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Emergence of two lamellas during impact of compound droplets

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A compound droplet consisting of water and silicone oil either spreads or splashes upon impacting a solid surface. We show that the transition from spreading to splashing of this type of compound droplet can be changed by varying the volumetric oil ratio in the droplets. In the spreading regime, we observe a surprising emergence of two lamellas spreading one after the other: the first spreading lamella consists of only oil and the trailing one water. We show that the two lamellas eventually meet, significantly affecting the spreading dynamics. Finally, we quantify the maximum spreading radius and show that it is a function of both the impact velocity and the volumetric oil ratio.

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Interactions between solid surfaces and multi-component droplets, which consist of two or multiple immiscible layers of fluids, are of great importance in a multitude of engineering applications such as fuel atomization,¹ food industry,² and drug delivery.³ For instance, utilizing water-in-oil encapsulation for internal combustion has been considered an important approach to reduce air pollution and improve combustion efficiency. This type of compound droplet induces micro-explosions, which create secondary breakups at a lower boiling point of oil and subsequently lead to significant enhancement in fuel spray atomization and combustion.^{1,4,5} Other examples include using compound droplets for protection or controlled release of bioactive compounds in the food industry.^{2,6}

Despite the industrial relevance of compound droplets, most studies of droplet-surface interactions^{7–19} have been focusing on single-component droplets. Only a few works^{20–24} devoted to the impact of compound droplets on solid surfaces. The residence time of water-in-oil compound droplets impacting superheated surfaces was investigated experimentally.²¹ Numerical simulations were conducted for spreading of compound droplets on flat surfaces²² and fluid coated surfaces.²³ The impact of the compound droplet on the liquid surface during cell printing was also reported.²⁴ There is, however, a lack of systematic understanding of the behaviors of compound droplets upon impacting solid surfaces, in particular, when the volumetric ratio of the participating components is varied.

In this paper, we focus on the impact behaviors of water-in-oil compound droplets on solid surfaces. We characterize the main impact behaviors with varying impact velocities and volumetric ratios of oil and water. In particular, we observe the emergence of two spreading lamellas and provide experimental evidences for the fluid component in each of these lamellas. Finally, we provide quantitative measurements of the spreading radius of impacting compound droplets in the spreading regime.

To generate compound droplets of water encapsulated in silicone oil (5 cst, Sigma-Aldrich), we use two syringe

pumps and two flat-tip needles of different outer diameters to independently supply oil and water. First, we generate a sessile oil droplet of volume Ω_o at the tip of the smaller needle (outer diameter 0.3 mm) using a Hamilton syringe (maximum volume 50 μ l). We then supply water through the other needle (outer diameter 0.51 mm) next to the small one to create a water droplet, which is quickly coated by the previously dispensed oil volume. The generated compound droplet detaches from both needles on its own weight. Although the volume of water in each droplet is not precisely controlled, compound droplets generated using this method have roughly the same radius $R_0 = 1.3 \pm 0.05$ mm. By adjusting the initial oil volume Ω_o , we vary the volumetric oil ratio $\alpha = \Omega_o/\Omega$ from 0.02 to 0.29. Here, Ω is the droplet total volume.

We vary the impact height of droplets from 50 mm to 500 mm, resulting in a variation of the impact velocity V_0 from 3.1 m·s⁻¹ to 2.6 m·s⁻¹. The Weber number, defined herein as the ratio of the total kinetic energy of the droplet and the surface energy of oil $We = 2[\alpha\rho_o + (1 - \alpha)\rho_w]V_0^2R_0/\sigma_o$, is used to characterize the initial kinetic energy of each impacting droplet. Here, V_0 and R_0 are the impact velocity and initial radius of a compound droplet, respectively. The density and surface tension are denoted as ρ and σ , respectively, with the subscripts “o” indicating oil and “w” water. The resulting Weber number varies from 124 to 1272. For each impact experiment, we use a dry and clean glass slide as the targeted surface. Each glass slide is cleaned with acetone and isopropyl alcohol (IPA) in an ultrasonic cleaner for 20 min.

We record the impact phenomenon using a high-speed camera (SA-X2, Photron) from the side view with the frame rate up to 30 000 frames per second. A combination of a Nikon microlens and an extension tube gives a resolution of 15 μ m per pixel. A light source (LS-M350, Sumita) provides backlight illumination.

To investigate the impact behaviors of compound droplets, we construct a characteristic phase diagram with varying impact velocities V_0 and volumetric oil ratios α (Fig. 1). We observe that there is a clear transition between the spreading and the splashing behaviors. Qualitatively, at low

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FIG. 1. Phase diagram of impact behaviors of compound droplets on solid surfaces. Two major characteristic behaviors are identified: splashing (triangles) and spreading (squares). In the spreading regime, either one-lamella spreading (open squares) or two-lamella spreading (solid squares) can be observed. In the splashing regime, droplets may splash and rebound (downward triangles) or simply splash and completely disintegrate (upward triangles). The solid line marks the transition between the spreading and splashing regimes, and the dashed line separates the splashing and splash-rebound behaviors. Representative snapshots of these characteristic behaviors are shown on the right. The scale bar represents 2 mm.

Weber numbers ($We \leq 200$), spreading prevails for all compound droplets with all tested values of α , whereas at high Weber numbers ($We > 200$), transition from spreading to splashing occurs with the increasing volume ratio α . Thus, by increasing α , we prompt splashing to occur at lower Weber numbers. In the splashing regime, we observe two different characteristic behaviors: an impacting droplet either retracts and rebounds after splashing or breaks up and deposits on the substrate. Therefore, we further categorize typical behaviors in this regime as “splashing and rebounded” and “splashing.” These behaviors are separated by the dashed line in Fig. 1.

More interestingly, we find that there exist two lamellas emerging during the spreading phase of a compound droplet for certain ranges of α and We (Fig. 1). The emergence of these spreading lamellas induces a remarkable feature in the spreading radius R . In Fig. 2(a), we show the dependence of the spreading factor $\beta = R/R_0$ versus the normalized time $t = T/(R_0/V_0)$. Here, T is the time after impact; $T=0$ is defined as the moment that an impacting droplet appears to touch the surface, as shown in Fig. 2(a). The plot β versus t is accompanied by a series of representative snapshots illustrating the emergence of two lamellas. We observe in Fig. 2(a) a profound “shoulder” in the spreading curve β versus t . The shoulder separates two spreading waves: the first wave starts from $t=0$ and ends at $t_b = T_b/(R_0/V_0)$, at which the shoulder reaches its peak, and the other wave then takes over after a transient period, after which the droplet continues to spread to its maximum spreading factor $\beta_m = R_m/R_0$, where R_m is the maximum spreading radius [See Fig. 2(a)]. From the accompanying snapshots, two lamellas appear around the transient stage [see enlarged snapshots in Fig. 2(a) for details]: the first spreading wave is caused by the first lamella and the second one emerges when the second lamella approaches the first one. The “shoulder” in the spreading curve therefore occurs as a result of the second lamella overtaking the first one.

FIG. 2. (a) Representative plot of $\beta = R/R_0$ vs. $t = TV_0/R_0$ showing a profound “shoulder” at $t_b = 1.25$. In this experiment, $\alpha = 0.15$ and $V_0 = 1.2 \text{ m} \cdot \text{s}^{-1}$, which translate to $We = 176$. Representative snapshots taken at various values of t are inserted to illustrate the emergence of the two lamellas. The scale bar of the snapshot at the bottom-left corner represents 1 mm. The snapshots in dashed-line boxes are shown with higher magnification indicated by the accompanying ratios. (b) Dependence of T_b on α for impacts having two clearly distinguishable spreading waves (or equivalently clear “shoulder”). The inset illustrates how the spreading curve (β vs. t) varies when changing α from 0.1 to 0.19, while keeping V_0 fixed at $V_0 = 1.2 \text{ m} \cdot \text{s}^{-1}$.

To quantify the occurrence of the “shoulder,” we extract the time T_b , measured as the duration from $T=0$ to the peak of the “shoulder,” for all the cases in which spreading waves are distinguishable. In Fig. 2(b), we show the dependence of T_b on α for all various values of impact velocity. We observe that varying V_0 from $1.0 \text{ m} \cdot \text{s}^{-1}$ to $1.4 \text{ m} \cdot \text{s}^{-1}$ has an insignificant effect on T_b . However, there is a marked dependence of T_b on α , signifying the crucial role of the oil-water volume ratio in the impact dynamics.

As the existence of the two lamellas is connected to the presence of the “shoulder,” we now focus on the two lamellas to gain more insight into the origin of the “shoulder.” We speculate that the first lamella consists of only oil and the second one of water because lamella spreading starts from the droplet’s outermost layer,^{16,25} which in this case consists of oil. In Fig. 3, we show top-view snapshots of the impact of a compound droplet with an oil-water ratio of $\alpha = 0.08$ and an impacting velocity of $V_0 = 1.4 \text{ m} \cdot \text{s}^{-1}$. In this experiment, we add dye (Methyl Blue, Sigma-Aldrich) to water (inner phase). Illuminating light is supplied from below of the transparent impacted surface. The added dye absorbs light, making the water phase dark, while the transparent oil

FIG. 3. Series of top-view snapshots of a compound droplet with velocity $V_0 = 1.4 \text{ m} \cdot \text{s}^{-1}$ and volumetric oil ratio $\alpha = 0.08$ impacting a glass substrate. Light absorbing dye (Methyl Blue) is added to water, making it dark, whereas oil is transparent and appears brighter. At $T = 0.4 \text{ ms}$, the first lamella appears much brighter than the second one, suggesting that it only consists of oil and the second one water. At 0.7 ms , water in the second lamella reaches the first lamella, evidenced by alternative dark and bright areas at the outermost rim. The scale bar is 1 mm .

phase appears much brighter. As shown in Fig. 3 at 0.4 ms , the rim of the first lamella is marked by the outermost ring, which appears dark due to the high curvature of the liquid surface at the rim. An inner bright ring next to the dark one indicates that the first lamella consists of only oil. The second lamella, however, consists of water, evidenced by the dark shade as light is absorbed by dye in water. At 0.7 ms , the water phase reaches and penetrates the oil phase, causing alternative dark and bright areas at the outermost rim. Observation of such contrasted areas at the rim demonstrates that this visualization method is capable of differentiating the oil phase from the water phase. Thus, we conclude that the first lamella consists of oil and the second lamella consists of water.

A simplified schematic of the impact phenomenon is depicted in Fig. 4 to qualitatively explain the existence of two lamellas. We assume that the outermost layer of the droplet first touches and covers the surface with oil. The water body therefore impacts the preceding covering oil layer [Fig. 3(b)]. As a result, the oil layer impacts the surface with the initial impact velocity V_0 of the compound droplet, while the water body subsequently impacts the oil-covering surface. These impacts generate two spreading waves, the first one from oil and the other one from water. The “shoulder” is generated as the second spreading wave catching up with the first one.

To obtain a quantitative description of the spreading of compound droplets, we measure the maximum spreading

FIG. 4. Schematics (not to scale) of the impact of a compound droplet on a surface. (a) At time $T=0$, the oil layer touches the surface. (b) At time $T = \Delta T$, the oil already spreads out and generates the first spreading wave, while the water just touches the surface. The spreading of water generates the second spreading wave that eventually catches up with the first one, causing the occurrence of the “shoulder” [Fig. 2(a)].

factor β_m for all impacts of droplets in the spreading regime. Note that for impacts of single-fluid droplets, the dependence of β_m on impact conditions has been the focus of intense research over the past few decades.^{9,18} Various correlations and theories have been proposed based on either energy or momentum conservations and mostly for impacts in the capillary or viscous regimes. Most recently, it was shown using the energy conservation approach that β_m depends on both the Weber number and the Reynolds number although such a dependence can be reduced to $\beta_m \sim \text{Re}^{1/5}$ in the viscous regime or $\beta_m \sim \text{We}^{1/4}$ in the capillary regime.^{15,25} Nonetheless, for investigations of impacting droplets at the lower limit of viscosity, using We as the only dimensionless number to describe variation in β_m is a common and simplified approach.^{7,26}

In Fig. 5, we show that rescaling β_m by We^γ , where $\gamma = 0.28$, collapses our data for a wide range of α ($0.02 \leq \alpha \leq 0.29$) values and various values of impact velocity ($1.0 \text{ m} \cdot \text{s}^{-1} \leq V_0 \leq 2.6 \text{ m} \cdot \text{s}^{-1}$). The exponent $\gamma = 0.28$ is found using the least-squares fitting method. Thus, our spreading data for compound droplets can simply be expressed as $\beta_m \sim \text{We}^{0.28} f(\alpha)$, which for a fixed value of α resembles the scaling $\beta_m \sim \text{We}^{1/4}$ for impacts of droplets in the capillary regime. For a fixed value of We , the spreading factor β_m is minimized at $\alpha \approx 0.15$, suggesting a method to effectively control the spreading radius by varying the volumetric ratio of compound droplets. The relationship between the spreading dynamics and the composition of the droplets, however, is not in the scope of the present study and merits further detailed investigations.

In summary, we report the behaviors of compound droplets upon impacting solid surfaces. By varying the impact velocity and the volumetric oil ratio α , we show that the impact behaviors mainly belong to two regimes: splashing and spreading. We experimentally show that the transition between the spreading and splashing behaviors depends not only on the Weber number but also on α . In particular, in the spreading regime, we reveal the emergence of two spreading lamellas that significantly affect the overall spreading behavior. We experimentally show that the preceding lamella consists of only oil and the trailing one water, thus shedding light on the mechanistic emergence and interaction of the two lamellas. We quantitatively characterize the spreading

FIG. 5. Dependence of $\beta_m / \text{We}^{0.28}$ on α for different V_0 values. Inset: log-log plot of β_m vs. We for different values of α .

radius of impacting compound droplets in the spreading regime and show that varying the volumetric ratio of oil and water provides a control method of the maximum spreading radius. Finally, as the oil phase acts as a spontaneous buffer layer preventing contact between water and the impacted surface, the impacting behaviors of compound droplets may be relevant to those whereby separation between water and the surface is possible, *e.g.*, impacts of droplets on Leidenfrost or superhydrophobic surfaces.^{7,12}

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