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ABSTRACT

Impacting dynamics of compound droplets consisting of oil and water–glycerol mixture transitions from spreading to splashing with an increasing volumetric oil-mixture ratio. The transition to splashing is directly related to the dynamics of the ejecting lamella upon impact. Here, we present a detailed study of the ejecting lamella at the early stage of the impact. We experimentally characterize the ejection velocity and time of the ejecting lamella for a wide range of impacting velocities, volumetric oil ratios, and viscosities of the mixture. We show that ejecting dynamics depends significantly on the volumetric oil-mixture ratio, while it is slightly affected by the viscosity of the mixture. We also propose and verify experimentally a predictive model for the ejection velocity and time of the lamella.

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The impact of droplets on solid surfaces is of great importance in diverse engineering applications, ranging from atomization, spray painting, coating, and spray cooling, to 3D ink-jet printing.^{1–4} Interactions between impacting droplets, in particular, those of a single fluid and solid surfaces, therefore, have become an exciting research area since the pioneering study by Worthington.⁵ Various phenomena that occur during and after an impacting droplet touches a solid surface have been visualized and studied extensively, including bubble entrapment, jetting due to cavity collapse,⁶ and instability caused by the expanding lamella.^{7,8} Subsequently, these studies have paved the way for obtaining better understanding and control of numerous quantities of practical interest such as the volume of entrapped bubbles,^{9–11} the maximum spreading diameter,^{12–15} and splashing threshold.^{16–19}

Nonetheless, impact dynamics of compound droplets consisting of two or multiple immiscible fluids remains largely unexplored, despite its increasingly important role in emerging applications such as spraying of blended fuels²⁰ and tissue engineering using ink-jet printing.²¹ The reported experimental studies on impacting compound droplets have focused on several aspects including the heat transfer between impacting water-in-oil droplets and superheated surfaces,²⁰ the resident time of water-in-diesel droplets on Leidenfrost surfaces,²² maximum spreading factor,²³ and the emergence of two-lamella during spreading of impacting water-in-oil droplets.²⁴ Numerical studies in this area include the deformation and impacting

behaviors of compound droplets on either flat surfaces²⁵ or fluid coated surfaces.²⁶ Although qualitative impacting behaviors of compound droplets have been explored, the lamella's ejecting dynamics, which was shown to be crucial in understanding the aerodynamic force responsible for lifting and eventual splashing in the case of single-component droplets,¹⁷ remains open.

In this paper, we experimentally investigate and theoretically analyze the ejection of the lamella upon impact of compound droplets, which consist of oil and water–glycerol mixture, on solid surfaces. As the ejected lamella consists of a thin oil layer coated over the water–glycerol mixture, its dynamical behaviors deviate from those observed from the impact of single component droplets. We experimentally characterize and theoretically analyze the dynamical behaviors including the ejection velocity and time to ejection of the expanding lamella for various impacting conditions and liquid ratios.

In our experiment, each compound droplet consists of a water–glycerol mixture core covered by a thin silicone oil coating. The weight ratio of glycerol in the tested mixtures ranges from 0% to 60%. Correspondingly, the viscosity μ_m of the mixture varies from 1.0 mPa · s to 11 mPa · s. To generate a compound droplet, we first use a glass syringe (maximum volume 50 μ l, Hamilton) to dispense 5 cSt ($\mu_o = 4.6$ mPa · s) silicone oil (Sigma-Aldrich) from a small flat-tip needle (outer diameter = 0.3 mm) to create a droplet of volume Ω_o . With this type of syringe, our syringe pump can supply liquid with a minimum volume rate of 57 pl · min^{–1}. Thus, we

were able to generate volumes of inner and outer fluids with a reproducibility, i.e., relative difference in volume between different experiments, of $\pm 0.05\%$ and an accuracy, i.e., relative difference in volume between the set volume and the actual dispensed volume, of $\pm 0.5\%$. Subsequently, we supply the water–glycerol mixture through another needle (outer diameter = 0.51 mm). The dispensed water–glycerol mixture is quickly coated by silicone oil, resulting in a compound droplet. By continuously pumping the water–glycerol mixture into the droplet, the compound droplet detaches from both needles when the gravitational force overcomes the surface tension. The surface tension of silicone oil is $18 \text{ mN} \cdot \text{m}^{-1}$, while the interfacial tension between inner and outer fluids is $32 \text{ mN} \cdot \text{m}^{-1}$.²⁷ We note that compound droplets generated this way have a consistent radius $R_0 = 1.3 \pm 0.05 \text{ mm}$. By adjusting the oil volume Ω_o in the range $0.1 \mu\text{l} \leq \Omega_o \leq 0.8 \mu\text{l}$, we vary the volumetric oil ratio $\alpha = \Omega_o / \Omega$ from 0.01 to 0.09. Here, Ω is the total volume of a compound droplet. As a result, the thickness H_c of the silicone oil layer therefore ranges from $5 \mu\text{m}$ to $38 \mu\text{m}$.

We vary the impact velocity V from $1.5 \text{ m} \cdot \text{s}^{-1}$ to $2.6 \text{ m} \cdot \text{s}^{-1}$ by adjusting the releasing height of the needles between 140 and 340 mm. We define the Weber number for a compound droplet by normalizing

its impact kinetic energy by the outer surface energy, giving $We' = 2[\alpha\rho_o + (1 - \alpha)\rho_m]V^2R_0/\sigma_o$, where ρ denotes the density, σ represents the surface tension, and the subscripts “o” and “m” indicate properties of oil and water–glycerol mixture, respectively. The resulting Weber number varies from 127 to 995. For each experiment, we use a dry and clean glass slide as the target surface; each glass slide is cleaned with acetone and isopropyl alcohol (IPA) in an ultrasonic cleaner for 20 min prior to experiment. We record the impact phenomena from the side view by a high-speed camera (SA-X2, Photron) with the frame rate up to 300 000 frames per second. A combination of a microlens (105 mm, Nikon) and extension tubes give a pixel size of $9.3 \mu\text{m}$. A light source (LS-M350, Sumita) provides backlight illumination.

In Figs. 1(a) and 1(b), we show representative series of snapshots illustrating how the impacting behaviors of compound droplets change by increasing the oil ratios α from 0.02 to 0.07, keeping the same impact velocity at $V = 2.1 \text{ m} \cdot \text{s}^{-1}$. We observe that while the lamella spreads without upward ejection for impact with $\alpha = 0.02$ [Fig. 1(a)], the lamella is lifted into the air for the case of higher oil ratio, $\alpha = 0.07$ [Fig. 1(b), $T = 0.24 \text{ ms}$]. The lifted lamella eventually destabilizes into small droplets at its advancing front ($T = 0.88 \text{ ms}$). It should be noted that the lamella may contain either the outer fluid, i.e., silicone oil, or both fluids, i.e., oil and the water–glycerol mixture. By comparing Figs. 1(a) and 1(b), and if one attributes the aerodynamic force to ejection of the lamella,¹⁷ it implies from this observation that increasing the oil ratio increases the ejection velocity, and subsequently, the aerodynamic force that causes lamella destabilization. Indeed, if one considers the splash threshold for impacting droplets of pure water, the threshold velocity V_s is $4 \text{ m} \cdot \text{s}^{-1}$ ($R = 1.63 \text{ mm}$, $We = 732$).¹⁷ By adding a layer of oil over the water droplets (the volumetric oil ratio is as small as $\alpha = 0.19$), the splashing threshold was reduced to $V_s = 1.4 \text{ m} \cdot \text{s}^{-1}$ ($We = 250$).²⁴

We note that although the splashing conditions for single-component droplets have been investigated extensively,^{17–19} a direct implementation of such conditions to the case of two-component droplets may not be trivial. Examples include using the so-called splash threshold β ,¹⁷ which is the ratio between the lifting force and the surface tension at the oil–air interface. The dependence of β on the impact velocity is shown in Fig. 2. It can be seen that β evaluated at the splash transition increases with the impact velocity, as opposed to

FIG. 1. (a) and (b) The impact of compound droplets on a dry flat solid surface at the same impact velocity $V = 2.1 \text{ m} \cdot \text{s}^{-1}$ for two values of oil ratio α . The impacting droplet with $\alpha = 0.02$ spreads smoothly on the surface, whereas the one with $\alpha = 0.07$ splashes. The scale bar represents 1 mm. (c) Schematics (not to scale) of a compound droplet at the initial impact point ($T = 0$) and shortly afterwards ($T > 0$). The radius of the neck and the radius of the spreading lamella are denoted A and R , respectively. The scale bar represents 1 mm.

FIG. 2. Splashing threshold β ¹⁷ vs impacting velocity V at the transition from spreading to splashing.

being a constant in the case of single-component droplets.¹⁷ This discrepancy suggests that taking the surface tension at the outermost interface as the sole resistant factor to splashing may be an incomplete approach to capture the splashing mechanism for two-component droplets: in addition to the interfacial tension between the inner and outer fluids, which is $32 \text{ mN} \cdot \text{m}^{-1}$,²⁷ the viscous effect may play a significant role. In other words, a more elaborate analysis to account for the resistant factors to splashing is required in deriving the splash transition for two-component droplets.

To gain more insight into dynamics of ejected lamellas, we measure the radius $R(T)$ of the spreading lamella and the distance $A(T)$ from the droplet's axisymmetric axis to the "neck," the indented region connecting the lamella and the main body of the droplet [Fig. 1(c)]. Here, T is the time duration measured from the initial moment, i.e., when the droplet appears to touch the surface [Figs. 1(b) and 1(c)]. We nondimensionalize the impacting time T as $t = TV/R_0$, and the two radii R and A as $r = R/R_0$ and $a = A/R_0$, respectively. In Fig. 3(a), we show the dependence of r and a on t for two representative cases having the same impacting velocity $V = 2.3 \text{ m} \cdot \text{s}^{-1}$ and different oil ratios, $\alpha = 0.01$ and $\alpha = 0.04$. We observe that increasing α does not cause a significant change in $a(t)$ but measurable variation in the rate of change in $r(t)$. The effect of α on the changing rate of $r(t)$ can be quantified at the so-called ejection time t_e , which marks the moment when a and r start deviating from one another. Note that for $t < t_e$, a and r are identical. At $t = t_e$, the corresponding ejection velocity v_e of the lamella can be measured experimentally [Fig. 3(a)]. The dependence of the ejection velocity v_e on α is shown in Fig. 3(b). It appears that changing α , although induces an insignificant effect on t_e , causes measurable variations in v_e [Fig. 3(b)]. We note that both v_e and t_e [inset of Fig. 3(b)] for single-component droplets (in this case, water droplets) have larger scatterings compared to those of compound droplets. This may result from a different contributing composition to the resistance of ejection dynamics: the ejection of the lamella is resisted by both surface tension and viscosity. In particular, the outer fluid of compound droplets has lower surface tension ($18 \text{ mN} \cdot \text{m}^{-1}$) but higher viscosity ($4.6 \text{ mPa} \cdot \text{s}$) compared to those of water. The change in relative contribution between surface tension and viscosity may result in smaller ejection time and less scattering compared to the case of water droplets.

To understand the observed effect of varying oil ratios, we adopt a simplified approach in which the impacts of the inner and outer fluids on the solid surface are decoupled, and thus, the impact is considered in two consecutive and independent steps (Fig. 4): the impact of the oil layer occurs first, followed by the impact of the inner fluid after a time gap $\Delta T \sim H_c/V$. Typically, ΔT varies from 2 to $25 \mu\text{s}$ as the thickness of the oil layer is relatively small ($5 \mu\text{m} \leq H_c \leq 38 \mu\text{m}$), resulting in $\Delta T < T_e$. In other words, the lamella is ejected after the second impact for the tested range of α in our experiment ($\alpha < 0.1$). Based on this observation, we postulate that the dependence of v_e on α originates from the time gap ΔT between the two hypothetical impacts. It should be noted that the analysis requires an assumption of a thin oil layer; if the oil layer is thick and the lamella is ejected before the second impact of the inner fluid, then the ejection dynamics may be considered similar to that of single-component droplets.

We now make several assumptions to estimate the lamella's ejection velocity of compound droplets. First, quantitative description for impacts of single-component droplets is still valid for each impact at

FIG. 3. (a) Representative profiles of the normalized "neck radius" $a(t)$ and the normalized spreading radius $r(t)$. The moment at which $r(t)$ starts deviating from $a(t)$ is defined as the ejection time t_e . The ejection velocity v_e is defined as dr/dt at $t = t_e$. (b) The dependence v_e on the volumetric oil ratio α for compound droplets with water as the inner fluid impacting with velocity ranging from 1.5 to $2.6 \text{ m} \cdot \text{s}^{-1}$. The inset shows that although t_e is affected by V , it is unaffected by α . The open markers represent the reference case of impacting water droplets ($\alpha = 0$), while the solid markers represent the case of impacting compound droplets.

the initial stage.¹⁷ Under this assumption and at $t = t_e$, the oil layer moves with velocity $v_o = \sqrt{3/2}t_e^{-1/2}$, whereas the inner fluid mixture moves with velocity $v_m = \sqrt{3/2}(t_e - \Delta t)^{-1/2}$ due to the time gap $\Delta t = \Delta T V/R_0 = H_c/R_0$ between the first impact of the oil surface and the second impact of the oil-mixture interface. As a result, the

FIG. 4. Schematic (not to scale) of the ejected lamella of a compound droplet. The ejected lamella consists of two layers advancing with a combined velocity v_e . The velocity of the oil layer is v_o and that of the mixture is v_m .

ejected lamella of a compound droplet consists of two components: the inner layer moving with velocity v_m and the outer layer moving with velocity v_o (Fig. 4). Second, the lamella's total thickness, which is not measured experimentally, follows the typical expression for the lamella thickness of single-component impacting droplets: $h = \sqrt{12}/(3\pi)t_e^{3/2}$ at $t = t_e$.¹⁷ Similarly, if the oil layer is sufficiently thin, the lamella thickness of the inner fluid is calculated using a similar expression, taking into account the time gap Δt , as $h_m = \sqrt{12}/(3\pi)(t_e - \Delta t)^{3/2}$. It follows that the thickness of the oil layer h_o is calculated by subtracting the total thickness of the lamella and that of the inner layer, resulting in $h_o = \sqrt{12}/(3\pi)(t_e^{3/2} - (t_e - \Delta t)^{3/2})$. Finally, the ejection velocity of the lamella of a compound droplet is calculated using momentum conservation of the involving components as

$$v_e = \frac{h_o}{h} v_o + \frac{h_m}{h} v_m. \quad (1)$$

From Eq. (1) and the expressions of h_m , h_o , and Δt , we establish the relation between the velocity v_e , the ejection time t_e , and the volumetric oil ratio α . The dependence of the ejection velocity on the kinetic characteristics of the involving components [Eq. (1)] should hold up to the ejection time t_e . If we use the experimentally measured values of t_e , both the velocities v_o and v_m can be calculated and used to predict the ejection velocity v_e using Eq. (1). As a test for this approach, which decouples the lamella's kinetic into those of individual components, we show in Fig. 5 several plots comparing the ejection velocity measured experimentally ($v_e - \text{measured}$) and the one predicted using Eq. (1) ($v_e - \text{predicted}$). In this comparison, we vary the viscosity of the mixture μ_m from 1.0 to 11 mPa · s, impacting velocity V from 1.5 to 2.6 m · s⁻¹, and volumetric oil ratio α from 0.01 to 0.09. The agreement between the predicted and measured values of v_e for wide ranges of control parameters suggests that our assumptions on kinetic characteristics of individual components of the lamella can be used to describe its expanding kinetics at the early stage of the impact.

FIG. 5. Measured values of lamella's normalized ejection velocity ($v_e - \text{measured}$) vs the predicted values ($v_e - \text{predicted}$) for V varying from 1.5 to 2.6 m · s⁻¹ and α from 0.01 to 0.09. Four mixtures of different viscosities were used: (a) $\mu_m = 1.0$ mPa · s, (b) $\mu_m = 3.7$ mPa · s, (c) $\mu_m = 6.0$ mPa · s, and (d) $\mu_m = 11$ mPa · s. The shaded areas indicate 5% deviation from predicted values.

We also infer an insignificant effect of the inner fluid's viscosity on the ejection velocity.

We note that the ejection time t_e remains a crucial parameter in the dynamics of lamella ejection. For a droplet of a single component, the inertia-driven ejection is resisted by both the capillary pressure gradient $We^{-1}h^{-2}$ and the viscous shear stress $Re^{-1}v h^{-2} \sim Re^{-1}t^{-1/2}h^{-2}$ in the spreading lamella.¹⁷ Here, the Weber number and the Reynolds number are defined as $We = \rho V^2 R_0 / \sigma$ and $Re = \rho V R_0 / \mu$, respectively. It was then hypothesized that lamella ejection is possible when its velocity exceeds the expanding velocity of the wetted area.¹⁷ The critical condition for ejection to occur, in dimensionless form, can then be derived and simplified as $c_1 Re^{-1}t^{-1/2} + We^{-1} = c^2 t^{3/2}$ at $t = t_e$, where $c_1 = \sqrt{3}/2$ and $c = 1.1$.¹⁷

For two-component droplets, the forces contributing to the ejection of the lamella include the viscous stress at the outer layer, $Re_o^{-1}v_e h^{-2}$, where $Re_o = \rho_o V R_0 / \mu_o$, the capillary pressure gradients at the oil-air interface, $We_o^{-1}h^{-2}$, where $We_o = \rho_o V^2 R_0 / \sigma_o$, and that at the oil-mixture interface, $We_m^{-1}h_m^{-2}$, where $We_m = \rho_m V^2 R_0 / \sigma_m$. Following the analysis for one-component droplets, and since the acceleration of the wetted area is $\dot{v}_o$, the required condition at the ejection time ($t = t_e$) of a compound droplet reads

$$We_o^{-1} \frac{1}{h^2} + We_m^{-1} \frac{1}{h_m^2} + Re_o^{-1} \frac{v_e}{h^2} = \dot{v}_o. \quad (2)$$

We numerically solve Eq. (2) to obtain the dependence of t_e on the impact conditions including the impact velocity V , the mixture's viscosity μ_m , and the volumetric oil ratio α . With each value of μ_m , we calculate and plot the dependence of t_e on Re_m for $\alpha = 0.01$ and $\alpha = 0.09$ (see Fig. 6), where $Re_m = \rho_m V R_0 / \mu_m$. Here, we note that the use of Re_m to plot the ejection time is to separate the datasets corresponding to four different inner fluids used in the experiment. We observe that most of the experimentally measured values of t_e fall within the boundaries generated by the limiting values of α , i.e., the lower and upper bounds correspond to $\alpha = 0.01$ and $\alpha = 0.09$, respectively. We note that although Eq. (2) only differs from the

FIG. 6. Comparison between measured values of the normalized ejection time t_e (solid markers) and the predicted values of t_e for four mixtures of different viscosities. For each value of mixture viscosity μ_m , the predicted values of t_e are represented by the shaded area bounded by two solid lines: the lower one corresponds to $\alpha = 0.01$ and the upper one 0.09.

critical condition for one-component droplets by the term representing the capillary pressure gradient at the oil-mixture interface ($We_m^{-1}h_m^{-2}$), the mixture implicitly affects the ejection time t_e through the dependence of v_e on the mixture thickness and velocity [as specified in Eq. (1)]. Nonetheless, the mixture viscosity μ_m does not appear to affect both t_e and v_e , indicating an insignificant effect of the mixture viscosity on the ejection dynamics. We note that the effect of viscosity of the outer fluid on the ejection dynamics is still an open question and merits a separate study.

In conclusion, the ejecting dynamics of the lamella for impacting compound droplets on solid surfaces is studied and quantitatively characterized with the emphasis on the ejection time and velocity. Besides the impact velocity, a trivial parameter affecting the splashing dynamics and particularly the transition from spreading to splashing, the volumetric oil ratio α was found to play a crucial role in determining the ejection time and velocity. The viscosity of the mixture, however, was found to have an insignificant effect on the ejection dynamics. We also highlight that our model decoupling the impacting dynamics of compound droplets into impacts of the participating fluid components reasonably predicts the ejection time and velocity, suggesting that this approach may be employed in predictive analysis of the splashing threshold for multiple-component droplets.

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