Rheologica Acta
https://doi.org/10.1007/s00397-023-01421-z

ORIGINAL CONTRIBUTION

Progress and challenges in suspension rheology

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Received: 24 October 2023 / Accepted: 26 October 2023

Abstract
Developments in the last century, and especially in the last 50 years, have advanced understanding of suspension rheology greatly. Here, a limited review of suspension work over this period is presented, emphasizing advances over the last three decades in understanding of the particle pressure and strong shear thickening, which were motivated by crucial experimental observations, computational advances, and a critical review, all from the 1980s. This review serves as a preview to some outstanding challenges in suspension mechanics. This article considers primarily dispersions of spherical particles, which serve not only as a model material for understanding the rheology of more complex fluids of practical relevance, but also as a basic system for the study of nonequilibrium statistical physics.

Keywords Suspensions · Particle pressure · Microstructure · Shear thickening · Friction

Introduction

This paper follows, with some added detail, the April 2022 Weissenberg Award Lecture at the Annual European Rheology Conference in Seville, Spain. As in the lecture, the objective of the paper is to distill out certain main themes of my own work, in order to give a sense of what seems most important after struggling with the questions for some years. A suggestion of a few major challenges in the field of suspension mechanics will also be presented.

Attention in this work will be confined to rheology and statistical physics, as an earlier perspective considered the fluid mechanics of suspensions (Morris 2020b). However, shear-induced particle migration plays a primary role in connecting nonlinear rheology to fluid mechanics, and this phenomenon will be discussed.

Suspensions and a few of their applications

A suspension is defined as particles immersed in a fluid, which for practical purposes means particles in a liquid. This is not quite a precise concept, so some qualifications are in order. It is assumed in applying the term suspension that the particles stay suspended for some significant period of time. For the case where gravitational settling is rapid and agitation is needed to keep particles suspended, "slurry" is the commonly used term. The term "dispersion" is used for both a suspension and an emulsion (drops in liquid), and often implies that the particles or drops are sufficiently small that they remain suspended due to Brownian motion, and is then a colloidal dispersion. We will consider noncolloidal suspensions and colloidal dispersions here, as the main difference is in the size of the particles and resulting change in the types of forces that dominate the particle motion. More precise definition of the issue will follow in the "System of interest: the near-hard-sphere suspension" discussion below.

Many industrial and natural examples of suspensions can be found. In fact, in our workplaces, we are surrounded by the residue of suspensions, as wall paints are composed of particles dispersed in liquid. Flowable cement suspensions (Roussel et al. 2010) are precursors to solid cement, which of course plays a major role in concrete used in building and highway construction. In nature, mud and crystal-bearing magma are examples of suspensions appearing in geophysics, while blood is a suspension primarily of red blood cells (RBCs) in liquid plasma. Deformability of the RBCs makes blood somewhat distinct from the cases considered here, and this highlights the fact that to sort out the science of complex fluid behavior, we require well-defined systems. We turn next to this point.

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Published online: 21 November 2023

System of interest: the near-hard-sphere suspension

Suspensions can be of widely varied form as suggested by the range of applications discussed above. To develop the mechanical framework, we focus on a basic form of suspension, which nonetheless exhibits the primary rheological phenomena seen in more complex suspensions. This is a suspension made up of spherical particles with very limited-range surface forces, slight deformability, or some surface roughness. Such a suspension is perhaps the simplest of complex fluids, described at leading order by the solid fraction ϕ, the particle size (radius a) distribution, and the suspending fluid properties. Consistent with most of my own research, the discussion will only consider results involving Newtonian suspending fluids (of viscosity η_0), which substantially reduces the parameter space and thus simplifies the description. Because the nonhydrodynamic forces considered are short-ranged, i.e., they are inconsequential beyond a particle separation small compared to the particle radius, or the deformability is slight, this is termed the near-hard-sphere (NHS) suspension. As opposed to the hard-sphere (HS) model of suspensions, with truly rigid spheres immersed in a continuum fluid, this implies that not only hydrodynamic and Brownian forces play a role. Either short-range conservative or contact forces, the latter implying deformation and possibly friction, must also be considered. More technical development of this point is given in the next section.

If the density of particles differs from that of the fluid, gravitational settling (or rising) must be considered. While this is an unavoidable factor in many applications, for purposes of understanding the suspension as a material—and this is the primary interest in rheology—it is best to consider neutrally buoyant suspensions, in which particle and liquid densities are matched.

Intellectual interest in the neutrally buoyant NHS suspension arises in large part because it plays a role for a class of mixtures that is in the spirit of the HS fluid, which serves as a very basic system for defining statistical mechanical understanding of gases and liquids. Through the ability to eliminate gravitational effects that lead to spatially varying concentration and stress, the suspension is a more flexible model than dry granular materials, to which it has significant similarity under high-ϕ (often termed "dense") conditions (Boyer et al. 2011). The statistical physics objectives include reduction from a many-body to a few-parameter description of behavior. This includes development of predictive understanding of the microstructure and its relation to properties from the particle-scale mechanics, both in equilibrium and—of more interest to rheologists—under flow, as well as development of understanding of the basis for particle migration described later in the work.

Owing to the variety of forces and surface features that may be considered, the NHS suspension actually represents a class of materials in itself yet it is simple to parameterize, realizable in the laboratory, and has been accessible by computer simulation for almost four decades: the first publication on the Stokesian Dynamics method was Bossis and Brady (1984). Thus, the NHS suspension is a basic nonequilibrium system whose behavior can be related to balances between the relatively few forces at play with most of these localized to the near-contact regions between particle pairs. Furthermore, in the HS limit, the system has a well-defined equilibrium thermodynamics, with the osmotic pressure π a key quantity (Russel et al. 1995). The osmotic pressure plays a major role in the developments here when we consider its extension to shear flow, Π, defined in "Rheological quantities" section below in the Introduction.

Depending on the size of the particles, it may be necessary to consider Brownian motion. This is characterized by the Stokes–Einstein-Sutherland diffusivity $D = kT/6\pi\eta_0 a$, with kT the thermal energy. The relative importance of thermal motion to flow is defined in terms of a Péclet number $\mathrm{Pe} = 6\pi\eta_0\,\dot{\gamma}a^3/kT$, where the shear rate of the flow is $\dot{\gamma}$. For $\mathrm{Pe} \to 0$, the suspension approaches thermodynamic equilibrium. For $\mathrm{Pe} \gg 1$, the suspension is far from equilibrium and its behavior depends on Pe (i.e., on the driving rate $\dot{\gamma}$), the solid volume fraction ϕ, and the details of those forces that make the system near-hard rather than truly hard sphere.

It is further assumed in the discussion here that the flow at rheologically relevant length scales is Stokes flow, i.e., the particle-scale Reynolds number $\mathrm{Re}_p = \rho\dot{\gamma}a^2/\eta_0 \ll 1$. The importance of being near-hard rather than truly hard (i.e., rigid and without any finite-range forces) arises in part from the fact that Stokes flow reversibility implies that the non-Brownian suspension, at $\mathrm{Pe}^{-1} \to 0$, should exhibit fore-aft symmetry in its microstructure. Thus, roughly speaking, a system that is prepared with structural isotropy should be able to be returned to isotropy simply by shearing forward and backward the same strain. In reality, this reversible nature of the fluid mechanical interactions is an important but not a controlling feature: irreversibility and the loss of fore-aft symmetry are observed after any significant strain of a reasonably concentrated suspension, even one very close to the HS limit at large Pe. The reasons for this in terms of finite-range surface forces and residual Brownian motion (Brady and Morris 1997) as well as roughness (Da Cunha and Hinch 1996; Rampall et al. 1997; Wilson 2005) have been explored, as have the implications in terms of chaotic dynamics of the sheared system (Dasan et al. 2002; Drazer et al. 2002).

Rheological quantities

Suspensions, even of this simplest NHS form, exhibit a wide range of rheological phenomena. The discussion will be limited to steady-state rheological properties in this paper, and the quantities discussed and the notation used will be defined here. For purposes of defining the rheological response of the material, it is common to describe the bulk stress in suspensions (SOR Nomenclature 2013) by $\boldsymbol{\Sigma} = \boldsymbol{\Sigma}^{\mathrm{F}} + \boldsymbol{\Sigma}^{\mathrm{P}}$, where superscripts F and P denote the fluid and particle contributions. To be consistent with the literature, we will use σ for an imposed shear stress or for the local continuum stress at a particle surface in several places, while the $\boldsymbol{\Sigma}$ notation is reserved here for analysis of the suspension stress where we specifically address either the particle–fluid nature or the averaging over the two phases to obtain a bulk property.

The rheological quantities that will be considered here are the shear viscosity and the normal stress response. The bulk stress in a shear flow with $u_1 = \dot{\gamma} x_2$ can be written

$$\Sigma = \begin{pmatrix} \Sigma_{11}^{\mathrm{P}} & \Sigma_{12} & 0 \\ \Sigma_{21} & \Sigma_{22}^{\mathrm{P}} & 0 \\ 0 & 0 & \Sigma_{33}^{\mathrm{P}} \end{pmatrix}$$

with 3 denoting the vorticity direction. Because the fluid is Newtonian, it does not contribute directly to the normal stress response. The standard rheometric functions are the apparent suspension viscosity, $\eta_s = \boldsymbol{\Sigma}_{12}/\dot{\gamma}$, and the normal stress differences:

$$N_1 = \Sigma_{11}^{\mathrm{P}} - \Sigma_{22}^{\mathrm{P}}, N_2 = \Sigma_{22}^{\mathrm{P}} - \Sigma_{33}^{\mathrm{P}}.$$

The viscosity is often expressed as the relative viscosity $\eta_r = \eta_s / \eta_0$. The mean particle normal stress, or particle pressure, is related to the osmotic pressure π and will also be discussed. This is given by

$$\Pi = -\frac{\Sigma_{11}^{\mathrm{P}} + \Sigma_{22}^{\mathrm{P}} + \Sigma_{33}^{\mathrm{P}}}{3}.$$

Outline

The growth of understanding of two particular topics that have occupied much of my time since about 1990 will be considered in "Recent decades in suspension rheology" section. To begin, however, earlier developments including both crucial technical advances as well as puzzling observations will be described in "A brief historical background up to 1990" section to set the stage. The work will conclude in the "Challenges" section with a discussion of some broad challenges in the study of suspensions.

A brief historical background up to 1990

The purpose in this section is to provide a brief general background and then highlight certain specific developments (numbered below in this section) that were critical foundations for my own work in suspension mechanics.

In opening a discussion of suspensions, it is common to recall that the analytical study of suspension properties dates to Einstein (1906). This work showed that the added dissipation due to a spherical particle immersed in a deforming fluid could be used to define an effective viscosity of the dilute suspension, yielding the celebrated result $\eta_E(\phi) = \eta_0(1 + 5\phi / 2)$. This was not an isolated calculation, but a part of a body of work developed to relate the transport and thermodynamic (osmotic pressure) properties of suspended particles. While a number of results, for example, for single particles in confinement and for pair interactions, appeared earlier and are gathered in the text of Happel and Brenner (2012), there was a gap of many years between the publication of Einstein's viscosity result and the development of a full stress-system analysis presented by Batchelor (1970). This work provides a foundation for description of the ensemble-averaged stress associated with the various mechanisms of stress generation—each associated with a specific type of force or surface traction—due to suspended particles. This general theory was coupled with dilute-ϕ flow-induced particle structure to describe the influence of the particles on the stress for non-Brownian (infinite-Pe) hard spheres in Batchelor and Green (1972a, b) and for weakly sheared Brownian ($\mathrm{Pe} \ll 1$) dilute dispersions in Batchelor (1977). This body of work by Batchelor laid the foundation for structural studies that relate closely to the normal stress response and more will be said about this topic in the "Recent decades in suspension rheology" section.

Published experimental studies of the flow properties of suspensions date at least to work on colloidal dispersions by Bingham and Robertson (1929), and this work was noted in the study of strong shear thickening (described by the term "dilatancy") exhibited by concentrated dispersions in work by Freundlich and Röder (1938). A significant part of suspension study for many years after that time focused on the ϕ dependence of the steady-shear viscosity, and from this arose the well-known formulas of Maron and Pierce (1956) and Krieger and Dougherty (1959), with the dependence as $\eta_s/\eta_0 \propto (\phi_{\mathrm{J}} - \phi)^{-\alpha}$ where α is typically near 2; here, ϕ_{J} is the maximum packing fraction, now more commonly called the jamming fraction, at which the apparent viscosity diverges. However, significant variation in the measured relative viscosity η_s/η_0 was seen, with order of magnitude variation at given ϕ between separate researchers, even though reproducibility by a single laboratory (working with a specific material) might be good (Thomas 1965). Furthermore, while the empirical forms captured the general behavior, different values of the maximum packing fraction were found for different realizations of suspensions composed of nominally monodisperse hard spherical particles: this was striking to me as a novice in suspensions, as it meant that a rheology depending on only the concentration variable ϕ and the distance from equilibrium Pe (if Brownian motion is relevant) was not sufficient. Clearly, other forces are at play, and thus, other dimensionless parameters than just ϕ and Pe must be considered. The HS model for suspensions implies hard spheres in a Newtonian continuum liquid so that only hydrodynamic and Brownian forces are active if a lubricating film avoids contact interactions. On the one hand, an essential point is that this model proves too restrictive to describe the actual observed rheology even of systems designed to approach the HS limit. But, on the other hand, working as close as possible to the HS model allows development of the most basic and general results, and this led to my focus on the NHS suspension. In simplest terms, the NHS suspension implies particles in which the suspended particles may have short-range forces, slight deformability (i.e., be slightly nonrigid) or have surface features such as roughness that make contact. Considering again the dimensionless description, roughness and contact can introduce a classical friction coefficient, itself a dimensionless parameter, or the ratio of roughness length scale to particle size. For the forces, the characteristic shearing force σa^2 can be compared to the short-range force scale F to form a group $\sigma a^2/F$. For deformable particles, a relevant balance is σ/G, where G is an elastic modulus of the particles, but this is not considered here. It is important to note that slight deformation induced when hard (but not truly rigid) particles are pressed together is important to create "flat" zones and enduring contacts relevant for frictional interactions, but highly deformable objects, such as RBCs, drops, or microgels, tend to have well-lubricated surfaces. The implications of this in concentrated suspensions, for wall slip (Cloitre and Bonnecaze 2017) and other rheological phenomena (Malkin et al. 2004; Seth et al. 2011; Malkin and Kulichikhin 2015), differ substantially from those of surface interactions of interest for the NHS suspensions considered here.

It is important for understanding the foundation on which rests much of the work described in this paper to highlight two critical research developments of the 1980s, and a review appearing at the end of the decade. Each played a major role in the advances seen in areas of suspension mechanics that have occupied much of my attention.

1. **Shear-induced migration.** Experimental evidence of striking non-Newtonian behavior that impacted upon the spatial distribution of the liquid and solid components of a suspension was published by Leighton and Acrivos (1987). This work described the observation of a slow evolution of viscosity of a concentrated suspension of nominally hard spheres, measured in a Couette (cup-and-bob) apparatus, and the authors showed the basis of this to be a shear-induced migration. The particles progressively moved from the high-stress region in the annular gap to the low-stress region under the bob, reducing ϕ in the annular gap and hence reducing the measured viscosity. The importance of this behavior to practical scenarios and the fundamental concept of a stress-driven flux considerably widened the scope of study in suspensions. Earlier work by Gadala-Maria and Acrivos (1980), showing the development of flow-induced structure and its impact on the measured viscosity in shear reversal experiments, was seen in retrospect to be related to these phenomena, as was work in the Gadala-Maria (1979) thesis on measurement of normal stress differences in concentrated suspensions.
2. **Stokesian Dynamics simulation**. The current understanding of the phenomena just noted was ultimately strongly influenced by simulation studies that spring from the development of Stokesian Dynamics (SD) (Bossis and Brady 1984; Brady and Bossis 1985, 1988) and related methods (Ladd 1988) appearing in the 1980s for the simulation of Stokes flow suspensions. The ability to simulate the motions of particles, even in the limited numbers accessible at that time, was eye opening. These simulation approaches have had influence on understanding of the rheology and dynamics of dispersions analogous to that of molecular dynamic (MD) simulations on understanding of statistical physics of gases and liquids, which began in the 1950s (Alder and Wainwright 1957). The SD method came much later, largely because of a need to describe the forces due to the continuum fluid between particles. To capture these hydrodynamic interactions in an efficient way that allows tracking only the macroscopic particle degrees of freedom, we now rely on the pair-particle resistance and mobility functions formalized 1980s by Jeffrey and Onishi 1984) and Jeffrey (1992), and used in SD implementations even before their publication.
3. **A critical review by** Barnes (1989) considered about 100 papers on shear thickening (ST) in concentrated suspensions. The phenomenon of ST, even extreme and discontinuous ST (or DST), had long been known (although terminology was different) as mentioned above (Bingham and Robertson 1929; Freundlich and Röder 1938). However, DST was shown to be observable in carefully synthesized spherical particle suspensions by Hoffman (1972), work that was seminal to development of understanding of ST. The systematic analysis by Barnes

established features of the shear thickening response, including a stress scaling for onset of thickening as $\sigma_c \sim a^{-2}$. This challenged the community to understand the force balance giving rise to such a scaling (Boersma et al. 1990; Maranzano and Wagner 2001).

Recent decades in suspension rheology

While the phenomena to be discussed here are rheological in nature, they involve application of thermodynamic ideas to far-from-equilibrium suspensions. Two types of change in the "state" of the material arising as the result of flow are emphasized.

The first of these changes in material state is the "demixing" with time under shear noted in point 1 of the prior section. When I started my own research career as a doctoral student in 1990, shear-induced migration (Leighton and Acrivos 1987) was a new concept. Although it was soon developed into a widely used model based on particle flux to regions of lower shear rate, $j \sim -\nabla\dot{\gamma}$, by Phillips et al. (1992), it was not based in familiar mass-transfer principles, e.g., the relation of species flux to a gradient in its chemical potential. The chemical potential of hard-sphere systems is closely related to osmotic pressure, and consideration of the nonequilibrium osmotic pressure, or particle pressure, is intertwined with my study of particle migration.

The second change in material state is rate dependence of the very abrupt form DST. In DST, the shear and normal stress (Seto et al. 2013) of a dense suspension undergo discontinuous changes at some critical rate $\dot{\gamma}_c(\phi)$. This phenomenon caught my attention rather sharply as a result of discussions with a post-doctoral groupmate, Dr. Willem Boersma, who in his PhD studies at Eindhoven had shown that this behavior was accompanied by extreme temporal fluctuations in the stress (Boersma et al. 1991) for $\dot{\gamma} \approx \dot{\gamma}_c$. This was suggestive of behavior seen at an equilibrium critical point and raised for me the compelling question of whether DST has features of a phase transition.

Shear-induced migration and the particle pressure

As noted above, shear-induced migration was clearly identified by Leighton and Acrivos (1987), and the particle flux resulting in a gradient in ϕ within an initially well-mixed suspension was described as $\mathbf{j} \sim -\nabla\dot{\gamma}_c$. With the mass conservation equation coupled to the momentum balance through the viscosity dependence $\eta_r(\phi)$, this work opened the way to suspension flow modeling that was intrinsically different from a generalized Newtonian model. The consequences of particle migration, as exemplified in Fig. 1 for a pressure-driven flow, could be significant, with large variation of ϕ, although the blunting of the velocity at this bulk $\phi \doteq 0.3$ is mild as seen at right in Fig. 1; this figure is adapted from Monsorno et al. (2017), whose work was based on two-fluid modeling (Morris and Boulay 1999; Miller et al. 2009).

The description of migration flux as $\mathbf{j} \sim -\nabla\dot{\gamma}_c$ does not allow one to make predictions of migration based on other measurable rheological properties of the suspension. This objection was addressed in work by Nott and Brady (1994), through a relation of flux to the divergence of the particle stress, $\mathbf{j} \sim \nabla \bullet \mathbf{\Sigma}^{P}$, or most simply to the pressure associated with the particle phase, $\mathbf{j} \sim -\nabla\Pi$. In this work, and later in Morris and Brady (1998), the particle pressure, as it has become known, was constitutively related to the particle fluctuational motion as $\Pi \sim T_s^{1/2}$, where the "suspension temperature" is $T_s = \langle \boldsymbol{u}'.\boldsymbol{u}' \rangle$ and $\boldsymbol{u}'$ is the fluctuation of a particle's velocity from the local affine motion. This concept was considered earlier in the context of suspensions by Jenkins and McTigue (1990) based on normal stress measurements linear in shear rate by Bagnold (1954), with the Stokes-flow scaling demanded by dimensional analysis $\Pi \sim \eta_0 T_s^{1/2}/a$. The suspension temperature is an analogy to the granular temperature, itself an analogy to the

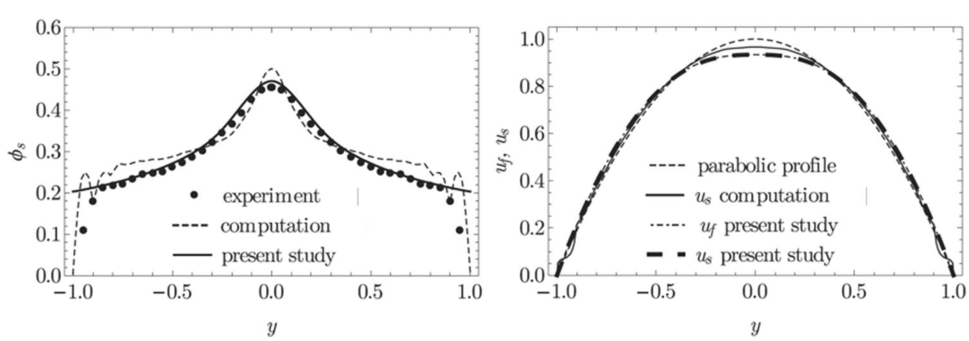

Fig. 1 Poiseuille flow of a suspension, $\phi_{bulk} = 0.3$; plots adapted with permission from Monsorno et al. (2017). At left is local solid fraction, denoted ϕ_s, and at right the velocity, with u_f for the fluid and u_s for the particles (solid). The curves labeled "present study" are based on two-fluid modeling (Morris and Boulay 1999; Miller and Morris 2006), while the "computation" results are from Yeo and Maxey (2011), which included the displayed experimental data of J. F. Gilchrist

thermodynamic temperature in kinetic theory of molecular fluids. In later work, the direct relation of the particle normal stress to the bulk shear rate (Morris and Boulay 1999), allowing for nonlocal-in-$\dot{\gamma}$ contributions (Miller and Morris 2006) to smooth the tendency toward maximum packing at places where the local mean shear rate vanishes (as used by Monsorno et al. (2017) with the desired effect at the centerline in Fig. 1a), was found to be effective and simpler as it reduces the number of fields and boundary conditions.

As argued in Yurkovetsky and Morris (2008), the (double) analogy to molecular kinetic theory is not the most fruitful relation for description of the viscous suspension particle pressure: the osmotic pressure is a more direct route. The osmotic pressure of a HS suspension at equilibrium, i.e., at $\mathrm{Pe}=0$, is a well-established thermodynamic quantity and for monodisperse spheres of radius a at volume fraction ϕ is $\pi = nkT\,[1+4\phi g(2a)]$; the pair distribution function at contact, $g(2a)$, represents the influence of excluded volume. In the early 1990s, Brady considered the relation of Brownian osmotic pressure (Brady 1993) to the hydrodynamic interactions between particles. This work made use of results that we (Jeffrey et al. 1993) developed to extend the hydrodynamic interaction functions relating the trace of the particle stress, the mechanically defined particle pressure $\Pi = -\Sigma^{\mathrm{P}}_{ii}/3$ described in the Introduction, to the configuration and the kinematics. The hydrodynamic functions are independent of the flow state, but the microstructure—the mean configuration, roughly speaking—depends on the interaction of flow with Brownian motion (Pe) and with various interparticle forces. We come to the microstructure in the following section.

The particle pressure was now well-founded in terms of the hydrodynamic traction moment (the symmetric part, or stresslet), $\Pi^{\mathrm{H}} \sim sym \int \boldsymbol{x} \bullet \sigma \bullet \boldsymbol{n} d$ S taken over the particle surface S_{p}, and moments of other interparticle forces $\Pi^{\mathrm{P}} \sim \boldsymbol{x} \bullet \boldsymbol{F}_P$, as well as Brownian motion. Thus it was possible, in Yurkovetsky and Morris (2008), to demonstrate that the same formulation applied to evaluation of Π both at equilibrium and in shear flow of Brownian hard spheres: the particle pressure is the thermodynamic osmotic pressure at $\mathrm{Pe}=0$, and is simply the nonequilibrium osmotic pressure under flow. The Brownian contribution Π^{B}/nkT as a function of ϕ for a large range of Pe is shown in Fig. 2a. At $\mathrm{Pe}=0.1$, this contribution is asymptotic to the hard-sphere result $\Pi^{\mathrm{B}}/nkT \sim \pi_{\mathrm{eq}}/nkT = 1+4\phi g(2a)$; because of the increasingly strong pair correlation near contact (see Fig. 4), Π^{B} increases with Pe. The hydrodynamic contribution, Π^{H}, is shown in Fig. 2b. Note that this quantity scales with $\eta_0\,\dot{\gamma}$, and $\Pi^H/\eta_0\dot{\gamma}$ begins increasing rapidly from a very small value at $\phi \approx 0.3$, consistent with the difficulty of measuring the small normal stress differences at $\phi < 0.3$ (Zarraga et al. 2000; Gamonpilas et al. 2016).

To measure the particle pressure requires discriminating between the phases. This is seen in the fact that a tendency of the particles to spread, implied by a positive Π, requires that the liquid tend to be sucked into the region as the particles tend to leave it. Thus, the two components, particle and liquid, are in opposing normal stress states. As a consequence, if we conceive of the suspension stress as a sum of the component contributions (as successfully done for the shear stress), the implications of the particle pressure are obscured. This is related to the mean pressure of the bulk suspension being indeterminate based on incompressibility of the materials (not of the phases, which may change density, e.g., ϕ), leading Batchelor (1970) to dismiss the isotropic stress in a suspension. However, the perspective is changed if we realize that the measurement of osmotic pressure, which owes its presence to material dispersed in a liquid, is performed by sampling the liquid response: consider a U-tube osmometer in which the pure solvent liquid is sucked into the solution (or dispersion) leg through a semipermeable membrane. This idea that multiphase pressures must discriminate between the phases was central in my own work to measure the shear-driven particle pressure (Deboeuf

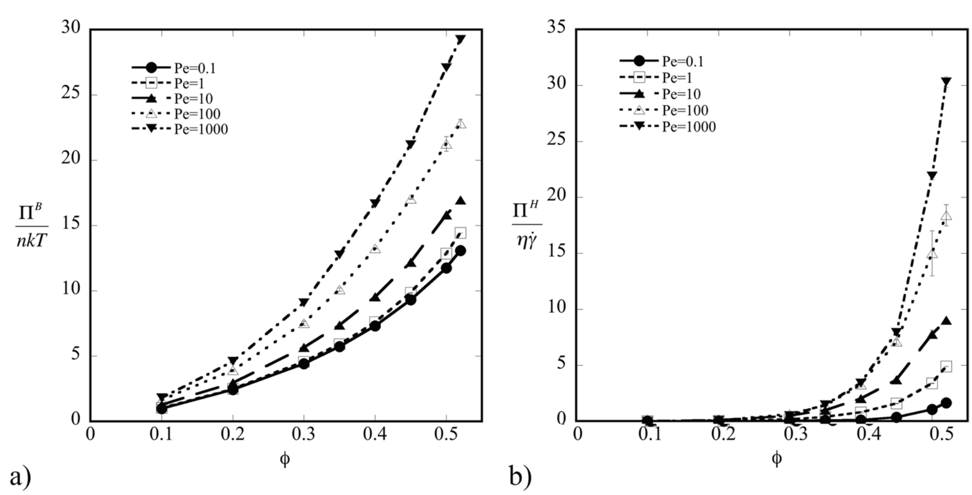

Fig. 2 Contributions to the particle pressure as a function of solid fraction ϕ in a hard sphere suspension. **a** Brownian contribution Π^B/nkT and **b** hydrodynamic contribution $\Pi^H/\eta^0\dot{\gamma}$ (in the plot, η is the pure fluid viscosity). Reprinted with permission from Yurkovetsky and Morris (2008)

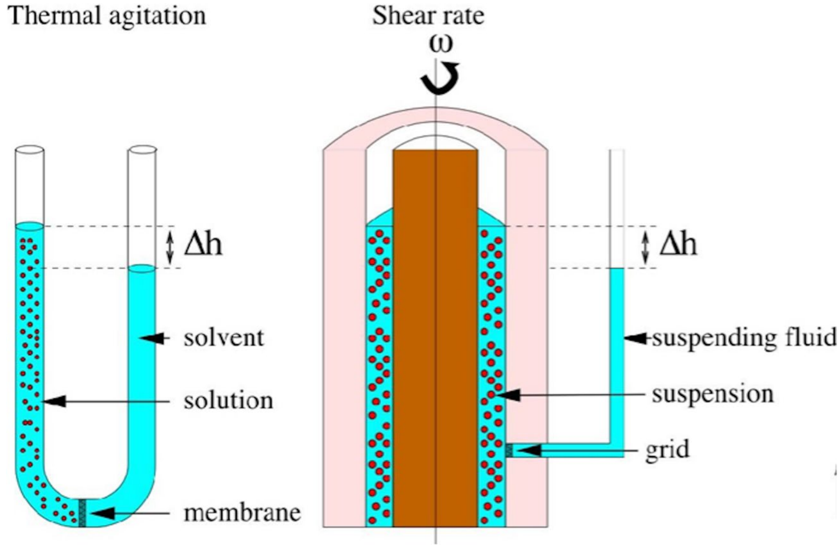

Fig. 3 Osmotic measurement of particle pressure. At left is a schematic of a U-tube osmometer, employing a semipermeable membrane to keep solute from passing, with Δh related to the thermally driven osmotic pressure, $\pi \sim nkT$; at right is a schematic particle-pressure measurement employing a grid (or screen) to maintain the particles in the suspension (Deboeuf et al. 2009; Garland et al. 2013) and Δh related to the particle pressure $\Pi \sim \eta_0 \dot{\gamma}$. Adapted with permission from Deboeuf et al. (2009)

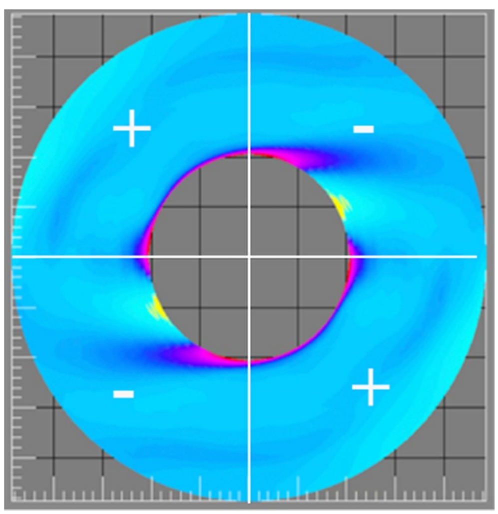

Fig. 4 Pair distribution function $g(r)$ of a strongly sheared, Pe = 1000, Brownian hard-sphere suspension of $\phi = 0.3$ with associated sign map for Π. The grey circle is at radius $r/a = 2$ with a the particle radius. The shear flow is to right at top and left at bottom. The elevated values in compressional (second and fourth) quadrants form a boundary-layer structure and correspond to the regions where $\Pi > 0$, while the depleted wake regions in extensional quadrants have $\Pi < 0$, leading to the observed bulk $\Pi > 0$. Adapted with permission from Morris and Katyal (2002)

et al. 2009; Garland et al. 2013). Here, the liquid suction pressure under various ϕ and $\dot{\gamma}$ was measured across a screen playing the role of a semipermeable membrane; see Fig. 3. A key earlier demonstration of the essential idea applied in this work was found in a study by Prasad and Kytömaa (1995), who used a porous plate to drive a shear flow, so that the liquid stress equilibrated and only the particle stress was measured; Boyer et al. (2011) used this approach to probe the rheology of suspensions up to the limit of jamming, thereby providing unifying understanding of granular and suspension properties through ideas of the internal or bulk friction given by the ratio of shear to normal stress $\mu = \sigma/\Pi$.

The idea of a flow-driven dispersed phase pressure in multiphase mixtures has a long history (Wallis 1969). However, over the last 30 years, its theoretical development and both conceptual and quantitative demonstration by experiment have established the particle pressure as a basic property in suspensions. When shear rate varies in a flow at an initially uniform solid fraction ϕ, as in Poiseuille flow of a suspension, the resulting $\nabla\Pi$ drives a particle flux toward the centerline to alleviate this gradient, by generation of elevated ϕ where $\dot{\gamma}$ is small as illustrated by Fig. 1. This simple idea captures the essence of an important multiphase phenomenon. As a consequence, the concept has found use in practical applications such as cross-flow filtration (Vollebregt et al. 2010). It is also seen in theoretical efforts to generalize the Stokes–Einstein-Sutherland diffusivity to sheared dispersions (Chu and Zia 2019). These two directions emphasize the point, alluded to in the opening of this section, that while its relation to normal stress differences is rheological, the influence of particle pressure on the shear-induced motion of particles is similar to a chemical potential and thus calls to mind thermodynamic concepts.

Particle microstructure

The development of shear-driven particle pressure or normal stress differences in a suspension of spheres requires loss of the fore-aft symmetry expected under pure hydrodynamic conditions, and in the current context, it seems appropriate to include some discussion of the microstructure here.

The strong microstructural asymmetry seen under strong shear (Pe = 1000) at $\phi = 0.3$ is illustrated by the pair distribution function in Fig. 4, with the sign of the dominant Π^{H} shown in each quadrant of the pair interaction. The near-contact structure is very strongly enhanced in the compressional (second and fourth) quadrants with $g > 300$ here. This is over 100 times its equilibrium value, and thus, there is a larger mean contact value than at $\mathrm{Pe} = 0$. The observed positive shear-driven pressure (due to asymmetry) as well as the basis for the enhanced Brownian contribution (due to the larger mean of $g(2a)$) can thus be rationalized. A distinct boundary layer (in the compressional quadrants) and wake (in extensional quadrants) structure of $g(\boldsymbol{r})$ is found for these conditions, as will be elaborated below.

The observation of the breaking of fore-aft symmetry and the development of non-Newtonian rheology were noted in the earliest SD simulations of two-dimensional (monolayer) suspensions (Bossis and Brady 1984), where the source of asymmetry was a short-ranged repulsive force. Exploration of the microstructure in suspensions requires considering varying rates of shear in relation to Brownian motion or such surface forces, and this rate dependence is a challenge less often (Hanley et al. 1987; Banetta and Zaccone 2019) considered for molecular liquids. While understanding of structure has expanded greatly as computational power has opened new possibilities in simulation, and analytical approaches have provided insight, this remains a topic where much remains to be developed at a predictive level.

In the study of particle microstructure, by which we mean the average spatial arrangement of particles, the large majority of work has focused on $g(\boldsymbol{r}) = P_{11}(\boldsymbol{r})/n^2$. Here, $P_{11}(\boldsymbol{r})$ is the probability of finding a second particle at a position $\boldsymbol{r}$ given the presence of a particle at the origin, normalized by the uncorrelated probability n^2 of finding two particles at these positions. For spherical or other isotropic-particle suspensions, $g(\boldsymbol{r})$ provides both information on the anisotropy and the radial accumulation. This work was initiated by Batchelor and Green (1972a, b), who considered the case of the pair probability of purely hydrodynamically interacting particles in extensional flow, rather than simple shear which leads to closed pair trajectories and an indeterminate probability distribution without a diffusive flux. This calculation showed that hydrodynamics alone results in a change in the radial distribution, in fact scaling at contact as $g \equiv p_\infty \sim (r/a - 2)^{-0.78}$ (this scaling arises from a combination of hydrodynamic functions), but isotropy is retained, i.e., $g(\boldsymbol{r}) = g(r)$ for "pure hydrodynamic" interaction at $\mathrm{Pe}^{-1} = 0$. The contact singularity in this pure hydrodynamic pair distribution is integrable, and thus, the influence on the rheology is finite (Batchelor and Green 1972b).

In the opposite limit, Batchelor (1977) considered weak-shear (small-Pe) perturbation of the equilibrium pair structure. Writing $g = g_{eq}(r)[1 + \mathrm{Pe} f_1(\boldsymbol{r}) + \ldots]$, with the leading-order term $f_1 = -h_1(r)\, \hat{\boldsymbol{r}} \cdot \hat{\boldsymbol{E}} \cdot \hat{\boldsymbol{r}}$, where $\hat{\boldsymbol{r}}$ is the unit vector along the line of centers of a pair of particles, and $\hat{\boldsymbol{E}}$ is the dimensionless strain rate. Owing to the quadrupolar symmetry of the disturbance to the isotropic g_{eq}, the leading correction does not impact on the viscosity. However, it does influence the normal stresses of the suspension at $O(\phi^2 \mathrm{Pe})$, as shown in Brady and Vicic (1995).

The low-Pe analyses reveal perturbations extending far from particle contact, with $f_1 \sim r^{-3}$ in the noted solution (Batchelor 1977). By contrast, Brady and Morris (1997) perturbed the Batchelor and Green (1972b) solution $p_\infty(r)$ with weak Brownian motion, i.e., $\mathrm{Pe}^{-1} \ll 1$ so that shear flow is dominant. This results in a classic narrow boundary-layer solution for $g(\boldsymbol{r})$ in the compressional quadrants of a pair interaction, which was found to scale as $g(2a) \sim \mathrm{Pe}$ over an $O(\mathrm{Pe}^{-1})a$ thickness layer. From this, we rationalized that the pure hydrodynamic limit is singular to essentially any perturbation, with significant implications for not only rheology, but also irreversibility and diffusion. The structural predictions were in qualitative agreement with experiments from Parsi and Gadala-Maria (1987). Simulations at large ϕ have shown similar boundary-layer structure (see Fig. 4), but with a weaker $g(2a) \sim \mathrm{Pe}^{0.7}$ scaling (Morris and Katyal 2002), and also show the pair-depleted wake.

The dilute-limit analyses noted above used the Smoluchowski (differential) equation (SE) to describe $g(\boldsymbol{r})$, and these provided important guidance to the development of my own colloidal rheology modeling (Frank et al. 2003). To reach large ϕ, later work by Nazockdast and Morris (2012) accounted for the influence of the surrounding bath of particles on the interaction of a pair. For this, an integro-differential form of the pair SE was developed; the integral portion captures the forces on the pair due to the bath. This allowed predictions in satisfying agreement with SD simulations of pair structure and the viscosity as well as normal stress response for $\phi \leq 0.55$ and $\mathrm{Pe} \leq 1000$. Related high-ϕ calculations of nonequilibrium structure and rheology include approaches of Brader et al. (2008) based on mode coupling and Scacchi et al. (2016) based on dynamic density-functional theory.

The finding of large pair correlation in compression shown in Fig. 4 suggested that true contact is highly likely and motivated the approach to shear thickening described in the "Extreme shear thickening" section. Two other studies providing crucial insight and motivation are noted. One is a study showing the difficulty of maintaining a liquid film between particles in a purely Stokes flow shearing motion at large ϕ, described as "lubrication breakdown" by Ball and Melrose (1995); a second key study, showing the role of even small induced roughness in causing major change in the rheological properties of concentrated suspensions, is described by Lootens et al. (2005). A change from classical lubrication to a contact interaction emerged as a valuable candidate for investigation.

When contact is allowed in a viscous suspension simulation, as illustrated in Fig. 5 for a low-stress (unthickened) and high-stress (thickened) state, the tenuous structures of

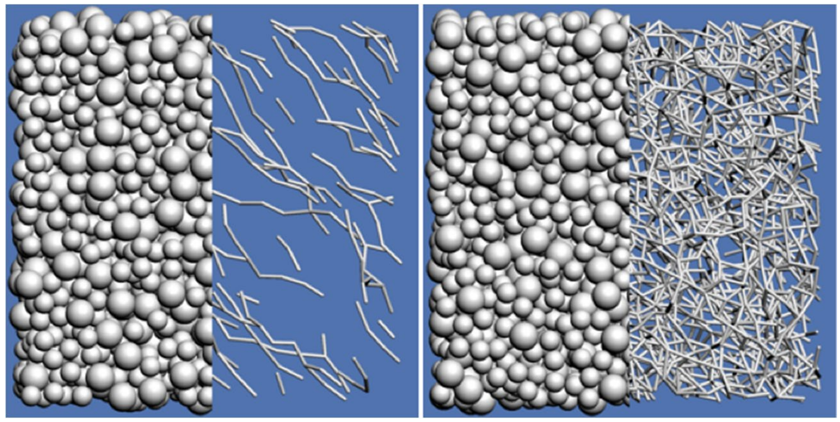

Fig. 5 Particle configurations and frictional contact force network from simulated suspension shear flow (to right at top and to left at bottom) at conditions ($\phi = 0.54$ and friction coefficient $\mu = 1$) displaying strong continuous shear thickening (Seto et al. 2013). At left, a low viscosity state at shear rate just below shear thickening, $\dot{\gamma} < \dot{\gamma}_c$, and at right a high viscosity state at $\dot{\gamma} > \dot{\gamma}_c$. Adapted with permission from Morris (2018)

contacts along the compressional direction (in agreement with the expected initial direction from elevated pair correlation in compression in Fig. 4) grow and proliferate to form a much denser and more isotropic network. A key observation is that, according to the simulation model (Seto et al. 2013; Mari et al 2014) detailed in the next section, some of the contacts in the system require hydrodynamic force and hence flow to occur. Thus, the percolating contact force network breaks and reforms continuously.

Extreme shear thickening

The extreme forms of shear thickening in dense suspensions, whether strong continuous shear thickening (CST) or DST (Freundlich and Röder 1938; Hoffman 1972; Bender and Wagner 1996; Cwalina and Wagner 2014), have long challenged our understanding of viscous suspension flow. The review of Barnes (1989) pointed out the need for a balance to explain the onset of thickening at a critical stress $\sigma_c \sim a^{-2}$. A simple way of understanding this is that a repulsive force of maximum magnitude F_R is effective in balancing the force due to the shear flow of σa^2 until $\sigma_c a^2 \sim F_R$, thus providing a microscopically defined critical stress scale $\sigma_c \sim F_R / a^2$.

Several studies provided detailed examinations of the possible basis for this scaling (Boersma et al. 1990; Maranzano and Wagner 2001; Kaldasch and Senge 2009). The predictions for the dependence of σ_c on a differ depending on whether the force balancing the external stress is due to surface electrostatic charge, steric effects arising from material grafted or adsorbed to the surface, or Brownian motion (Morris 2020a). Since Brownian motion alone (i.e., without other forces) is not sufficient to explain the observations (Morris and Katyal 2002; Mari et al. 2015a), while the repulsive force of steric or electrostatic origin has size dependence, it remains unclear why such a scaling with a seemingly constant F_R works.

Nonetheless, the scaling developed by Barnes (1989) based on a large body of early work has been more recently confirmed, for example, by Guy et al. (2015). In addition to a critical stress, there is a need for a qualitative change in the dominant mechanism of stress generation to explain DST. The key idea that has been developed over the last decade is that, for $\sigma > \sigma_c$, this takes the form of a frictional interaction, so that the shear thickening involves a lubricated-to-frictional transition in the particle interactions (Morris 2018). In simplest form, which happens to be the commonly used simulation approach (Mari et al. 2014, 2015a), this means that the ratio of tangential to normal force at the contact $F_{tan}/F_{nor} \leq \mu$, with μ a friction coefficient. The contact forms in this model when the shear force driving a pair together is $\sigma a^2 > F_R$. The range of behavior that may be captured by this description is illustrated in Fig. 6. Most NHS suspensions exhibit some shear thinning, and recent work has emphasized that this may be a result of a stress-dependent

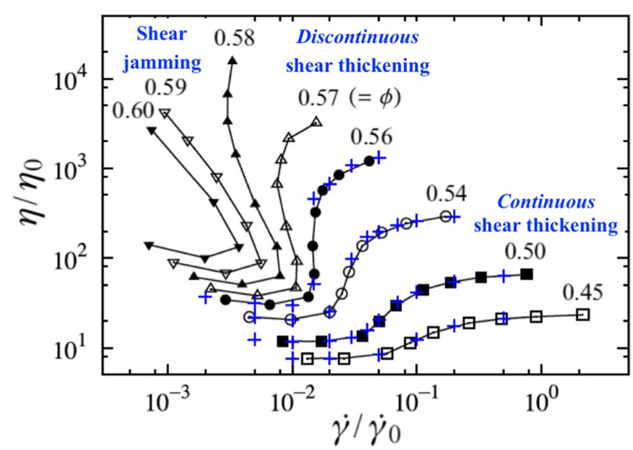

Fig. 6 Range of flow curves found in the lubricated-to-frictional scenario (Mari et al. 2014; Morris 2018), here for an interparticle friction coefficient of $\mu = 1$ and equal parts by volume particles of radii a and $1.4a$. From low to high ϕ, the system displays continuous shear thickening, discontinuous shear thickening, and shear jamming. Black symbols with lines are at controlled stress; blue + symbols are at controlled shear rate. Adapted with permission from Mari et al. (2015b)

interparticle friction coefficient (Lemaire et al. 2023), with measurements showing that the measured friction coefficient may depend in a qualitative way on the suspending fluid (or "solvent") (Le et al. 2023). A friction coefficient with stress dependence has been used in simulation to capture complex flow behavior (Khan et al. 2023).

It is worthwhile to consider the meaning of lubrication in discussion of a frictional interaction. For a lubricated contact between two spheres of radius a at surface separation ϵa with $\epsilon \ll 1$, the force depends on the pair relative motion with $F_{\mathrm{tan,lub}} \sim \eta_0 U_{\parallel} a \ln \epsilon$ and $F_{\mathrm{nor,lub}} \sim \eta_0 U_{\perp} a \epsilon^{-1}$ for tangential and normal velocities of the pair $U_{\parallel}$ and $U_{\perp}$, respectively. This indicates that $\mathrm{F_{tan,lub}/F_{nor,lub}} \sim \epsilon \ln \epsilon$, and as the name implies, lubricated tangential motion is much easier than normal motion. A different way to achieve an O(1) ratio of tangential to normal resistance to motion is to recognize that surface asperities on otherwise smooth particles approach with a normal motion when the carrying sphere motion is tangential. Thus, for very small separations, the tangential resistance for two spheres also scales as ϵ^{-1}, resulting in an O(1) effective friction coefficient as $\epsilon \to 0$ (Wang et al. 2020). This has the advantage that existing Stokesian Dynamics algorithms can be very simply altered to capture the effects of friction, but it is unclear whether this model can capture the static jamming state, as it retains lubricating layers.

The introduction of contact friction between particles reduces the jamming solid fraction, consistent with the fact that the number of contacts per particle needed for jamming of spheres is $Z = 6$ if the contacts have only a normal force, whereas $4 \leq Z_{\mathrm{fric}} < 6$ for frictional contacts, with small values needed for larger friction coefficient (Papanikolaou et al. 2013). This is a crucial element in the modeling of the rheological transition at DST by Wyart and Cates (2014), as the viscosity is modeled as $\eta/\eta_0 \sim [\phi - \phi_m(\sigma)]^{-2}$, with $\phi_m(\sigma) = f(\sigma)\phi_J^{\mu} + [1 - f(\sigma)]\, \phi_J^0$ with $\phi_J^{\mu}$and ϕ_J^0 the frictional and frictionless jamming fractions. The interpolation of the stress-dependent jamming fraction makes use of the "fraction of frictional contacts," denoted $f(\sigma)$ and shown to have a sigmoidal form going from $f = 0$ at low stress to $f \to 1$ at high stress (Mari et al. 2014); the original model of Wyart and Cates defined f in terms of the particle pressure, but an O(1) ratio of shear to normal stress makes the forms interchangeable.

Simulations (Seto et al. 2013) and theory (Wyart and Cates 2014) have converged on an interaction that has the appearance of contact forces inclusive of friction as the dominant stress-generating mechanism at stresses that overcome a repulsive interparticle "barrier" force. Experimental evidence of the role of contact is largely indirect, but includes influence of roughness (Hsiao et al. 2017; Hsu et al. 2018), behavior in strain reversal (Lin et al. 2015), and the influence of oscillatory orthogonal (Lin et al. 2016). The behavior under strain reversal and orthogonal shear presumably results from breaking of contact networks, such as that illustrated in Fig. 5.

Challenges

The perspective presented in the work described here is that NHS suspensions have a special role in development of our understanding of complex fluids, because they are the simplest of this class of materials. As such, they allow us to explore in detail the basis for non-Newtonian rheology and its impact on mixture fluid mechanics, while also providing a tractable system for study of nonequilibrium statistical physics issues, for example, the basic issue of the microstructure under shear. Numerous directions of study could be mentioned, but here the discussion is limited to three broad challenges:

1. **Development and validation of continuum models of suspensions.** This area of study has significant scope for advancement, and the motivations for advancements are strong from both fundamental and practical perspectives. Modeling the flow of suspensions began in earnest as a result of the clear description of shear-induced migration, and this modeling is challenging on multiple levels. These include the fact the materials may vary from fluid to soft solids with extreme rate dependence, the multiphase nature of suspensions becomes a key factor because of migration, and the difficulty in establishing boundary conditions on suspension flows can also be noted. At a level that has great practical importance, the behavior of suspensions has significant dependence on details of the particle size distribution and this challenges even our understanding of the rheological properties (Pednekar et al. 2018; Guy et al. 2020; Malbranche et al. 2023), to say nothing of the relative motion of the different particle sizes due to migration phenomena (Lyon and Leal 1998; Semwogerere and Weeks 2008). This discussion exposes just a few issues and does not at all address the difficult and important issue of particle shape, but in scraping the surface, it suggests the noted scope for further work. Because advances in this direction provide tools that allow exploration of the predictive power of existing models, a valuable feedback channel from practitioners to theorists is opened by testing existing theories against the behavior of practically relevant materials. This is exemplified nicely by work of Lee et al. (2020) to explore how the lubricated-to-frictional constitutive models (Singh et al. 2018) apply to more complex suspensions.
2. **Development of understanding of contact and tribological impacts on rheology**. Recent work has

developed a description of the ST transition in terms of contact with friction. The challenge is then to answer, what is happening in this "contact" zone? More generally, what are the forces at the surfaces that matter most, and how generic are the bulk outcomes to the range of specific surface forces? As these forces are often short-ranged, experimental access to the small length scales is a clear challenge, while trying to probe the forces by MD simulation may lead to difficulty in reaching sufficiently long time scales to be representative of particle interactions when contact takes place. A consideration of understanding gained by tribologists may allow classification of the influence of forces based on the constraints placed on particles' relative motions (Guy et al. 2018).

3. **Nonequilibrium statistical physics of dispersions**. The relationship of microstructure, from pair level to networks of contacts, to the properties of suspensions opens a range of questions in nonequilibrium statistical physics. The NHS suspension is relatively simply defined and, because it has limiting equilibrium behavior of the HS fluid, is a well-grounded system for such study. The physical system can be composed readily and its properties can be probed with standard rheometry in many laboratories. With scattering methods, the structure under flow even for colloidal dispersions is accessible (Gurnon and Wagner 2015). Combining this experimental access with the various simulation approaches (Seto et al 2013; Banchio and Brady 2003; Heussinger 2013; Ness 2021) available, the theoretical challenge of establishing principles for far-from-equilibrium systems may find an important testing ground in suspensions.

A closing remark

Taken together, these challenges span from the nanometer-scale contacts to the bulk scale. Establishing how these scales interact, for example, through the two-way coupling of forces related to physics and chemistry at the contacts driven by forces imposed at the bulk scale, and development of physically based models and occasional contributions to theories have been the goals in my work. The Weissenberg Award leading to this paper was a deeply felt honor, but it becomes clear in considering these challenges that it is the work of others that will follow that matters most.

Acknowledgements The research described here was in large measure the accomplishment of students and collaborators with whom I have been fortunate to work. For my start in suspensions and the push in the direction of their statistical physics, I owe John Brady (Caltech) a debt of gratitude, and for our many years of deeply stimulating work together, I owe a similar debt to Élisabeth Guazzelli (CNRS). The lecture upon which this work is based was dedicated in memoriam to James Swan and Yevgeny Yurkovetsky, two rheologists from whom I learned a great deal and who passed away far too soon.

Funding The preparation of this paper was supported by NSF CBET-2228680.

Declarations

Conflict of interest The author declares no competing interests.

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