

Drop Impact Dynamics of Complex Fluids: A Review

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Abstract

The impact of fluid drops on solid substrates has widespread interest in ink-jet printing, agricultural pesticides, and many other coating and spraying applications due to its relevance to a variety of industrial processes. Drop impact dynamics has been an active field of soft-matter research for over a century. A large body of experimental and theoretical work has been done to understand the impact dynamics of Newtonian fluids, yet we are far from understanding the drop impact of complex fluids. In this review, we outline recent experimental, theoretical, and computational advances in the study of impact dynamics of complex fluids on solid surfaces. Here, we provide an overview of this field, with our discussion segmented by rheological behavior. Throughout, we highlight promising future directions, as well as ongoing experimental and theoretical challenges in the field.

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I. INTRODUCTION

The impact of fluid drops on solid surfaces is ubiquitous in many natural and industrial processes such as geological erosion, spraying and coating of surfaces, ink-jet printing, and agrochemical delivery. In addition to the impact conditions (impact velocity, drop size, impact angle), the dynamics of drop impact depends on the properties of the fluid (density, surface tension, viscosity), the properties of the surface (contact angle with the fluid, surface roughness and hydrophobicity), and the properties of the surrounding gas (density, pressure) [1, 2].

Even at moderate impact velocities (~ 1 m/s), fluid drops experience relatively high shear stresses upon impact onto a solid surface. The overall shear rate in the drop at impact can be estimated as the ratio of impact velocity and drop diameter. For instance, a fluid drop of diameter $d_0 = 3$ mm impacting a surface at a velocity of $u = 5$ m/s would undergo a shear rate of $\dot{\gamma} = \frac{u}{d_0} = 1.67 \times 10^3$ s⁻¹. This is approaching the operating limit of most shear rheometers. Drop impact experiments can therefore provide a bridge to expand our understanding of flow properties of complex fluids into the high-stress regime. Furthermore, drop impact allows an exploration of these flow behaviors in a system with a deformable free surface, rather than the bulk measurements one typically encounters in rheometry data. As we will discuss, this allows for observation of new and unusual behaviors that only appear at high shear rates.

Drop impact of Newtonian fluids has been an active field of research for more than a century [6]. In recent years, the rapid development of high-speed imaging and image analysis techniques has re-invigorated the field. Complex fluids exhibit an even wider variety of behaviors when impacted on solid substrates [Fig. 1]. While the impact dynamics of Newtonian fluids have been studied extensively, the body of work done to understand the impact of complex fluid drops has been quite limited to date.

The goal of this review is to provide a comprehensive summary of the work on the impact of complex fluid drops. Much of the work summarized here is confined to impact on solid surfaces—the most straightforward impact scenario to interpret. This area of research is both vast and largely unexplored, and past works have pursued very disparate avenues. Here, we attempt to classify the existing work into physically relevant categories. We hope it will provide a useful reference point for future studies and help move towards a more

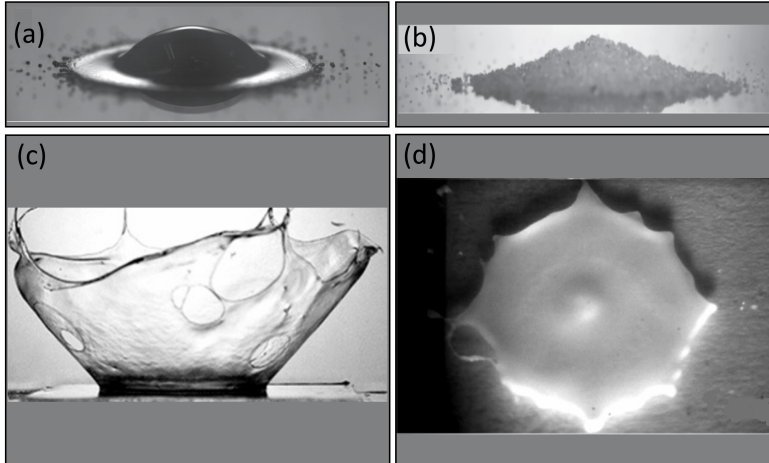


FIG. 1. Post-impact snapshots of (a) a drop of mercury (a Newtonian fluid) on glass (Adapted from [2]), (b) a drop of dense granular suspension on glass (Adapted from [3]), (c) a drop of 0.25% Carbopol in water on a wet substrate (Adapted from [4]), and (d) a cornstarch drop on a hydrophobic, micro-patterned substrate (Adapted from [5]).

complete understanding of the physics of complex fluids under high stresses.

The review is structured as follows: Section I A provides an overview of the current state of the Newtonian drop impact field, followed by section II summarizing the relevant concepts of fluid rheology. We organize the following sections according to different complex fluid behaviors. Section III outlines the impact dynamics of granular suspensions. In section IV, we summarize both experimental and numerical results for the impact of yield-stress drops. The advances in the impact dynamics of shear thinning fluids are outlined in section V A, followed by shear thickening fluids in section V B. Section VI concludes the review by discussing the current challenges in the field, and also suggests several directions of research that would lead to a comprehensive understanding of the impact of complex fluids on solid substrates.

A. Advances in Newtonian drop impact

Compared to studies of complex fluids, the drop impact of Newtonian fluids is a relatively more mature field of study. Despite many key differences between the impact dynamics of Newtonian and complex fluids, Newtonian drop impact studies have provided the foundation for their complex fluid counterparts. In this section, we summarize the state of the Newto-

nian drop impact field, before we dive into the rheology and impact dynamics of complex fluids. This is not intended to be a comprehensive review of Newtonian drop impact. For a deeper discussion of this field, we refer the interested reader to the following reviews on the subject [1, 2].

The relative importance of various quantities is quantified in terms of dimensionless numbers. Here we outline the most relevant dimensionless numbers for Newtonian drop impact:

1. Reynolds number: ratio of inertial and viscous stresses. $Re = \frac{\rho U d_0}{\mu}$
2. Weber number: ratio of inertial and surface stresses. $We = \frac{\rho U^2 d_0}{\sigma}$
3. Ohnesorge number is sometimes used instead of Reynolds and Weber numbers, and is given by $Oh = \frac{\sqrt{We}}{Re}$. The Ohnesorge number compares the effect of viscosity with the combined effect of surface tension and inertia, making it an ideal dimensional entity for use in studies where inertial, viscous, and surface stresses may all be relevant.

Here ρ is the fluid density, U is the drop velocity, d_0 is the drop diameter before impact, μ is the dynamic viscosity, and σ is the fluid's surface tension. These and other dimensionless numbers are often modified to describe complex fluids. We will introduce these modifications as needed.

So far, the work in this field has studied the stages of early impact, the relatively longer-timescale spreading and receding dynamics, and also the transition from spreading to splashing [1, 2]. At early stages of contact, an air layer has been observed under the spreading fluid, and this air layer has been experimentally characterized with a variety of techniques including interferometric and total internal reflection imaging [7–11]. Despite extensive studies, the role of this air layer in splashing dynamics has not been resolved. For some splashing regimes such as low velocity impacts, it appears to strongly control spreading, whereas in other regimes such as high velocity or viscous fluid impact, the air layer appears to play no role in controlling spreading. Furthermore, while ambient pressure is known to control the splashing transition [12], it remains an open question how this phenomenon is connected to the air layer entrapped at impact.

Over longer timescales, the spreading and receding dynamics of the drop arise as a balance between the drop inertia, viscosity, and surface tension. The thickness of the expanding liquid film is not always uniform, the lamella at the boundary is often thicker than rest

of the disc. Additionally, the structure and wetting properties of the solid surface play a large role in the spreading and receding dynamics. Since both intrinsic fluid properties and the fluid-substrate interaction affect the impact dynamics, decoupling the two effects is essential for a better understanding of this process. To this end, experiments on small targets are a very effective way to minimize substrate effects. Such targets are typically comparable in size to the drop diameter, but are much smaller than the maximum spread of the drop, eliminating most of the fluid-surface interaction during spreading. It should be noted that gravity plays a negligible role over the impact timescale (a few milliseconds), therefore small target studies still produce a horizontally spreading film. Scaling laws have been fairly successful in characterizing the spreading over time in both viscous and inertial regimes [2]. Despite having a good understanding of how the spreading diameter grows with time, current predictions are less effective when the drop is close to the splashing transition. This is due to the difficulty in clearly demarcating when spreading stops and splashing begins.

The transition to splashing has been observed to be governed by the parameter $K = We(Re)^{\frac{1}{2}}$, where higher values of K lead to splashing [13, 14]. This characterization of the splashing threshold incorporates only the fluid properties. In addition to liquid parameters, the quantitative value of the splashing threshold has been shown to depend both on the details of the surface [15] and the surrounding gas properties [12]. Thus, while the parameter K has been useful in providing a state diagram for this problem, the phase space it covers is limited. The combined effect of surface roughness and gas properties has attracted a lot of interest, and recently theoretical progress has been made in understanding how these parameters control spreading and splashing [16]. A complete understanding is still to emerge. Quantitative studies of the number of fingers and the size distribution of ejected droplets have also been performed, but more work is needed to achieve a consolidated framework.

In many applications, drops impact on inclined or soft surfaces, for example pesticides sprayed onto leaves. These surface modifications have been explored in a variety of studies. The large asymmetry in the problem of a drop impacting onto a tilted surface makes it challenging yet interesting. Recent studies have explored splashing in case of oblique and translating surfaces [17–19], and have found that in some cases splashing can be suppressed in the ‘uphill’ direction, while occurring in the ‘downhill’ direction due to the differing effective impact velocity experienced by the drop. It has generally been observed that impact onto a

more compliant surface lowers the splashing threshold [20]. A recent work studied in more detail the effect of solid elasticity on splashing and observed that decreasing the Young's modulus of the substrate suppressed splashing, and linked these changes to the increase of energy dissipation in the softer substrate [21]. In addition to shedding more light on the role of the substrate in drop impact, these studies might open up novel ways to control splashing and secondary droplet ejection in a variety of applications.

The relevant parameter space for Newtonian drop impact is large, since it includes properties of the liquid, the substrate, as well as the surrounding gas. Despite this, several scaling parameters have been constructed for the variety of behaviors in fluid drop impact. These empirical scaling laws are very successful for specific regimes, but we do not have a unified framework for the whole impact process.

Even though a lot of experimental data and empirical models are available, we are far from achieving a cohesive theoretical understanding of the drop impact phenomenon. Though progress has been made [22, 23], simulation and theory in this area are particularly difficult due to the vast range of timescales and lengthscales at which different processes take place [2]. An additional complication in this problem is the role of the dynamic contact angle [24, 25], which differs during the spreading and receding phases, and also with the fluid-surface combination [26]. The results of simulations are sensitive to the assumptions made about contact angles, making it harder to reconcile results of simulations with experiments. Furthermore, it remains a challenge to measure the microscopic dynamic contact angle experimentally. Additionally, most experimental data are at intermediate values of Reynolds numbers, a regime that is the most challenging to study theoretically due to the competing forces of inertia and viscosity.

In addition to the need for more theoretical studies, there are multiple experimental avenues that deserve more exploration. Although there have been some studies about substrate properties [27, 28], more experiments with micro-patterned substrates (with both regular and random patterns) would be beneficial to a number of industrial applications. There are many unanswered questions about oblique drop impact too, which are highly relevant to real-life impact processes.

II. RHEOLOGY OF COMPLEX FLUIDS: OVERVIEW

What makes drop impact of complex fluids so rich and interesting is the large variety of flow behaviors of complex fluids. By definition, complex fluids are ones whose flow behavior is a non-constant function of both the applied strain rate and of different timescales. This is in contrast with Newtonian fluids that exhibit a constant viscosity over a range of strain rates. Rheology is the study of material behavior as a function of applied stress [29]. Drop impact studies allow us to access regimes of very high strain rates, potentially opening new avenues for the development of high-stress rheological techniques.

In this section, we summarize the concepts of rheology that are relevant to the work presented in later sections. Therefore, we will mostly discuss the fundamentals of shear rheology and yield-stress rheology. The flow behavior of complex fluids is a vast field of research, and details of many other phenomena including viscoelasticity and thixotropy are outside the scope of this review [30, 31].

The most commonly encountered non-Newtonian flow behaviors are shear thinning and shear thickening. Shear thinning is a phenomenon where fluid viscosity decreases as the shear rate is increased. Paint and whipped cream are shear-thinning fluids, since they flow more easily when sheared using a brush or a spraying can. Shear thickening fluids are the exact opposite, i.e. they show a higher viscosity and hence a higher resistance to flow at higher shear stresses. The most commonly known example of a shear thickening fluid is oobleck, a mixture of corn starch and water. Oobleck acts like a semi-solid blob in response to applied shear, but flows again once the shear is removed.

Many colloidal, granular, and polymeric fluids show both shear thinning and thickening behaviour, depending on the particle/polymer concentration and the value of applied shear stress [34, 35]. Shear thinning is believed to be the result of the microscopic rearrangement of particles or detangling of polymers due to applied shear. In colloidal suspensions, the origins of this behavior has not been fully resolved, but it is thought that partial phase separation and hydrodynamic friction might also play a role in the shear-thinning. Shear thickening typically occurs at high shear rates, when contact forces between particles overcome the lubrication between particles and fluids. Fig. 2(b) outlines the possible mechanisms of shear-thinning and thickening behavior. There are two types of shear-thickening behavior: Continuous Shear Thickening (CST) and Discontinuous Shear Thickening (DST). Continu-

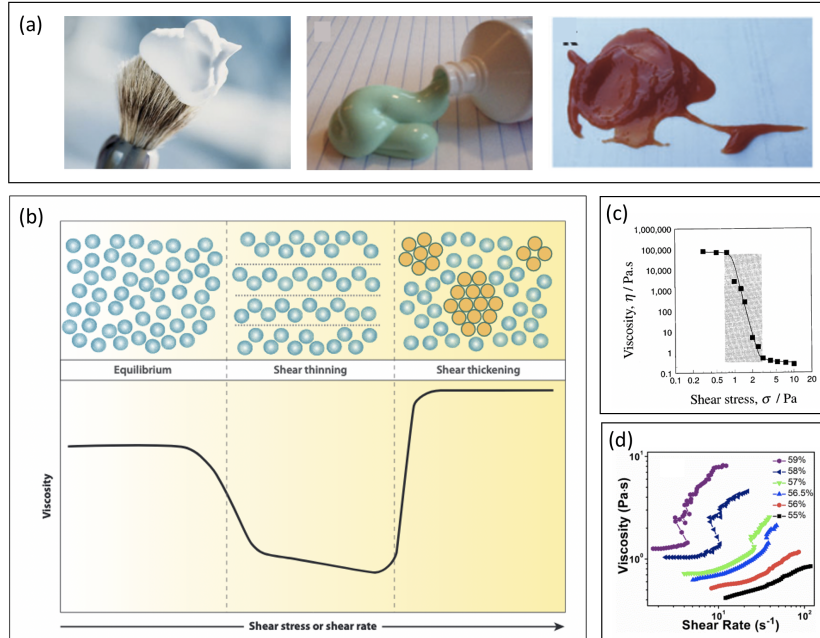


FIG. 2. (a) Many everyday fluids such as shaving foam, toothpaste, and ketchup show an initial yield-stress and shear-thinning behavior. (b) A schematic of the microscopic origins of shear-thinning and thickening behavior in particulate suspensions. At zero stress, the Brownian collisions among particles gives the fluid a viscosity. At non-zero shear rates, the particles rearrange in the fluid flow, and the bulk viscosity drops. At even higher rates of shearing, the contact forces among particles become significant, and formation of particle clusters causes shear-thickening. Adapted from [29]. (c) Viscosity vs. shear stress plot for an iron oxide suspension in mineral oil. As shown by the shaded region, the viscosity drops by 5 orders of magnitude with a small change in shear stress. Adapted from [32]. (d) Viscosity vs. shear rate plots for PMMA suspensions in NaI and water. As the particle volume fraction increases, the fluid transitions from Continuous Shear Thickening to Discontinuous Shear Thickening. Adapted from [33].

ous Shear Thickening(CST) is when viscosity increases continuously with shear rate, while the sudden jump observed in viscosity at high shear rates is known as discontinuous shear thickening. Discontinuous Shear Thickening(DST) often happens near ‘jamming’ of particles, beyond which flow is not possible without fracture [Fig. 2(d)]. Other mechanisms of DST below the jamming concentration, such as a transition from lubrication-mediated contacts to friction-mediated contacts, have also been proposed [36–38].

Another commonly observed rheological behavior is the observation of a ‘yield stress’.

‘Yield stress’ is the critical stress at which a fluid transitions from solid-like behavior to liquid-like flow properties [32, 39, 40]. To unpack this a little more, if we subject a material to a slowly increasing shear rate, the material initially shows very high viscosity (for all practical purposes acting as a solid). However, above the critical value of stress (yield stress), the viscosity dramatically drops and the material transitions to more liquid-like behavior [Fig. 2(c)]. This behavior is observed in many commonly encountered fluids: toothpaste, peanut butter, and wet cement to name a few.

The yield-stress property is quantified by the well-known Bingham model [41–43]. This model assumes that the shear rate is zero under the critical stress, i.e. the material acts like a solid, and the shear rate grows linearly above the critical stress, i.e. the material acts like a Newtonian fluid. Although convenient, this framework is an oversimplification. Many materials show steady deformation even below the yield-stress value, and the general consensus now is that ‘everything flows’, though this sometimes occurs over quite long timescales or with very little material deformation. However, the concept of a yield-stress is still very useful, since many materials do exhibit a dramatic change in flow behavior at a critical shear stress. Here, the materials we consider ‘yield-stress fluids’ — also sometimes referred to as ‘viscoplastic fluids’ — are the ones whose critical stress values and timescales of deformation lie within the experimentally accessible range.

For shear stresses higher than the critical value, yield-stress fluids often exhibit shear-thinning behavior. Some fluids can also transition to shear thickening at high shear. The Herschel-Bulkley model [44] is widely used to theoretically capture both the yield-stress and the shear rheology of a fluid:

$$\tau = \tau_o + k\dot{\gamma}^n$$

Here, τ is the shear stress, $\dot{\gamma}$ is the shear rate, τ_o is the yield-stress, k is the consistency coefficient, and n is the power law index. For $\tau < \tau_o$, the fluid behaves as a rigid solid, and for $\tau > \tau_o$ it transitions to fluid behavior. Newtonian behavior corresponds to $n = 1$, $n < 1$ being shear-thinning and $n > 1$, shear thickening. Even though this model captures most of the features of yield-stress and shear rheology, it should be noted that it does not completely capture the behavior of fluids that may transition from shear-thinning to shear-thickening at larger shear rates.

Complex fluid rheology is, of course, not limited only to shear rheology and yield-stress

behaviors; these materials exhibit many intriguing flow behaviors other than the ones mentioned above. One such property is viscoelasticity: the property of materials to behave like either elastic solids or viscous liquids. Viscoelastic materials respond differently to different timescales of applied stress, i.e. the same material can show an elastic response on shorter timescales but more viscous fluid-like behavior over a longer time. This is different than viscoplastic or yield-stress fluids, where the material primarily responds to the amount of shear. Another phenomenon worth a mention is thixotropy: the change in material properties such as viscosity as the material ages [31]. Lastly, elongational rheology studies material responses to elongational stresses (as opposed to shear stresses) [45]. Although all these factors must play an important part in the drop impact process, their roles are highly active fields of research, and are outside the scope of this review.

III. GRANULAR SUSPENSIONS

Suspensions comprising of particles larger than the Brownian limit ($\geq 10 \mu m$ in this review), or granular suspensions, are expected to behave in a unique manner, since inter-particle interactions can generally be neglected in granular suspensions as opposed to Brownian (colloidal) suspensions. The study of granular suspensions is also unique because the particles in this case are large enough to be detected without a microscope, yet about an order of magnitude or more smaller than the typical drop diameters chosen in impact studies.

Granular particles are large enough to escape the surface forces of the surrounding fluids and individually escape, which makes the drop impact of granular fluids fundamentally different from that of Newtonian and colloidal fluids [Fig. 3(a)]. Naturally, the dimensionless parameters typically used to study Newtonian and colloidal drop impact ($We = \frac{\rho U^2 d_o}{\sigma}$, $Re = \frac{\rho U d_o}{\mu}$, and $Re_{eff} = \frac{\rho U d_o}{\mu_{eff}}$) need to be modified in order to understand granular drop impact.

A. Spreading and receding dynamics

Multiple studies have investigated the spreading dynamics of granular drops at moderate to high volume fractions [3, 46–50]. The maximum drop diameter d_{max} is observed to decrease with increasing particle volume fraction, but is not significantly affected by indi-

vidual particle size. The drop diameter d grows as a square root of time, very similar to the spreading of Newtonian drops [3]. For low and moderate particle concentration, spreading dynamics can be quantitatively understood in terms of the steady effective viscosity of the suspension. As effective viscosity grows with particle concentration, d_{max} decreases.

Jorgensen et al. [46] used rheological measurements to measure effective viscosity and compared their findings with the spreading of Newtonian drops of similar viscosity values. The agreement between the data verifies that the spreading dynamics for granular suspensions is primarily controlled by the bulk effective viscosity. At a given shear rate, a complex fluid can be effectively treated as a Newtonian fluid with a certain viscosity. This value is the fluid’s ‘bulk effective viscosity’ and can be inferred from the rheology data of the fluid. Numerically, the bulk effective viscosity is an extension of the Einstein viscosity beyond the linear term. Other quantifiers of drop deformation, such as the steady-state height of the drop have also been observed to be controlled by the effective viscosity [49]. Additionally, over longer timescales (a few seconds), the drops are observed to relax due to interaction with the substrate.

When the particle concentration approaches the critical volume fraction for jamming (ϕ_c), the effective viscosity diverges. For high volume fraction drops, even though the effective viscosity framework predicts zero deformation, non-zero deformation was observed after impact. One proposed explanation for this has been the rearrangement of particles leading to a variable volume fraction during impact. However, a quantitative understanding of the impact in the high-concentration limit is yet to be reached.

The retracting phase on the other hand is observed to be more affected by substrate properties and the viscosity of the suspending liquid. At low fluid Re , the oscillations of the drop diameter are suppressed, in turn affecting the steady-state particle distribution [48]. Long after impact, the particles are concentrated in an annular region for a high- Re suspending fluid, while they exhibit a more or less uniform distribution for a low- Re suspending fluid [Fig. 3(c)].

In order to minimize dissipation due to fluid-surface interactions, Raux et al. [50] performed impact experiments on small targets. This study showed that the d_{max} and t_{max} are independent of particle size, indicating that even on small targets, particles only affect the drop spreading dynamics through the bulk effective viscosity. The retraction, on the other hand, is slowed down by the presence of particles. Larger particles are observed to make

the film unstable during the receding phase, leading to rupture and a decrease of the film lifetime. The ejected droplets were clusters of particles held together by liquid, as opposed to individual particles in case of dense suspensions.

Close to the critical volume fraction ϕ_c , the effective viscosity framework cannot explain impact dynamics. Lubbers et al [51] studied the inertia dominated splat of dense suspensions that resulted in a particle monolayer. The growth of the monolayer was observed to be fundamentally different than the growth of a liquid drop on a surface. This high- ϕ , inertia-dominated phenomenon was explained by the particle-based Weber number We_p and liquid Stokes number St . The authors proposed a particle-based chain model and viscous drag model, both of which make predictions that agree with their experimental findings.

To sum it all up, impact of low- and moderate- volume fraction drops can be understood using the bulk effective viscosity of the suspension, but for high concentration drops, the liquid merely acts as an agent that holds together the drop, and the impact dynamics are mostly governed by particle-based parameters.

B. Splashing threshold of dense granular suspensions

Peters et al. [53] studied the splashing threshold of dense granular drops. ‘Splashing’ in this case comprised of individual particles ejecting out of the edge of the drop. They observed that this splashing threshold is better characterized in terms of the particle-based Weber number $We_p = \frac{\rho_p r_p U^2}{\sigma}$. This suggests that splashing occurs when an individual particle can overcome the surface energy of the surrounding liquid and escape. They observed the onset of splashing for $We_p \geq 14$, independent of the substrate wettability and roughness.

This We_p dependence predicts that larger and denser particles splash at lower drop velocities because of their high inertia. This has been verified in consequent studies [52, 54]. But in the case of bimodal suspensions comprising of two particle sizes, Peters et al [53] found that smaller particles were ejected more easily than larger particles. Although seemingly contradictory to the particle-based Weber number predictions, this was explained by considering the distribution of velocities in particle-particle collisions. Due to momentum conservation, smaller particles end up with higher velocities than the larger ones, enabling them to be ejected earlier. Therefore, the bigger picture that emerges is that dense granular drop impact is mostly governed by the collisions of particles with the surface and each other,

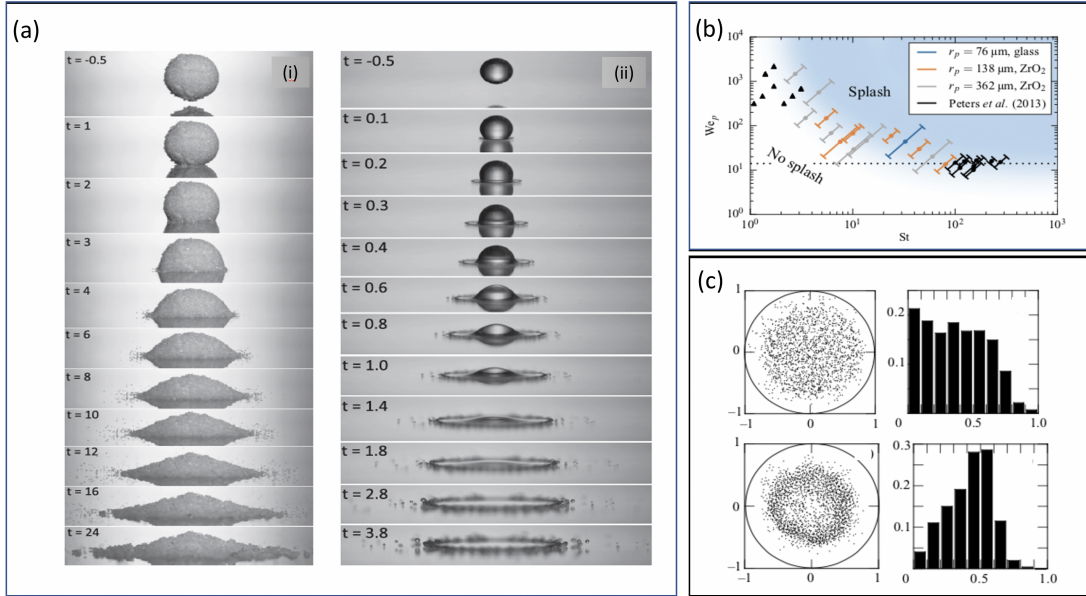


FIG. 3. (a) Comparison of impact of a 26 mm granular drop (left) and a 3.6 mm water drop (right) onto a hydrophobic glass plate. Adapted from [3]. (b) Granular drop impact can be divided into splashing and non-splashing regimes with respect to the particle-based Weber number and the Stokes number of the suspending liquid. Adapted from [52]. (c) Radial distribution of particles in the spread droplets is uniform for low Reynolds number of surrounding fluid, and annular for high Re . Adapted from [48].

the liquid primarily providing the surface energy to bind the drop together.

Consequently, Schaarsberg et al. [52] experimentally investigated the effect of liquid viscosity on dense granular splashing. They found that the individual particle splashing threshold increases with the viscosity of the suspending fluid. They showed that the phase space defined by We_p and Stokes number of the particles in the suspending fluid, St , is cleanly divided into splashing and non-splashing regimes [Fig. 3(b)]. Thus, it seems that the interaction of particle inertia and viscous interactions between particle and suspending liquid both have roles to play in the splashing threshold, as opposed to the bulk rheological properties of the suspension that govern the spreading dynamics.

Marston et al. [3] tracked individual particles ejected after splashing and found that the maximum particle velocity is typically twice that of the drop velocity. Image analysis in the same study also suggested that when a suspension drop touches a surface, it acts as a compressible fluid and particles rearrange. They observe that particles were ejected when

the volume fraction was close to jamming; they primarily use the observed area of the drop for this analysis, assuming an axisymmetric drop. For a more comprehensive understanding of this correlation, the elastic energy of the jammed network needs to be taken into account, since near the jamming concentration long-range correlations among the particle structure must be significant.

Most existing works have used spherical particles in impact studies. It would be interesting to explore the effect of anisotropy in particle shape on the spreading and splashing dynamics. Particle shape not only affects the critical volume fraction ϕ_c [55, 56], but the realignment of particles due to shear can also affect bulk flow properties of suspension. Additionally in the dense suspension limit where the bulk viscosity framework is inadequate, using particles that are index-matched with the liquid phase and introducing a few tracer particles could be a way to visualize the rearrangements within the drop during impact.

IV. POLYMERIC FLUIDS

Polymer additives are used in a variety of industrial applications, both to achieve desired fluid properties and to control the interaction of fluids with solid surfaces. The two most convenient parameters to experimentally study polymeric fluids are polymer concentration and the molecular weight (and in turn, chain length) of the polymer. But changing either of these quantities can affect multiple properties of the fluid: the yield-stress, the shear-thinning coefficient, and the zero-shear viscosity to name a few. Thus, numerical studies are key to gain an understanding of how each fluid property separately affects the behavior of impacting polymer drops. Hence, we will first outline the experimental results in polymeric drop impact, followed by the current numerical studies providing more insight into the physics of the process.

A. Experimental results

In coating, spraying, and agricultural pesticide industries, suppressing bounce and splash is important for the efficient use of chemicals. Bergeron et al. [57] observed that a small amount of polymer additives suppressed bounce of liquid droplets on hydrophobic surfaces. This result was observed to be independent of the type of hydrophobic surface, and this

suppress in bounce due to polymer additives has since been confirmed by many other studies [58–62]. It has also been shown that the rebound suppression was more pronounced when higher molecular weight polymers were used [62]. Rheological data has suggested that the fluid shear viscosity was not affected by the presence of these additives. The bounce suppression, therefore, was attributed to the rise in elongational viscosity [57, 61].

Some later studies disagree that the elongational viscosity causes bounce suppression, since both the spreading and receding should be affected by higher elongational viscosity [58]. The experimental data, on the other hand, suggest that the spreading phase is unaffected by the presence of small concentration of polymer additives, but the retraction is slowed down. Further complicating matters, for more concentrated polymeric fluids, both the spreading and the receding phases were suppressed [63]. Multiple other explanations besides an increased elongation viscosity have been suggested to explain the bounce suppression. Smith and Bertola [59] performed Particle Image Velocimetry with fluorescent particles, and observed that the contact line velocity is lower in polymer drops. As a result, they suggested that dissipation at the contact line suppresses bounce. Normal stresses have also been suggested to play a part in bounce suppression [58].

Luu and Forterre [64, 65] observed that carbopol (a yield-stress fluid) drops spread to a much larger extent on rough substrates; the spread was found to be even larger on hydrophobic surfaces. Their data collapsed well when represented in terms of the elastic Mach number ($M = \frac{U}{\sqrt{G/\rho}}$, where U is the fluid velocity and $\sqrt{G/\rho}$ is the elastic velocity of the fluid), indicating that the fluid elasticity plays an important role in this phenomenon [Fig. 4(a)]. They also suggested that for rough surfaces and high Weber numbers, the impact timescale was too short for fluid to seep through the surface troughs, effectively reducing the contact area and in turn, frictional dissipation. Guemas et al. [5], on the other hand, did not observe this super-spreading on surfaces of similar roughness. The only different parameter between these studies was drop diameter. Hence to understand this phenomena better, more work as a function of drop size and roughness is needed. Microscopic imaging and experiments with regular 3-d printed surfaces might also shed more light on the dynamics of this process.

As with Newtonian fluids, the interaction of the impacting drop with the surface can be minimized and hence decoupled from fluid processes via experiments on small targets. Rozhkov et al. [66] observed that the normalized drop diameter vs. time behave similarly

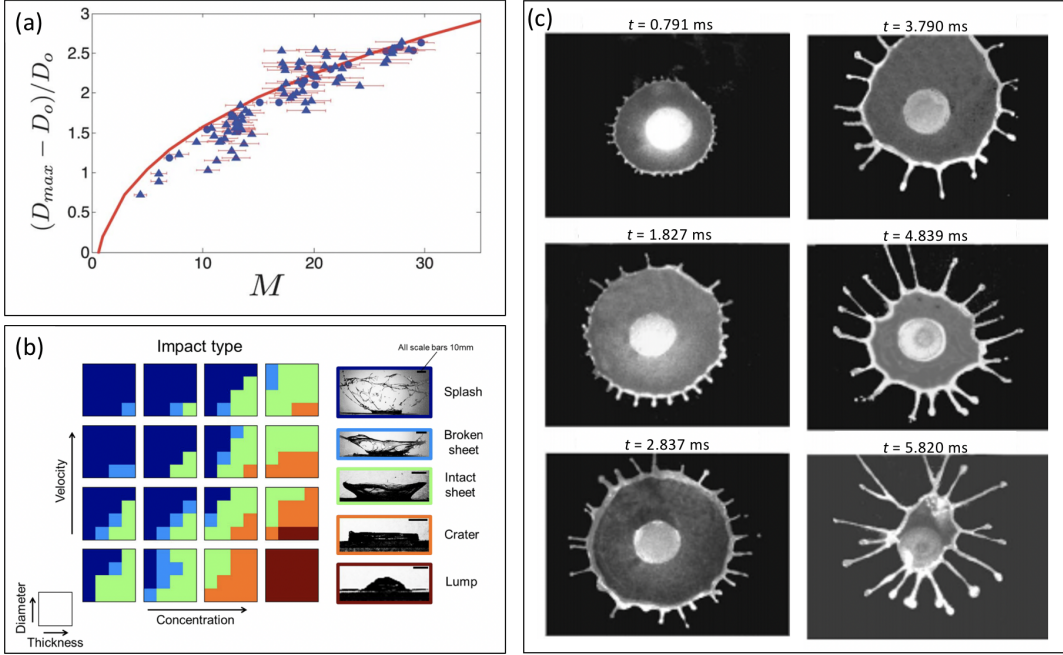


FIG. 4. (a) Spreading factor of polymeric drops on partially wetting surfaces depends on the elastic Mach number, as predicted by Luu and Forterre [64]. Adapted from [5]. (b) Phase diagram of the impact behaviors of polymeric drops impacting on a wet surface, as a function of four parameters: impact velocity, polymer concentration, drop diameter, and thickness of the liquid layer on the substrate. Adapted from [4]. (c) Timelapse of a polymeric drop after impact on a small target. Adapted from [66].

for water and polymer solutions on small targets. Slower retraction of polymer drops in other experiments, therefore, must be due to the fluid-surface interaction. Additionally, the rupture of ejected films and length of protruding fingers was suppressed by polymer additives [Fig. 4(c)]. This is consistent with the stabilizing effect of polymers on splash and bounce.

Even though impacting drops on dry surfaces is the simplest case to be experimentally studied, drops often impact on an already wet surface in industrial processes. Blackwell et al. [4] studied such a system of carbopol in water, where the surface was already coated with the same liquid. In addition to splashing being suppressed for higher concentrations of carbopol, they observed that it was also suppressed in presence of thicker liquid layers on the surface. They identified the ratio of coating thickness and drop diameter as a key dimensionless factor. Another relevant parameter was the ratio of inertial and dissipative stresses, including yield stress and viscosity. In the space of these dimensionless parameters,

the data separated itself into sticking and splashing regimes [Fig. 4(b)]. In a further study, Sen et al. [67] studied the effect of thixotropic aging on carbopol and Laponite suspensions, finding that the impact of these fluids could still be characterized using the same dimensionless groups. Even though these dimensionless parameters were useful for interpreting the experimental data in this case, their range is somewhat limited, as they would have to be modified when surface tension and elasticity effects are not negligible. More experimental data spanning a larger range of coating thicknesses is also needed to obtain a complete picture of polymeric drop impact on wet surfaces.

Many mechanisms have been suggested for bounce suppression in low-concentration polymeric drops, but a strong consensus is yet to emerge. In order to zero in on a better understanding, more experiments that incorporate direct measurements of shear, elongational, and normal stresses within the impacting drop might be useful. Additionally, the super-spreading phenomenon observed by Luu and Forterre could have fascinating applications in coating and should be further explored; a large body of systematic experiments on substrates of varying roughness and with drops of different sizes is needed in order to understand the parameters governing this super-spreading.

1. Numerical studies

In experiments involving polymeric drop impact, the natural choice for the independent parameter is the concentration of polymer or particulate additives. The fluid's yield stress increases with increasing polymer concentration, but so does the effective viscosity and rate of shear thinning. This makes it challenging to decouple the effects of yield stress and shear rheology. It is also difficult to experimentally form spherical drops of fluids with high polymer concentration. To eliminate the concerns about initial geometry and understand the effects on yield stress and rheology separately, numerical studies of yield-stress drop impact are crucial.

Kim and Baek [68] performed a numerical study of impacting yield-stress drops using the Herschel-Bulkley rheological model. They varied the yield-stress, viscosity, shear-thinning, and surface tension parameters separately to study the effects on the spreading and retraction phases. They observed that the spreading is mostly dominated by inertia and effective viscosity, but is not affected by the yield-stress or surface tension. Retraction, on the other

hand, was inhibited by both yield stress and capillarity. Past experimental works indicating that higher polymer concentration affects both spreading and retraction are consistent with this data. Additionally, the effect of yield stress and rheology are better understood in a decoupled way through this numerical investigation. Even though results of this study qualitatively agree with experimental data, failure to get a quantitative agreement is suspected to be due to assumptions about contact angles, which, just as with Newtonian fluids, vary during the spreading and retracting phases. The authors also observed locally different viscosity values in different parts of the drop after impact, which might be a direct result of local variation in fluid velocity, and in turn, shear stresses.

Recently, Oishi et al. [69] investigated both normal and oblique impacts numerically, also taking into account thixotropic effects. Thixotropy, the change in material properties over longer timescales, was modeled by introducing a delay between applied stresses and the resulting structural changes in the fluid; their numerical data qualitatively matches the experimental results of Luu and Forterre [64]. Oishi et al. treat the thixotropic timescale as a fitting parameter for their numerical data for carbopol and kaolin droplets, and their study considered a phase space of the elastic Ohnesorge number Oh_e , and a parameter Y consisting of the elastic Mach number, the plastic number, and the thixotropic timescale. This phase space was clearly divided between the sticking, bouncing, and rolling (in case of inclined planes) behaviours of the impacted drop.

Existing numerical studies additionally shed light on the dynamics of drop impact on a microscopic scale, such as varying structure factors and viscosities across the spreading/retracting drop. More numerical studies, especially on wet or thinly coated surfaces, would be helpful to build an understanding of how macroscopic polymeric drop impact behaviors (sticking, bouncing, etc.) are coupled to the microscopic interactions between the drop and the substrate. Numerical works have so far been effective in decoupling the physical parameters affecting polymeric drop impact. To understand their microscopic origins, fitting parameters such as the thixotropic timescale during impact need more theoretical attention. The suggested mechanisms behind experimentally observed bounce suppression could also be tested more conveniently in future numerical work. Additionally, the connection between non-uniform fluid velocities, and how this is linked to spreading and receding behaviors is best explored in a numerical context.

V. SHEAR-THINNING AND THICKENING FLUIDS

Fluids that show a decrease in bulk viscosity as the shear rate is increased are known as shear-thinning fluids. Some of these fluids also show an increase in viscosity at very high shear rate. In that range, they are said to undergo ‘shear thickening’. Thus, the same fluid can exhibit both shear-thinning and thickening behavior depending on the applied shear rates (see Section II for more details). Many fluids in this category are colloidal suspensions: suspensions of micron-sized or smaller particles in Newtonian fluids such as water. As opposed to granular particles, colloidal particles are small enough to be held together by the surface tension of the surrounding fluid, enabling us to measure the suspension’s bulk rheological properties. At high concentrations and shear rates, the particle-particle interactions also become significant. The particle size also introduces a new lengthscale, making the impact problem even more complex.

A. Shear-thinning suspensions

Shear thinning liquids have a relatively high viscosity at low shear rates, that decreases with increasing shear rates and plateaus at the infinite-shear viscosity value. Because of the variation of local fluid velocity in the drop impact process, the local shear viscosity in the drop varies temporally as well as spatially. Despite shear thinning fluids being very common, there have been surprisingly few studies of drop impact of shear-thinning fluids.

Surprisingly, according to the few existing studies in this area, shear-thinning drops show a qualitatively similar behavior to Newtonian fluids [5, 70]. Shear thinning fluids typically have a high consistency coefficient (which quantifies the average viscosity) in addition to the viscosity variation with shear rates. As compared to the expanding phase, the consistency coefficient is believed to have a larger effect in arresting drop spread. An and Lee [71] compared the quantitative behavior of impacting shear-thinning and Newtonian drops. They assumed that the average viscosity in the drop oscillates between the fluid’s zero-shear viscosity and infinite shear viscosity during spreading, receding, and oscillating phases [Fig. 5(a)]. To understand this assumption, let us consider the spreading and receding phases separately. During the spreading phase, the drop’s surface energy gets converted to kinetic energy and the fluid velocity increases. At the end of the spreading phase, the

fluid velocity must become zero before switching direction, i.e. entering the receding phase. Similarly, the velocity reaches a maximum and goes down to zero while moving from the receding phase to the next oscillation (if any) of the fluid film. Since shear rate can be assumed to be proportional to the fluid velocity, it must also oscillate over the impact process. An and Lee [71] observed that qualitatively the impact looks similar to that of Newtonian fluids, with a fluid viscosity defined by an average of the infinite shear viscosity and the zero-shear viscosity. Thus, by assuming an average shear viscosity for the shear-thinning liquid, one can capture its spreading behaviour with existing Newtonian models.

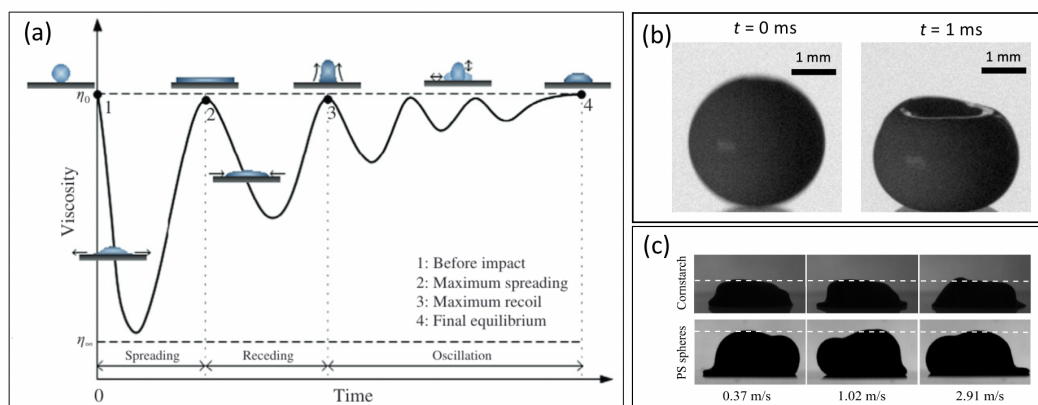


FIG. 5. (a) Schematic of the hypothesized variation in viscosity during the impact of shear-thinning drops. Adapted from [71]. (b) A drop of dense colloidal suspension (49% by volume) before and after impact on a glass substrate. After impact, a dimple forms at the top, indicating that the drop is jammed at the bottom but still liquid-like at the top. Adapted from [72]. (c) Snapshots of cornstarch and polystyrene suspension drops after impacting on a solid surface. The drop height h was observed to be independent of impact velocity. Adapted from [49].

De Goede et al. [73] studied the impact of blood (well-known to be a shear thinning fluid) on different solid surfaces, and observed that substrate properties have little effect on the splashing threshold of blood, just like Newtonian fluids. They also observed no qualitative differences between the spreading dynamics of blood and Newtonian fluids, which is consistent with previous works on shear thinning fluids.

In future works on shear-thinning drop impact, extensive use of techniques to measure local stresses in an impacting drop is key. It would also be interesting to explore drop impact in the range of shear rates where some fluids transition from shear thinning to

shear thickening, since in this region, one might expect to observe packets of localized shear thickening and jamming. More numerical and theoretical work that incorporate the spatial variation of shear stresses during phases of drop impact could provide a necessary phenomenological basis for more experiments.

B. Shear-thickening Fluids

Shear thickening fluids typically show an increase in viscosity and then jamming at very high shear rates. Studying the drop impact of such fluids is especially interesting, since we can access regimes of large shear rates by increasing the impact velocity. Despite being a field with great potential to expand our knowledge of complex rheology, drop impact studies of shear thickening fluids have been few and far between.

Since concentrated suspensions have a high effective viscosity, the drop's surface energy is lost quickly through viscous dissipation; thus most studies ignore the effects of surface tension. Therefore at high impact speeds, inertia and fluid viscosity are the dominating parameters. On impact, such dense suspension drops have been observed to undergo jamming [49, 72]. A recent work by Arora and Driscoll [72] observed shear-thickening drops to undergo partial jamming, where the bottom of the drop jammed but the top portion deformed like a fluid [Fig. 5(b)]. At even higher impact velocities, the drop did not deform at all, and even bounced back. This points towards a potential mechanism for the jamming dynamics: jamming happens soon after the bottom of the drop impacts the surface, and then a jamming front propagates upward through the drop. Future research focused on local stress measurements is needed to test this hypothesis. Additionally, Boyer et al. observed that shear-thickening drops showed a maximum deformation that was independent of the impact velocity, and the drops stayed frozen at the maximum deformation for much longer than the characteristic impact timescale [Fig. 5(c)].

One study has reported that drops impacted over hydrophobic PTFE surfaces stayed jammed for days [74]. This suggests that the interaction between the drop and the surface plays a key role in the long-timescale drop dynamics. Similar experimental work on hydrophilic surfaces exploring the relaxation time of jammed drop over long timescales would be of great interest.

To understand the physics of localized shear jamming, systematic measurements of shear

stresses in the impacted drop are crucial. Overall shear stresses in impacting drops can be estimated and measured, but localized information is much more difficult to obtain experimentally. However, this information is key to understanding observed partial jamming processes, and development of new experimental techniques to obtain this information should be prioritized.

Additionally, it has been shown in bulk rheological measurements that the critical volume fraction for jamming varies with particle shape, in particular particle aspect ratio [55, 56]. Thus, it would be interesting to conduct impact experiments for fluid suspensions with varying aspect ratio particles to explore the role of shape asymmetry in drop impact. Furthermore, this would aid in connection to industrial processes, where the component particles in slurries and suspensions are often far from spherical.

Drops jammed at impact have been observed to relax and spread like a fluid over much longer timescales. Investigation of the scaling laws of these spreading dynamics and comparison with other complex fluids is needed. Though quite challenging in the dense suspension regime, numerical simulations are crucial to link current macroscopic observations to microstructural rearrangements.

VI. CONCLUSION

In this review, we have summarized current work on the drop impact of complex fluids on solid substrates. Drop impact studies at high velocities enable us to study fluid properties at very high stresses, often beyond the range of typical rheometers, and in the absence of boundary conditions. This may lead to a significant expansion in our understanding of complex fluid properties.

Being a relatively new field, the studies so far have followed very disparate avenues. Nevertheless, we have attempted to classify the findings according to different flow behaviors. Dense granular suspensions have been observed to splash in a fundamentally different manner than Newtonian fluids, and their splash is controlled by the particle-based Weber number and the Stokes number with the surrounding fluid. Polymer additives seem to suppress bounce of impacting drops, the effect being more pronounced for both higher molecular weight and higher concentration of polymers. The physical origin of bounce suppression is an active field of investigation. The impact of colloidal suspensions that show shear-thinning

and shear-thickening behavior can be studied in terms of bulk effective viscosity of the fluid, as long as the concentration is not too close to the critical volume fraction (ϕ_c) for jamming. Shear thickening fluids have also exhibited fascinating behaviour such as dilatancy of the particle phase (in granular suspensions) and partially-jammed states after impact with high velocities (in colloidal suspensions).

The experimental studies so far have shown a wide range of interesting behaviors of impacting complex fluid drops, and scaling laws have been fairly successful in predicting fluid behaviors in certain regimes. However, a unified description correlating the available rheological data with impact experiments is still far out of our reach. More numerical and theoretical studies are necessary to understand the role of each fluid property in a decoupled way. Additionally, more studies with local measurements of fluid velocities and stresses after impact need to be performed in order to understand the microscopic origins of the rich array of observed fluid behaviors. Here, we outline the key theoretical challenges and suggest a number of avenues for future experimental studies that will take us closer to a more holistic description of complex fluid drop impact.

A. Theoretical and computational challenges

Capturing processes which occur on a large range of scales of length and time is the biggest challenge in modeling Newtonian drop impact dynamics. This problem is only augmented further in case of complex fluids, since particle size and the interaction timescales of the solid and liquid phases compete with the already existing scales such as drop size and surface roughness. Modeling the effect of such a large space of parameters is no small task. Many dimensionless parameters have been identified to understand drop impact dynamics better, but thus far, they are only effective to specific conditions and a generalized picture still eludes us. Given ever-improving computational capabilities, as well as the renewed trend in parallel computing, more detailed simulation of drop impact processes, guided by existing models should be possible.

Another effect that is difficult to capture in simulations is that of evaporation. Evaporation changes the overall, and local, particle volume fraction of the suspension. It can also set off additional flows on the surface and in the bulk of the drop. These flows and local heterogeneity in volume fraction might have nontrivial effects on the impact dynamics of

dense suspensions, which are generally disregarded in theoretical and computational models. Decoupling these effects from the impact dynamics, both before and after impact, is key to the success of theoretical predictions.

Our understanding the behavior of impacting complex fluid drops is inevitably built on the current understanding of the rheology of these fluids. Although a large amount of rheological data is available and new data is being added over a wide range of timescales and shear rates, a relatively small body of work has been done on the constitutive models that explain rheological behavior. Thus, the empirical nature of much of our current rheology knowledge is another significant roadblock in developing theoretical frameworks for complex fluid drop impact.

B. Open questions and/or perspectives

While there is some data available about the effect of substrate properties on the drop impact of complex fluids, more systematic studies are necessary to form a full picture. In particular, the effect of surface roughness, both patterned and random, deserves more attention since it is a pragmatic way of controlling droplet splashing and deposition. The effect on the advancing and receding contact angles of droplet properties such as particle shape, size, and effective surface tension also deserves further attention, as it will also guide computational studies in the right direction. The effect of competing lengthscales of surface roughness and particle sizes on effective contact angles is a further level of complexity which needs to be examined.

Though much of the work in this field is dominated by the study of drop impact on a dry surface, impact on wet surfaces is becoming the focus of more and more studies. This is an especially important avenue of research, since impact on a surface which is already wet with a coating of the complex fluid is a more realistic scenario in industrial spraying and coating processes. A larger range of the liquid layer thicknesses should be explored as a parameter, since the physics of complex drop impact could vary substantially with the ratio of liquid layer thickness and drop size.

Shear thickening fluids have recently showed some fascinating behavior, such as localized or complete jamming, when impacted on a surface. In almost all studies on colloidal and granular drops, spherical particles have been used, despite the fact that anisotropic particle

suspensions are much more common in industrial processes. The effect of particle shape on impact dynamics as well as the jamming threshold would contribute to our understanding of jamming in complex fluids. Furthermore, the use of anisotropic particles adds another parameter to the microscopic dynamics inside the drop, namely the particle orientation. Rheological studies of non-spherical particles have indicated that they jam at a much lower volume fraction [55, 56], and statistical studies of the rearrangement of rods inside the impacting drop could enhance our knowledge about the interaction between the solid and liquid phases.

While the variety of competing lengthscales and timescales enriches the drop impact problem, it also makes it extremely challenging. The availability of a number of microscopy techniques, and advances in high-speed imaging and data storage capacity make this an exciting time to attack this problem. There have already been some soft matter studies that combine these tools; for example refractive-index matching, fluorescent or dyed particle tagging, and particle-image velocimetry further enrich our ability to extract more localized information from drop impact studies. The time is ripe for widespread use of these techniques to get more spatio-temporally resolved data, that in turn will provide direction to further theoretical and computational studies.

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