

Annual Review of Fluid Mechanics Hydrodynamic Interactions Among Bubbles, Drops, and Particles in Non-Newtonian Liquids

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Annu. Rev. Fluid Mech. 2018. 50:505-34

First published as a Review in Advance on October 23, 2017

The Annual Review of Fluid Mechanics is online at fluid.annualreviews.org

https://doi.org/10.1146/annurev-fluid-122316-045114

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Keywords

particles, bubbles, shear thinning, viscoelasticity, pairwise interaction, negative wake, bubble velocity discontinuity, clustering

Abstract

The understanding of hydrodynamic forces around particles, drops, or bubbles moving in Newtonian liquids is modestly mature. It is possible to obtain predictions of the attractive-repulsive interaction for moving ensembles of dispersed particulate objects. There is a certain intuition of what the effects of viscous, inertial, and surface tension forces should be. When the liquid is non-Newtonian, this intuition is gone. In this review, we summarize recent efforts at gaining fundamental understanding of hydrodynamic interactions in non-Newtonian liquids. Due to the complexity of the problem, most investigations rely on experimental observations. However, computations of non-Newtonian fluid flow have made increasingly significant contributions to our understanding of particle, drop, and bubble interactions. We focus on gravity-driven flows: rise or sedimentation of single spheroidal objects, pairs, and dispersions. We identify the effects of two main rheological attributesviscoelasticity and shear-dependent viscosity-on the interaction and potential aggregation of particles, drops, and bubbles. We end by highlighting the open questions in the subject and by suggesting future directions toward the fundamental physics as well as applications.

1. INTRODUCTION

The hydrodynamics of bubbles, drops, and particles is at the heart of two-phase flows. For their scientific interest as well as their technological importance, two-phase flows have been the focus of intense research in the past few decades. The subject has reached a degree of maturity, as attested by a well-established journal, the *International Journal of Multiphase Flow*, and several monographs that have become classics (e.g., Happel & Brenner 1965, Kim & Karrila 1991, Clift et al. 2005).

In many applications involving bubbles, drops, or particles dispersed in a suspending fluid, the fluid is non-Newtonian. For example, hydraulic fracturing slurries for oil and gas exploration are typically dense suspensions of particles in a polymer solution (Shah 1993). Processing of polymer blends involves deforming droplets of the dispersed component into fibrils embedded in an immiscible polymer matrix to act as in situ reinforcement (Folkes & Hope 1993). Airlift bioreactors rely on swarms of bubbles stirring a tank of non-Newtonian fluids containing various biomacromolecules (Cerri et al. 2008). Microbubbles have recently found applications in medical imaging, where the air–liquid interface acts as a contrasting agent for ultrasounds (Blomley et al. 2001). The suspending biofluids, such as blood, are almost always non-Newtonian.

Starting in the 1970s, pioneering work has shown that solid particles can behave in drastically different ways in non-Newtonian fluids (Gauthier et al. 1971a,b; Bartram et al. 1975). This was initially understood from perturbation analysis that generated insights into how the normal stresses of viscoelastic fluids affect particle motion (Leal 1979, Brunn 1980). As the subject of rheology developed (Barnes et al. 1989), deeper knowledge about viscoelasticity, shear-dependent viscosity, and memory effects was integrated into the study of the hydrodynamics of particles, drops, and bubbles in viscoelastic fluids (De Kee & Chhabra 2002, McKinley 2002, Chhabra 2006). The deformable interface of drops and bubbles introduces more complications to the hydrodynamic problem, in terms of both the location of the interface, which is a priori unknown, and the boundary conditions on them. In recent years, experimental and computational techniques have developed rapidly in multiphase flows and non-Newtonian fluid mechanics. At the intersection of these two fields, the dynamics of particles, drops, and bubbles in non-Newtonian fluids has seen much activity and our understanding has progressed.

From both the fundamental and the applied perspectives, two-phase flow situations are of special interest. In the first, the suspended objects are carried in an external flow, with negligible settling or rising of the dispersed phase. Depending on the nature of the external flow—shear, extensional, or a mixed type—the individual particles, drops, or bubbles will interact in different ways. Among those, shear flows have received the most attention for their direct connection to rheological measurements. The results have been summarized in recent reviews of solid particles in sheared suspensions (e.g., Denn & Morris 2014, D'Avino & Maffettone 2015). For drops, in turn, most studies have focused on single-drop deformation and breakup in non-Newtonian fluids (Guido 2011). There have also been continuum models for the rheology of sheared emulsions (Lee & Park 1994). To our knowledge, the drop interaction in shear flows of non-Newtonian fluids has not been studied.

The second problem concerns the settling or rising of particles, drops, or bubbles under gravity or buoyancy force in a quiescent non-Newtonian fluid. D'Avino & Maffettone (2015) have provided a clear summary of solid particles sedimenting in viscoelastic liquids. As far as we know, no systematic review has appeared of bubbles and drops rising in non-Newtonian fluids. Chhabra (2006) has examined at length the rise velocity and the associated drag coefficient of a single drop or bubble. This book remains a good entry point to the literature on this subject. The bubble velocity discontinuity (BVD), a critical issue in this subject, was clarified only recently (Fraggedakis et al. 2016c). Moreover, newer work has appeared regarding the interaction between two or more rising bubbles and their swarming behavior (Vélez-Cordero & Zenit 2011, Vélez-Cordero et al. 2014).



A schematic depiction of the general case of an object of size D settling or rising due to gravity g or buoyancy in a quiescent fluid of density ρ_{ext} and viscosity $\mu_{\text{ext}}(\vec{\gamma})$. The object could be solid or fluid with density ρ_{int} and viscosity μ_{int} . Because the two phases are immiscible, a surface tension σ has to be considered.

Given the current state of the literature on particulate hydrodynamics in non-Newtonian fluids, this review focuses on the interaction between particles, drops, and bubbles that are sedimenting or rising under gravity or buoyancy in the absence of external flow. This limited scope means that we do not discuss shear flow, which has been reviewed recently (Guido 2011, D'Avino & Maffettone 2015), nor several other flow situations, e.g., bubble oscillation driven by an external pressure pulse, cavitation and the collapse of bubbles, bubble formation and the growth at orifices and spargers, and the impingement and spreading of drops on solid surfaces. In terms of fluid rheology, we focus on shear thinning and viscoelasticity, leaving aside other important effects such as yield stress, thixotropy, and rheopexy.

It is convenient to define the important dimensionless parameters of the problem. First, consider the steady-state sedimentation or rising of an object in a quiescent fluid. The general case is depicted schematically in **Figure 1**. The object of size *D* is either solid or fluid with density ρ_{int} and is surrounded by another fluid ρ_{ext} . If the object is deformable, its shape and linear dimensions may change. Then we define its equivalent diameter $D = (6V/\pi)^{1/3}$ from its volume *V*. The internal fluid is Newtonian with viscosity μ_{int} , but the external fluid could be either Newtonian or non-Newtonian. In this review, we focus on two distinct non-Newtonian effects: viscoelasticity and shear-dependent viscosity. The two phases are immiscible; therefore, a surface tension σ has to be considered. However, neither variations of surface tension around the object (Marangoni effects) nor thermal effects on any of the material properties are discussed. The unknown quantity of the greatest interest is the terminal velocity *U* of the object as it settles or rises steadily.

The case in which the external fluid is also Newtonian has been addressed extensively in the literature (Clift et al. 2005). From the parameters mentioned above, we can define a nominal terminal velocity from the Stokes drag of a solid sphere:

$$U_{\infty} = \frac{|\rho_{\text{ext}} - \rho_{\text{int}}|D^2g}{18\mu_{\text{ext}}}.$$

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Using this characteristic velocity, we can construct the four dimensionless groups that characterize the problem as such:

$$Re = \frac{\rho_{\rm ext} U_{\infty} D}{\mu_{\rm ext}},$$

$$Mo = \frac{g\mu_{\text{ext}}^4}{\rho_{\text{ext}}\sigma^3},$$
3.

$$\Pi_1 = \frac{\rho_{\rm int}}{\rho_{\rm ext}},\tag{4}$$

$$\Pi_2 = \frac{\mu_{\rm int}}{\mu_{\rm ext}}, \qquad 5.$$

where Re is the Reynolds number, M_0 the Morton number, Π_1 the density ratio, and Π_2 the viscosity ratio. Note that M_0 , Π_1 , and Π_2 are material properties of the given combination of external/internal fluids. For the case of solid particles, M_0 and Π_2 tend to infinity. For the case of bubbles, Π_1 are Π_2 are practically zero. For the case of drops, no simplifications can be made. Now the terminal velocity U can be expressed as a function of the four dimensionless groups: $U/U_{\infty} = f(Re, M_0, \Pi_1, \Pi_2)$.

Clearly, this set of dimensionless parameters is not unique. There are other common groups that are used in the literature, depending on the convenience needed for a particular subject. Among the most often used, we mention:

$$We = \frac{U_{\infty}D^2\rho_{\rm ext}}{\sigma},\tag{6}$$

$$Fr = \frac{U_{\infty}^2}{gD},$$
7.

$$Ga = \frac{\rho_{\text{ext}}^2 D^3 g}{\mu^2},$$
8.

$$Ca = \frac{U_{\infty}\mu_{\text{ext}}}{\sigma},$$
9.

$$Eo = \frac{(\rho_{\rm ext} - \rho_{\rm int})gD}{\sigma},$$
 10.

where We is the Weber number, Fr the Froude number, Ga the Galileo number, Ca the capillary number, and Eo the Eötvös number.

If the external fluid is viscoelastic (see the sidebar titled Viscoelasticity) with an elastic relaxation time λ , we can define the Deborah number

$$De = \frac{\lambda U_{\infty}}{D}$$
 11.

to compare the response of the fluid to the characteristic time of the motion. Note that some researchers use the Weissenberg number $Wi = \bar{\gamma}\lambda$, where $\bar{\gamma}$ is the mean shear rate, interchangeably with *De*. Strictly speaking, these two dimensionless quantities are not the same (Dealy 2010). Another dimensionless group often used to characterize the relevant importance of inertial effects

VISCOELASTICITY

A viscoelastic fluid simultaneously exhibits viscous and elastic properties (Barnes et al. 1989). A perfectly elastic material such as rubber deforms under applied stress but returns to the initial undeformed state once the stress is removed. A purely viscous fluid such as water or glycerol flows under stress, but the flow ceases immediately once the stress is no longer applied. A viscoelastic fluid flows under stress and exhibits partial recovery toward its undeformed state once the stress is removed.

with respect to elastic ones is the so-called elastic Mach number (Liu & Joseph 1993):

$$Ma = U_{\infty} \sqrt{\frac{\rho_{\text{ext}}\lambda}{\mu_{\text{ext}}}}.$$
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Note that $De = Ma^2/Re$.

For the case of shear-thinning fluids (see the sidebar titled Shear-Thinning and Shear-Thickening Fluids), we can use the power index n as a dimensionless measure of the variation of viscosity with shear rate. Its definition comes from the Ostwald-de Waele relationship that assumes a power-law shear-dependent viscosity:

$$n = 1 + \frac{\log(\mu_{\text{eff}})_{\text{ext}} - \log \kappa}{\log \bar{\dot{\gamma}}},$$
13.

where κ is the consistency coefficient. Some authors (De Kee & Chhabra 2002) use a dimensionless number called the Carreau number *Cu*, which compares the shear rate at which the shear-dependent viscosity first appears to the characteristic time of the flow. However, this number does not quantify the amount of viscosity reduction/increase of the liquid; it only indicates when these effects become important for a given flow. Evidently, the problem of determining the terminal velocity of an object in a non-Newtonian fluid is vastly complex due to the large number of parameters involved in the process.

For larger *Re* and *We* numbers, the object no longer rises in a rectilinear trajectory, evolving into a zigzag, spiral, and eventually chaotic motion (Magnaudet & Eames 2000, Ern et al. 2012). Generally speaking, the loss of the steady state is caused by the wake becoming three-dimensional (3D) and unsteady behind the moving object. To our knowledge, the effect of non-Newtonian rheology of the surrounding fluid on the onset and evolution of unsteady sedimentation or rise

SHEAR-THINNING AND SHEAR-THICKENING FLUIDS

The viscosity of a fluid may change depending on the magnitude of the applied shear. For Newtonian fluids, the viscosity is constant. For many fluids, the value of viscosity decreases with shear: These fluids are called shear thinning or pseudoplastic. If the fluid viscosity increases with shear, the fluid is called shear thickening or dilatant (Barnes et al. 1989). A simple model that captures the shear-dependent fluid viscosity μ_{eff} is the so-called power law:

$$\mu_{\rm eff} = \kappa \, \bar{\dot{\gamma}}^{n-1}$$

where κ and n are the consistency and power indices. If n < 1, the fluid is shear thinning; conversely, if n > 1, the fluid is shear thickening. If n = 1, the fluid is Newtonian.

has not been investigated in depth; at the moment, one has to rely on the Newtonian results (e.g., Clift et al. 2005). We do not discuss this aspect further in this review.

When two or more objects settle or rise in close proximity, their hydrodynamic interaction becomes the most interesting question. This leads not only to changes in the rise velocity and the drag force, but also to modified trajectories and even the formation of clusters. The amount of objects in the liquid is commonly quantified by the volume fraction ϕ , which is the ratio of volume occupied by the objects to the total volume:

$$\phi = \frac{V_{\text{objects}}}{V_{\text{total}}}.$$
14.

When $\phi \neq 0$, interaction forces may arise among the objects in addition to the drag. These forces will depend on the spatial distribution of the objects. Most importantly, the interaction forces may be attractive or repulsive. Consequently, the objects may cluster or remain dispersed as they ascend or descend. For the case of solid particles in Newtonian fluids, the particles remain dispersed for low *Re*, and the sedimentation velocity is a function of ϕ (Guazzelli & Hinch 2011). For pairs of millimetric bubbles ascending side by side in water, the interaction force transitions from repulsive to attractive as the Reynolds number increases (Legendre et al. 2003). Bubbly flows at moderate *Re* remain well dispersed as they ascend in quiescent Newtonian fluids (Martinez-Mercado et al. 2007).

In this review, we start from the state of the art for single objects sedimenting or rising under the action of gravity. Then we address the current understanding of pairwise interactions on the behavior, focusing on the effects of non-Newtonian properties in the attractive–repulsive nature of the interaction forces. Lastly, we discuss studies that have considered the motion of a dispersed collection of objects.

2. GRAVITY-DRIVEN MOTION OF A SINGLE SPHEROIDAL OBJECT

We now discuss the motion of isolated spheroidal objects sedimenting or ascending in a non-Newtonian liquid. We restrict the discussion to objects whose shape does not deviate significantly from a sphere. Therefore, flows with large values of Re and We are not considered. Although ever present in experiments, the wall effects are not discussed here. McKinley (2002) and Chhabra (2006) considered the effects of walls in detail. The combined effects of inertia and viscoelasticity on the motion of objects are not discussed either; Joseph & Liu (1993), Liu & Joseph (1993), and Joseph et al. (1994) have addressed this issue at length.

The discussion centers around the terminal velocity and the drag coefficient. However, because the flow field around the object is important for determining the drag, we also examine the structure of the wake.

2.1. Solid Particles

As described above, the speed of a spheroidal object immersed in a fluid as a result of gravity depends on several dimensionless groups. For the case of particles, the number of parameters is reduced because the shape of the object does not depend on the flow around it, and also because the interfacial condition is no-slip. For Newtonian fluids, the drag coefficient only depends on Re. For non-Newtonian fluids, De and n are also important. It is relevant to note that, experimentally, the effect of the wall proximity is always present (and is especially important when Re < 1).

As discussed by McKinley (2002), for creeping flows, the effect of viscoelasticity on the drag coefficient for solid spheres can be significantly different (can either increase or decrease with respect to the Newtonian case) for apparently similar Boger fluids (see the sidebar titled Boger

BOGER FLUIDS

Viscoelastic fluids with constant (or nearly constant) viscosity are called Boger fluids (James 2009). Experimentally, it is not generally easy to prepare a fluid that has large viscoelastic effects (measurable first normal stress difference) but has a constant shear viscosity. Most researchers follow well-known recipes from the literature. Similarly, for inelastic shear-thinning fluids, one must rely on empirical recipes (e.g., Barnes et al. 1989).

Fluids). **Figure 2** shows a typical behavior of the drag coefficient for a sphere as a function of Weissenberg number for two Boger fluids with similar properties. To rationalize the effect of fluid rheology for spheres, and for any other particles, drops, or bubbles, one needs to analyze the properties of the test fluid in great detail. McKinley's (2002) review provides a detailed discussion of this issue in particular, including the effects of the solvent and solute molecular weights, extensional viscosity, wall effects, and shear thinning of the fluid. When, in addition to viscoelasticity, the fluid has a shear-dependent viscosity, the drag coefficient decreases with n, but it also depends on the value of the Carreau number Cu (Rodrigue et al. 1996b). More details on the effect of shear-thinning viscosity are provided by Chhabra (2006).

As may be expected, the effect of the non-Newtonian nature of the fluid is also observed in the flow around a solid sphere. Early studies showed a shift in the flow streamlines (in comparison with the Newtonian flow in creeping flow) close to the solid surface, either down- or upstream of the sphere (Mena et al. 1987, Chhabra 2006). For high–Weissenberg number sedimentation in Boger fluids, Fabris et al. (1999) found that the bi-extensional flow in the leading face of the sphere and the extensional flow in the wake exacerbate the non-Newtonian reaction of the liquid, leading to significant changes in the structure of the flow around the sphere.



Figure 2

Normalized drag coefficient X_e as a function of Weissenberg number We for the flow around a solid sphere. Open red circles represent a polyisobutylene/polybutene Boger fluid and solid blue triangles represent a polyacrylamide/corn syrup Boger fluid. Adapted with permission from Solomon & Muller (1996), copyright Elsevier.

NEGATIVE WAKE

In his seminal investigation, Hassager (1979) reported that the flow behind a bubble ascending in a polymeric solution seemed to move in a direction opposite to the bubble motion. This flow reversal behavior has been observed in the wake of spheres, bubbles, and drops, and it has been associated with the viscoelastic nature of the fluid—in particular, with its extensional viscosity.

When, in addition to viscoelasticity, the fluid also exhibits a shear-thinning viscosity, a negative wake may appear downstream from the sphere (see the sidebar titled Negative Wake): The flow moves in a direction opposite to that of the sphere. A historical review of this phenomenon is provided by Arigo & McKinley (1998). **Figure 3** shows the characteristic features of the flow around a sedimenting solid sphere. Qualitatively the appearance of this phenomena is readily explained by the inherent memory of viscoelastic fluids: Some time after the fluid has been stretched or sheared, the material tends to recoil to its initial undisturbed state. However, the detailed conditions for the appearance of the negative wake are not yet fully understood (Mendoza-Fuentes et al. 2009). Interestingly, despite the appearance of the negative wake, the drag coefficient or the sedimentation velocity does not seem to be greatly affected (Arigo & McKinley 1998, Mendoza-Fuentes et al. 2010).

Clearly, the significantly different structure of the flow behind a sphere in which a negative wake appears may lead to different hydrodynamic interaction forces. Bird et al. (1987) hypothesized that negative wakes may explain the separation of two solid spheres settling in a line, reported by Riddle et al. (1977). However, Verneuil et al. (2007) concluded that the negative wake did not modify the interaction significantly. For the case of bubbles or drops, this issue has not been studied to date. The motion of bubbles or drops may be more sensitive to changes in the surrounding flow due to their small inertia.



Figure 3

Sketch of the flow around a moving sphere in which a negative wake appears, showing the axial fluid velocity u_z as a function of axial position z. Upstream of the particle, the fluid velocity is in the same direction as the particle. Some distance downstream of the particle, the direction of the fluid motion is opposite to that of the particle.

2.2. Drops and Bubbles

When the object is deformable (i.e., a drop or a bubble), its shape will evolve as the sedimentation velocity increases. Therefore, the drag coefficient will depend, in addition to De, n, and Re, on the Weber number We and the density and viscosity ratios. For creeping flow in Newtonian liquids, the drag coefficient for a spherical object is (Hadamard 1911, Rybczyński 1911)

$$C_{\rm D} = C_{\rm D}|_{\rm sphere} \frac{\frac{\mu_{\rm int}}{\mu_{\rm ext}} + 2/3}{\frac{\mu_{\rm int}}{\mu_{\rm ext}} + 1}.$$
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For shear-thinning liquids, a flattening of the rear side of the bubble or drop is observed as the size increases (Soto 2008, Prieto 2015). **Figure 4***a* shows an example of the evolution in shape in a shear-thinning inelastic fluid. When the fluid is viscoelastic, with constant viscosity or with shear-thinning effects, the object develops a characteristic teardrop shape with a pointed rear end. **Figure 4***b* shows a typical evolution of the shape of bubbles as the size increases in a shear-thinning viscoelastic fluid (Chehata 2004). The tip of the cusp progressively becomes sharper as the size increases, and eventually, for larger sizes, the topology of the tip transitions into a knife-edge shape (Liu et al. 1995, Soto et al. 2006, Ohta et al. 2015).

2.3. Velocity Discontinuity

For bubbles and drops, as for the case of particles, as the size increases, the velocity increases too. As a result, the viscoelastic and shear-thinning effects gain importance. Particularly for the case of bubbles, a peculiar behavior occurs: The velocity increases with size in an apparently discontinuous manner. At a certain critical size, the velocity increases abruptly (the so-called BVD). **Figure 5** shows a typical example of the phenomenon. This behavior was first reported by Astarita & Apuzzo (1965) and has since been investigated by many (for a recent review on the subject, see Chhabra 2006). This behavior is particularly notable when the fluid is both viscoelastic and shear thinning. For liquid drops or solid particles, no discontinuous jump in the terminal velocity has been observed with increasing drop or particle volume. However, Rodrigue & Blanchet (2006) and Ortiz et al. (2016) have reported a less dramatic discontinuity for liquid drops; at a critical volume, the velocity–volume curve changes slope instead of being discontinuous.

The understanding of the appearance of the discontinuous behavior of the bubble terminal velocity has progressed slowly over the years. Only recently have researchers reached a clear physical picture. In particular, Fraggedakis et al. (2016c) recently significantly contributed to our understanding of the BVD. There are several features that characterize this phenomenon: The bubble's shape transitions when it surpasses the BVD critical volume (Herrera-Velarde et al. 2003); above the critical volume, a negative wake appears (Herrera-Velarde et al. 2003, Pillapakkam et al. 2007); the BVD has been observed in viscoelastic fluids with both constant (Vélez-Cordero et al. 2014) and shear-thinning viscosities, but the increase of velocity is significantly larger when the fluid is shear thinning; and it has been argued that the change of surface properties in the bubble may influence the appearance and magnitude of the BVD (Rodrigue et al. 1996a, Vélez-Cordero et al. 2014). Attempts to predict the critical volume include those by Rodrigue et al. (1996a), Soto et al. (2006), Pillapakkam et al. (2007), Pilz & Brenn (2007), Soto (2008), and Vélez-Cordero et al. (2014). In their numerical investigation of the problem, Fraggedakis et al. (2016c) found that terminal velocity hysteric loops appear depending on which of the above features are present. In particular, they discovered two separate hysteresis loops, one associated with the velocity jump and the other with the appearance of the shape change and the negative wake. When the



Sequence of images of the evolution of bubble shape with size. (*a*) Shear-thinning inelastic fluid. The volume ranges from 40 to 4,000 mm³, from the smallest to the largest bubble shown. The fluid is a mixture of ethylene glycol and carbopol. Adapted with permission from Soto (2008), copyright Universidad Nacional Autonoma de México. (*b*) Shear-thinning viscoelastic fluid. The volume ranges from 24 to 410 mm³. The fluid is a solution of polyacrylamide in a water–glycerin mixture. The bubble velocity discontinuity occurs at a critical volume of 68 mm³, which is between the third and fourth images on the top row. Adapted with permission from Chehata (2004), copyright Universidad Nacional Autonoma de México.

shear-thinning effect was important, the two loops merged into one, leading to the appearance of a large velocity increase, i.e., the BVD.

Integrating the findings of Fraggedakis et al. (2016c) with the many other previous investigations, we propose an explanation of the BVD, as depicted schematically in **Figure 6**. The mechanism can be summarized as follows.

As the bubble size gradually increases, a monotonic increase of the bubble speed is expected because of an increase in the buoyancy force (step 1 in **Figure 6**). This is true in general. One exception would be when the bubble is too deformable and elongated in the transverse direction for



Bubble velocity as a function of bubble volume. The fluid is a viscoelastic, shear-thinning polymer solution. The different symbols show experiments recorded at different heights. Adapted with permission from Pilz & Brenn (2007), copyright Elsevier.

large–Weber number values (Legendre et al. 2012). In this case, the drag increases, and therefore, the bubble velocity does not increase.

As a result of the velocity increase, the rates of extension and shear around the bubble also increase, as both scale with U_{∞}/D (step 2 in **Figure 6**). In particular, the rate of bi-axial extension increases near the front end of the bubble, and the rate of extension increases near the rear end of the bubble. Around the equator of the bubble, the rate of shear increases.

As a result of the increase of the extension and shear rates around the bubble, three main mechanisms may occur. First, there is an elastic response (step 3a in **Figure 6**). The elongational and shear flows around the bubble tend to elongate the bubble in the direction of the flow, as is obvious from the first three frames of **Figure 4b**. The elongational flow near the front and back stagnation points pulls the bubble while the shear flow and the resultant hoop stress squeeze the bubble inward at the equator. This change of shape reduces the drag on the bubble. A more drastic shape change occurs when the bubble volume crosses the critical value. The appearance of the pointed tip at the rear of the drop (fourth frame in **Figure 4b**) can cause a drag reduction on the order of 50% according to the calculations of Soto (2008). Another potential effect of the strong elongation downstream of the bubble is the appearance of the negative wake (Herrera-Velarde et al. 2003), which may produce a thrust on the bubble (Pillapakkam et al. 2007). Thus, elasticity can deform the bubble in different ways that contribute to a drag reduction and an increase in its rise velocity.

Second, there is a viscous response (step 3b in **Figure 6**). If the fluid is shear thinning, in addition to being viscoelastic, the viscous component of the drag will be reduced as a result of the thinning of the viscous field around the bubble (Vélez-Cordero et al. 2011). Such drag reduction will also contribute to an increase in the speed of the bubble. The recent work of Fraggedakis et al. (2016c) has shown that the viscosity reduction is an important factor in the BVD.



Schematic view of the mechanism associated with the appearance of the bubble velocity discontinuity. (1) Increase in velocity due to buoyancy. (2) Increase in shear and extension rates. (3*a*) Elastic response. (3*b*) Viscous response. (3*c*) Surface response. (4) Bubble velocity discontinuity.

Third, there is a surface response (step 3c in **Figure 6**). It is known that, as a result of the velocity increase, the surface of the bubble may clean itself from possible surface active agents (Rodrigue et al. 1996a). Effectively, the boundary condition on the surface of the bubble may change from a no-slip condition (fully contaminated) to a free-slip condition (clean interface). For a spherical particle, the reduction of drag from this change of boundary condition can be as large as 33.3% (Clift et al. 2005).

All of the mechanisms described above induce reductions of the drag force on the bubble and, acting together, may give rise to the BVD. If one of the effects does not occur, the BVD may not appear or it may manifest itself as a weaker change in velocity. For bubbles, the magnitude of the velocity jump is much smaller in Boger fluids than in shear-thinning viscoelastic fluids (Soto et al. 2006, Pilz & Brenn 2007, Vélez-Cordero et al. 2014). For solid particles, there is no shape deformation or change in boundary conditions, and therefore, no discontinuous change in falling speed has been observed in any fluid (Arigo & McKinley 1998, Mendoza-Fuentes et al. 2009). For the case of drops, a few studies have reported a discontinuous change in the slope of the velocity–volume curve (Rodrigue & Blanchet 2006, Ortiz et al. 2016): In such a case, the rise or fall velocity is typically much smaller than for bubbles, and this makes the velocity discontinuity

less likely. It also important to note that the recent work by Fraggedakis et al. (2016a,b) has shown that the BVD is also observed in yield-stress fluids, where both viscoelasticity and shear-thinning viscosity effects are present.

3. PAIR INTERACTIONS

When two particles, drops, or bubbles are moving in close proximity, they interact hydrodynamically in a way that depends on the rheology of the surrounding liquid and their relative distance and orientation. This puts pairwise interaction at the center of the problem. Nevertheless, our literature survey finds that the pairwise interaction in non-Newtonian fluids is far from clear. A main complication is that multiple rheological attributes—shear thinning, normal stress differences, and memory effect—can potentially influence pairwise interaction, and it is difficult to distinguish their effects in experimental studies. Theoretical analysis is feasible only for the simplest of constitutive equations, e.g., the second-order fluid model, and thus has not offered enough insight into the different rheological attributes. Finally, numerical solutions of moving boundary problems in non-Newtonian fluids have remained challenging to date.

Perhaps not surprisingly, most of the studies on pairwise interactions have dealt with solid particles. Only a handful of studies have considered drops and bubbles; the deformability of the latter adds an additional layer of complexity to the problem. From these few studies, it is not easy to determine to what extent the results can be explained by the same mechanisms as for solid particles, and which features are distinctly due to the deformable and potentially slip-prone interface of drops and bubbles. Therefore, we first discuss the interaction between a pair of settling solid particles. On the basis of this discussion, we then examine the more complex interactions between deformable drops and bubbles.

3.1. Interaction Between Two Settling Solid Particles

Two initial configurations have been studied: two particles released side by side and one atop the other. The first case seems to be well understood, with little controversy. Thus, let us discuss it first.

3.1.1. Side by side initial configuration. Both experiments and numerical computations have been conducted on the side by side initial configuration. Some theoretical studies also exist but are limited to simple rheological models.

Joseph et al. (1994) published the first comprehensive experiments on how two spheres dropped side by side interact. In viscoelastic polymer solutions, the spheres attract each other if the initial separation is not too wide; otherwise, they fall straight down and demonstrate no interaction. **Figure 7** shows an image from Joseph et al. (1994) that depicts the sideways attraction. This forms an interesting contrast to inertia-based repulsion for a similar pair settling in Newtonian fluids. As the two spheres approach each other laterally, their line of centers rotates toward the vertical direction, with the pair eventually settling one on top of the other. The two particles may come into contact to form a doublet or stay separated during the rest of the sedimentation. Pairwise interaction in this vertical configuration is the focus of Section 3.1.2. Gumulya et al. (2011) later confirmed these observations using other polymeric solutions and also addressed the competition between the viscoelastic and inertial effects. Joseph & Liu (1993) and Liu & Joseph (1993) had earlier explored such an effect in a wider context.

Joseph et al. (1994) proposed an explanation for side by side attraction between two sedimenting spheres based on the first normal stress difference N_1 . In particular, they correlated the



An image showing the pairwise interactions of two spheres initially released side by side in a viscoelastic fluid. Adapted with permission from Joseph et al. (1994), copyright Elsevier.

normal-to-shear stress ratio N_1/τ with the tendency of sideways attraction. This explanation is informed by theoretical analysis that preceded Joseph et al.'s (1994) experiments. Leal (1979) and Brunn (1980) pioneered the asymptotic analysis of particle motion in non-Newtonian fluids using the second-order fluid model in the limit of vanishing Deborah number ($De \rightarrow 0$). Later, numerical computations were carried out using the second-order fluid model (Phillips 1996, Ardekani et al. 2008, Khair & Squires 2010). The conclusion is clear: The two particles attract each other regardless of their relative position, and they do so in the special case of the horizontal side by side configuration. Besides, the horizontal configuration is unstable to rotation of the line of centers toward the vertical direction. These have confirmed Joseph et al.'s (1994) observations and supported their N_1 -based explanation.

To explore the behavior of particle pairs beyond the limitations of a second-order fluid, Feng et al. (1996) carried out 2D computations using the Oldroyd-B model for finite Deborah numbers, De = O(1). They reproduced the same qualitative features previously observed in experiments and shown for second-order fluids. By analyzing the pressure and stress distributions on the surface of the particles, they further demonstrated that viscoelasticity affects the particle motion mostly by modifying the pressure distribution on the particles. The direct contribution of the normal stress difference N_1 to the attraction force is small. This appears to be a rather general feature of sedimentation in viscoelastic fluids (Feng et al. 1995). Binous & Phillips (1999) and Goyal & Derksen (2012) carried out 3D simulations using finitely extensible nonlinear elastic (FENE) models to probe particle interaction in Boger fluids, and reached the same conclusion regarding the behavior of a pair of spheres released side by side.

From the studies reviewed above, a more or less coherent picture emerges regarding the interaction of two particles settling side by side in the horizontal configuration. Viscoelasticity causes the two to attract and approach each other. This is mostly due to the first normal stress difference N_1 that modifies the pressure distribution on the particle surface (Feng et al. 1996). Furthermore, as the two particles approach, their line of centers turns toward the vertical, due to essentially the same viscoelastic mechanism that rotates a long particle's axis parallel to the direction of fall (Leal 1979, Joseph & Liu 1993). There have been suggestions of a separate role of shear thinning in causing particles to attract sideways, but existing evidence is scarce and contradictory (Joseph et al. 1994, Gheissary & van den Brule 1996). We return to the effect of shear thinning below in discussing particle interaction in the vertical configuration, where it plays a more easily identifiable role.

3.1.2. Initial configuration with one particle atop the other. Arguably, the vertical configuration is more important than the horizontal one because it is directly related to the formation of vertical chains in sedimentation (Joseph et al. 1994). Moreover, the side by side configuration typically evolves into the vertical one as the two particles interact. As alluded to earlier, the vertical configuration is less well understood than the horizontal. There have been multiple inconsistencies in experimental observations and in their interpretations.

In **Table 1**, we compile a list of representative experimental studies of particle interactions during sedimentation in the vertical configuration. Although most authors reported particle attraction and aggregation, others documented apparent repulsion and separation (Riddle et al. 1977, Gheissary & van den Brule 1996, Bot et al. 1998). Moreover, there are disagreements among this latter group of studies. Whereas Riddle et al. (1977) documented short-range attraction and long-range repulsion, Bot et al. (1998) saw the opposite (short-range repulsion and long-range attraction). Gheissary & van den Brule (1996) recorded repulsion in Boger fluids at all particle separations.

One naturally attempts to sort these experiments by the rheological properties of the fluids used. **Table 1** comprises a wide variety of fluids that manifest some of these rheological attributes: shear thinning, normal stress differences, long memory, yield stress, and pronounced elongational viscosity. Joseph et al. (1994) proposed two separate mechanisms that can produce particle attraction: (*a*) shear thinning plus memory and (*b*) elastic normal stresses. Each alone can account for

Table 1 Experimental	observations of the interaction betw	veen two particles settling one	atop the other (vertical confi	guration) in a variety of
non-Newtonian fluids				
Reference	Fluids and rheology	Parameters	Main observations	Interpretation
Eisenberg et al. 2013	Polyisobutylene solution	$0.4 < De < 3.5, Re \ll 1$	Attraction and formation of	Due to elastic normal stresses; no
	(viscoelastic and shear thinning)		doublet	negative wake; shear thinning
				unimportant
Gumulya et al. 2011	Polyacrylamide solution	$De \approx 2,000$ (based on shear	Attraction and formation of	Due to memory of reduced viscosity

Reference	Fluids and rheology	Parameters	Main observations	Interpretation
Eisenberg et al. 2013	Polyisobutylene solution (viscoelastic and shear thinning)	$0.4 < De < 3.5, Re \ll 1$	Attraction and formation of doublet	Due to elastic normal stresses; no negative wake; shear thinning unimportant
Gumulya et al. 2011	Polyacrylamide solution (viscoelastic, shear thinning, and with yield stress)	$De \approx 2,000$ (based on shear relaxation time), 0.003 < Re < 0.084	Attraction and formation of doublet	Due to memory of reduced viscosity; elasticity effect should also exist but is never mentioned
Verneuil et al. 2007	Xanthan solution (shear thinning and with memory)	$21 < De < 512, Re \ll 1$	Attraction and formation of doublet	Likely due to memory of shear thinning; negative wake unimportant
Gueslin et al. 2006	Laponite suspension (shear thinning and with memory and yield stress)	De not given, $Re\ll 1$	Attraction and formation of doublet	Due to memory of shear thinning; negative wake unimportant
Daugan et al. 2002a	Xanthan solution (shear thinning and with memory)	$45 < De < 200, Re \ll 1$	Attraction and formation of doublet	Due to memory of shear thinning; validates corridor of reduced viscosity
Bot et al. 1998	Polyacrylamide-based Boger fluid	$2 < De < 30, Re \approx 0.1$	Attraction at large separation and repulsion at small separation	Long-range viscoelastic attraction and short-range modulation of elongation-dominated wake
Gheissary & van den Brule 1996	Inelastic and elastic shear-thinning fluids; Boger fluid	De not given, $Re < 0.05$	Attraction with shear thinning and repulsion for Boger fluids	Attraction due to memory of shear thinning; repulsion due to elasticity
Joseph et al. 1994	Inelastic and elastic shear-thinning fluids; Boger fluid	$10^{-3} < De < 6,$ $10^{-3} < Re < 0.4$	Attraction and formation of doublet in all cases	Memory of shear thinning and elasticity as separate mechanisms for attraction
Riddle et al. 1977	Viscoelastic, shear-thinning fluids	1 < De < 30, Re < 0.05	Attraction at small separation and repulsion at large separation	Far-field repulsion due to negative wake (Bird et al. 1987)

Abbreviations: De, Deborah number; Re, Reynolds number.

Table 1

the outcome of particle aggregation, although both may coexist in an experiment. This framework seems to explain most of the results in **Table 1**, with some exceptions.

For fluids with strong shear thinning and a long memory, the passage of the leading sphere will leave the fluid in its wake in a state of reduced viscosity. Upon entering this corridor of reduced viscosity, the trailing particle will experience lower viscous drag, attain a higher velocity, and catch up with the leader, thus giving the appearance of attraction. This provides an explanation for all experiments using inelastic shear-thinning fluids that show negligible normal stress differences (Joseph et al. 1994, Gheissary & van den Brule 1996, Daugan et al. 2002a, Gueslin et al. 2006, Verneuil et al. 2007). Even for polyacrylamide solutions with both shear thinning and elasticity, Gumulya et al. (2011) considered this the dominant mechanism.

For elastic fluids with normal stress differences and little shear thinning, particle–particle attraction is also observed. Joseph et al. (1994) concluded that the normal-to-shear stress ratio N_1/τ correlates well with the tendency of particle aggregation. Eisenberg et al. (2013) observed that two spheres approach each other to form a doublet in a shear-thinning viscoelastic fluid. However, the velocity of approach is quantitatively predicted by calculations using a second-order model (Phillips & Talini 2007), which has no shear thinning. This led the authors to conclude that the attraction between spheres is driven by elasticity. More generally, particle–particle attraction in shear-thinning viscoelastic fluids (Joseph et al. 1994, Gumulya et al. 2011) is likely the result of a combined effect of the two mechanisms. Finally, Haward & Odell (2004) measured the drag force on two spheres fixed in a uniform flow of a Boger fluid, and found that the downstream sphere experienced significantly (up to 40%) less drag than the one upstream. This provides indirect support for the elasticity-based attraction between two particles.

Numerous computations using the second-order fluid model (Brunn 1980, Phillips 1996, Phillips & Talini 2007, Ardekani et al. 2008, Khair & Squires 2010) have all come to the same conclusion of particle attraction in the vertical configuration. In fact, the attraction is observed regardless of the orientation of the two particles relative to the direction of fall, although the attraction is greatest in the vertical configuration (Brunn 1977). The finite-*De* simulations of Feng et al. (1996), using the Oldroyd-B model, have also reached the same conclusion regarding attraction between particles sedimenting one on top of the other. More recently, Yu et al. (2006) employed a constitutive model that integrates shear thinning, memory, and viscoelasticity to simulate the sedimentation of two or more particles. They essentially demonstrated both of the above mechanisms: Shear thinning plus memory is sufficient to induce vertical chains of particles, but elastic normal stresses—via the Oldroyd-B model in this case—are necessary to stabilize such chains and aggregates against inertia-induced dispersion.

In shear thinning as well as in Boger fluids, a vertical doublet falls at a faster speed than that of a single particle. In the experiment of Eisenberg et al. (2013), where two particles attract mainly because of fluid elasticity, the average velocity of the particles increases as the two approach, achieving a maximum around 1.6 times that of a single sphere. In a shear-thinning xanthan solution, a vertical doublet falls at roughly twice the speed of a single particle (Daugan et al. 2002a). This increase in fall velocity is anticipated from a theory for second-order fluids (Brunn 1977) and has been confirmed by numerical simulations for Boger and shear-thinning fluids (Feng et al. 1996, Yu et al. 2006).

The above framework cannot explain the observations in Boger fluids that the two particles tend to separate if they were initially close (Gheissary & van den Brule 1996). Given the attraction at large separations, the two spheres thus approach a steady-state separation regardless of the initial separation (Bot et al. 1998). The near-field repulsion, Bot et al. (1998) argued, may be related to the highly elongational flow in the wake of a single sphere, which produces a drag enhancement on the sphere. As the trailing sphere enters the wake of the leading sphere, it disrupts the latter's wake and reduces its elongation-based drag, causing it to accelerate and move farther ahead. Using

the FENE model as a representation of a Boger fluid, Binous & Phillips (1999) reproduced the approach to a steady-state separation and essentially confirmed the explanation of Bot et al. (1998). Their simulation demonstrates an appreciable elongational effect in the wake of falling particles, and the particle–particle interaction can be traced to the elongation of dumbbells in different regions of the flow. However, not all Boger-fluid experiments have shown near-field repulsion between particles; some showed only attraction (Joseph et al. 1994, Haward & Odell 2004). In these cases, there may not have been sufficient elongation in the wake of the leading sphere to produce the short-range repulsion, either because the fluid is only weakly elastic (Joseph et al. 1994) or because the Deborah number is too low (Haward & Odell 2004).

Finally, the early experiment of Riddle et al. (1977) remains unexplained. With five shearthinning fluids with more or less elasticity, these authors observed near-field attraction and farfield repulsion between two spheres, in apparent contradiction to the observations of Bot et al. (1998) in Boger fluids. The near-field attraction is probably due to shear thinning plus memory in the fluids. For the far-field repulsion, Bird et al. (1987) suggested negative wake as an explanation: An upward flow in the wake of the leading particle would push the trailing particle upward. But newer evidence has disproved this idea. Gueslin et al. (2006) and Verneuil et al. (2007) observed particle attraction with and without a negative wake, and concluded that the latter feature has little effect on the particle interaction. Thus, the far-field repulsion reported by Riddle et al. (1977) remains a mystery.

To conclude, we have identified three mechanisms that govern the interaction between a pair of spheres settling in a non-Newtonian fluid. First, the viscoelastic normal stresses tend to push the particles toward each other and promote their aggregation regardless of their spatial arrangement. For particles settling side by side, this is the only mechanism at work, and it causes the two to attract each other sideways and their line of centers to turn into the vertical. For particles settling one atop the other, two more mechanisms may manifest themselves. The so-called corridor effect due to shear thinning plus memory causes the trailing particle to experience reduced viscosity and catch up with the leader. Moreover, in Boger fluids exhibiting strong elongational effects, the trailing particle entering the wake of the leading particle may cause the latter to accelerate, thus separating the two. In this situation, the two particles attract when far apart, separate when close, and attain a steady-state separation in a Boger fluid. These three mechanisms explain all prior observations except that of Riddle et al. (1977), who found that two particles settling one on top of the other in shear-thinning viscoelastic fluids approach each other if initially close but separate if sufficiently far apart initially. Because this observation has never been replicated by later experiments, it could be an artifact of the experimental setup or conditions.

3.2. Interaction Between a Pair of Bubbles or Drops

Compared with the interaction of sedimenting spheres, there have been far fewer studies of the interaction between bubbles rising in a quiescent non-Newtonian fluid. To our knowledge, no relevant studies exist for the case of drops. We therefore focus only on bubbles in the rest of this section.

From the knowledge of the solid particle pairwise interaction, one may approach the current problem by asking the following question: How does the deformation of bubble shape modify the way that bubbles move and interact, so that they present novel features unseen with solid particles?

Generally speaking, two rising bubbles interact in ways similar to the interaction between two settling particles. De Kee et al. (1990) released bubbles of different sizes simultaneously from orifices placed side by side in shear-thinning viscoelastic liquids. The larger bubble rises faster, and draws the smaller bubble into its wake. Then the trailing bubble usually catches up with the leader to coalesce. This resembles the attraction between particles released side by side as discussed

above (Joseph et al. 1994). More recently, Fan et al. (2009) showed that in carboxymethylcellulose solutions, two bubbles released side by side attract each other if their initial separation is small and their line of centers turns vertical during the rise. If their initial separation is large, however, the two tend to repel each other. This recalls similar observations of sedimenting particles (Gumulya et al. 2011). If the analogy holds, the repulsion at larger distances is due to inertia. Li et al. (2001) and Frank et al. (2005) observed bubbles rising in line and coalescing in shear-thinning viscoelastic polymer solutions. The authors ascribed the attraction to the corridor of reduced viscosity in the wake of the leading bubble, in very much the same way as for solid particles (Joseph et al. 1994). Lin & Lin (2003, 2009) confirmed the entrainment of the trailing bubble into the wake of the leading bubble, and cited both shear thinning and elasticity in the wake of the leader as contributing factors.

Vélez-Cordero et al. (2011) used inelastic shear-thinning fluids to examine the competition between inertia and shear-thinning during the rise of a pair of bubbles under conditions of no coalescence. **Figure 8** shows a typical example of this type of interaction. As noted by others, shear thinning enhances the entrainment of the trailing bubble into the wake of the leading one. Afterwards, a process similar to the drafting-kissing-tumbling scenario for solid particles occurs if inertia dominates. The bubbles touch and tend to tumble into a horizontal orientation, a well-known inertial effect. However, if shear thinning is strong, the reduced viscosity in the region enveloping the bubbles keeps the pair from tumbling and separating. Instead, they either maintain a vertical configuration or a staggered configuration with the line of centers oscillating about the horizontal. In general, large bubble deformation and low inertia favor bubble pairing (Vélez-Cordero et al. 2011). In this situation, as is generally true for bubble columns, the rise velocity is higher than for a single bubble (Vélez-Cordero & Zenit 2011, Vélez-Cordero et al. 2011).

In summary, two rising bubbles interact in mostly the same way as two solid particles. One novel feature due to the bubble deformation is oscillations of single and bubble pairs (Vélez-Cordero et al. 2011). Another is the potential of bubble–bubble coalescence. Indeed, coalescence is a feature that has received much attention for bubbles interacting in a non-Newtonian fluid. Earlier work (Acharya & Ulbrecht 1978, De Kee et al. 1986) noted that elasticity in the liquid tends to hinder film drainage and delay coalescence. Conceivably, this may have to do with the high elongational viscosity that is activated by the squeezing flow during film drainage. Recent studies have painted a more nuanced picture: Viscoelasticity may hinder or promote film drainage depending on the bubble size and strain rates in the film (Dreher et al. 1999, Yue et al. 2005).

4. ENSEMBLES

Based on the previous discussion, we can now address the issue of the motion of assemblies of particles, drops, or bubbles in a non-Newtonian fluid. As with two-body interactions, much more work has been done for the case of solid particles than for bubbles. To our knowledge, there are no studies for the case of drops. In general, a shear-thinning viscosity and viscoelasticity induce aggregation and cluster formation for both solid particles and bubbles.

4.1. Ensembles of Solid Particles

Experimentally, the sedimentation of suspensions of spherical particles has been investigated in fluids that are shear thinning inelastic, elastic with constant viscosity, and shear thinning viscoelastic (Bobroff & Phillips 1998; Daugan et al. 2002b, 2004; Mora et al. 2005). **Figure 9** illustrates a concentration instability in this type of experiment: The formation of particle-rich vertical columns is observed after a certain time, starting from uniformly distributed particles. Bobroff & Phillips (1998) observed such dense vertical columns, separated by particle-free liquid gaps of comparable



A pair of bubbles, one atop the other, ascending (*left*) in a Newtonian fluid (Eo = 0.9, Re = 1.3) and (*right*) in a shear-thinning inelastic fluid (Eo = 10.7, Re = 4.5, n = 0.7). The pair is shown in different time instants. Adapted with permission from Vélez-Cordero et al. (2011), copyright Elsevier.

width, in shear-thinning viscoelastic fluids. For the case of inelastic shear-thinning fluids, Daugan et al. (2002b) observed the formation of doublets and triplets for sets of three particles. In some cases, the doublet would leave a particle behind, as its sedimentation speed would be larger. In a settling suspension, Daugan et al. (2004) observed the formation of vertical streaks, but in contrast to Bobroff & Phillips (1998), no sedimentation fronts were detected. Using fluids that were also inelastic and shear thinning, Mora et al. (2005) confirmed the formation of vertical columns and discussed the nature of the concentration instability.

Many advances have been made via numerical simulations (Singh et al. 2000, Yu et al. 2006, Phillips & Talini 2007, Hao et al. 2009, Phillips 2010, Goyal & Derksen 2012). Most of these





are extensions of the studies for pair interactions; hence, they only consider tens of particles at the most. Considering 11 2D particles sedimenting in an Oldroyd-B, Singh et al. (2000) and Hao et al. (2009) observed vertical chaining for sufficiently high values of the elasticity number. The chaining could be prevented if enough inertia was present (Mach number higher than 1). The same general trend was observed by Phillips & Talini (2007), who extended their study of pair interactions in a second-order fluid. They also observed the appearance of vertical particle doublets. The more recent computational study of Goyal & Derksen (2012), who considered a FENE-Chilcott-Rallison fluid, find similar results for up to eight sedimenting particles.

Yu et al. (2006) conducted simulations for more than two particles but also included the effect of a shear-thinning rheology. They argue that the combined shear-thinning and viscoelastic effects produce vertical particle doublets. With sufficiently large inertia, the doublets can tumble. Simulations comparing settling in Newtonian, shear-thinning plus memory, Oldroyd-B, and shear-thinning viscoelastic fluids show stable aggregates only in the last fluid. They also found a dramatic increase in the fall velocity of aggregates of particles.

In summary, for solid particles, both elasticity and shear-thinning viscosity induce the formation of vertical chains. These structures are stable for sufficiently high elasticity numbers, provided that Ma < 1 (small inertia). When the inertia is sufficiently large, the arrays may rotate toward the horizontal direction and even disperse.

4.2. Ensembles of Bubbles

In a manner similar to the sedimentation of solid particles, ensembles of gas bubbles ascending in non-Newtonian fluids become clustered. Vertical doublets and chains of bubbles have been reported (Vélez-Cordero & Zenit 2011; Vélez-Cordero et al. 2012, 2014). However, as discussed below, dispersion can also be observed in some circumstances. One significant difference between the behavior of bubble ensembles and particle ensembles is the possibility of coalescence (Buchholz et al. 1978). In extreme cases, the topology of the flow may change into a slug-type flow (Dziubinski et al. 2004).

Figure 10 shows images of the different distributions that have been observed for Newtonian and non-Newtonian bubbly flows. First, in a Newtonian liquid (Figure 10*a*), the bubbles are dispersed and no significant aggregation is observed. For shear-thinning inelastic fluids (Figure 10*b*), the bubbles tend to form vertical doublets and large aggregations. For viscoelastic fluids with nearly constant viscosity (Figure 10*c*), the bubbles also aggregate but, in this case,



Supplemental Material

Rising swarms of bubbles in (*a*) a Newtonian fluid, (*b*) an inelastic shear-thinning fluid, and (*c*,*d*) a Boger fluid. In panels *c* and *d*, the bubbles are smaller and larger, respectively, than the critical volume at which bubble velocity discontinuity is observed in this particular fluid. In all four images, the gas volume fraction is approximately the same, $\phi \approx 0.05$. Videos of each of these cases are also shown in **Supplemental Videos 1–4**. Adapted with permission from Vélez-Cordero & Zenit (2011), copyright Elsevier, and Vélez-Cordero et al. (2012), copyright Elsevier.

very distinct vertical chains form. If the bubble size is large, as discussed below, they ascend in a dispersed manner in this Boger fluid (**Figure 10***d*). Videos of each of these cases are also shown in **Supplemental Videos 1–4**.

For bubbles rising in inelastic shear-thinning fluids, Vélez-Cordero & Zenit (2011) observed significant clustering; both vertical bubble doublets and large, roughly horizontal bubble aggregations appeared. Based on the results of Vélez-Cordero et al. (2011), Vélez-Cordero & Zenit (2011) argued that the clustering was triggered by a shear-thinning wake behind each bubble: The bubbles that ascend in the low viscosity wake would be less likely to disperse sideways into a high viscosity region. Once formed, such vertical doublets and triplets rise faster than single bubbles and capture more bubbles. As the cluster grows, so do its rise speed and inertia. Eventually, the cluster becomes a large bubble aggregate that assumes an oblong shape and a nearly horizon-tal orientation. However, excessive inertia coupled with small bubble deformability (i.e., under large-Re and low-Eo conditions) tends to hamper bubble clustering and to promote dispersion (Vélez-Cordero & Zenit 2011).

Vélez-Cordero & Zenit (2011) also reported an increase of the mean terminal bubble velocity with volume fraction (**Figure 11**). This observation contrasts with the Newtonian case where a decrease of mean bubble velocity is observed as the gas volume fraction increases (Martinez-Mercado et al. 2007). The increase of mean velocity was observed for both dispersed and clustered flows. The largest increase was mainly a result of clustering because bubble aggregates rise faster than dispersed bubbles. Moreover, because the shear rate around the bubbles increases with gas fraction, part of the increase of the mean bubble velocity results from the thinning viscosity. This was predicted by Gummalam & Chhabra (1987), who considered a cell model in which the bubbles remain dispersed. Vélez-Cordero et al. (2012) observed this behavior for dispersed bubbly flows.

In Boger fluids, bubbles may aggregate into distinctive vertical chains, as Vélez-Cordero et al. (2012) have reported. In their study, instead of varying the gas volume fraction for a given bubble size, they conducted experiments for two distinct bubble sizes at a fixed volume fraction. Interestingly, they found that whereas small bubbles tend to cluster, forming vertical bubble chains, larger bubbles ascend in a dispersed manner. This change of behavior coincides with the BVD: The two sizes straddle the critical value for the BVD in the particular fluid used in that study. **Figure 10***c*,*d* shows typical images of the two types of configurations Vélez-Cordero et al. (2012) observed. The authors argued that the formation of chains was not simply due to the normal stress, as proposed by Joseph et al. (1994). Two bubbles do not show such attraction under the same conditions because the normal stress is too weak. Rather, Vélez-Cordero et al. (2012) claim that it



Normalized mean bubble velocity U_b/U_∞ as a function of gas volume fraction ϕ for shear-thinning fluids with different values of the power index n. U_∞ is the terminal velocity of an isolated bubble. The horizontal dashed line separates the two types of flows observed: dispersed bubbles (below the *dashed line*) and clusters (above the *dashed line*). Predictions from Gummalam & Chhabra (1987) for n = 1 (*solid blue line*) and for n = 0.3 (*dashed-dotted blue line*) are also shown. Adapted with permission from Vélez-Cordero & Zenit (2011), copyright Elsevier.

is the accumulation of normal stresses, as the liquid is deformed by a successive chain of bubbles, that ensures the chaining (Li et al. 1998). The reason why the bubbles disperse for sizes larger than the critical size is not well understood. Large bubbles have experienced the BVD. According to the arguments of Joseph et al. (1994), such a velocity increase would increase the inertia of the flow, which could break the clustering. Moreover, the wake behind these larger bubbles could show a negative wake, as discussed in Section 2.3. Such flow structures may induce the dispersion of the bubble clusters, but this effect has not been investigated to date. Additionally, the interface of fast bubbles may also become mobile; such an effect could reduce the accumulative normal stress effect, leading to dispersion.

More recently, Vélez-Cordero et al. (2014) studied the dynamics of preformed clusters in non-Newtonian liquids. They observed that the BVD was not observed for clusters as it was for individual bubbles. Moreover, a weaker negative wake was observed. Interestingly the study shows that vertical chains of bubbles prevail in shear-thinning viscoelastic liquids, whereas in Boger fluids, the clusters are less elongated. This seems consistent with observations of solid particles in sedimentation (Singh et al. 2000, Yu et al. 2006, Hao et al. 2009). For the case of particles, the clusters formed during the sedimentation, whereas for bubbles the clusters were preformed.

In summary, for shear-thinning inelastic fluids, the formation of small and large bubble clusters is observed. Small clusters appear as vertical bubble doublets; large bubble aggregations have an irregular shape but are roughly aligned horizontally. For viscoelastic fluids with nearly constant viscosities, there is a transition from vertical chain aggregation to dispersion as the bubble volume increases. The transition seems to coincide with the velocity discontinuity of a single rising bubble. For fluids that are both shear thinning and viscoelastic, bubbles form vertical columns, as solid particles do in sedimentation. This results from the cooperative effects of shear thinning and elastic normal stresses. Bubble deformability is also expected to influence the behavior of bubble swarms and aggregation because it is an important factor in bubble–bubble interaction (Vélez-Cordero et al. 2011). However, this issue has not been investigated in detail to date.

5. CONCLUSIONS AND OUTLOOK

For particles, drops, and bubbles that settle or rise in a non-Newtonian fluid, the non-Newtonian rheology changes the nature of the hydrodynamic interaction significantly from what is expected in a Newtonian fluid. In this review, we have identified four rheological factors that modify their interaction and potential aggregation. (*a*) Shear thinning produces low-viscosity regions in the wake and vicinity of a moving object and thereby encourages neighboring objects to aggregate and discourages dispersion. (*b*) Memory coupled with shear thinning can produce the so-called corridor effect, in which a rising or falling object leaves a low-viscosity corridor in its wake. (*c*) Normal stresses promote aggregation of neighboring objects and also tend to elongate drops and bubbles in the flow direction. (*d*) Elongational viscosity can produce a negative wake behind a single object. The strong elongation can also deform drops and bubbles by producing a pointed tip on the downstream side.

As discussed throughout this review, clustering is the dominant feature in this type of flow. Importantly, it is not yet possible to predict when clusters will form. In other words, critical values of the Deborah number (for viscoelastic fluids) or the power index (for shear-thinning fluids) have not yet been determined. The identification of such conditions is of crucial importance for practical applications. The performance of devices handling flows of this nature will be strongly influenced by the presence of clusters; the heat and mass transfer rates will be significantly lower than those expected for a well-dispersed system. Given the state of current understanding, engineers and designers should assume that clusters will form and make design choices accordingly.

In general, we have a much better understanding of the behavior of solid particles than that of bubbles and drops. The deformable interface adds to the complexity of the problem. For numerical simulations, additional measures to track and model the interface are needed, adding to the difficulty of producing physically meaningful results. Experimentally, when bubbles or drops are considered, the number of parameters increases significantly, making the experimentation long and tedious. In most of the studies conducted so far, the conditions and parameter regimes are such that the behavior of drops and bubbles largely resembles that of solid particles. In other regimes, we can expect the special features of drops and bubbles, e.g., interfacial deformation and coalescence, to produce novel dynamics not shared by solid particles.

SUMMARY POINTS

- 1. A pair of objects rising or falling side by side in a viscoelastic fluid attract each other due to the normal stress effect. As the two approach, the line of their centers rotates toward the vertical direction.
- 2. A pair of objects rising or falling one atop the other may attract each other through viscoelastic normal stresses, the corridor effect in fluids with shear thinning and memory, or both. In a Boger fluid with sufficient elasticity, two particles may experience a near-range repulsion such that the two approach an equilibrium separation regardless of their initial separation.

- 3. A suspension of settling particles exhibits a concentration instability in shear-thinning inelastic and shear-thinning viscoelastic liquids, by which particle-rich vertical columns form and alternate with columns of clear liquids.
- 4. A swarm of rising bubbles tends to aggregate in shear-thinning and viscoelastic fluids, although the configuration and orientation of the aggregates differ. In shear-thinning fluids, the aggregates tend to take on an oblong shape and a more or less horizontal orientation. In Boger fluids, long vertical chains prevail if the bubble size is below the threshold for the BVD. Larger bubbles tend to disperse.
- 5. These non-Newtonian effects are prominent in low–Reynolds number flows with small inertia. As the fall or rise velocity increases, inertia becomes important and tends to oppose the non-Newtonian effect, e.g., by rotating chains and elongated clusters toward the horizontal direction and even causing dispersion.

FUTURE ISSUES

- 1. The non-Newtonian rheology affects the rise of a single bubble or drop, e.g., via unsteady complex wake dynamics and interfacial deformation. Of particular interest is how rheology of the surrounding fluid modifies the thresholds between the various regimes of bubble or drop dynamics that are known in Newtonian fluids.
- 2. The deformability of bubbles and drops influences their interaction and especially aggregation. For the special case of Boger fluids, preliminary experiments offered tantalizing clues on how the appearance of the pointed rear tip accompanies the transition from chaining to dispersion. A more systematic investigation of this question will likely be fruitful.
- 3. Despite qualitative indications of how bubble aggregation can drastically modify the average bubble rise velocity, there have been few quantitative measurements, e.g., on how the average rise velocity depends on the size of aggregates and in turn on the non-Newtonian rheology of the suspending fluid.
- 4. Surfactants are ubiquitous in experimental systems, either as a result of contamination or a means of controlling surface dynamics and transport. In Newtonian fluids, the role of surface contamination on bubble dynamics has long been recognized (Stone & Leal 1990, Takagi et al. 2008, Hallez & Legendre 2011). In non-Newtonian fluids, relatively little has been done, including on Marangoni effects.
- 5. Numerical simulations have played a major role in elucidating the dynamics of sedimenting particles. For bubbles and drops, the interfacial mobility and deformability add to the numerical challenges. But recent progress has greatly widened the range of problems that can be simulated, and we expect the computation of bubble and drop interactions to yield new insights in the near future.
- 6. More detailed and better-controlled experiments are needed both to validate numerical simulations and to explore new regimes and behaviors. This is true for solid particles as well as for drops and bubbles.

7. With our increasing knowledge of the hydrodynamic interaction and aggregation of particles, drops, and bubbles in non-Newtonian fluids, we can apply the scientific insights and techniques to increasingly complex industrial applications, for instance, on various aspects of bubble column reactors and polymer blending and processing.

DISCLOSURE STATEMENT

The authors are not aware of any biases that might be perceived as affecting the objectivity of this review.

ACKNOWLEDGMENTS

We greatly thank D. Legendre, J.R. Vélez-Cordero, and J. Tsamoupoulos for useful comments on the manuscript. R.Z. has been supported by CONACYT Mexico grant CB-2008-102527 and by the PAPIIT-DGAPA-UNAM research program grants IN103900, IN102303, and IN101312. J.J.F. has been supported by the Natural Sciences and Engineering Research Council (NSERC) of Canada grant 05862.

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