

Splashing Characteristics of Monodisperse Sprays with Significant Viscosity Differences Impacting a Flat Surface

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Drop impingement and splashing as a function of viscosity and, to a lesser extent, temperature are examined here. The working fluid is a mixture of water and glycerin with relative mass percentages varying from 0 to 100%, which spans a viscosity range of three orders of magnitude. First, a criterion that separates "on" and "off" for splashing as a function of glycerin percentage is expressed in terms of both Weber and Reynolds numbers and its highly nonlinear behavior is a function of the change in fluid viscosity. Next, the complex splashing characteristics of a rather simple monodisperse spray injected at a pressure of 2 bars onto a flat, 2-mm-diameter aluminum cylindrical rod are examined. Spatial variations in the fraction of splashed liquid, Sauter mean diameter, splashed droplet size distribution, and splash volume fraction as a function of radial distance for these mixtures are reported.

Keywords Dissipation energy; Glycerin viscosity; Splash criteria; Spray coating

INTRODUCTION

Drop impact phenomena are encountered in numerous industrial applications such as spray drying,^[1-6] pharmaceutical coating,^[7–9] inkjet printing, painting, spray cooling, fire suppression,^[10,11] and fuel injection systems of internal combustion engines.^[12] It is also relevant to spray-drying applications where powders are produced from suspensions and solutions that are dispersed with various types of atomizers. Another area of application is found in thin-film coating for solar cell production, which is in rapidly increasing demand due to green technologies.^[13] Instead of using a high-cost vacuum physical vapor deposition (PVD) or chemical vapor deposition (CVD) method, non-vacuum spray coating at room temperature is now considered an economical way to produce cost-competitive solar cells. In this solar cell spray-coating application, liquid drops are used as a carrier and are dispersed and deposited onto a substrate. These sprayed solvent drops, often mixed with highly viscous additives, contain semiconducting nanoparticles to enhance coating quality by decreasing drop splashing.

During this deposition process, well-known impact phenomena like bouncing, spreading, and splashing can occur. When the surface tension is high, bouncing can occur. With increasing dynamic force (kinetic or impact energy), spreading and eventually splashing become the predominant mechanisms. Though surface tension effects are described by the Weber number (We), impact phenomena are also influenced by viscosity and hence the Reynolds number (Re) is also an important nondimensional parameter. The combination of these two parameters comprise the impingement parameter, which is defined as $K = We^{a}Re^{b}$, where a and b are constant^[14] and K is a useful nondimensional parameter that reflects the effects of kinetic energy, viscosity, and surface tension effects on the action of drop impact and can be used to define incipient splashing. Our previous work underscored the importance of viscosity^[15] in splashing, and this effort extends the analysis to further evaluate viscosity effects.

To better understand the deposition processes, we focus on a detailed analysis of postsplashing mechanisms subject to significant viscosity changes for a water–glycerin mixture including moderate temperature changes. In a real solar cell coating process, the substrate temperature may be up to 600°C, which may induce the Leidenfrost effect that prevents a drop from depositing because of sudden vapor formation between the drop and substrate. However, this work will not address the issue of elevated substrate temperature (therefore, no phase change is included) so that we can focus only on splash characteristics by varying liquid properties. Moreover, although nearly all real sprays are polydisperse, a monodisperse spray is considered here to simplify this analysis.

An energy balance is invoked to describe how the total initial energy is distributed across various regimes after impact. Three major postimpact modes are noted: rebounding, spreading, and splashing. Rebounds are atypical, do not splash, and are not observed here. Spreading is a prerequisite for splashing where jets or fingers at the rim of a crown separate from the spreading crown given sufficient energy. Because total energy is conserved, balance requires

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that the incoming drop's kinetic and surface tension energies (KE_i and SE_i) are equal to the postimpact energies, which include surface tension energy of a spreading disk (SE_{disk}), kinetic and surface tension energies (KE_{sat} and SE_{sat}) of splashed satellite droplets, and energy lost to viscous dissipation ($DISS_{visc}$):

$$KE_i + SE_i = SE_{disk} + DISS_{disk} + N_{sat}(KE_{sat} + SE_{sat})$$
(1)

 N_{sat} represents the number of splashed or satellite droplets, which can be estimated from the mass balance as in $N_{\text{sat}} = f_{\text{sp}}/(D_{\text{sat}}/D_i)^3$ where and f_{sp} , D_{sat} , and D_i stand for the mass fraction of the splashed liquid, splashed satellite diameter, and the incoming drop's original diameter. This energy balance suggests that the postimpact satellite's kinetic energy, KE_{sat} , is small if the dissipation energy is large or vice versa. According to Chandra and Avedisian,^[16] this dissipation energy is directly proportional to viscosity. In other words, viscosity largely controls the postimpact phenomena, which are quantified in our experimental studies with splashed fraction, Sauter mean diameter, dispersion coefficient, and the line volume concentration for splashed satellite droplets.

Our unique data describe how much mass is splashed and deposited as a function of the liquid's viscosity and temperature. Also, we report the average size and distribution of the splashed "droplets" (*drop* refers to the impacting drop and *droplet* refers to the satellite or splashed liquid) as well as how far these splashed droplets travel after impact. The working fluid is a water–glycerin mixture with *mass* percentage of glycerin varying from 0 to 100%, which yields a significant viscosity change; see Fig. 1. Liquid drops of nearly uniform size (monodisperse) were injected at a pressure of 2 bars onto a 2-mm-diameter flat aluminum cylindrical rod.



FIG. 1. Viscosity as a function of glycerin percentage.

EXPERIMENTAL SETUP

A schematic diagram of the experimental apparatus is shown in Fig. 2. Liquid was supplied to the atomizer using pressurized nitrogen. A pressure transducer and thermocouples were installed in the liquid supply pipe to measure injection (or operating) pressure and temperature to within 1%. The volumetric (or mass) flow rate was measured using a gear displacement flow meter with accuracy also within 1%. The temperature controller varied the liquid temperature from 20 to 35°C, which in turn slightly varied the liquid viscosity, density, and surface tension. The monodisperse spray injector, subject to an injection gauge pressure of 2 bar, was positioned 7 cm above the top of the impact rod; see Fig. 3. An initial or injected drop size was $D_i = 235 \pm 5 \,\mu\text{m}$ and the maximum injection speed was 14 $< U_i < 15 \text{ m/s}$, depending on the glycerin mass percentage. This injection speed remains nearly constant across the 7 cm until impact and, thus, we assume it as the impact speed. Though the Weber number change is relatively small, increasing the glycerin fraction in the mixture yields a viscosity increase of nearly 1,000-fold, which in turn reduces the Reynolds number by a factor of 1,000. Upon impact onto the rod, splashed droplets are ejected with horizontal momentum and the remaining nonsplashed liquid is collected from drippings off the bottom of the rod. Horizontally ejected droplets are also collected using a patternator shown in Fig. 2.

To measure droplet characteristics, a 10-mm-diameter laser beam transects various x (0 < x < 8 cm) and y (0 < v < 4 cm) locations at spatial intervals of 5 mm as shown in Fig. 3. A Malvern particle sizer measures the Sauter mean diameter (SMD) using Fraunhofer diffraction from a monochromatic light beam. Splashed droplet diameters, D_s, were fit to a Rosin-Rammler distribution and they are typically $20 < D_s < 110 \,\mu\text{m}$. Based on previous experience,^[17] droplet size uncertainty is within 5% because the detector monitors a statistically reliable average of the light-scattering characteristics across several million droplets. A high-speed CCD camera (500 to 10,000 fps) was used to capture images of the splashing droplets with stroboscopic illumination. All work was conducted at room temperature, although additional lighting could raise local air temperature slightly above 300 K. Thermal evaporation of the splashed droplets was assumed negligible; the important parameters governing splashed droplet characteristics are the fluid's viscosity change because of glycerin fraction and the moderate temperature variation, 20 to 35°C.

RESULTS AND DISCUSSION Single Drop Impact

In this section, the splash criterion that separates on and off for single-drop splashing as a function of glycerin



FIG. 2. Schematic of the experimental setup.

percentage is expressed as a function of Weber and Reynolds numbers.

In an effort to find the critical impingement parameter describing incipient splashing, $K_{\rm crit}$, for a given glycerin percentage, the drop impact speed was varied by changing the drop release height without any injection pressure, which gives the impact speed of $U_i = \sqrt{2gh}$. Here, $K_{\rm crit}$ is defined as $K_{\rm crit} = We^{0.5} Re^{0.25}$, according to Mundo et al.^[14] The diameters of released drops remain fairly constant as $D_i = 4.760 \,\mathrm{mm}$ or $4,760 \,\mathrm{\mu m}$ (much larger drop compared



Measuring Point

FIG. 3. Droplet measurement points.

to the diameter of the monodisperse spray, $D_i = 235 \pm 5 \,\mu\text{m}$); the impacting substrate was aluminum with a nondimensional surface roughness on the order of $R^* = R/D_i \sim 10^{-5}$ or less.

Figure 4 displays the snapshots taken 400 µs after a single drop's initial impact for various glycerin percentages. The drop release height and the impact velocity were h = 1.9 mand $U_i = 6.1 \text{ m/s}$, respectively. Between 0 and 20% glycerin, moderate splashing is observed. Within this glycerin range (which we define as Regime 1), the dynamic motion of the spreading disk is impeded by viscous (or shear) forces at the substrate wall; there is competition between the dynamic force and viscous force. In Regime 1, when viscosity is higher, the shear stress is higher, which slows the spreading motion and results in a smaller spreading disk. Moreover, it is more difficult for a drop to splash (or destabilize) at higher viscosities because viscosity tends to stabilize disturbances from impact. As a result, the critical impingement parameter, which distinguishes on and off for splashing, is higher at higher viscosity; see Fig. 5. Regime 1 is therefore referred to as the viscous stability regime. Overall, the threshold value of K_{crit} appears to be relatively high compared to those of Mundo et al.,^[14] perhaps because of the small roughness of the substrate used herein.

When glycerin is in the range of 20 to 60%, the splashing phenomenon takes an unexpected turn: splashing actually increases with increasing viscosity. This new regime, Regime 2, is referred to as the *transient* or *viscous instability* regime. Though it is generally assumed that viscosity stabilizes flow, it can also destabilize flow when an adverse



0% (*Re*=32621) 10% (*Re*=25754) 20% (*Re*=20611) 30% (*Re*=12247) 40% (*Re*=10212)

50% (Re=6482)



60% (Re=3789) 70% (Re=1989) 80% (Re=750) 90% (Re=223) 100% (Re=38)

FIG. 4. Snapshots taken 400 μ s after the single drop's initial impact for glycerin mass percentages varying from 0 to 100%. The corresponding impingement parameter, *K*, of these snapshots is plotted in Fig. 5 with circles. The drop release height and the impact velocity are h = 1.9 m and $U_i = 6.1$ m/s, respectively.

pressure gradient at the wall causes a rollup motion (primary instability), which eventually transforms into a hairpin vortex (secondary instability).^[18,19] In this viscous instability regime, increasing the viscosity causes a larger adverse pressure gradient due to increased shear stress at the contact wall. For flows in general, increased shear stress at the boundary layer expedites the incipient turbulence. This source of turbulence is described by the Orr-Sommerfeld equation,^[20–22] which identifies the dominant wavelength arising from that viscous instability. For 20



FIG. 5. Splash criterion that separates on and off for splashing with changing glycerin percentage. Red circles represent the values of *K* from Fig. 4. The first seven data points, up to 60% glycerin, show splashing because their *K*s are greater than K_{crit} (black squares). K_{crit} was not available for glycerin percentage greater than 60% because drop impact speeds of greater than 10 m/s were not achievable with our experimental setup.

to 60% glycerin, the primary source of instability is the viscous shear stress, which yields a highly nonlinear velocity profile near the interface that eventually leads to rollup motion and that yields a highly nonlinear velocity profile near the interface that eventually leads to rollup motion and splashing. Within this viscosity range, splashing is noted for relatively small K_{crit}. Above 70% glycerin, splashing is completely suppressed and measurement of $K_{\rm crit}$ was not possible because it was too large to be measured using our experimental apparatus. This is Regime 3, where viscosity again acts as a stabilizing force and splashing is inhibited. Note that according to Brown et al.,^[23] a 10-cm-diameter, high-glycerin-content drop with an impact speed of $\sim 100 \,\mathrm{m/s}$ does not splash even at We ber and Reynolds number of $We \approx 2 \times 10^5$ and $Re \approx 10^4$, yielding $K = We^{0.5} Re^{0.25} = 5 \times 10^3$, which was not achievable with our current experimental setup.

It is noteworthy that the impingement parameter, *K*, can be defined in various forms. For example, Mundo et al.^[14] defined it as $K_{\text{Mundo}} = Re^{1.25}$ Oh or $We^{0.5} Re^{0.25}$ where $Oh = We^{0.5} Re^{-1.0}$. Cossali et al.^[24] used a slightly modified version, $K_{\text{Cossali}} = We^{1.0} Oh^{-0.4} = We^{0.8} Re^{0.40}$, with exponents substantially larger than those of Mundo et al.^[14] Yarin and Weiss^[25] introduced the nondimensional impact velocity, $u^* = Ca \lambda$, where the capillary number is defined as $Ca = \rho U_i \nu / \sigma = We/Re$, and $\lambda = (\nu / f)^{1/2} / (\rho \nu^2)$ with frequency $f = (3/2)(U_i / \varphi)(d/\varphi)^2$; nozzle diameter *d*; and $\varphi \approx 2d$. Yarin and Weiss^[25] noted that the nondimensional impact velocity, in the range of $17 \le u^* \le 18$, is the splashing threshold for the wide range of λ and *Ca* for the impact of continuously injected drops over an initially dry substrate that became wet due to impact of the preceding drops. Here, u^* can be reduced to $K_{\text{Yarin-Weiss}} = u^* = We^{0.25} Re^{0.125}$ for the case of single drop impact if $f = U_i/D_i$. It is interesting that the K parameter of various authors retains the form of $We^a Re^b$, where b = a/2, and the influence of the Weber number is always more prominent than that of the Reynolds number. The threshold range of $17 \le u^* \le 18$ seems to be applicable to our case also when the glycerin percentage is relatively low, as shown in Fig. 6 (note that $K_{\text{Yarin-Weiss}}$ is not the same as K_{Mundo}). However, upon increasing the glycerin percentage, this splashing threshold no longer holds, suggesting that viscosity is a critical factor not sufficiently considered in deriving the empiricism that b = a/2. Perhaps this ratio should be investigated further when large changes in viscosity are possible, but such an effort is beyond the scope of this current study. Moreover, differences in splashing criterion between our results and those of Yarin and Weiss^[25] could arise because our experiment is for a single drop onto a "dry" substrate, whereas Yarin and Weiss examined monodisperse spray onto a "wet" substrate (though initially dry), yielding various uncertainties such as liquid film thickness and surface roughness.

Monodisperse Drop Impact on a Cylinder

Figure 7 presents snapshots of glycerin–water mixture drops impacting onto an aluminum cylindrical rod at various liquid temperatures. Drops splash upon impact and form crowns, which transform into fingers and satellite droplets of various sizes. Because the surface tension does not vary much with glycerin fraction, the Weber number remains fairly constant, 643 < We < 822, whereas the Reynolds number ranges from Re = 631 to 4,688 (Fig. 7). Snapshots for glycerin >60% are not shown because there was no splashing for the impact velocity and temperatures considered here.

A temperature decrease induces a viscosity increase, which, in turn, decreases the Reynolds number. Based on



FIG. 6. The splash criterion ($K_{\text{Yarin-Weiss}}$) or the nondimensional impact velocity (u^*) of Yarin and Weiss^[25] as a function of glycerin percentage, which is different from $K_{\text{Mundo}} = We^{0.5} Re^{0.25}$.

the snapshots, it is difficult to discern the effect of a moderate temperature change (20 to 35° C) on splashing phenomena. However, it is apparent that the glycerin fraction alters splashing. In general, when glycerin percentage increases, there is less splashing because of the increase in viscosity.

It is noteworthy that the higher glycerin percentages yield larger splashed droplets, thicker crown, and more uniform size distributions of splashed droplets. When the viscosity is low (low glycerin percentage), less energy is lost through viscous dissipation and more kinetic energy is available for the splashed droplets, yielding a broader size distribution of splashed droplets. Conversely, when the glycerin percentage is high, much of the incoming drop's kinetic energy is lost through viscous dissipation and, thus, less energy is available for the splashed droplets.

Splash Fraction

Figures 8 and 9 plot the fraction of splashed droplets as functions of liquid temperature and glycerin percentage, respectively. Injected drop diameters remain fairly constant for all cases. Splash fraction is calculated as

$$f_{sp} = \frac{(\dot{m}_o - \dot{m}_f)}{\dot{m}_o},\tag{2}$$

where \dot{m}_o is the injected mass flow rate and \dot{m}_f is the mass flow rate collected from drippings off of the bottom of the rod.

Figure 8 indicates that a moderate temperature increase induces a slight increase in splash fraction, which becomes more evident when the glycerin percentage is high. When glycerin percentage is low or zero, the moderate temperature change does not induce a change in the splash fraction, as shown in Fig. 8. It is interesting that the temperature trend becomes apparent when glycerin is greater than 20%.

For a given liquid temperature, the effect of glycerin percentage on splash fraction is shown in Fig. 9. With increasing percentage, less splashing is observed, nearly independent of temperature. At zero glycerin percentage, about 0.65 of the total mass splashed. However, at a glycerin percentage of 50%, only 0.15–0.25 of the total mass splashed. There is no splash observed for larger glycerin fractions; that is, >60%.

Sauter Mean Diameter

Figure 10 shows the SMD, taken at the vertical location of the top of the impact rod, for various glycerin percentages, liquid temperatures, and horizontal distances. In general, the SMD of the droplets increases as the radial distance increases for all glycerin percentages and temperatures. Because a larger SMD is observed at larger radial distance, this indicates that the larger droplets of the



FIG. 7. Snapshots of water-glycerin mixture at various glycerin percentages and temperatures.

distribution travel farther and have more momentum. This does not, however, suggest that all large droplets travel far, only that the largest droplets in the distribution travel farther than their smaller counterparts. It is also clear that an increasing glycerin percentage yields larger satellite droplets for all temperatures. This trend is expected



FIG. 8. Splash fraction as a function of liquid temperature.



FIG. 9. Splash fraction as a function of glycerin percentage.



FIG. 10. SMD variation for various glycerin percentages and temperatures: (a) 20° C, (b) 25° C, (c) 30° C, and (d) 35° C.

because it is thought that droplet size is inversely proportional to the Reynolds number;^[26] more glycerin \rightarrow decreased Reynolds number \rightarrow larger droplet size. Temperature increases narrow the SMD gap between the

results. This effect is primarily a result of the 50% glycerin case approaching the other cases as temperatures increase. In other words, SMD is most sensitive to temperature change when glycerin is at 50%. Figure 11 summarizes the effect of liquid temperature on SMD. As expected, temperature increases, which induce viscosity decrease, yield an overall decrease in SMD.

Rosin-Rammler Distribution of Postimpact Droplets

Figures 12 and 13 display the distribution (or dispersion coefficient) of the Rosin-Rammler distribution for various glycerin percentages and horizontal distances. The larger the N, the more uniform the distribution (or smaller width in the PDF approaching monodisperse droplets). The Rosin-Rammler probability distribution function is

$$PDF(D_S) = \frac{ND_S^{N-1}}{\overline{D}^N} \exp\left[-\left(\frac{D_S}{\overline{D}}\right)^N\right],\tag{3}$$

where D_s is the splashed droplet diameter and \overline{D} is the characteristic or mean droplet size^[19,27] defined as:

$$D_{10} = \overline{D}\Gamma\left(\frac{1}{N} + 1\right),\tag{4}$$

or

$$D_{32} = \frac{\overline{D}^3 \Gamma(3/N+1)}{\overline{D}^2 \Gamma(2/N+1)}.$$
(5)

Here, D_{10} and D_{32} are the arithmetic and SMD, respectively, and Γ is the gamma function. Based on Figs. 12 and 13, at the furthest detectable droplet distance in the horizontal direction, N is fairly large, N > 20; the droplets



FIG. 11. SMD variation for a 50% glycerin mixture.



FIG. 12. Rosin-Rammler distribution for various glycerin percentages and temperatures: (a) 20°C, (b) 25°C, (c) 30°C, and (d) 35°C.

that traveled furthest (largest horizontal distance) approach monodispersity. N increases with distance because near the impact, the measurement includes all splashed droplets and, farther away, only the larger, faster



FIG. 13. Rosin-Rammler distribution for a 50% glycerin mixture.

droplets remain, leaving a more narrow distribution of (larger) droplets to measure. This suggests that distribution of the postimpact kinetic energy for satellite droplets is distributed in a way that the larger droplets receive a larger portion of the kinetic energy.

In general, it is fair to state based on results from Fig. 12 that a larger glycerin fraction yields a more uniform or less dispersive size distribution at all temperatures. When a drop impacts, waves ripple across its surface as it distorts into a spreading disk. These variously sized waves contribute to a distribution of sizes for splashed droplets. With increasing viscosity, both magnitude and wavelength of these waves are reduced, with consequent narrower distribution of splashed droplet sizes (larger Rosin-Rammler N; see Fig. 14).

Volume Distribution

Splashed droplets are collected using a patternator with bins located at the bottom of the cylinder; see Fig. 2. Because the patternator is a line bin, only the line volume of the splashed droplet is collected into the bins. These collected volumes are normalized by the entire volume of the corresponding splashed liquid, which are plotted in Fig. 15. The maximum of the normalized volume is less than 1.5%, indicating that only a small fraction of the entire splashed liquid is collected in the bins.

Captured volume maximum and minimum values are seen near the cylinder center (at 12.5 cm) and at the furthest radial distance, respectively. At 50% glycerin, the slope of the normalized volume with respect to the radial distance is steepest because not much liquid is splashed due to the high viscosity. Conversely, the slope becomes more gradual with reduced glycerin percentage, indicating that fluid is splashed farther with decreasing viscosity.



FIG. 14. Comparison of drop size distributions obtained at various glycerin percentages. Note that the Rosin-Rammler PDF model is used with the size data obtained at 20° C and radius location of 3 cm from Figs. 10a and 12a.

It is noteworthy that the splashed fraction from 50% glycerin is comparable to that of 0% glycerin (or 100% water) near the cylinder center. This trend is also reflected in the snapshots for Re = 631 from Fig. 7. When viscosity is increased, energy lost through viscous dissipation is significant and less kinetic energy remains for the splashed droplets. Consequently, most high-glycerin-content splashed droplets (even though they are, in general, larger) fall near the cylinder because they do not have sufficient kinetic energy to travel farther. This does not contradict our observation that the largest droplets (for all viscosities and Reynolds numbers) travel farther than their smaller



FIG. 15. Normalized volume distribution for various glycerin percentages at 20° C.

counterparts (Fig. 10). More glycerin (increased viscosity) yields larger droplets, in general, with more energy lost during impact to viscous dissipation, but those large droplets of the splashed distribution still travel farther than the smaller ones. Small droplets generated from large-glycerin-fraction impacts have, in general, the shortest travel distances.

CONCLUSION

The critical impingement parameter, $K_{\rm crit}$, describing incipient splashing was defined. Next, the postimpact characteristics of splashed droplets from uniform drop impacts onto a cylindrical rod were studied experimentally. These characteristics include splash fraction, SMD, dispersion coefficient, and the line-volume concentration. The working fluid was a water–glycerin mixture where glycerin mass varied from 0 to 100%, which facilitated a viscosity change of nearly 1,000-fold. With increasing viscosity, smaller amounts of splashing liquid were observed and the splashed droplets tended to be more uniform and larger but traveled shorter distances due to increased energy lost through viscous dissipation.

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