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Impact of concentrated colloidal suspension drops on solid surfaces

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ABSTRACT

When droplets of high concentration wet powder impact on a solid surface, the large stresses that build up upon impact may convert them to a stable system of dry granules. Dilation/jamming has been proposed to explain such powder granulation processes. Stress causes dilation of particles through the droplet surface, against capillary pressure, roughening the surface on the scale of the constituent particles. Under the right conditions of stress magnitude and particle concentration, the droplet jams internally in response to capillary pressure, forming a mechanically stable granule. This remains a tentative model of granulation, which despite its importance in process industries ranging from minerals to foods to detergents, is still imperfectly understood.

This work presents the preliminary results of drop impact experiments of a suspension of near hard-core colloidal particles, with the purpose to investigate the impact morphology in the presence of shear thickening or jamming, which may be induced by the large velocity gradients arising upon drop impact. In particular, drops of a suspension of nearly hard-core particles in octadecene (volume fraction: approximately 60%) impacting on substrates of different wettability are studied experimentally by high-speed imaging, for impact Weber numbers ranging between 26 and 262. Upon impact, these drops do not exhibit inertial spreading, which is observed for other Newtonian and non-Newtonian fluid drops. On wettable surfaces (glass), impact is followed by capillary-driven spreading at the same rate observed in power-law fluids (Starov's law), while on less wettable surfaces (PTFE) the colloidal suspension drops relax to achieve the shape of a spherical cap, but do not spread. This peculiar impact morphology, and in particular the absence of inertial spreading, is interpreted as a consequence of dilatancy and jamming occurring upon impact.

1. Introduction

The impact of single and clouds of drops on a solid surface is of great scientific interest and practical importance. An improved understanding of drop impact is needed in applications as diverse as spray coating, spray painting, delivery of agricultural chemicals, spray cooling, ink-jet printing, soil erosion due to raindrop impact, turbine wear, soldering, powder granulation and even forensics (blood spatter). The phenomenology of macroscopic drop impact behaviour (shape deformation, spreading, rebound) can be described in terms of characteristic dimensionless groups such as the Reynolds number ($Re = \rho u D_0 / \mu$, where ρ is the fluid density, u the impact velocity, D_0 the equilibrium drop diameter, and μ the dynamic viscosity), describing the relative roles of viscosity and inertia, and the Weber number ($We = \rho u^2 D_0 / \sigma$, where σ is the surface tension of the fluid), which represents the competition between kinetic energy and surface energy. Since the pioneering observations of Worthington [1] more than 100 years ago many studies have been carried out on a range of fluids and surfaces, in a range of conditions [2,3]. However, there remains limited fundamental understanding of the roles of fluid parameters and surface factors, especially in the practically most relevant case of fluids with complex non-Newtonian rheology. Non-Newtonian or so-called complex fluids [4] such as polymer solutions and melts [5–9], surfactant solutions or phases [10–12], polymer gels and dispersions [13–16], and pastes and slurries, show stress-dependent response, e.g. shear- and extensional thinning and thickening, frequency-dependent viscoelasticity, significant normal stress differences, and history-dependent and geometry-dependent response. These rheological features are a consequence of mesoscopic or macromolecular-scale structure (polymers, micelles, colloids) with relaxation times comparable to the typical flow timescale: hence flow can generate significant mesoscopic structural deformation (e.g. aligned polymers, ordered colloid phases) taking systems far from mechanical equilibrium. In droplet impact, stress and strain rates also vary temporally and spatially throughout the event, therefore the structural/rheological response of the droplet can become exceptionally complex [17].

Particulate systems are an important class of non-Newtonian fluids. Particulates include colloidal suspensions, pastes and slurries (wet granulars), and emulsions (deformable particle systems), and are thus relevant across many deposition processes, such as spray-painting, surface treatment, soldering, and granulation. While dilute concentrations

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of colloids/particles (<few % by volume) may have little rheological or dynamic effect beyond increase in effective viscosity and/or decrease in diffusion rate, highly concentrated particulates such as colloidal glasses and granular slurries show complicated rheology including yielding (the system behaviour is solid-like below a critical stress, or yield stress, and fluid-like above), jamming (the system behaviour is fluid-like below a critical stress, and solid-like above), and complex glassy dynamics [18,19]. None of these phenomena are fully understood, but substantial research efforts over the past decade and significant advances in techniques such as direct observation by confocal microscopy, computational methods, and theoretical methods such as mode-coupling, soft glassy models, and shear-transformation zones [18–22], have seen a rapid improvement in basic understanding.

Jamming is often associated with dilation: as shown for granular media by Reynolds [23], and more recently investigated in colloids and pastes [24,25], above a critical concentration a system of particulates cannot deform macroscopically without local microscopic dilation, i.e. local decrease in particle volume fraction. When an incompressible solvent is involved, local dilation of the particle system requires either increase in local volume fraction elsewhere (and hence fluid flow) or, in the case of a droplet with a free surface, expansion of the total droplet volume may be possible by the particles deforming the surface. The dilation/jamming mechanism has been proposed to explain powder granulation processes where droplets of high concentration wet powder are converted by applied stress to a stable system of dry granules [26]. Stress causes dilation of particles through the droplet surface, against capillary pressure, roughening the surface on the scale of the constituent particles. Under the right conditions (stress, concentration) the droplet jams internally in response to capillary pressure, forming a mechanically stable granule. This remains a tentative model of granulation, which despite its importance in process industries ranging from minerals to foods to detergents, is still poorly understood.

Given this growing baseline of fundamental dynamics and rheological theory, there is now real potential to link droplet impact behaviour in particulates to fundamental system properties, via systematic quantitative data from controlled model experiments. A comprehensive, well-controlled study of droplet impact will feed back to better understanding of the consequences of glassy, yielding and jamming rheology. Furthermore, due to the high stresses and velocity gradients arising during impact, well beyond the maximum values achievable in conventional rheometric instruments, impacting drops represent a unique system to explore the phenomenology of yielding and jamming under extreme conditions.

This work presents the preliminary results of drop impact experiments of a suspension of near hard-core colloidal particles, with the purpose to investigate the impact morphology in the presence of shear thickening or jamming, which may be induced by the large velocity gradients arising upon drop impact.

2. Experimental method

2.1. Material preparation and characterisation

A model colloidal suspension characterised by nearly hard-core (i.e., purely repulsive) interactions was prepared with poly-methylmethacrylate (PMMA, density: 1180 kg/m^3) spheres (radius r approximately 604 nm, 5% polydispersity) sterically stabilised by poly-12-hydroxy stearic acid (PHSA) chemically grafted onto their surface [27], and suspended in octadecene (density: 789 kg/m^3; surface tension: 0.03 N/m). Suspensions were centrifuged at 2000 rpm for 12 h to create a sediment, which was assumed to have a concentration of 64% (random close packing or maximum random packing) [28]. The sediment was then diluted with octadecene to a concentration of approximately 60%, corresponding to an average density of 1024 kg/m^3.

A correct choice of the fluid medium is essential to ensure that particles interact as hard spheres. In particular, the fluid must be a poor solvent for PMMA, in order to limit particle swelling, but a good solvent for PHSA so that polymer chains stretch out. When two particles come into contact, the repulsive energy given by the superposition of the two polymer layers rapidly increases as the distance between particles reduces [29]. Among the various suitable solvents, octadecene was selected because of its very low volatility.

According to the equilibrium thermodynamics phase diagram of ideal hard-spheres [30], for volume fractions between 54.5% and 74% (maximum crystalline close packing fraction) the system is crystalline. However, experiments showed that model suspensions of sterically-stabilized colloidal hard-spheres failed to crystallize for volume fractions greater than 58%, remaining in a non-equilibrium phase known as the colloidal glass [31].

Fig. 1 shows the flow curve for a dense hard-sphere suspension at a volume fraction of approximately 0.6, obtained using a Thermo Mars II rotational rheometer equipped with stainless steel cone-plate geometry, in controlled-stress mode. At shear rates $\lesssim 2\ s^{-1}$, the fluid behaviour is initially shear-thinning, with a small yield stress (with magnitude of the order of approximately 1 Pa), however a strong shear thickening can be observed at shear rates between 2 s^{-1} and 3 s^{-1}. Such shear-thickening could be an indication of jamming inside the fluid sample. Indeed, dilatancy and jamming can be directly related to shear-thickening during extensional rheology experiments [25]. The shear-thickening part of the flow curve can be approximated by a power law, where coefficients are obtained by least-square best fit of experimental data:

$$\tau_{xy} = 0.038\dot{\gamma}^{5.22\pm0.18}. \quad (1)$$

For the sake of comparison, a Newtonian fluid with a viscosity of approximately 1 Pa·s was obtained by preparing a 98% solution of glycerol (density: 1250 kg/m^3) in water.

2.2. Experimental apparatus and procedure

Drops were generated using a syringe with blunt hypodermic needle (gauge 21, 0.495 mm i.d.) driven by a micrometric screw, and detached under their own weight. The needle was suspended above two substrates of different surface energy (glass and PTFE). To change the impact velocity, the drop release height was adjusted between 2 and 18 cm using a Vernier height gauge, which corresponds to theoretical free-fall velocities of the drop upon impact with the substrate between 0.6 and 1.9 m/s. The competition between inertia and surface forces was characterised through the Weber number, $We = \rho v^2 D_0 / \sigma$, where ρ is the suspension density, σ is the solvent surface tension, v is the drop

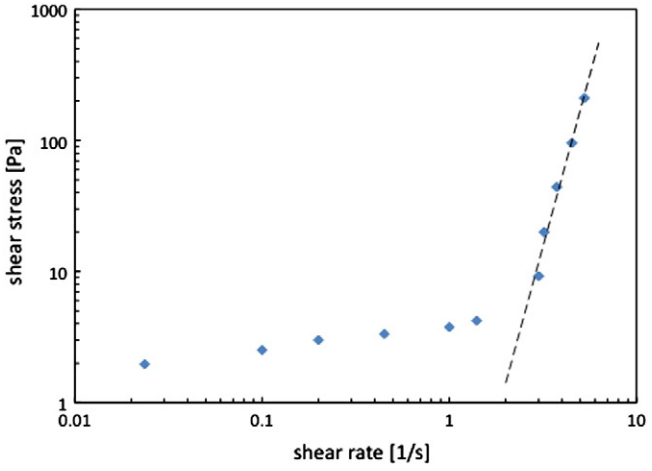

Fig. 1. Flow curve of a colloidal suspension with volume fraction of 0.603, measured in a rotational rheometer with cone-plate geometry at controlled stress. The dashed line represents a least-squares best fit to the last five points of the flow curve (Eq. (1)).