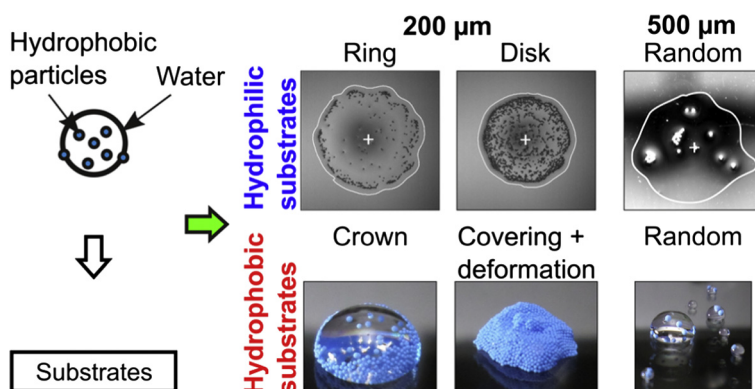


Regular Article

Impact of particle-laden drops: Particle distribution on the substrate

Viktor Grishaev^{a,b}, Carlo Saverio Iorio^b, Frank Dubois^b, A. Amirfazli^{c,*}^a Center for Design, Manufacturing and Materials, Skolkovo Institute of Science and Technology, Skolkovo Innovation Center, Building 3, Moscow 143026, Russia^b Service de Chimie-Physique EP, CP165-62, Université Libre de Bruxelles, 50 Av. F.D. Roosevelt 1050, Brussels, Belgium^c Department of Mechanical Engineering, York University, Toronto, ON M3J 1P3, Canada

GRAPHICAL ABSTRACT



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ABSTRACT

The splat morphology after the impact of suspension drops on hydrophilic (glass) and hydrophobic (polycarbonate) substrates was investigated. The suspensions were mixtures of water and spherical hydrophobic particles with diameter of 200 μm or 500 μm. The impact was studied by side, bottom and angled view images. At Reynolds and Weber numbers in the range $150 \leq We \leq 750$ and $7100 \leq Re \leq 16,400$, the particles distributed in a monolayer on the hydrophilic substrates. On hydrophobic substrates, many particles were at the air-water interface and 200 μm formed a crown-like structure. The current study for impact of particle-laden drops shows that the morphology of splats depends on the substrate wettability, the particle size and impact velocity. We developed correlations for the inner and outer diameter of the particle distribution on the hydrophilic substrates, and for the crown height on hydrophobic substrates. The proposed correlations capture the character of the particle distributions after drop impact that depends on particle volume fraction, the wettability of both particles and the substrate, and the dimensionless numbers such as Reynolds and Weber.

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1. Introduction

Many technologies are associated with the impact of particle-laden drops such as additive manufacturing [1–3], printed

electronics [4,5], plasma coating technology [6] and spraying of liquid friction modifiers [7,8]. For these technologies, it is crucial to understand the role that particles and substrates play on drop impact phenomena as well as the fate of particles after impact.

It is well known that for pure liquids, the impact morphology depends on Weber $We = \rho D_0 U^2 / \sigma$ and Reynolds $Re = \rho D_0 U / \mu$ numbers. These non-dimensional numbers include the main

* Corresponding author.

E-mail addresses: v.grishaev@skoltech.ru (V. Grishaev), ciorio@ulb.ac.be (C.S. Iorio), frdubois@ulb.ac.be (F. Dubois), alid2@yorku.ca (A. Amirfazli).

parameters of the process: the drop diameter, D_0 ; drop impact velocity, U ; drop density, ρ ; drop viscosity, μ , and surface tension of liquid–air interface, σ . There are also many other parameters such as the substrate roughness, ambient pressure, etc. to affect drop impact (details can be found in Refs. [9] and [10]).

For particle-laden drops, significant changes occur depending on the particles' size, volume fraction, ϕ , and the substrate and particle wettability. Particle-laden drops can be in the form of liquid marbles and suspensions. Liquid marbles are liquid drops covered fully by particles in contrast to suspension drops where particles are locating inside of drops as well as drop surfaces. In this paper, we will study suspension drops, and henceforth will focus the discussion on such systems.

Addition of particles can suppress the appearance of singular jet when drop retracts after impact on hydrophobic substrates [11] as well as partial drop rebound [11,12]; drop break-up during rebound from superhydrophobic substrates can also be suppressed in particle-laden drops [13] so as rebound [14]. Also, it was reported that particles can lead to drop splashing on hydrophilic [11,15,16] and hydrophobic [11] substrates. The splashing of suspensions can happen far away from the drop contact line, a phenomenon remarkably different from prompt or corona splash for pure drops [11]. Furthermore, the addition of particles can lead to drop fragmentation as a result of receding break-up or rupture of the drop's lamella [11].

Particles not only lead to the suppression or the appearance of new phenomena in drop impact, but can also change the drop spreading. It has been shown that the addition of particles can reduce the maximum drop spreading factor, D_{max}/D_0 , where D_{max} is the maximum diameter of the drop contact area during spreading [11,12,14,15,17]. Nicolas [15] proposed that it could be explained by effective viscosity but it can be questionable for systems where distinct particles exist (see below).

The effective viscosity was calculated by the Krieger-Dougherty model, and was used in the estimation of maximum spreading factor [15]. The maximum spreading factor was found assuming that kinetic energy is dissipated mostly by viscous forces at $Re \gg 1$ and $We \gg 1$. For particle volume fractions much smaller than random close packing (which was taken as 0.68), the formula for maximum spreading factor was:

$$\frac{D_{max}}{D_0} \approx \left(\frac{Re}{12}\right)^{1/4} [1 - 0.69 \cdot \phi] \quad (1)$$

The used value for the random close packing ($\phi = 0.68$) is questionable (see for example, Jaeger and Nagel [18]). Nevertheless, this may not be important, because Eq. (1) was obtained using the particle volume fraction much smaller than random close packing.

Nicolas [15] found that this formula works for hydrophilic glass substrates in the range of Reynolds and Weber numbers: $79 < Re < 6000$ and $10 < We < 370$, respectively. However, Eq. (1) was not applicable to high Reynolds and Weber numbers $6000 < Re < 10000$ and $370 < We < 1276$. At these conditions, the maximum spreading factor increased with an increase of particle volume fraction. This observation is contradictory to the effect of an increase in viscosity that should result in reduced spreading; it was explained, however, by the non-circular shape of the splats caused due to a distorted contact line, or the drop break-up. Nevertheless, it seems that the effective viscosity is not quite able to explain the changes in the maximum spreading factor.

Concerning splat morphology and particles' arrangement in the splat, limited studies have been done. The limited studies that exist have examined the splat morphology for dense (particle volume fraction $\phi \geq 0.5$) and dilute suspensions ($\phi < 0.5$), separately.

The splat morphology for dense suspension drops on hydrophilic glass substrates was studied in the work of Lubbers et al. [19].

The suspensions were mixtures of water or silicone oils seeded with hydrophilic particles of zirconium dioxide (average diameter: 250 μm). It was found that the suspension drops rapidly expanded to a monolayer at Weber numbers equal or greater than 1862 while the particles grouped into clusters separated by particle-free regions, i.e. a mesostructure was formed. The development of the mesostructure was quantitatively explained by using models deduced from the balance of forces acting on the individual particles. The forces acting on moving particles were viscous drag and surface tension of deformed air–liquid interface near particles. No analytical expression (or correlations) for the maximum splat diameter or splat shape, and the particle distribution after the impact, was provided.

In the case of dilute suspensions, the splat morphology and the particle distribution have been considered in [15], where a ring distribution was seen for particles with diameter, d_p , of 380 μm , and 720 μm at Reynolds numbers $3184 < Re < 3513$, and $Re \approx 5000$, respectively; on the contrary, for $Re < 807$ and particles with $d_p = 640 \mu\text{m}$, a disk-like distribution of particles was observed. The occurrence of the ring distribution was explained in [15] by the liquid oscillations (explained as the liquid movement towards the centre during recoil), which became larger as Reynolds number increased. When the disk or ring distribution was observed, the particles were absent near the drop contact line at a dimensionless distance from the centre of the drop impact (a distance divided by the equivalent radius of a drop contact area with a substrate) larger than 0.7. The absence of particles was explained by thin liquid film in this region, which prevented the particles moving closer to the contact line. There was no analytical relationship provided for the particle distributions.

To summarize, although a number of valuable works on splats of suspensions with hydrophilic particles on hydrophilic substrates exist, no correlations for the particle distributions or analytical expressions exist. Therefore, the following questions remain: how particle distribution may be affected by substrate wettability, particle size, particle volume fraction, and drop impact velocities. Another question is that whether or not one can find an analytical relationship for the particles' distribution in the splat.

This experimental study aims to investigate mainly dilute suspensions that are less studied. Furthermore, it focuses on the hydrophobic particles that are scarcely studied to date. The idea is to gain a first impression of overall behavior of particles and their distribution in the splats upon impact of a particle-laden drop onto surfaces of different wettabilities.

2. Material and methods

Main parameters of the experiments are presented in Table 1. For drop generation, water (deionized reagent grade III, Acros Organics) and a dispersion of polyethylene particles in water were used. The density and dynamic viscosity of water at room temperature were considered to be equal to 1 g/cm^3 and 0.890 mPa s , respectively. Surface tension of water was measured by the pendant drop method (drop shape analyser DSA30S, KRÜSS) and found as 72.8 mN/m at room temperature. Blue polyethylene spherical particles with diameter 180–210 μm (BLPMS-1.00 180–212 μm , Cospheric) and 425–500 μm (BLPMS-1.00 425–500 μm , Cospheric) were used in preparing the complex drops.

The single particle wettability was characterized by measuring the contact angle of particles as they floated at the air–water interface. The procedure to conduct the measurements was to form a puddle of DI water on a polycarbonate plate with an approximate size of 70 \times 70 \times 71 ($W \times L \times H$) mm. A camera and the diffused light source of the drop shape analyzer (KRÜSS DSA30S) allowed for implementing a shadowgraphy technique. The particles were

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