Rupture of thin films formed during droplet impact

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Rupture of liquid films formed during droplet impact on a dry solid surface was studied experimentally. Water droplets (580 ± 70 μm) were photographed as they hit a solid substrate at high velocities (10–30 m s⁻¹). Droplet–substrate wettability was varied over a wide range, from hydrophilic to superhydrophobic, by changing the material of the substrate (glass, Plexiglas, wax and alkylketene dimer). Both smooth and rough wax surfaces were tested. Photographs of impact showed that as the impact velocity increased and the film thickness decreased, films became unstable and ruptured internally through the formation of holes. However, the impact velocity at which rupture occurred was found to first decrease and then increase with the liquid–solid contact angle, with wax showing rupture at all impact velocities tested. A thermodynamic stability analysis combined with a droplet spreading model predicted the rupture behaviour by showing that films would be stable at very small or at very large contact angles, but unstable in between. Film rupture was found to be greatly promoted by surface roughness.

Keywords: droplet impact; film rupture; wettability; superhydrophobic surfaces

1. Introduction

When a liquid droplet impacts on a dry solid surface, it spreads radially outwards until its kinetic energy has been dissipated, creating a thin liquid film whose radius and thickness depend primarily upon initial droplet diameter, impact velocity and liquid properties (Pasandideh-Fard et al. 1996; detailed reviews on the general phenomena of droplet impact on solid and liquid layers have been given by Lesser & Field 1983; Rein 1993; Yarin 2006). Many engineering applications that use spray deposition require the deposited liquid layer to remain stable and adhere to the substrate. For example, in spray painting or coating, a thin, uniform liquid film is applied on a substrate and it must stay intact until it dries. During pesticide spraying on plants, the liquid should remain on the leaf surface and not drip to the soil where it can contaminate underground water. A contiguous liquid layer is often difficult to achieve because droplets impinging with sufficient velocity to spread them into a thin film may disintegrate into smaller fragments that bounce off.

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Owing to the practical importance of the subject, many studies (e.g. Stow & Hadfield 1981; Mehdizadeh et al. 2004b; Xu et al. 2005; Deegan et al. 2008) have been conducted to investigate the cause of droplet break-up during impact. The focus of these studies has been the instability along the edges of the thin film created by the impacting droplet (Allen 1975; Kim et al. 2000; Mehdizadeh et al. 2004b) and the effect of factors such as surface roughness (Stow & Hadfield 1981), surrounding gas pressure (Xu et al. 2005, 2007), substrate elasticity (Pepper et al. 2008) and solidification (Dhiman & Chandra 2005) on droplet splashing. Most of these studies have been conducted at relatively low impact velocities (1–10 ms$^{-1}$), in which fluid instabilities around the edge of the spreading droplet caused the formation of long fingers that detached to form satellite droplets (Allen 1975; Kim et al. 2000). At higher impact velocities, up to 40 ms$^{-1}$, photographs of water droplets impacting a polished stainless steel surface showed that the liquid film became so thin that it ruptured internally and then, as the holes expanded owing to surface tension, disintegrated completely (Mehdizadeh et al. 2004b). When the substrate was heated sufficiently to trigger nucleate boiling, so that vapour bubbles punctured the liquid film at a large number of points, it disintegrated into a cloud of fine droplets (Mehdizadeh & Chandra 2006). Most practical spray applications use high impact velocities, and it is likely that a major cause of droplet break-up is internal rupture rather than edge instabilities.

The formation of holes in a stationary liquid film has been the subject of many studies (Padday 1970; Taylor & Michael 1973; Sharma & Ruckenstein 1989). Padday (1970) measured the critical thickness below which water films ruptured on a variety of surfaces and found that the critical thickness increased with the liquid–solid contact angle. Taylor & Michael (1973) investigated the formation of a hole in water and mercury films by blowing an air jet onto the film. They applied the Young–Laplace equation of capillarity to the hole profile and found that for a given film thickness and contact angle, there exists a critical hole size: larger holes grow, whereas smaller ones heal. Sharma & Ruckenstein (1989) presented a thermodynamic analysis which considered the difference in free energy of an intact liquid film and one with a hole in it. Their predictions of a critical film thickness, below which films became unstable when punctured by a hole of a given diameter, matched well with the observations of Padday (1970). Kheshgi & Scriven (1991) investigated the mechanisms responsible for hole formation and argued that the nucleation of a hole is preceded by a film thinning disturbance that locally thins the film. Redon et al. (1991) studied the rate of hole growth and found that their velocity is independent of film thickness, but critically dependent on the receding contact angle formed by film liquid on the surface.

In a previous paper (Dhiman & Chandra 2008), we investigated the internal rupture of radially spreading films formed by the normal impact of a water jet on a horizontal plate. Combining a simple mathematical model with the thermodynamic analysis of Sharma & Ruckenstein (1989) to predict film thickness, we developed a criterion to predict film rupture by calculating a critical Reynolds number ($Re$) for the impacting jet, above which rupture would occur. The critical $Re$ depended on both the initial diameter of the hole initiated in the film and the liquid–solid contact angle.

Our objective in this study was to determine conditions under which an impacting droplet, flattening into a thin film, would rupture internally. We varied substrate wettability by using four different surface materials (Plexiglas, glass,
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wax and alkylketene dimer (AKD)) for which advancing liquid–solid contact angle varied from 58° to 164°. Wax substrates with two different values of surface roughness were tested: 0.13 and 1.98 μm. Impact velocity was varied from 10 to 30 m s⁻¹, giving Reynolds and Weber (We) number ranges of 5800–17 400 and 800–7200, respectively. We developed a simple criterion, based on the analysis of Sharma & Ruckenstein (1989), to predict conditions under which the liquid film would rupture.

2. Experimental method

To produce a liquid film thin enough to rupture while spreading, small droplets with high impact velocities are required. This was achieved by using a droplet generator that produced sub-millimeter size droplets, which collided with a substrate mounted on the rim of a 0.4m diameter, rotating flywheel. By synchronizing the position of the rotating substrate with the ejection of droplet, the substrate hit the falling droplet at a high velocity while a fast-shutter charge-coupled device (CCD) camera captured the dynamics of impact. Figure 1 shows a schematic of the experimental apparatus, which has been described in detail by Mehdizadeh et al. (2004a).

Droplets of distilled water, 580 ± 70 μm in diameter, were produced using a pneumatic drop-on-demand droplet generator (Cheng & Chandra 2003), which works by applying a gas pressure pulse on the free surface of water contained in a cavity, forcing a single droplet out of a small hole (250 μm) in its bottom. The ejected droplet fell vertically downwards with a velocity less than 1 m s⁻¹ towards the centre of the substrate. The test surface was glued to an aluminium coupon (38.1 × 25.4 × 1 mm thickness) that could be screwed onto an aluminium plate bolted to the flywheel rim. The rotational speed of the flywheel was varied from 470 to 1410 r.p.m., giving linear velocities of 10–30 m s⁻¹, which were maintained within ±1% by a digital motion controller. The position of the revolving substrate was detected by an optical sensor. The signal was then sent to a time delay unit (DG535; Stanford Research Systems, CA, USA), which produced three signals in reference to the input signal. These signals were sent to a CCD camera, an electronic flash unit and the droplet generator, so that as the falling droplet was hit by the rotating substrate, the camera and flash were triggered to capture a single image of an impacting droplet. The CCD camera (Sensicam, Optikon Corporation, Kitchener, Ontario, Canada) used to capture the images of droplet deformation during impact could be shuttered as fast as 0.1 μs with high resolution (1280 × 1024 pixels). Illumination during impact was provided by an electronic flash of 10 μs duration.

By varying the delay between droplet generation and triggering the flash, different stages of droplet deformation were captured and impact dynamics reconstructed from a sequence of these photographs. This technique, commonly known as single shot photography, yielded higher-resolution images than possible with typical high-speed video cameras and allowed us to clearly observe tiny holes that formed in films. However, the repeatability of droplet impact was less than the time interval between images, and therefore there are no times indicated next to individual frames. There were two reasons for this drop-to-drop variation: the instant of droplet impact varied slightly owing to air turbulence caused by
substrate rotation; and droplet recoil, which is driven by capillary forces, was affected by local variations in surface wettability and roughness that randomly alter droplet shape during receding. This lack of timing information, however, was not a serious limitation as we did not make any time-dependent measurements from the images.

Four different substrate materials were used in this study: glass, Plexiglas, wax and AKD, which is a superhydrophobic surface (Mohammadi et al. 2004). Two wax surfaces, with differing roughness, were prepared. Molten wax was poured onto a mirror-polished aluminium plate; the upper surface of the wax, which solidified in contact with air, was much rougher than the lower surface. All surfaces were characterized by measuring their roughness and contact angles (advancing, receding and equilibrium) formed with water droplets. Roughness was measured with an interferometric microscope that scanned each surface and produced a three-dimensional profile of it. Each surface was scanned at five different locations, and the results were averaged to obtain an average for the entire surface. Average surface roughness $R_a$ was $0.01 \mu m$ for the glass
Table 1. Characteristics of the solid surfaces used.

| surface     | equilibrium contact angle $\theta$ (°) | advancing contact angle $\theta_a$ (°) | receding contact angle $\theta_r$ (°) | average roughness $R_a$ (μm) |
|-------------|----------------------------------------|----------------------------------------|---------------------------------------|-----------------------------|
| glass       | 47                                     | 58                                     | 20                                    | 0.01 ± 0.003                |
| Plexiglas   | 71                                     | 80                                     | 40                                    | 0.02 ± 0.006                |
| smooth wax  | 105                                    | 107                                    | 84                                    | 0.13 ± 0.02                 |
| rough wax   | 102                                    | 106                                    | 85                                    | 1.98 ± 0.62                 |
| AKD         | 160                                    | 164<sup>a</sup>                        | 147<sup>a</sup>                       | 1.25 ± 0.17<sup>a</sup>     |

<sup>a</sup>Mohammadi <i>et al.</i> (2004).

substrate, 0.02 μm for Plexiglas, 0.13 μm for smooth wax and 1.98 μm for rough wax. Average roughness of the AKD samples was 1.25 μm (Mohammadi et al. 2004). In all cases, surface roughness was an order of magnitude less than the thickness of the liquid film, which was typically approximately 20 μm.

The procedure followed to measure the advancing and receding contact angles was that described by Johnson & Dettre (1993). A micrometre-controlled syringe was used to dispense small water droplets on a surface through a needle. With the needle still inside the sessile droplet formed, water was added slowly with the syringe until the liquid–solid contact line at the edge of the droplet began to advance. The motion of the droplet was recorded with a CCD camera. Images of the moving contact line were imported into image analysis software (IMAGEJ; National Institutes of Health, USA), and the advancing contact angle was measured. Receding contact angles were measured by reversing the flow of the liquid, sucking water back into the syringe until the contact line began to recede. The results obtained for the contact angles for all surfaces used along with the roughness measurements are shown in table 1. All experiments were performed at 25 °C, controlled by a thermostat-monitored room air conditioning system, and 1 atm pressure.

3. Results and discussion

Figure 2 shows three different sequences of the impact of water droplets on a smooth glass surface at three different impact velocities: 10, 20 and 30 m s<sup>-1</sup>. Glass had the lowest contact angle of the four surfaces tested, $\theta = 47°$. Each vertical column shows successive stages of impact at one of the velocities, which yielded Reynolds numbers ($Re$) of 5800, 11 600 and 17 400, respectively. Droplets flattened into a thin film after impact as they reached their maximum extension (figure 2b), which was followed by retraction until they eventually attained equilibrium. The diameter $D_{\text{max}}$ of the films at maximum extension increased with $Re$ (figure 2b), and hence their thickness decreased. At $Re = 5800$ and 11 600 the films remained stable, but at $Re = 17 400$ the film ruptured and several holes became visible (figure 2c), which continued to grow afterwards (figure 2d, e) rendering the film unstable.
Reducing the wettability of the substrate changed the impact dynamics significantly and promoted hole formation. Figure 3 shows the impact of water droplets on a Plexiglas surface that has a contact angle higher than that on glass ($\theta$ for Plexiglas = 71°). The impact at $Re = 5800$ was similar to the one seen on glass surfaces. However, droplets impacting at both $Re = 11600$ and 17400 now became unstable and ruptured. At $Re = 17400$, where $D_{\text{max}}$ was the greatest, the number of holes formed was also larger than that observed at $Re = 11600$.  

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Impact on a smooth wax surface, whose contact angle is even larger than that on Plexiglas ($\theta$ for wax = $105^\circ$), showed rupture at all $Re$ in our experiments, as shown in figure 4. In addition, holes on wax surfaces are much larger, unlike those on glass and Plexiglas surfaces, because they grow rapidly owing to their high contact angle with the solid. The rate at which holes grow has been shown to be critically dependent on the receding contact angle of the liquid with the solid (Redon et al. 1991). As a result, the area covered by the liquid after the impact of water drops on wax surfaces was the least compared with both glass and Plexiglas surfaces.

The dynamics of droplet impact on a wax surface with roughness $R_a=1.98 \mu m$ was quite different from that on smooth ($R_a=0.13 \mu m$) wax and is shown in figure 5. A large number of holes appeared in the water film almost immediately.

Figure 3. Impact of 580 $\mu$m diameter water droplets with different $Re$ on smooth Plexiglas surfaces.
after impact along with long fingers at their periphery (figure 5a). The holes grew until their boundaries met those of the neighbouring holes (figure 5b). The final state consisted of a number of small droplets resting on the surface (figure 5c). The diameter $D_{\text{max}}$ of the films at maximum extension and the number of holes formed increased with $Re$. However, compared with previous images on glass, Plexiglas and smooth wax surfaces, $D_{\text{max}}$ was approximately 25% smaller, which may be due to material loss through splashing triggered by surface roughness before the films reached their maximum extension.

Increasing the contact angle further by using a superhydrophobic surface (AKD) revealed very different impact behaviour. This is shown in figure 6 for the three values of $Re$ investigated. The AKD surface had the highest liquid–solid contact angle ($\theta = 160^\circ$) among all surfaces tested. Figure 6 shows that splashing from the edges of the droplet is visible at all $Re$, unlike the

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other surfaces (figures 2–5), which mostly exhibited internal rupture. Long fingers formed around the edges of the drop, which then detached as satellite droplets, especially as the droplet receded (figure 6c–e), leaving only an array of small droplets on the surface. Hole formation on the AKD sample, even though it was rough ($R_a = 1.25 \mu m$), was minimal at $Re = 5800$ and $11600$, but occurred at $Re = 17400$. In contrast, the rough wax surface, which had almost the same roughness, showed extensive hole formation at all $Re$. This behaviour suggests that hole formation is suppressed on superhydrophobic surfaces. Films on AKD surfaces become unstable through continuous shedding of satellite droplets from their edges during retraction (figure 6e), whereas on the other surfaces tested, fragmentation of liquid films commenced with growth of internal holes.

High liquid–solid contact angles are also encountered during the impact of molten metal droplets, which have high surface tension. Mehdizadeh (2002) photographed the impact of $600 \mu m$ diameter molten tin droplets on a mirror-polished stainless steel surface held at a temperature above that of the droplet melting point, so that impact was isothermal and droplets did not solidify during impact. Aziz & Chandra (2000) measured dynamic liquid–solid contact angles for molten tin on stainless steel and found both advancing and receding contact angles to be near $140^\circ$. Droplet impact velocity was varied between 10 and $30 \text{m s}^{-1}$ giving $Re$ values from $2.3 \times 10^4$ to $6.9 \times 10^4$. Figure 7 shows photographs taken by Mehdizadeh (2002) at three different impact velocities: two images are
Figure 6. Impact of 580μm diameter water droplets with different Re on superhydrophobic AKD surfaces.

shown at each velocity, showing droplets at maximum extension (figure 7a) and in the final configuration (figure 7b). Hole formation can be seen (figure 7) at all impact velocities tested, eventually leading to internal rupture of droplets.

Water films in the present experiments were approximately 20μm thick and liable to get punctured by air bubbles trapped underneath, or surface protuberances, such as dust particles. Once a hole is nucleated, it may grow or
Figure 7. Impact of 600 μm diameter molten tin droplets with different $Re$ on polished stainless steel surfaces (data of Mehdizadeh (2002), reproduced with permission). Surface temperature $= 240^\circ C$ ensures that the impacting droplet does not solidify during spreading.

close, depending upon whether the liquid–solid contact line around the periphery of the hole recedes or advances. Which of these two outcomes transpires can be predicted using a stability analysis (Sharma & Ruckenstein 1989). If holes expand, the film will eventually disintegrate, whereas if they spontaneously heal, the film will remain stable.

Sharma & Ruckenstein (1989) developed a stability analysis to predict the stability of a thin liquid layer on a solid surface. The surface energy of a liquid film of surface area $A$, whose upper and lower surfaces are in contact with air and a solid substrate, respectively, is:

$$F_{\text{film}} = (\gamma_{lg} + \gamma_{sl})A,$$

where $\gamma$ represents the interfacial tension between the two interfaces indicated by the subscripts (l, liquid; g, gas; s, solid). If the film is punctured with a hole that has radius $r_1$ at the lower surface in contact with the solid substrate and $r_2$ on the upper surface in contact with air (figure 8a), its energy becomes:

$$F_{\text{hole}} = \gamma_{lg}(A - \pi r_2^2) + \gamma_{sl}(A - \pi r_1^2) + S\gamma_{lg} + \pi r_1^2\gamma_{sg},$$

where $S$ is the area of the liquid meniscus lying between $r_1$ and $r_2$. The change in free energy owing to the formation of the hole is:

$$\Delta F = F_{\text{hole}} - F_{\text{film}} = -\pi r_2^2\gamma_{lg} + \pi r_1^2(\gamma_{sg} - \gamma_{sl}) + S\gamma_{lg}.$$

Using Young’s equation

$$\gamma_{sg} - \gamma_{sl} = \gamma_{lg} \cos \theta,$$
we have an expression for the free energy change:

$$\Delta F = \Delta \gamma \left( S - \pi r_2^2 + \pi r_1^2 \cos \theta \right).$$  \hspace{1cm} (3.5)

The stability of the film depends on the magnitude of $\Delta F$: if $\Delta F < 0$, the hole reduces the energy of the film and will grow; if $\Delta F > 0$, the hole will spontaneously close and the film will remain intact. Therefore, for film stability, we require $S$ to be large enough to outweigh the negative term on the right-hand side of equation (3.5). *Sharma & Ruckenstein* (1989) derived the following expression for the surface area $S$ of the liquid meniscus by solving the Young–Laplace equation of capillarity:

$$S = \frac{1}{2} \pi r_1^2 \left\{ (1 + \cos^2 \theta) \sinh \left( \frac{2h}{r_1} \frac{1}{\sin \theta} \right) + 2 \cos \theta \left[ \cosh \left( \frac{2h}{r_1} \frac{1}{\sin \theta} \right) - 1 \right] + 2 \frac{h}{r_1} \sin \theta \right\}. \hspace{1cm} (3.6)$$

$r_2$ was given as:

$$r_2 = r_1 \left[ \cosh \left( \frac{h}{r_1} \frac{1}{\sin \theta} \right) + \sinh \left( \frac{h}{r_1} \frac{1}{\sin \theta} \right) \cos \theta \right]. \hspace{1cm} (3.7)$$

$\Delta F$ is a function of $h/r_1$ and $\theta$. In general, though, $S$ is large when $\theta$ approaches either 0° or 180°, as shown in figure 8a,c. Both high and low contact angles produce a meniscus with large surface area when a hole is created in the liquid film. The large surface energy associated with this exposed area will inhibit further expansion of the hole, making it close. In contrast, intermediate contact angles ($\theta \sim 90^\circ$) produce a meniscus with small surface area (figure 8b), rendering such a hole energetically favourable and making it grow.
The boundary between film stability and instability can be determined by setting $\Delta F = 0$ in equation (3.5), combined with equations (3.6) and (3.7), to obtain the critical film thickness, $h_c$, below which the film would be unstable when a hole of radius $r_1$ was created:

$$2 \left( \frac{h_c}{r_1} \right) \frac{1}{\sin \theta} = 1 + \frac{(1 - \cos \theta)^2}{\sin^2 \theta} \left[ \cosh \left( 2 \frac{h_c}{r_1 \sin \theta} \right) - \sinh \left( 2 \frac{h_c}{r_1 \sin \theta} \right) \right].$$  (3.8)

The ratio $h_c/r_1$ is an implicit function of the contact angle, $\theta$.

To apply this stability criterion to droplet impact, we need a way of estimating film thickness $h$ formed following impact. An estimate of $h$ may be obtained by using mass conservation of the droplet before and after impact when it resembles a thin disc (figure 2b). The mass of the droplet when equated before and after impact (at maximum spread) yields:

$$\rho \frac{\pi D_o^3}{6} = \rho \frac{\pi }{4} D_{\text{max}}^2 h,$$

where $\rho$ is the density of the film liquid, $D_o$ is the diameter of droplet before impact and $D_{\text{max}}$ is the diameter of the film at maximum spread of the droplet. Simplifying equation (3.9) gives:

$$h = \frac{2D_o}{3 \xi_{\text{max}}^2},$$  (3.10)

where $\xi_{\text{max}} = D_{\text{max}}/D_o$ is the maximum spread factor of the impacting droplet. To calculate $\xi_{\text{max}}$, an energy balance model developed by Pasandideh-Fard et al. (1996) is used. According to this model, $\xi_{\text{max}}$ is given as:

$$\xi_{\text{max}} = \frac{D_{\text{max}}}{D_o} \sqrt{\frac{We + 12}{3(1 - \cos \theta_a) + 4(We/\sqrt{Re})}},$$  (3.11)

where $\theta_a$ is the advancing contact angle formed by droplet liquid with the solid surface. Pasandideh-Fard et al. (1996) further showed that equation (3.11) may be simplified as:

$$\xi_{\text{max}} = 0.5 Re^{0.25},$$  (3.12)

if $We \gg \sqrt{Re}$. This condition is satisfied in the present experiments (table 2). The predictions for $\xi_{\text{max}}$ from equation (3.12) were compared with the corresponding values measured from the photographs. Once $\xi_{\text{max}}$ has been calculated, $h$ can be estimated from equation (3.10). An alternative method of calculating an average value of $h$ directly from experiments was to measure $D_{\text{max}}$ from photographs and setting:

$$h_{\text{exp}} = \frac{2D_o}{3 \xi_{\text{max,exp}}^2},$$  (3.13)

where $\xi_{\text{max,exp}} = D_{\text{max,exp}}/D_o$.

Table 2 compares predicted (from equations (3.10) and (3.12)) and measured (equation (3.13)) values of spread factors and film thicknesses and shows that they predict film thickness quite well. Film thickness decreases as impact $Re$ is increased.
Table 2. Comparison of measured spread factors and film thicknesses with predictions.

| Re     | We  | maximum spread factor $\xi_{\text{max}}$ | film thickness $h$ (μm) |
|--------|-----|-----------------------------------------|------------------------|
|        |     | model | experiment | model | experiment |
| 5800   | 800 | 4.4   | 3.9        | 21    | 27         |
| 11600  | 3200| 5.2   | 4.9        | 15    | 17         |
| 17400  | 7200| 5.8   | 5.6        | 12    | 13         |

The calculated film thickness may be used in the criteria for film stability (equation (3.8)), written in the dimensionless form as:

\[
2 \left( \frac{h_c^*}{r_1^*} \right) \frac{1}{\sin \theta_r} = 1 + \frac{(1 - \cos \theta_r)^2}{\sin^2 \theta_r} \left[ \cosh \left( 2 \frac{h_c^*}{r_1^*} \frac{1}{\sin \theta_r} \right) - \sinh \left( 2 \frac{h_c^*}{r_1^*} \frac{1}{\sin \theta_r} \right) \right],
\]

where $h_c^* = h_c/D_o$ and $r_1^* = r_1/D_o$. Note that the receding contact angle $\theta_r$ is used in the above equation because holes in thin films have been shown to expand at a rate which is critically dependent on $\theta_r$ (Redon et al. 1991). Using equation (3.10), we can write:

\[
h^* = \frac{2}{3 \xi_{\text{max}}^2},
\]

where $h^* = h/D_o$. Substituting $\xi_{\text{max}}$ from equation (3.12) into equation (3.15) gives:

\[
h^* = \frac{8}{3 \sqrt{Re}}.
\]

Setting $h^* = h_c^*$ and substituting equation (3.16) into equation (3.14), the criterion for film rupture during droplet impact is:

\[
\left( \frac{16}{3 r_1^* \sin \theta_r} \right) \frac{1}{\sqrt{Re_c}} = 1 + \frac{(1 - \cos \theta_r)^2}{\sin^2 \theta_r} \left[ \cosh \left( \frac{16}{3 r_1^* \sin \theta_r} \frac{1}{\sqrt{Re_c}} \right) - \sinh \left( \frac{16}{3 r_1^* \sin \theta_r} \frac{1}{\sqrt{Re_c}} \right) \right],
\]

where $Re_c$ is the critical Reynolds number of impact at which the film ruptures through formation of holes. If $Re > Re_c$, film rupture is likely to occur. $Re_c$ is a function of two variables: receding contact angle, $\theta_r$, and hole radius, $r_1^*$. For a given liquid, receding contact angle depends on the substrate: measured values are listed in table 1. The initial hole radius is more difficult to determine since only rough, order-of-magnitude estimates can be made from photographs. Holes appear to be triggered by air bubbles trapped at the liquid–solid interface and increasing surface roughness resulted in larger holes, possibly because of more air entrapment in surface crevices.

Figure 9 shows the variation of $Re_c$ with $\theta_r$, for three different values of $r_1^*$. Films are always stable for very large or very small contact angle, which results in a very large hole surface (figure 8), and are most likely to rupture at $\theta_r = 118^\circ$. 

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Figure 9. Critical Reynolds number $Re_c$ of impact, above which films would rupture, as a function of $\theta_r$ for different values of dimensionless hole radius, $r_1^*$. Experimental data: square, glass; circle, Plexiglas; inverted triangle, smooth wax and rough wax; diamond, AKD; triangle, molten tin. Bold line, $r_1^* = 0.05$; dashed line, $r_1^* = 0.1$; dotted line, $r_1^* = 0.15$.

which corresponds to the minimum on each $r_1^*$ curve. Experimental data for the four surfaces used in our study, glass, Plexiglas, wax and AKD, are marked on the figure. Since $\theta_r$ for smooth and rough wax surfaces was approximately the same, their data points overlap. In addition, the experimental data of Mehdizadeh (2002) obtained from the impact of molten tin droplets on stainless steel surfaces are also shown. Solid symbols in figure 9 represent cases in which liquid films ruptured, whereas hollow symbols show stable films.

Droplets on glass ($\theta_r = 20^\circ$) and Plexiglas ($\theta_r = 40^\circ$) surfaces did not become unstable until $Re = 1.74 \times 10^4$ and $1.16 \times 10^4$, respectively (figures 2 and 3), whereas those on smooth wax ($\theta_r = 84^\circ$) and rough wax ($\theta_r = 85^\circ$) were unstable at all $Re$ investigated (figures 4 and 5). This behaviour is best described by using the line for $r_1^* = 0.1$ to separate stable (below the line) and unstable (above the line) regions. Assuming $r_1^* = 0.15$ suggests that droplets with even the lowest impact velocity would rupture on the Plexiglas surface, which we did not observe. This would imply that holes with a diameter of 116 $\mu$m (corresponding to $r_1^* = 0.1$), but not as large as 174 $\mu$m (corresponding to $r_1^* = 0.15$), formed during impact. Experimental observations of the earliest stages when holes were visible appeared to confirm this observation: holes that were visible were approximately 100 $\mu$m in diameter. The model is also able to predict hole formation seen in the photographs taken by Mehdizadeh (2002) of molten tin droplets impacting on
stainless steel surfaces ($\theta_r = 140^\circ$). The holes seen in the films (figure 7) were approximately 200 $\mu$m in diameter, corresponding to $r^*_1 = 0.15$, and therefore $Re \gg Re_c$ in all cases.

Film rupture becomes less likely once $\theta_r > 118^\circ$, according to the model. We observed that there was much less film rupture (figure 6) on the superhydrophobic AKD surface ($\theta_r = 147^\circ$) than on the rough wax ($\theta_r = 85^\circ$), even though both had surface roughness of the same order of magnitude. The line $r^*_1 = 0.05$ better predicts the behaviour of AKD surfaces in figure 9. However, the model cannot make predictions that are quantitatively more accurate since it is very sensitive to the value of contact angle and initial hole diameter. It does predict the trend that film rupture is suppressed on very hydrophobic surfaces.

4. Conclusions

We investigated the rupture of thin liquid films formed during the impact of 580 $\mu$m water droplets onto a solid substrate for a range of impact velocities (10–30 m s$^{-1}$). Four different substrates, of glass, Plexiglas, wax and superhydrophobic AKD, with equilibrium liquid–solid contact angles of 47°, 71°, 105° and 164°, respectively, were used. Two different wax surfaces were prepared with different roughnesses. Photographs of droplet impact showed that films ruptured through the formation of holes as they became thinner owing to an increase in impact velocity. The impact velocity at which rupture occurred was found to depend on the liquid–solid contact angle: films with low (glass) or high (AKD) contact angles ruptured only at the highest impact velocity tested (30 m s$^{-1}$), whereas films with intermediate contact angles (wax) ruptured at all impact velocities. A thermodynamic stability analysis combined with a droplet impact model was used to explain film break-up behaviour by showing that films remain stable at very small or very large contact angles, but are unstable in between. Film rupture was greatly promoted by increasing surface roughness.

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References

Allen, R. F. 1975 The role of surface tension in splashing. J. Colloid Interf. Sci. 51, 350–351. (doi:10.1016/0021-9797(75)90126-5)

Aziz, S. D. & Chandra, S. 2000 Impact, recoil and splashing of molten metal droplets. Int. J. Heat Mass Transf. 43, 2841–2857. (doi:10.1016/S0017-9310(99)00350-6)

Cheng, S. & Chandra, S. 2003 A pneumatic droplet-on-demand generator. Exp. Fluids 34, 755–762. (doi:10.1007/s00348-003-0629-6)

Deegan, R. D., Brunet, P. & Eggers, J. 2008 Complexities of splashing. Nonlinearity, 21, C1–C11. (doi:10.1088/0951-7715/21/1/C01)

Dhiman, R. & Chandra, S. 2005 Freezing-induced splashing during impact of molten metal droplets with high Weber numbers. Int. J. Heat Mass Transf. 48, 5625–5638. (doi:10.1016/j.ijheatmasstransfer.2005.05.044)
Rupture of droplet impact films

Dhiman, R. & Chandra, S. 2008 Rupture of radially-spreading liquid films. *Phys. Fluids* **20**, 092104. (doi:10.1063/1.2978186)

Johnson, R. E. & Dettre, R. H. 1993 In *Wettability* (ed. J. C. Berg), pp. 10–13. New York, NY: M. Dekker.

Khesghi, H. S. & Scriven, L. E. 1991 Dewetting: nucleation and growth of dry regions. *Chem. Eng. Sci.* **46**, 519–526. (doi:10.1016/0009-2509(91)80012-N)

Kim, H. Y., Feng, Z. C. & Chun, J. H. 2000 Instability of a liquid jet emerging from a droplet upon collision with a solid surface. *Phys. Fluids* **12**, 531–541. (doi:10.1063/1.870259)

Lesser, M. B. & Field, J. E. 1983 The impact of compressible liquids. *Annu. Rev. Fluid Mech.* **15**, 97–122. (doi:10.1146/annurev.fl.15.010183.000525)

Mehdizadeh, N. Z. 2002 Droplet impact dynamics: effect of varying substrate temperature, roughness and droplet velocity. PhD thesis, University of Toronto, Toronto, Ontario, Canada.

Mehdizadeh, N. Z. & Chandra, S. 2006 Boiling during high velocity impact of water droplets on a hot stainless steel surface. *Proc. R. Soc. A* **462**, 3115–3131. (doi:10.1098/rspa.2006.1722)

Mehdizadeh, N. Z., Raessi, M., Chandra, S. & Mostaghimi, J. 2004a Effect of substrate temperature on splashing of molten tin droplets. *J. Heat Transf.* **126**, 445–452. (doi:10.1115/1.1737778)

Mehdizadeh, N. Z., Chandra, S. & Mostaghimi, J. 2004b Formation of fingers around the edges of a drop hitting a metal plate with high velocity. *J. Fluid Mech.* **510**, 353–373. (doi:10.1017/S0022112004009310)

Mohammadi, R., Wassink, J. & Amirfazli, A. 2004 Effect of surfactants on wetting of superhydrophobic surfaces. *Langmuir* **20**, 9657–9662. (doi:10.1021/la049268k)

Padday, J. F. 1970 Cohesive properties of thin films of liquids adhering to a solid surface. *Spec. Discuss. Faraday Soc.* **1**, 64–74. (doi:10.1039/sd9700100064)

Pasandideh-Fard, M., Qaio, Y. M., Chandra, S. & Mostaghimi, J. 1996 Capillary effects during droplet impact on a solid surface. *Phys. Fluids* **8**, 650–659. (doi:10.1063/1.868850)

Pepper, R. E., Courbin, L. & Stone, H. A. 2008 Splashing on elastic membranes: the importance of early-time dynamics. *Phys. Fluids* **20**, 082103. (doi:10.1063/1.2969755)

Redon, C., Brochard-Wyart, F. & Rondelez, F. 1991 Dynamics of dewetting. *Phys. Rev. Lett.* **66**, 715–718. (doi:10.1103/PhysRevLett.66.715)

Rein, M. 1993 Phenomena of liquid drop impact on solid and liquid surfaces. *Fluid Dyn. Res.* **12**, 61–93. (doi:10.1016/0169-5983(93)90106-K)

Sharma, A. & Ruckenstein, E. 1989 Dewetting of solids by the formation of holes in macroscopic liquid films. *J. Colloid Interf. Sci.* **133**, 358–368. (doi:10.1016/S0021-9797(89)80044-X)

Stow, C. D. & Hadfield, M. G. 1981 An experimental investigation of fluid flow resulting from the impact of a water drop with an unyielding dry surface. *Proc. R. Soc. Lond. A* **373**, 419–441. (doi:10.1098/rspa.1981.0002)

Taylor, G. I. & Michael, D. H. 1973 On making holes in a sheet of fluid. *J. Fluid Mech.* **58**, 625–639. (doi:10.1017/S0022112073002375)

Xu, L., Zhang, W. W. & Nagel, S. R. 2005 Drop splashing on a dry smooth surface. *Phys. Rev. Lett.* **94**, 184505. (doi:10.1103/PhysRevLett.94.184505)

Xu, L., Barcos, L. & Nagel, S. R. 2007 Splashing of liquids: interplay of surface roughness with surrounding gas. *Phys. Rev. E* **76**, 066311. (doi:10.1103/PhysRevE.76.066311)

Yarin, A. L. 2006 Drop impact dynamics: splashing, spreading, receding, bouncing... *Annu. Rev. Fluid Mech.* **38**, 159–192. (doi:10.1146/annurev.fluid.38.050304.092144)