Wettability-Dependent Breakup of Thin Films Formed by Droplet Impact

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Abstract

The effect of droplet-substrate wettability on droplet splashing and breakup during impact at high velocities was studied experimentally. 0.6 mm diameter water droplets were produced by a pneumatic drop-on-demand droplet generator. High velocities of impact (V_o=10-30 m/s) were achieved by mounting the substrate on the rim of a rotating flywheel, giving impact Reynolds and Weber numbers between 6000-18000 and 800-7300, respectively. Droplet-substrate wettability was varied over a wide range, from hydrophilic to superhydrophobic conditions, by changing the material of the substrate (glass, Plexiglas, wax, and Alkylketene Dimer (AKD)). Both smooth and rough wax surfaces were tested. Photographs of impact showed that the droplets spread into a thin film at maximum extension, followed by a receding phase. However, as the impact velocity increased and film thickness decreased, films became unstable and ruptured internally through formation of holes. Furthermore, the impact velocity at which rupture occurred was found to first decrease and then increase with the liquid-solid contact angle θ: on glass (θ=47°), rupture was observed only at the highest impact velocity, whereas on wax (θ=105°), rupture occurred at all impact velocities tested. On the AKD surface (θ=164°), films did not rupture until V_o=30 m/s, but showed significant splashing from edges at all V_o, especially during the receding phase. On the rough wax surfaces, extensive film rupture occurred at all impact velocities with the formation of a large number of holes. The above results suggest that during high velocity impact, internal rupture through formation of holes may be the dominant mechanism preventing droplets from complete deposition on a solid surface.

Introduction

Many technological applications involve impact of liquid droplets on solid surfaces, examples being thermal spray coating, spray painting, and pesticide spray on plant leaves. An important parameter that determines the effectiveness of such applications is the deposition efficiency, which is the fraction of sprayed material that remains deposited on the substrate. During impact, the droplet may splash, causing material wastage and environmental pollution. Many studies [1-7] have examined splashing of droplets during their impact with solid surfaces. However, most studies have focused on the splashing from the spreading edge of an impacting droplet as a result of surface roughness [1], fluid instabilities [2-4], surrounding gas pressure [5], substrate elasticity [6], or solidification [7], in case the droplet solidifies during impact. Photographs [4] of water droplets impacting a stainless steel surface at 40 m/s revealed that the droplet film becomes so thin that it ruptures internally through formation of several holes. In such a case, the droplet film may no longer adhere to the substrate and cause material wastage.

Formation of holes in stationary thin films has been studied by many researchers [8-12]. Padday [8] 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 and Michael [9] 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 while smaller ones heal. Sharma and Ruckenstein [10] did a thermodynamic analysis on hole formation by considering the free energy change associated with the transition of film from an intact state to one when it had developed a hole inside. Their predictions about the critical film thickness matched well with the observations of Padday [8]. Kheshgi and Scriven [11] 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*. [12] studied the rate of hole growth and found that their velocity varies as θ^3, independently of film thickness. Recently, Dhiman and Chandra [13] investigated hole formation is radially-spreading films produced by the normal impact of a water jet on a solid surface. Combining a simple mathematical model to predict film thickness together with the thermo-

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dynamic analysis of Sharma and Ruckenstein [10], they developed a criterion to predict film rupture by calculating a critical Reynolds number for the impacting jet.

In this paper, our objective was to investigate the rupture of thin films formed during droplet impact and to study the effect of liquid-solid contact angle on film rupture. We made 600 μm water droplets impact a solid surface at high velocities (10-30 m/s) by mounting it on the rim of a rotating flywheel. Droplet-substrate wettability was the key parameter varied, from hydrophilic to superhydrophobic, by changing the material of the substrate.

Materials and Methods

Figure 1 shows a schematic of the experimental apparatus used. It consists of a pneumatic drop-on-demand droplet generator that produced uniform size water droplets of 600 μm diameter repeatably. The substrate was mounted on the rim of a flywheel whose rotational speed was controlled precisely by a digital motion controller. To hit a falling droplet with a rotating substrate and photograph its impact, a timing circuit was built to synchronize the release of the droplet and triggering of a fast-shutter CCD camera and flash with the position of the substrate. A detailed description of the apparatus is given elsewhere [4]. Substrates of different materials (glass, Plexiglas, wax, and Alkylketene Dimer (AKD)) were used to study the effect of liquid-solid contact angle on the rupture of droplet impact films. Both rough and smooth wax surfaces were tested. Each substrate was characterized by measuring its surface roughness and contact angles formed with water droplets. Roughness was measured with the help of an interferometric microscope at five different locations on each surface and average of those five values was taken to obtain a representative value for the entire surface. Equilibrium, advancing, and receding contact angles formed by water droplets on each surface were measured following the procedure described by Johnson and Dettre [14]. Table 1 lists the surface roughness and contact angles of each substrate tested in the present study.

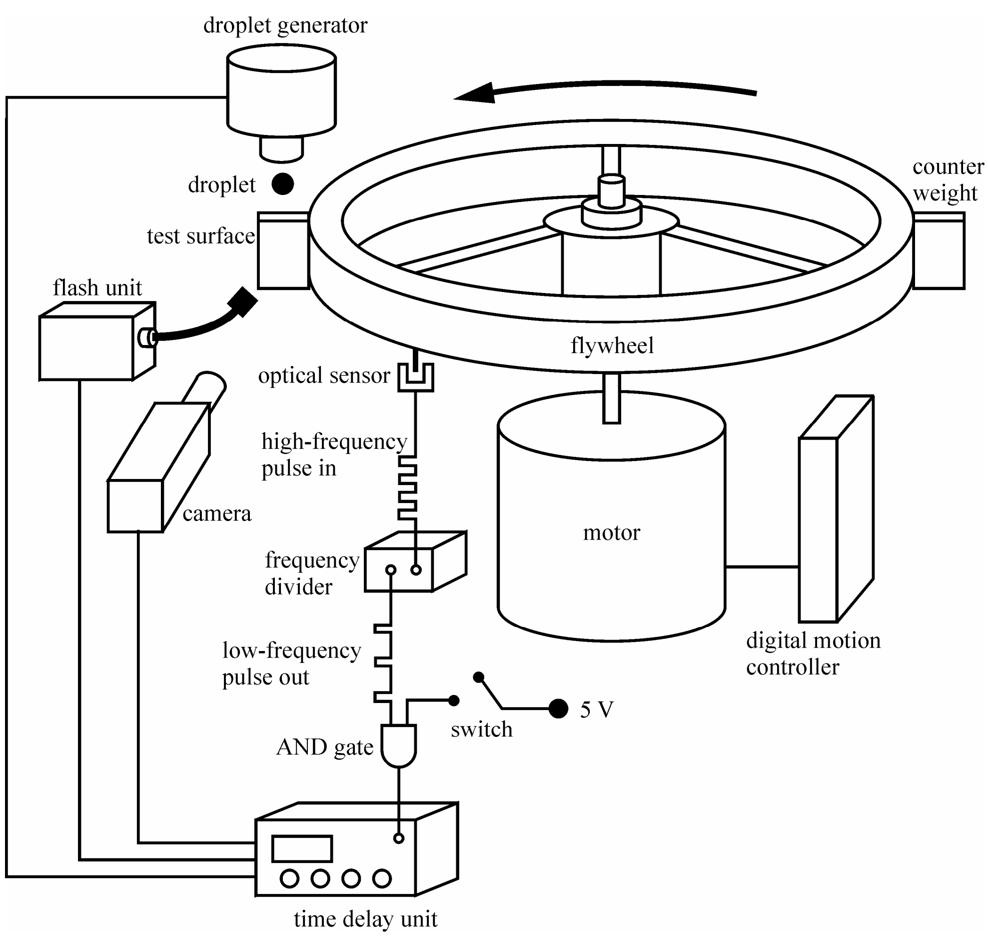

Figure 1. A schematic diagram of the experimental apparatus

Table 1. Characterization of the different substrates used

Surface	Equilibrium contact angle (θ), deg.	Advancing contact angle (θ_a), deg.	Receding contact angle (θ_r) , deg.	Roughness (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	-	164 [14]	147 [14]	1.25±0.17 [14]

Results and Discussion

Figure 2 shows photographs of water droplets during impact on different substrates at different impact Reynolds numbers ($Re = \rho V_o D_o/\mu$, where ρ, μ, V_o and D_o are droplet liquid density, dynamic viscosity, impact velocity and diameter, respectively) tested in this study. Each column shows photographs at one of the three impact *Re*, whereas each row shows photographs at one of the four substrate materials investigated.

As seen in Fig. 2, films remain stable on glass at *Re* = 6000 & 12000, but rupture at *Re* = 18000 through formation of several holes, which continue to grow afterwards rendering the film unstable. Changing the wettability of the solid changed the rupture behavior: on a Plexiglas surface, which had a contact angle higher than glass (θ for glass = 47^o, Plexiglas = 71^o), droplets impacting at *Re* = 12000 & 18000 both now become unstable and rupture. Impact on smooth wax surfaces (Fig. 2, third row), which had water contact angles ($\theta = 105^o$) much larger than both glass and Plexiglas, showed rupture at all three *Re*. This behavior suggests that the tendency to hole formation increases as the contact angle increases. In addition, holes on smooth wax surfaces are much larger as compared to those on glass and Plexiglas surfaces possibly because they grow rapidly due to their high contact angle. This behavior is in conformity with the observations of Redon *et al.* [12], which showed that the rate at which holes grow increases as θ^3. The rapid growth of holes causes the covered area (or, the deposition) of water drops on wax surfaces to be the least as compared to the glass and Plexiglas surfaces.

Changing the wettability further by means of a superhydrophobic surface (AKD) revealed very different rupture behavior (see Fig. 2 last row). The AKD surface had the highest liquid-solid contact angle ($\theta_a = 164^o$, $\theta_r = 147^o$) amongst all surfaces tested. Hole formation on the AKD surface, even though it was rough (R_a= 1.25 µm), did not occur until *Re* = 18000; instead, at all *Re* droplet films exhibited extensive splashing from the edges, unlike all other surfaces, especially during the receding phase leaving no deposition on the surface. The suppression of hole formation on the AKD surface suggests that the tendency to film rupture decreases at high values of the liquid-solid contact angle.

The above change in film rupture behavior with the liquid-solid contact angle was demonstrated by Dhiman and Chandra [13] in radially-spreading films produced by the normal impact of a water jet on a solid surface. As shown in Fig. 3, when the contact angle is very small or very large, the surface area *S* of the hole is also large. The surface energy of the system increases, and the holes close spontaneously. On the other hand, at intermediate values of contact angle, *S* is small and the total energy of the system is reduced by formation of the hole: it grows larger, rendering the film unstable. Using a thermodynamic analysis [10], Dhiman and Chandra [13] showed that films would be stable at very small or very large contact angles, but unstable in between, similar to the behavior observed in the present experiments.

The dynamics of film rupture on the rough wax surface (R_a= 1.98 µm) was quite different. This is shown in Fig. 4. A large number of holes form inside water films along with the formation of long fingers at their periphery (see Fig. 4*a*). The holes grow until their boundaries meet those of the neighboring holes (see Fig. 4*b*) and the equilibrium state corresponds to a number of satellite droplets resting on the surface (see Fig. 4*c*). The diameter D_{max} of the films at maximum extension and the number of holes formed increased with *Re*. However, compared with glass, Plexiglas, and smooth wax surfaces, D_{max} was about 25% smaller. This behavior might be due to the material loss through splashing triggered by surface roughness before the films arrive at their maximum spread. Hole formation on the rough wax surface is quite different from the AKD surface although both had surface roughness values of the same order: whereas on the rough wax surface, a large number of holes form in the film at all *Re*, on the AKD surface, only few holes form at *Re* = 18000.

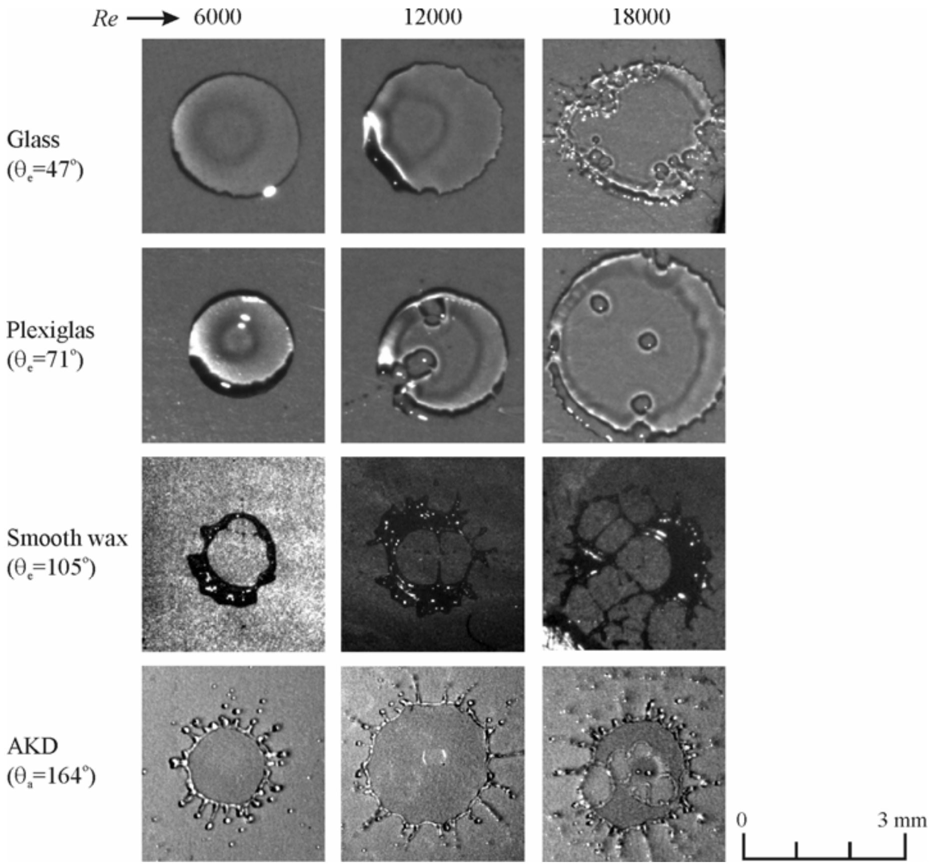

Figure 2. Photographs of water droplets during impact on different substrates

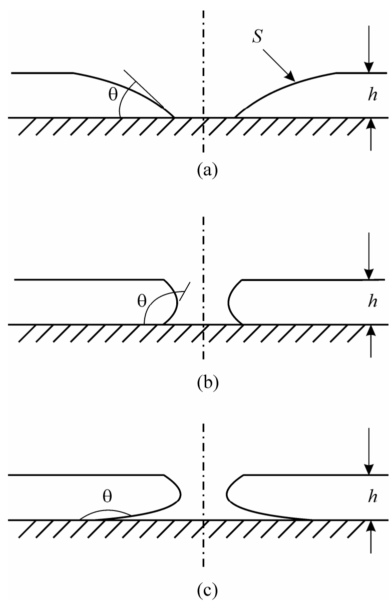

Figure 3. Schematic of hole profile at different contact angles: (a) small θ, large S, film stable, (b) intermediate θ, small S, film unstable, (c) large θ, large S, film stable. h is film thickness.

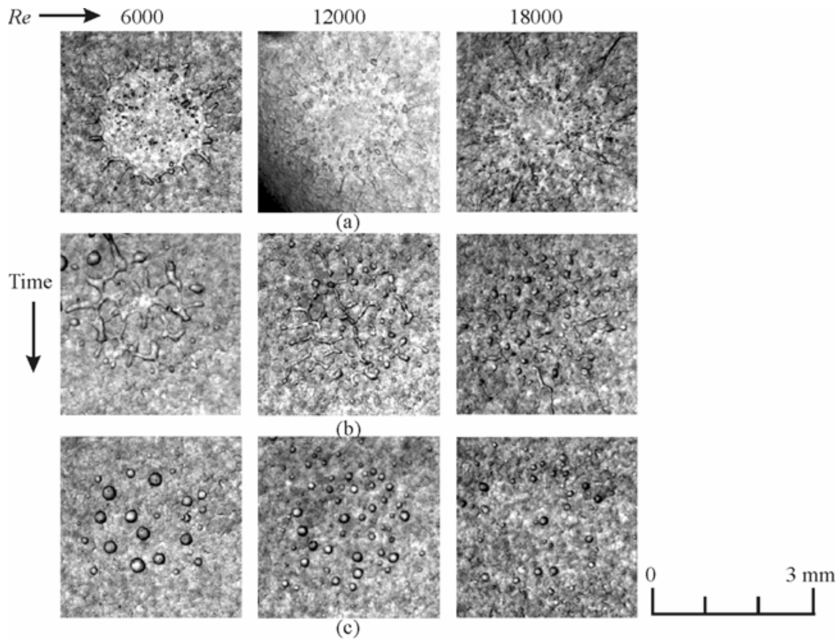

Figure 4. Photographs of water droplets during impact on rough wax substrates

Conclusions

We investigated the breakup of liquid films produced by the high-speed impact of water droplets onto a solid substrate. The droplet diameter before impact was 600 μm and the droplet impact velocity was varied from 10-30 m/s. Droplet-substrate wettability was varied over a wide range by changing the material of the solid surface (glass, Plexiglas, wax, and AKD, with equilibrium liquid-solid contact angles of 47°, 71°, 105°, and 164°, respectively). Two different wax surfaces were prepared with different roughness. Photographs of impact showed that the films ruptured through formation of holes as they became thinner due to the increase in the impact velocity. However, the velocity at which films ruptured first decreased and then increased with the contact angle. Film rupture was also promoted by increasing surface roughness. The results suggest that internal film rupture through formation of holes may be the dominant mechanism preventing complete deposition during high-speed droplet impact.

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