Analogies between colloidal sedimentation and turbulent convection at high Prandtl numbers

P. Tong, B. J. Ackerson, Department of Physics, Oklahoma State University, Stillwater, Oklahoma 74078, USA,
ptong@osuunx.ucc.okstate.edu

In a recent experiment, Segrè et al. [1] used the particle imaging velocimetry (PIV) technique to measure the spatial correlation function, $C(\ell) = \langle \delta v(r) \delta v(r+\ell) \rangle$, of the velocity fluctuation δv in a sedimenting suspension of non-Brownian particles over a wide range of particle concentrations and sample sizes. They found that the measured $C(\ell) \sim \exp(-\ell/\xi)$, where the velocity correlation length ξ depends on the particle radius a and volume fraction ϕ_0 in a non-trivial power-law form $\xi \simeq a\phi_0^{-1/3}$. In this paper we propose a new set of coarse-grained equations of motion to describe concentration and velocity fluctuations in a dilute sedimenting suspension of non-Brownian particles. With these equations we find that colloidal sedimentation is analogous to high Rayleigh number, high Prandtl number turbulent convection [2,3]. Our model explains the experimental results by Segrè et al. and also provides a coherent framework for the study of sedimentation dynamics in different colloidal systems.

To understand the basic principles governing the colloidal sedimentation, we consider a simple case of a dilute sedimenting suspension of hard spheres in a long cylindrical tube of radius L. To separate the velocity fluctuation δu from the mean settling velocity $\bar{v}$, we choose a uniform suspension with $\bar{v} = 0$ as our reference system. It has been suggested [1,4] that velocity fluctuations in a sedimenting suspension may arise from fluctuations of the local particle concentration. Therefore, we model the colloidal sedimentation with a coarse-grained Navier-Stokes equation. The fluid velocity $\delta \mathbf{u}$ and pressure δp at a point $\mathbf{x}$ satisfy the creeping flow equation [5]

$$\nabla \delta p(\mathbf{x}) - \eta \nabla^2 \delta \mathbf{u}(\mathbf{x}) = \mathbf{f} \delta n(\mathbf{x}), \tag{1}$$

where η is the viscosity of the fluid and δn [$= n(\mathbf{x}) - \bar{n}$] represents the fluctuation of the particle number density $n(\mathbf{x})$ about its mean $\bar{n}$. In the above, $\mathbf{f} = (4\pi/3)a^3 \Delta\rho \mathbf{g}$ is the buoyancy force acting on a particle of radius a, where $\mathbf{g}$ is the gravitational acceleration and $\Delta\rho = \rho_p - \rho_s$ is the density difference between the particle (ρ_p) and the solvent (ρ_s). In writing Eq. (1) we have assumed that the fluid volume element δV is a coarse-grained volume, which is large enough to contain many particles but is small enough such that the particle distribution inside δV is uniform. In this case, we have $\mathbf{f}\delta n(\mathbf{x}) = \Delta\rho \mathbf{g}[\phi(\mathbf{x}) - \phi_0]$, where $\phi(\mathbf{x})$ is the particle volume fraction and ϕ_0 is its mean value.

Nondimensionalizing Eq. (1) with respect to the length L, the time L^2/D, and the concentration ϕ_0, we have

$$-\frac{1}{\sigma}\nabla \delta p(\mathbf{x}) + \nabla^2 \delta \mathbf{u}(\mathbf{x}) = Ra\,\phi(\mathbf{x})\hat{\mathbf{z}}, \tag{2}$$

where the unit vector $\hat{\mathbf{z}}$ is directed upward opposite to the direction of $\mathbf{g}$, and the dynamic pressure δp has included a term, $-\Delta\rho g \phi_0 z$, to absorb contributions from the constant forcing term $-\Delta\rho \mathbf{g} \phi_0$. In Eq. (2) the Rayleigh number Ra is defined as

$$Ra = \Delta\rho g \phi_0 L^3/(\eta D), \tag{3}$$

where D is an effective diffusion constant of the particles. The Schmidt number σ is given by $\sigma = \nu/D$ with ν being the kinematic viscosity of the fluid. For a dilute suspension of small colloidal particles, D is approximately equal to the particle self diffusion constant $D_s = k_B T/(6\pi\eta a)$, where $k_B T$ is the thermal energy. For large non-Brownian particles, however, the effect of thermal agitations is negligible and their diffusion-like motion is produced by the hydrodynamic interactions between the particles [6]. Nicolai et al. have shown [7] that the hydrodynamic diffusivity has the form $D_h \simeq 5aU_0$, where $U_0 = 2a^2\Delta\rho g/(9\eta)$ is the Stokes velocity.

With the hydrodynamic diffusivity D_h, Eq. (3) becomes

$$Ra = 0.9\phi_0(\frac{L}{a})^3. \tag{4}$$

It should be mentioned that while it is cancelled out in Ra, $\Delta\rho g$ is needed so that D_h can be used to describe the hydrodynamic diffusion of the settling particles at small length scales. Equation (2) together with the continuity equation for an incompressible fluid

$$\nabla \cdot \delta \mathbf{u} = 0 \tag{5}$$

and the advective mass diffusion equation

$$\partial_t \phi + (\delta \mathbf{u} \cdot \nabla)\phi = \nabla^2 \varphi \tag{6}$$

complete the description of concentration and velocity fluctuations in colloidal sedimentation.

It is evident that Eqs. (2)-(6) are the same as those for buoyancy-driven convection [3]. Velocity and concentration fluctuations in colloidal sedimentation are therefore analogous to those in buoyancy-driven convection, and they are completely controlled by the two dimensionless parameters Ra and σ, once the boundary conditions are specified. We now estimate typical values of Ra and σ in colloidal sedimentation. In the experiment by Segrè et al. [1], the particle's radius $a \simeq 8\ \mu m$, Stokes velocity $U_0 \simeq 6.5\ \mu m/s$, volume fraction $\phi_0 \simeq 0.05$, and the characteristic sample size $L \simeq 1\ cm$. With these experimental values, we find $D_h \simeq 2.6 \times 10^{-6}\ cm^2/s$, $Ra \simeq 8.8 \times 10^7$, and $\sigma \simeq 3800$. The Schmidt number σ

is equivalent to the Prandtl number in thermal convection. Colloidal sedimentation is, therefore, associated with high Rayleigh number, high Prandtl number turbulent convection.

To understand the sedimentation dynamics, it is helpful to distinguish two characteristic length scales in convection: the viscous dissipation length δ_v and the diffusive dissipation length δ_d. The values of δ_v and δ_d are determined, respectively, by the transition Reynolds number $Re_c = \widetilde{\delta u}\delta_v/\nu$ and the transition Peclet number $Pe_c = \widetilde{\delta u}\delta_d/D_h$. Here $\widetilde{\delta u}$ is the rms value of the velocity fluctuation δu averaged over a volume of δ_v^3 (or $\delta_d{}^3$). It is the ratios of these lengths to each other and to the sample size L that determine the flow state of the system [2]. For high-Ra, high-σ turbulent convection, one anticipates that the flow consists of three different regions: (i) $a < \ell < \delta_d$, (ii) $\delta_d < \ell < \delta_v$, and (iii) $\delta_d < \ell < L$. In Region (i), molecular viscosity and hydrodynamic diffusivity determine the momentum and mass transport processes, respectively, and hence the particle distribution remains uniform without any large fluctuations. In Region (ii) turbulent (or eddy) diffusivity and molecular viscosity are dominant, and thus large fluctuations in particle concentration are expected but the velocity field remains relatively smooth. Finally, in Region (iii) turbulent diffusivity and viscosity both dominate over the corresponding hydrodynamic and molecular processes. In this case, one expects to see large fluctuations both in particle concentration and in velocity at different length scales.

We first discuss the length δ_d, above which velocities become large and concentration fluctuations are transported by convection. This occurs when the local Peclet number $Pe = \widetilde{\delta u}\ell/D_h$ becomes larger than Pe_c. Recent thermal convection experiments have shown [8] that while turbulent mixing creates on average an isothermal fluid in the turbulent bulk region, large temperature fluctuations still remain in the region and the characteristic length scale associated with these fluctuations is of the order of δ_d. Therefore, the velocity correlation length ξ is determined by δ_d in Region (ii). According to Kraichnan's theory [2],

$$\delta_d \simeq \left(2\pi^2 Pe_c^2\right)^{1/3} L Ra^{-1/3}, \qquad (7)$$

where the power law amplitude is expressed in terms of the numerical value of Pe_c. Priestley [9] first gave a direct argument for the $Ra^{-1/3}$ scaling. He argued that when Ra is large enough, δ_d should be a new length scale independent of the sample size L. With Eqs. (7) and (4), we immediately have $\xi \sim \delta_d \simeq LRa^{-1/3} \simeq a\phi_0^{-1/3}$. The mapping of colloidal sedimentation to turbulent convection, therefore, explains the experimental finding that $\xi \simeq 11a\phi_0^{-1/3}$. It also provides a physical interpretation for the existence of a velocity cut-off length, which prevents hydrodynamic dispersion coefficients from being divergent.

We now discuss the velocity variance $\widetilde{\delta u}$ in colloidal sedimentation. According to Kraichnan's theory [2],

$$\widetilde{\delta u} \simeq \frac{Pe_c D_h}{\delta_d} \simeq \frac{Pe_c D_h}{\left(2\pi^2 Pe_c^2\right)^{1/3} L Ra^{-1/3}}. \qquad (8)$$

Eq. (8) states that at the transition Peclet number Pe_c, the mass flux due to hydrodynamic diffusion, $D_h\widetilde{\delta\phi}/\delta_d$, is approximately equal to that by convection, $\widetilde{\delta u}\widetilde{\delta\phi}$. Because $\delta_d \simeq a\phi_0^{-1/3}$ and $D_h \simeq aU_0$, we find from Eq. (8) that $\widetilde{\delta v} \sim \widetilde{\delta u} \simeq D_h/\delta_d \simeq U_0\phi_0^{1/3}$, which is independent of the sample size L. This result agrees well with the experimental finding that $\widetilde{\delta v} \simeq 2U_0\phi_0^{1/3}$ [1].

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