditional average perturbation velocity of particle 1 due to hydrodynamic interactions. In this problem, a suspension of hardspheres inside a cylinder is driven by a flow along the x direction and hence, owing to the symmetry of the cylindrical geometry, both the steady-state average flow profile and the particle density distribution would depend only on the radial position, and therefore, $\langle \mathbf{v}_1^0 \rangle_1 = \langle v_1^0 \rangle_1 \hat{\mathbf{e}}_x$ together with $\nabla \cdot [\rho(\mathbf{r}) \langle \mathbf{v}_1^0 \rangle_1(\mathbf{r})] = \nabla \cdot [\rho(r) \langle v_1^0 \rangle_1(r) \hat{\mathbf{e}}_x] = 0$. Therefore, $\nabla \cdot \rho(\mathbf{r}) \langle \mathbf{v}_1 \rangle_1(\mathbf{r}) = \nabla \cdot \rho(\mathbf{r}) \langle \mathbf{v}_1' \rangle_1(\mathbf{r}) \neq 0$, suggesting that the steady-state one-body density distribution would be altered in the presence of hydrodynamic interactions. This conclusion is consistent with that of ref 41, where the hardsphere density profile was investigated in the presence of an infinite-flat-wall-bounded simple shear flow. In our DDFT framework, since the convective term is expressed in terms of a combination of $\langle \mathbf{v}_1 \rangle_1$ and a regular perturbation approximation, it is crucial to choose an “appropriate” average flow field felt by the chosen particle in order to correctly capture the hydrodynamic effects. While for dilute suspensions the unperturbed average flow field $\langle \mathbf{v}_1^0 \rangle_1$ may simply be described by a Poiseuille profile, for denser suspensions it would be more appropriate to consider a blunted velocity profile such as the Casson profile. At finite Reynolds numbers, the chosen particle translating in a unidirectional flow parallel to a wall boundary would feel a wall-induced lift force in the direction perpendicular to the wall. In the hydrodynamically-dilute limit, if we neglect the disturbance to $\langle \mathbf{v}_1^0 \rangle_1$ due to the presence of a second particle, the inertial migration effect may be considered at the single particle level and be captured through $\langle \mathbf{v}_1' \rangle_1$. Consequently, in this limit, we may write $\langle \mathbf{v}_1' \rangle_1 = \langle v_1' \rangle_1 \hat{\mathbf{e}}_r$, where $\langle v_1' \rangle_1$ is the magnitude of the mean migrational velocity for particle 1. At steady state, Eq. 8 for the r direction may be simplified as

$$\frac{\partial}{\partial r}[\rho(r)\langle v_1' \rangle_1(r)] = \frac{\partial}{\partial r} \left[\frac{D_{\perp}}{k_B T} \frac{\partial}{\partial r} \frac{\delta \mathcal{F}[\rho(r)]}{\delta \rho(r)} \right], \quad (10)$$

where $D_{\perp}$ is the r -component of the self-diffusivity tensor $\mathbf{D}_s$ and represents the particle diffusivity perpendicular to the wall. Whereas for spherical particles in an unbounded medium $\mathbf{D}_s = D_0 \mathbf{I}$ with $\mathbf{I}$ being the identity tensor, the confining cylindrical wall breaks the symmetry of $\mathbf{D}_s$ and increases the resistivity of the particle, thus making $D_{\perp} \neq D_0$. Noting that $\rho(r)$ should be symmetric with respect to $r=0$, integrating Eq. 10 twice with respect to r yields

$$K = \int_0^r \frac{f_L(r')}{k_B T} dr' - \ln \rho(r) \Lambda^3 \frac{1}{k_B T} \frac{\delta \mathcal{F}^{\text{ex}}}{\delta \rho(r)} - \frac{U_{\text{ext}}(r)}{k_B T}, \quad (11)$$

where K is an undetermined coefficient, and f_L is the lift force exerted on the particle given

by $f_L = \frac{k_B T}{D_{\perp}} \langle v_1' \rangle_1$ according to linearized hydrodynamics. This integral of the lift force also suggests that in the DDFT framework, the effect of the external flow may be viewed as an additional work done by the lift force.

Before closing this subsection, we summarize the hydrodynamic interactions considered in the DDFT framework in the following: (i) The chosen particle feels an average flow field generated due to the collective hydrodynamic interactions of the rest of the $N-1$ particles. Hence, in the case of dilute suspensions, the flow field experienced by the chosen particle is described by a Poiseuille flow, whereas in the case of dense suspensions, the flow field is approximated by a blunted velocity profile, (ii) Although we approximate the diffusivity tensor as $\mathbf{D}_{ij} \approx \mathbf{D}_s \delta_{ij} = D_{\perp}$, the particle diffusivity $D_{\perp}$ is a function of its distance from the wall due to wall-hydrodynamic interactions.⁴⁶ (iii) The particle diffusivity $D_{\perp}$ is also a function of the local volume fraction $\phi(r)$, and the gradient of the diffusivity with respect to r can contribute to a drift velocity, as evident in Eq. 10.

3.1 $\rho(r)$ in the μVT ensemble

In Eq. 8, the term involving the variation of free energy with respect to the particle distribution denotes the net driving force acting on the particle and it can be expressed as

$\frac{\delta \mathcal{F}[\rho(\mathbf{r}, t)]}{\delta \rho(\mathbf{r}, t)} = \mu(\mathbf{r}, t)$, where $\mu(\mathbf{r}, t)$ is the chemical potential at $(\mathbf{r}, t)$. In conventional DFT, for a system in equilibrium, the chemical potential is a constant throughout the system. It also

follows immediately that when both U_{ext} and $\langle \mathbf{v}_1 \rangle_1$ are independent of time, $\frac{\delta \mathcal{F}[\rho(r)]}{\delta \rho(r)} = \mu$ is a constant. In the case where the cylindrical sub-system under consideration is in equilibrium with a particle reservoir, the chemical potential in the cylinder would be equivalent to that in the bulk reservoir, and the number of particles inside the cylinder would vary accordingly. As U_{ext} and f_L both vanish in the bulk, from Eq. 11 we obtain

$K = -\ln \rho_b \Lambda^3 - (k_B T)^{-1} [\partial \mathcal{F}^{\text{ex}}(\rho_b) / \partial \rho_b]$ with ρ_b being the particle number density in the bulk. As a result, the one-body density profile is determined as

$$\rho(r) = \rho_b \exp \left\{ -\frac{1}{k_B T} \frac{\delta \mathcal{F}^{\text{ex}}[\rho(r)]}{\delta \rho(r)} + \frac{1}{k_B T} \frac{\delta \mathcal{F}^{\text{ex}}(\rho_b)}{\partial \rho_b} - \frac{U_{\text{ext}}(r)}{k_B T} + \int_0^r \frac{f_L(r')}{k_B T} dr' \right\}. \quad (12)$$

For particulate flows through a porous medium in contact with a bulk suspension, the particle density distribution within the cylinder may be described by Eq. 12.

3.2 $\rho(r)$ in the *NVT* ensemble

Alternatively, if the number of particles in the cylinder is a constant but the chemical potential is allowed to vary, as is typically the case in a microfluidic chamber with a supply of a fixed number of particles, the Helmholtz free energy would be determined by a Lagrange function,

$$\frac{\mathcal{L}[\rho(\mathbf{r}, t)]}{k_B T} = \frac{\mathcal{F}[\rho(\mathbf{r}, t)]}{k_B T} - \lambda \left[\int_V \rho(\mathbf{r}, t) d\mathbf{r} - N \right], \quad (13)$$

where λ is a Lagrange multiplier. As $t \rightarrow \infty$, the steady-state density distribution $\rho(\mathbf{r})$ would be the one that minimizes $\mathcal{L}$, i.e. $\delta \mathcal{L} / \delta \rho = 0$, which yields $(k_B T)^{-1} \delta \mathcal{F} / \delta \rho = \lambda$. This is equivalent to the statement that K in Eq. 11 plays the same role as a Lagrange multiplier. As a result, with λ or K determined from the normalization condition, $\int_V \rho(\mathbf{r}) d\mathbf{r} = N$, we obtain

$$\rho(r) = \rho_b \frac{e^{-\frac{1}{k_B T} \frac{\delta \mathcal{F}^{\text{ex}}[\rho(r)]}{\delta \rho(r)} - \frac{U_{\text{ext}}(r)}{k_B T} + \int_0^r \frac{f_L(r')}{k_B T} dr'}}{2 \int_0^R e^{-\frac{1}{k_B T} \frac{\delta \mathcal{F}^{\text{ex}}[\rho(r)]}{\delta \rho(r)} - \frac{U_{\text{ext}}(r)}{k_B T} + \int_0^r \frac{f_L(r')}{k_B T} dr'} r dr}. \quad (14)$$

3.3 Approximation of lift force

As we noted in the Introduction, exact expressions for the lift forces are only available for simple flow profiles and some bounding geometries. To evaluate f_L using a known expression, in the region of the parabolic flow of length L , we herein adopt the solution given by Cox and Hsu who have solved for the radial velocity of a freely rotating sphere near an infinite plane using the method of matched asymptotic expansions.¹⁸ Their expression for the velocity due to inertial migration $\mathbf{v}_L$ is given by

$$\mathbf{v}_L(h) = \frac{5}{288} \frac{av_{\text{max}}^2}{v} \left(\frac{a}{L} \right)^2 \left(1 - \frac{h}{L} \right) \left(22 - 73 \frac{h}{L} \right) \hat{\mathbf{h}}, \quad (15)$$

where h is the distance of the particle center of mass from the wall and $\hat{\mathbf{h}}$ is the wall-surface unit normal vector pointing in the direction of $-\hat{\mathbf{e}}_r$. Writing $\mathbf{v}_L = -\langle \mathbf{v}'_1 \rangle_1$, the force acting on

the sphere is then given by $\mathbf{f}_L(h) = \frac{k_B T}{D_\perp} \mathbf{v}_L(h)$ with $k_B T/D_\perp$ being the mobility of the sphere in the direction perpendicular to the cylinder wall. In principle, $D_\perp$ would depend on the local particle volume fraction. If we assume that the leading order hydrodynamic effect is captured through the average flow field felt by the chosen particle, we may approximate $D_\perp$ as $D_0/\beta_\perp$, where $\beta_\perp$ denotes the wall-induced resistivity factor, and is given by

$\beta_\perp = \frac{6(z-1)^2 + 9(z-1) + 2}{6(z-1)^2 + 2(z-1)}$ with $z = h/a$.⁴⁶ Finally, the lift force felt by a given particle is determined by

$$\frac{\mathbf{f}_L(h)}{k_B T} = \text{RePe} \beta_\perp \frac{5}{288} \left(\frac{a}{L^2} \right) \left(1 - \frac{h}{L} \right) \left(22 - 73 \frac{h}{L} \right) \hat{\mathbf{h}}. \quad (16)$$

In the above expression, the velocity v_{max} and the radius of the sphere a are included in the product RePe . Through incorporating the work done by such an inertial lift force, to leading order, the disturbance on the microstructure due to flow is of $\mathcal{O}(\text{RePe})$, in contrast to the $\mathcal{O}(\text{Pe})$ effect obtained in other theoretical investigations on inertialess conditions, i.e. $\text{Re} \rightarrow 0$.^{41,47,48} Therefore, the factor of $\mathcal{O}(\text{RePe})$ represents the inertia-augmented distortion due to shear. According to Eq. 16, the fixed points of the lift force are obtained at $h/L = 1$ and $22/73$. For a Poiseuille flow in which $L = R$, the non-trivial fixed point is at $h = 22/73R$, or equivalently at $r_C = R - h \sim 0.7R$. This is comparable to the location of the Segré-Silberberg annulus⁴⁹ at $r_S \sim 0.6R$. Therefore, we would expect that a particle experiencing the lift force given by Eq. 16 and moving in the central region $0 < r < r_C$ will move towards the wall to the position r_C , whereas a particle in the region $r_C < r < R$ will move away from the wall towards r_C .

It should be noted that in the definition of this model, we have made a few assumptions. We have ignored higher order hydrodynamic interactions between the particles and considered only the mean-field hydrodynamic effect on the individual particles. The work done by the lift force combined with the hardsphere free energy functional implies that the interparticle interactions are limited to excluded volume interactions. A second assumption is that we ignore the distortion of particle distribution due to the wall-induced drag force experienced by the particles in the axial direction, but only focus on the effect of particle migration in the radial plane. Thirdly, the expression for the lift force adopted is given for a sphere near a single planar wall. Recent studies have shown that a sphere near a cylindrical wall experiences enhanced lift forces as compared to a sphere near a planar wall,⁵⁰ and that increased curvature (for narrower pipes) correspond to increased magnitudes of lift forces that are experienced.⁵¹

3.4 Determination of excess free energy

In both Eqs. 12 and 14, it is necessary to specify the dependence of the excess free energy on the particle distributions. Here, in order to capture the effect of the hardsphere potential between particles on the density distribution within the cylinder, we apply the smoothed density approximation (SDA).^{29,31} This sets the Carnahan–Starling equation of state⁵² as the

reference to the prevailing local thermodynamics, and has been shown to yield satisfactory predictions for the equilibrium microstructure of simple fluids in the bulk as well as in confined geometries.^{35,36,53} In the SDA, the excess free energy for the hardsphere fluid is written as

$$\mathcal{F}^{\text{ex}}[\rho(\mathbf{r})] = \int_V \rho(\mathbf{r}) \Delta\psi(\bar{\rho}(\mathbf{r})) d\mathbf{r}, \quad (17)$$

where ψ is the excess free energy per particle calculated from the Carnahan–Starling equation of state on the smoothed density $\bar{\rho}(\mathbf{r})$, which is defined as

$$\bar{\rho}(\mathbf{r}) = \int_V \rho(\mathbf{r}') w(|\mathbf{r} - \mathbf{r}'|; \bar{\rho}(\mathbf{r})) d\mathbf{r}' \quad (18)$$

Here, $w(|\mathbf{r} - \mathbf{r}'|; \bar{\rho}(\mathbf{r}))$ is the normalized, density-dependent weight function. The density dependence of the weight function is approximated in a power series given by

$$w(r, \rho) = w_0(r) + w_1(r)\rho + w_2(r)\rho^2 + \cdots, \quad (19)$$

where the coefficients w_0 , w_1 , and w_2 are determined from the consistency requirement between the resulting two-body direct correlation function and that predicted by the Pereus–Yevick approximation⁴⁵ for a homogeneous hardsphere fluid. They have the following forms:

$$w_0(r) = \frac{3}{4\pi\sigma^3} \Theta(\sigma - r), \quad (20)$$

$$w_1(r) = \begin{cases} 0.475 - 0.648 \left(\frac{r}{\sigma}\right) + 0.113 \left(\frac{r}{\sigma}\right)^2, & r < \sigma \\ 0.288 \left(\frac{\sigma}{r}\right) - 0.924 + 0.764 \left(\frac{r}{\sigma}\right) - 0.187 \left(\frac{r}{\sigma}\right)^2, & \sigma < r < 2\sigma \\ 0, & r > 2\sigma \end{cases}, \quad (21)$$

and

$$w_2(r) = \frac{5\pi\sigma^3}{144} \left[6 - 12 \left(\frac{r}{\sigma}\right) + 5 \left(\frac{r}{\sigma}\right)^2 \right] \Theta(\sigma - r), \quad (22)$$

where $\Theta(x)$ is the Heaviside step function.

Given the expressions for the weight function, the resulting $\frac{\delta \mathcal{F}^{\text{ex}}}{\delta \rho}$ terms in Eqs. 12 and 14 are derived as^{35,36,54}

$$\frac{\delta \mathcal{F}^{\text{ex}}[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r})} = \Delta \psi(\bar{\rho}(\mathbf{r})) + \int_V \rho(\mathbf{r}') \frac{\partial \Delta \psi}{\partial \bar{\rho}(\mathbf{r}')} \frac{\delta \bar{\rho}(\mathbf{r}')}{\delta \rho(\mathbf{r}')} d\mathbf{r}', \quad (23)$$

where

$$\frac{\delta \bar{\rho}(\mathbf{r}')}{\delta \rho(\mathbf{r})} = \frac{w(|\mathbf{r} - \mathbf{r}'|; \bar{\rho}(\mathbf{r}))}{\left\{ 1 - \int_V \rho(\mathbf{r}') \frac{dw(|\mathbf{r} - \mathbf{r}'|; \bar{\rho}(\mathbf{r}))}{d\bar{\rho}(\mathbf{r})} d\mathbf{r}' \right\}} = \frac{w(|\mathbf{r} - \mathbf{r}'|; \bar{\rho}(\mathbf{r}))}{1 - \bar{\rho}_1(\mathbf{r}) - 2\bar{\rho}_2(\mathbf{r})\bar{\rho}(\mathbf{r}')}, \quad (24)$$

with

$$\bar{\rho}_i(\mathbf{r}) = \int_V \rho(\mathbf{r}') w_i(|\mathbf{r} - \mathbf{r}'|) d\mathbf{r}', \quad (25)$$

for $i = 1$ or 2 .

3.5 Numerical methods

In solving Eqs. 12 and 14, we choose equally-spaced grids in the r direction within the cylinder with the number of grid points being in the range 50 – 100. The two equations can therefore each be discretized into a coupled set of nonlinear integral equations $\mathbf{F}(\boldsymbol{\rho}) = 0$ with $\mathbf{F}$ and $\boldsymbol{\rho}$ denoting the entire vectors of the target functions F_j and variables ρ_j , respectively, and j being the index of the grids. At each grid of r_j , U_{ext} enforces zero probability of finding

a particle center at $R - \frac{\sigma}{2} < r \leq R$, and this constraint is applied throughout the numerical procedure. The integrals involving $\mathcal{F}^{\text{ex}}$ are evaluated by Eqs. 23 and 24 using a multidimensional Romberg integration⁵⁵ with adaptive trapezoidal method until an accuracy of 10^{-4} relative error is met. At any radial position not on the grid, the values of r and ρ are interpolated from those of the neighboring two grid points. The integral of f_L is evaluated analytically. In the absence of flow and/or for dilute suspensions, both the equations can be solved iteratively using a Picard-type method with the mixing parameter to ensure convergence to a predefined level of accuracy.⁵⁶ However, in the presence of flow, the additional work done by the lift force may distort the equilibrium correlations significantly, thus much finer grids and much smaller iterative step sizes than available in the Picard framework are required. In order to more efficiently solve the two sets of nonlinear equations, we employ a globally-convergent Newton's method.⁵⁵ where at each iterative step a line search with an adaptive step size is performed to check if the Newton step minimizes

the target function, $\frac{1}{2} \mathbf{F} \cdot \mathbf{F}$. If not, we backtrack along the Newton step direction and try a smaller step size. This procedure is repeated until an acceptable step is found. The Jacobian matrix in the Newton step is evaluated using a numerical forward difference method with $\delta = 10^{-12}$. The convergence is achieved when the relative absolute error for ρ_j between the two consecutive steps is less than 10^{-7} .

4 Monte Carlo simulations

We compare the results obtained using DDFT with results obtained by employing a complementary method based on MC simulations. The MC simulations are started with hardspheres of diameter σ randomly placed inside the cylinder with a volume fraction ϕ . We assume that the spheres interact with each other as well as with the wall of the cylinder via the hardspheres potential, $U_{\text{hardspheres}}$

$$U_{\text{hardspheres}}(r) = \infty, \quad r \leq \sigma, \quad = 0, \quad r > \sigma. \quad (26)$$

Overlap between the hardspheres is avoided by ensuring that the distance between centers of the hardspheres satisfies the requirement $r_{ij} \geq \sigma$. Since the wall of the cylinder is rigid, overlaps between the wall and the hardspheres are avoided by ensuring the radial distance from the center $r < R - \sigma/2$. A periodic boundary condition is imposed in the x-direction. The potential energy $U_L(h)$ due to the lift force $\mathbf{f}_L(h)$ is calculated in terms of a change in the energy when a particle is moved from its position at a distance h from the wall, to a position where the lift-force acting on the particle vanishes. This change in energy for the i^{th} particle is given by:

$$U_L(h_i) = \int_{h_i}^L \mathbf{f}_L(h) \cdot d\mathbf{h}. \quad (27)$$

The potential energy of the N -particle system due to the lift-force is then given by:

$$U_L(N) = \sum_{i=1}^N U_L(h_i) = \sum_{i=1}^N \int_{h_i}^L \mathbf{f}_L(h) \cdot d\mathbf{h}. \quad (28)$$

To obtain the equilibrium configurations of hardspheres in a fluid flow in a cylinder, with dilute suspensions, we use both grand canonical and canonical MC simulations, while we use canonical MC simulations for denser suspensions. Our implementation of the NVT and μVT methods follows standard recipes.⁵⁷

In the canonical (NVT) ensemble, the number of particles N , the volume V and the temperature T of the system are constants. The MC moves consist of attempts to displace the particle with an acceptance probability P_{disp} . An attempt is made to move a particle from its old position $\mathbf{r}_o$ to a new position at $\mathbf{r}_n = \mathbf{r}_o + [\text{rand}]$. The random number $[\text{rand}] \in [-1, 1]$. The displacement is chosen such that 50% of the moves are accepted. The new position is checked for overlaps with neighboring particles and the cylindrical wall of the cylinder. If an overlap is present, the move is rejected. Otherwise, the move is accepted with a probability P_{disp} defined by:

$$P_{\text{disp}}(h_o \rightarrow h_n) = \min[1, \exp(-\beta\{U(h_n) - U(h_o)\})]. \quad (29)$$

Here, h_o and h_n are the distances of the particle from the wall at the old and the new positions respectively, $U(h_o)$ and $U(h_n)$ are the corresponding potential energies due to the lift-forces exerted on the particle (see eq 27). We execute $N \times 10^6$ moves for equilibration and an equal number of moves for production runs.

In the grand canonical (μVT) ensemble, the suspension of hardspheres inside the cylinder is in equilibrium with an external reservoir with volume fraction ϕ , chemical potential μ and temperature T . Particles are exchanged between the reservoir and the cylinder with the chemical potential μ , volume V and temperature T held constant. The chemical potential μ is defined by $\mu = \mu_{ex} + \mu_{id}$. Here, $\mu_{id} = \ln \rho \Lambda^3$ is the chemical potential of an ideal gas. The excess chemical potential for hardspheres is given by the Carnahan-Starling equation:⁵²

$$\mu_{ex}/kT = \phi(8 - 9\phi + 3\phi^2)/(1 - \phi)^3. \quad (30)$$

In addition to the displacement moves as carried out in NVT simulations, attempts are made to insert particles in the cylinder with an acceptance probability P_{add} and to remove particles with an acceptance probability P_{del} . When a move to insert a particle at a random position inside the cylinder is attempted, the new position is checked for overlaps with neighbors and the cylindrical wall. In the absence of overlaps, the move is accepted with a probability P_{add} given by

$$P_{add}(N \rightarrow N+1) = \min\left[1, \frac{V}{\Lambda^3(N+1)} \exp(\beta(\mu - U_L(N+1) + U_L(N)))\right]. \quad (31)$$

Here, $U_L(N+1) - U_L(N)$ is the change in the potential energy due to lift-forces on adding a particle to the cylinder (eq. 28). Similarly, a randomly chosen particle is removed with a probability P_{del} given by:

$$P_{del}(N \rightarrow N-1) = \min\left[1, \frac{\Lambda^3 N}{V} \exp(-\beta(\mu + U_L(N-1) - U_L(N)))\right]. \quad (32)$$

The MC moves consist of (a) M_D attempts to displace the hardspheres with an acceptance probability P_{disp} and (b) M_A attempts to insert hardspheres with a probability P_{add} and (c) M_R attempts to remove the hardspheres with a probability P_{del} . To satisfy detailed balance, the number of attempts to add and remove the particles was chosen to be the same. If the total number of MC moves is $M = M_D + M_A + M_R$, the particles have been added and removed with a probability $M_A/M = M_R/M = 0.15$. The simulation was run for $N \times 10^6$ MC moves for equilibration and $N \times 10^6$ MC moves for production runs.

The standard deviation of the reduced densities $\rho\sigma^3$ have been estimated by dividing the production run consisting of $N \times 10^6$ MC steps into 5 blocks. The average reduced densities for each of the block was calculated, and the standard deviation was estimated for the average densities obtained.

5 Results & Discussion

In this section, we discuss the results obtained from DDFT and MC simulations. We observe that the presence of inertial migration in a suspension of hardspheres due to flow effects changes the structural ordering of hardsphere suspensions. As mentioned above, we separately employ the grand canonical framework and the canonical framework to capture the effects seen with the different physical systems of interest.

5.1 The grand canonical (μVT) ensemble

We begin our discussion with the results obtained in the grand canonical framework for dilute suspensions, where the reservoir volume fraction is fixed to be in the range $\phi_r \in [0.05, 0.2]$.

5.1.1 Effect of flow on the local density distribution—In Fig. 2(a), the reduced densities $\rho\sigma^3(r)$ at various radial positions r obtained from our calculations using DDFT for a reservoir volume fraction $\phi_r = 0.05$ and for various values of the product RePe are plotted as symbols, while those obtained using grand canonical MC simulations are plotted as lines. In the simulations, the reduced densities $\rho\sigma^3(r)$ at various r are calculated by counting the number of centers of particles $n(r)$ within a cylindrical shell volume centered at r and normalized by the volume of the shell. The number densities obtained from the simulations are noted to match extremely well with those obtained using DDFT within the limits of statistical error. This agreement between the independent predictions of DDFT and MC simulations provides credibility to the results presented in this paper.

In the absence of flow ($\text{RePe} = 0$), hardspheres are seen to preferentially localize on the wall resulting in the distribution showing a peak at $r/R = (R - a)/R$, the position of the center of the hardsphere nearest to the wall. This is a well known result attributed to depletion interactions induced by the wall in the absence of a flow.⁵⁶ For a finite RePe , the number density of the particles in the proximity of the wall reduces as a consequence of the lift force felt by the particles which tends to push them away from the wall (see Fig. 3(a)). This is also evident in Fig. 2(b), where the number density at the wall $\rho_w\sigma^3$ is plotted as a function of RePe for volume fractions $\phi = 0.05, 0.1, 0.2$. The near wall-number density falls exponentially as a function of RePe , indicative of a characteristic velocity scale. In Fig. 2(b), the exponential fit $\rho_w\sigma^3 = n_w(\phi)\exp(-\text{RePe}/144)$ is displayed as a dashed line. The characteristic energy scale RePe_c appears to be $\text{RePe}_c = 144$ which can also be extracted from the lift force expression in Eq. 16. With an increase in flow velocity, the formation of a depleted region near the wall is simultaneously accompanied by a decrease in the number density at $r = 0$. The particles in the central region migrate towards the fixed point r_C .

In Fig. 2(c), for various reservoir volume fractions $\phi_r = 0.05, 0.1, 0.2$, we plot the position of the peak of the distribution r_p normalized by the radius of the cylinder R as a function of RePe . We note that for all the cases, with increasing flow velocity (higher RePe), the peak of the distribution moves away from its initial position at $r_p/R = (R - a)/R$ at $\text{RePe} = 0$. The position of the peak of the distribution is closer to the wall for denser suspensions. Interestingly, in the dilute case (i.e., $\phi_r = 0.05$), the shift appears to be non-monotonous, wherein the position of the peak first moves away from the wall, and then moves back

towards the wall eventually settling to a steady location at r_p^+/R as RePe is increased. For high RePe, the position of the peak of the distribution appears to converge to $r_p^+/R=0.81 \pm 0.1$ for reservoir volume fractions $\phi_r \in [0.05, 0.2]$. This is different from the location of the Segré-Silberberg annulus which forms at $r_s/R = 0.6$.

The shift in equilibrium location of the annulus noted in our investigations is attributable to the increased volume fractions inside the cylinder at higher RePe. Fig. 2(d) shows the average volume fractions ϕ_m inside the cylinder for various RePe and reservoir volume fractions $\phi_r \in [0.05, 0.2]$. With an increase in the flow velocity (higher RePe), more hardspheres are exchanged between the cylinder and the reservoir in order to maintain the chemical potential between the two at equilibrium. As a result, higher volume fractions occur inside the cylinder. As a consequence, an increasingly blunted average velocity profile (much different from the imposed Poiseuille flow) is expected as is shown in experiments conducted with denser suspensions.^{22,23} This in turn would affect the equilibrium location of the annulus, and we will further explore this feature with suspensions of higher densities in section 5.2.2. We would like to point out that the velocity distribution corresponding to the obtained density distribution can be computed¹⁷ and imposed on the suspension to arrive at the steady-state density distribution. We plan to explore this approach in our future studies. Another important observation is that with increasing RePe, an increased structural order in the suspensions is noted. These aspects will be discussed in section 5.1.2.

5.1.2 Formation of layers with increase in flow velocity—As discussed earlier, in the μVT ensemble, i.e., at a fixed reservoir chemical potential μ , the number densities are observed to increase with increase in flow velocities at all radial positions away from the wall (see Fig. 2(d)). This is accompanied by an onset of structural ordering in the suspension, as is evidenced by the formation of multiple annuli with increase in RePe. While not commonly described in flow systems, the induction of layers are well known in confined systems.⁹ In Fig. 3(a)–(c), for a reservoir volume fraction $\phi_r = 0.05$ and RePe = 400, 1000, 4000, we plot a single snapshot of the $y - z$ positions of the center of the hardspheres in the radial plane. We note the appearance of dense concentric bands with increasing RePe. The denser bands appear distinct and well-separated at the highest value investigated, RePe = 4000 (see Fig. 3(c)). In Fig. 3(d), we plot the reduced density as a function of the radial distance from the center r . With increasing RePe, additional peaks appear at locations closer to the central axis. These peaks become increasingly distinct as RePe is increased. However, there is an upper limit to the number of peaks that can be formed, limited by the ratio R/σ . The distance between the peaks also decreases as the flow velocity increases as is shown in Fig. 3(f). In this state, the suspension reaches its densest state. One such configuration for RePe = 4000 is plotted in Fig. 3(e), where the diameters of the hardspheres and the cylindrical pipe are drawn to scale. We see that the hardspheres are densely packed, yet remarkably the energetics of this system line the centers of the hardspheres in distinct annuli.

In the grand canonical framework, a ‘governing’ chemical potential μ in conjunction with the flow enables the influx of particles until a minimum energy state is achieved. We will

next discuss the canonical case in section 5.2, where the number of particles N inside the cylinder is constant and we can discern the effect of flow on the suspension.

5.2 The canonical(NVT) ensemble

To compare with results of experiments, where the number of particles inside the cylinder is maintained to be a constant, we investigate the cases of both dilute and moderate suspensions ($\phi = 0.2$) using DDFT and MC simulations under the canonical framework. The details of the method were presented in section 3 and 4.

5.2.1 Dilute suspensions—With dilute suspensions, we assume the velocity profile in the cylinder to remain parabolic and is not affected by the presence of hardspheres. Both DDFT calculations and MC simulations in the canonical (constant NVT) framework consider the suspension with a volume fraction $\phi = 0.05$ inside a cylinder of radius $R = 6\sigma$. In Fig. 4(a)–(b), we plot ten snapshots of the locations of the centers of the hardspheres on the radial plane in the flow medium. The RePe values considered are plotted for $\text{RePe} = 10^2$ and 10^4 . At $\text{RePe} = 100$ (i.e., at low values of RePe), the hardspheres are diffusely spread on the radial plane, while at $\text{RePe} = 10^4$ (i.e., at high values of RePe), the hardspheres are concentrated in a single annulus. The reduced densities $\rho\sigma^3$ for this case obtained using DDFT (symbols) and MC (lines) are plotted in Fig. 4(c). The reduced density at the center of the cylinder is noted to decrease with increasing RePe. The formation of a single peak in the density distribution is also noteworthy. The location of the peak of the density distribution at $r_p \sim 0.7R$ is comparable to the position of the fixed-point r_C predicted by the Cox and Hsu expression (see Eq. 16). We note that we can make direct comparisons with results obtained using this ensemble and results obtained in experiments. The formation of annulus in this ensemble is remarkably similar to the Segré-Silberberg annulus.¹³

We may also draw comparisons with results of experiments in three-dimensional microchannels conducted by Choi-Lee,⁵⁸ where they have studied the inertial migration of a dilute suspension of spheres ($\phi = 0.0005$ in a cylinder of radius $R = 6\sigma$). Their study is for channel Reynolds numbers defined by $\text{Re}_C = 2v_{\max}R/\nu = 1.6, 9.7$ and 58.1 . These values of Re_C correspond to particle Reynolds number $\text{Re}_P = \text{Re} = av_{\max}/\nu = \text{Re}_C(a/2R) = 0.065, 0.40$ and 2.4 , a range of Re we have also been interested in. Fig. 4(d)–(e) show the spatial distribution of particles in the radial plane obtained for $\text{Re}_C = 1.6, 58.1$ which correspond to $\text{Re}_P = 0.065, 2.40$ respectively. The spatial distribution in Fig. 4(d)–(e) look similar to the snapshots in Fig. 4(a)–(b). The probability distribution function of the spatial positions obtained in their experiments shows a peak at $r_p \sim 0.6R$ for $\text{Re}_C = 1.6$ and at $r_p \sim 0.7R$ for $\text{Re}_C = 58.1$, which is comparable to the location of the annulus obtained in our simulations (Fig. 4(c)).

Based on these very good comparisons between our predictions and available experimental results, our NVT calculations of flow-driven suspensions in confinement sufficiently and accurately capture the behavior of hardsphere suspensions seen in experiments. In section 5.2.2, we study the minimum energy configuration obtained using NVT ensemble for denser suspensions.

5.2.2 Denser suspensions—Evidence from experiments indicates that when a denser suspension of spheres flows in a cylinder, the effect of the suspension on the flow is such as to change the velocity distribution in the flow to a Casson-like profile.^{22,23} We here compare our predictions with the velocity distributions presented in Hampton et al,²³ and choose the region of parabolic flow in the cylinder as $L = 0.7R$ for $\phi = 0.2$ and $L = 0.6R$ for $\phi = 0.3$ (see Fig. 1(b))

The reduced densities $\rho\sigma^3$ obtained from NVT calculations for various RePe and $\phi = 0.2, 0.3$ are displayed in Figs. 5(a)–(b). Since the number of particles is a constant in these calculations, the area under the curve is a constant even when the flow velocity is increased. For RePe = 0, the reduced density at the center of the cylinder (i.e., at $r = 0$) is the same as initially prescribed. However, with an increase in RePe, the reduced density at the center of the cylinder decreases, as is noted in Fig. 5(c) where the reduced density obtained in the central region for $\phi = 0.2$ (Fig. 5(a)) has been replotted. The depletion in the number of hardspheres in the central and near-wall region is accompanied by the formation of a peak in the vicinity of the fixed point r_C . As RePe is increased further, the location of this peak shifts until it reaches a fixed point at $r_P/R = 0.8$ and $r_P/R = 0.85$ for $\phi = 0.2, 0.3$ respectively. In our calculations, the location of the outer peak depends on the extent of the parabolic region. With denser suspensions, as the volume fraction is increased, the extent of the blunt (core) region increases accompanied by a decrease in the extent of the parabolic region. In the expression for lift force given by Cox and Hsu (eq. 16), the location of the fixed point r_C depends on the extent of the parabolic region L . With a decrease in the extent of the parabolic region associated with denser suspensions, the location of the outer peak is closer to the wall of the cylinder (see Fig. 5(d))

Also remarkable is the formation of an inner peak closer to $r \sim 0.6R$ for $\phi = 0.2, 0.3$ at intermediate values of RePe. The location of the inner peak moves towards the outer peak as the flow velocity (or RePe) is increased, and eventually it merges with the outer peak. However, at a higher volume fraction such as $\phi = 0.3$, the inner annulus does not merge with the outer annulus even at large values of RePe, indicating a competition between the inertia-induced lift-force which promotes the formation of the annulus and the excluded volume interactions which limit the number of hardspheres that can be packed in a single annulus. We find similarities between these results and the formation of multiple peaks noted in the experiments of Matas et al, who reported multiple rings for $Re > 700$.²⁰ They have also reported a shift in the position of the outer peaks towards the wall with an increase in the Reynolds number. A similar effect was seen in our calculations carried out in the grand canonical framework (Fig. 3), where an increase in RePe resulted in an increase in the average density of the hardspheres inside the cylinder accompanied by a formation of multiple peaks. The presence of annuli in dense suspension could be attributed to substantial hydrodynamic and thermodynamic interactions between the suspended particles.

Although we have generally good agreement between DDFT and MC in dilute suspensions, in more concentrated systems the deviations between the two methods are observed. We have invoked the assumption that the equilibrium correlations between particles always apply, but it is anticipated that this assumption would be increasingly invalid as the many-body effects become important and the applied flow field significantly perturbs the

equilibrium microstructure, as noted for $\phi = 0.2 - 0.3$. Given the convergence condition stated in section 3.5 for the DDFT study, the computational time also increased significantly for investigating denser suspensions. For example, the CPU time at a given number of RePe is 10 – 15 times longer for $\phi = 0.3$ than for $\phi = 0.05$.

6 Conclusions

In this paper, we have studied the effects of inertial migration due to flow on the steady-state behavior of a suspension of hardspheres using DDFT and MC simulations. With initially dilute suspensions in equilibrium with a reservoir chemical potential, we observe that the density of suspension inside the cylinder increases with an increase in flow velocity. This increase in density is coincident with increased structural ordering of the hardspheres inside the cylinder as is evidenced by the formation of distinct layers at increased flow velocities. In the case of more concentrated suspensions studied in the canonical framework, we observe that the position of the Segré-Silberberg annulus depends on the radial velocity profile inside the cylindrical pipe, which in turn is dictated by the density of the suspensions. At high concentrations of the hardspheres, more than one annulus can be observed at large flow velocities, indicating a competition between the forces due to inertial migration and excluded volume interactions which limit the packing of the number of particles in a given annulus.

These results have consequences for the rheology of the suspension. It will be interesting to explore the effect of the structural ordering and associated build-up of spatial correlations on the apparent viscosity of the suspension, and whether the reorganization of the hardspheres into minimum energy configurations leads to a shear-thinning vs shear-thickening behavior. This study can be further extended to include anisotropic rigid bodies, where the inertial lift also depends on the orientation of the rigid body, and collective behavior of a large number of these anisotropic particles in the presence of hydrodynamic interactions can give rise to interesting rheological and structural properties.⁵⁹ A physiological system where these questions become very relevant are in the context of blood flow in the capillaries, where the red blood cells are deformable capsules experiencing significantly enhanced lift forces. The segregation of the constituents of the blood flow and the dependence of viscosity on the hematocrit and diameter of the cylindrical confinement (famously known as the Fahraeus-Lindqvist effect)⁶⁰ is exploited by pharmaceutical companies for microfluidic cell sorting as well as for targeted drug delivery.

In this study, we have ignored the effect of near-field hydrodynamic interactions between the particles, which play a major role in influencing the rheological properties of the suspension.^{41,43} A second assumption in our study is to ignore the effect of the curvature of the cylindrical wall on the inertial forces acting on the particle. In our future studies, we plan to include these effects and obtain a clearer picture of the collective particle dynamics in the suspension.

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Figure 1.

(a) A representative configuration of hardspheres of diameter σ in a cylinder of radius $R = 4\sigma$ and height $H = 4R$. The arrows denote the lift-force $\mathbf{f}_L$ acting on the hardspheres due to presence of a flow. (b) Velocity profiles in the radial direction of the cylinder. The dotted line represents a Poiseuille flow, and the solid line represents a blunted velocity profile with a parabolic flow in the region $R - L < r < R$, where r is the radial distance from the center of the cylinder.

Figure 2.

(a) Reduced density $\rho\sigma^3$ obtained using μVT ensemble for $RePe = 0, 20, 60, 100, 200$ for reservoir volume fraction $\phi_r = 0.05$. The lines represent results from MC simulations and symbols represent results from the DDFT calculation. Vertical dotted line indicates the position of r_C , the fixed point solution given by Cox and Hsu.¹⁸ The standard deviation of $\rho\sigma^3$ calculated using block averages is estimated to be 0.001 for $RePe = 200$. (b) Reduced density at the wall ($\rho_w\sigma^3$) plotted as a function of $RePe$ on a semi-log scale. The dashed line is an exponential fit to the data of the form $\rho_w\sigma^3 = n_0(\phi) \exp(-RePe/144)$. (c) Position of the peak of the distribution r_p as a function of $RePe$. (d) The average volume fraction ϕ_m plotted as a function of $RePe$. Panels (b–d) share the same legend.

Figure 3.

(a)–(c) A single snapshot of y and z positions of hard spheres obtained using μVT simulations for reservoir volume fraction $\phi_r = 0.05$ and $\text{RePe} = 400, 1000$ and 4000 respectively. (d) Reduced density $\rho\sigma^3$ plotted as a function of distance from center, r/R . The symbols represent results obtained using DDFT and lines represent results obtained using MC. The peaks have been labeled from 1 – 4. Vertical dotted line indicates the position of r_C . (e) The configuration obtained for $\text{RePe} = 4000$ shown to scale. (f) Distance between the various labeled peaks plotted as a function of RePe .

Figure 4.

(a), (b) Each panel represents ten overlaid snapshots of $y - z$ positions of hard spheres in a cylinder of radius $R = 6\sigma$ obtained using *NVT* simulations for volume fraction $\phi = 0.05$ and $\text{RePe} = 100$ and 10000 respectively. (c) Reduced density $\rho\sigma^3$ of hard spheres obtained for the configurations in (a)–(b). The symbols and lines represent results obtained using DDFT and MC, respectively. Vertical dotted line indicates the position of r_C . (d)–(e) spatial distributions of particles in the radial plane obtained in experiments by Choi and Lee⁵⁸ on flow-driven hard sphere suspension in a cylinder for channel Reynolds number $\text{Re}_C = 1.6$, 58.1 , (f) shows the density distribution of the particles obtained in the experiments by Choi and Lee for parameters shown in Fig. (d)–(e)

Figure 5.

Reduced density $\rho\sigma^3$ obtained using NVT ensemble for hard spheres subjected to blunted velocity profile in the cylinder for $RePe = 20, 100, 400, 1000, 4000, 8000$ and (a) volume fraction $\phi = 0.2$ and extent of parabolic flow $L = 0.7R$, (b) volume fraction $\phi = 0.3$ and extent of parabolic flow $L = 0.6R$. The points and lines represent results obtained using DDFT and MC respectively. Vertical dotted line indicates the position of r_C . (c) Reduced density $\rho\sigma^3$ obtained in (a) replotted for radial distances $r/R < 0.6$. Decrease in density in the central region with increase in flow ($RePe$) is evident. (d) The position of the peak of the distribution r_p/R plotted as a function of $RePe$ for volume fractions $\phi = 0.05, 0.2, 0.3$.