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The impalement of water drops impinging onto hydrophobic/superhydrophobic graphite surfaces: the role of dynamic pressure, hammer pressure and liquid penetration time

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ABSTRACT

Droplet impingement experiments at low Weber numbers were conducted by digitizing silhouettes of impacting water drops onto unlike graphite substrates, typified by different advancing water contact angles (θ_a): 140 and 160°. The relaxation of wetting diameter, dynamic contact angle, and drop shapes were measured. The purpose was to carefully investigate the phenomenology and possible causes of the failure of the superhydrophobicity. During impact and spreading phases, all the drops impinging onto both graphite substrates showed a similar behavior. Then, after an initial free recoil, drops impinging at lower impact velocities onto graphite substrates characterized by $\theta_a = 140°$ clearly exhibited time intervals in which the wetting diameter appeared to be almost constant. The duration of this pinned phase was observed decreasing with increasing the impact height and almost completely disappearing for drops impinging at higher impact velocities. This behavior has never been reported before, and, contrariwise, water droplets impinging at lower impact velocities onto hydrophobic and superhydrophobic surfaces have been generally observed more freely retracting, and ultimately rebounding, compared to drops impacting at higher velocities. In the present study, this latter behavior was recorded just for drops impinging onto graphite surfaces characterized by $\theta_a = 160°$. A theoretical description of the experimental results was proposed, specifically investigating the role of dynamic pressure, hammer pressure and liquid penetration time during the impact, spreading and recoil stages.

1. Introduction

In the last decade, surfaces that could mimic the hydrorepellency of lotus leaves [1] have been the subject of an intensive research due to their decisive role in numerous applications, such as self-cleaning, anti-contaminating and anti-sticking applications [2–6]. Generally, this distinctive wetting behavior is driven by chemical and morphological characteristics of the surface. Various studies have been conducted in manufacturing, designing, modeling and testing the superhydrophobicity of textured and randomly rough surfaces in static or quasi-static processes [7,8]. However, it has been observed that, for sundry applications which involved dynamic and not stationary cases, such as in the presence of mechanical vibrations [9], strong decrease of the drop volume [10–12], external pressures [13], or drop impact [14–32], the water repellency of these peculiar surfaces could be partially or totally lost.

Many applications for superhydrophobic surfaces involve impinging drops scenarios. Therefore, during the past several years, a great attention has been placed in exploring the properties of these surfaces when used with impacting droplets [14–35]. Specifically, it has been reported that, in these cases, the main cause of the failure of the hydrorepellency is due to a partial or total liquid penetration into the rough or textured surfaces [14–32]. This imbibition leads to an increase of the adhesion of the droplet to the substrate and prevents the drop to detach from the surface.

For droplets impacting on microtextured surfaces, Bartolo et al. [16], and Jung and Bhushan [23] introduced similar semi-quantitative models describing two possible events: a completely wetting state, in which the liquid follows the topography of the solid surface (Wenzel state) and the drop is stuck on it; or a bouncing case, in which the drop rebounds on the substrate, touching just the top of the pillars of the textured surface, with air pockets remain trapped into the micropattern (Cassie–Baxter state). Afterwards, Reyssat et al. [17], reported that the wetting state could be

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just partial, with a water penetration noticeable only in a region near the impact zone. However, all these works [16,17,23] indicated uniquely the dynamic pressure as responsible for pushing the liquid interface into the microtextured surface.

In 2009 Deng et al. [24] pointed out that the use of the sole dynamic pressure could not entirely explain partial wetting states observed in several experiments and it could actually lead to an inaccurate design of superhydrophobic surfaces for impacting droplet applications. They, thus, introduced an additional wetting pressure, the effective water hammer pressure, which they regarded as the main cause of the water penetration at the impact stage. The imbibition during the spreading stage was considered, instead, as exclusive consequence of the dynamic pressure. Afterwards, this model was used by Kwak et al. [26], Chen et al. [30] and J.B. Lee and S.H. Lee [31].

However Deng et al. [24], as, previously, Bartolo et al. [16], Jung and Bhushan [23], and Reyssat et al. [17], did not take into account the important role of the penetration time during the drop spreading and receding phases. Impact, spreading and retracting stages are, in fact, highly dynamic processes. Therefore, modeling them just after static considerations could be inaccurate, especially for substrates with micro- (but not nano-) scaled roughnesses and drops impinging at low impact heights. Erroneously, e.g., it could be concluded that, with decreasing the impact velocity of drops impinging onto these microrough substrates, a decrease of the partially wetted area (and, eventually, a complete Cassie–Baxter state) should be observed. Contrariwise, as we illustrated in the present study for the first time, drops impacting at lower impinging heights could actually present a stronger sticky configuration.

The purpose of this work was to carefully investigate these wetting behaviors for drops impinging at low impact velocities, analyzing, especially, phenomenology and possible causes of the failure of the superhydrophobicity. We analyzed impacting, spreading, retracting, sticky and rebounding behaviors of water drops impinging onto two unlike graphite surfaces typified by different advancing water contact angles, 140 and 160°, respectively. The impact dynamics were measured in terms of the variation of wetting diameter, contact angle, and drop shape, digitalizing silhouettes of impinging droplets with a highly precise video enhanced image technology.

In this study we termed as "*pinning*" a triple line anchoring scenario in which surface pores in the nearest vicinity of the triple line are partially or totally filled with liquid. Generally speaking, the remaining pores under the drop could or could not be partially or totally filled with liquid. We termed "*impalement*" an anchoring scenario in which most of the partially or totally filled substrate pores could be detected closer to the impact region. It is worth to note that pinning configurations could be due also to random defects of the surface, such as dilute defects, impurities or ridges, so there could be pinning without any impalement [36]. Similarly, if just a specific zone far from the triple line is impaled, there could be impalement without any pinning. Finally, during the drop recoil, if the triple line (free to move) reaches the impaled zone, the liquid adhesion prevented the drop to move further and the detected pinning configuration is actually due to the impalement. The absence of drop rebound, i.e. a *sticky* configuration of the drop, could be caused by either pinning or impalement.

2. Experimental

2.1. Apparatus

A similar apparatus of the droplet impingement imaging system detailed in Wang et al. [37] was used to do the following: create silhouettes and top-view images of impacting and spreading drops; take video-images of the silhouettes; and digitize the images. The apparatus consisted of a halogen light source, a plano-convex lens system for generating a collimated beam, an objective lens, and two solid-state cameras, as schematized in Fig. 1. The video image system digitized the pictures into 128 lines × 128 pixels (for CCD_1, PhotonFocus DS1-D1024-160-CL-10) each of which was assigned a grey level value with an eight-bit resolution. The rate of image acquisition was 6770 images per second.

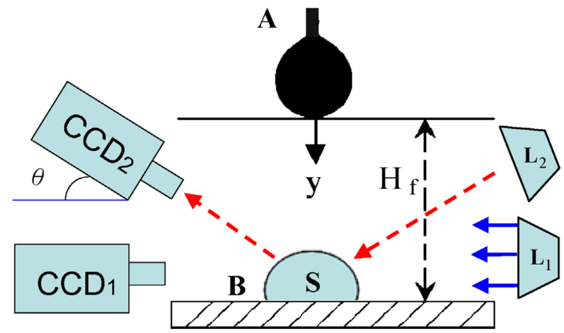

Fig. 1. Experimental design for studying the impingement behavior of a free falling droplet. A: needle; B: solid substrate; CCD: high-speed video camera; H_f: height of the droplet before falling; L: parallel light surface.

An edge detection routine was devised in the following way. The change in the grey level ranged from the black inside (0 level) to the bright outside (255 level) in a few pixels. The change was not a step increase from 0 to 255 but instead was continuous. The variation was symmetric at around 127.5, and therefore, the edge was defined at the position with 127.5 grey level.

Therefore, the drop edge was obtained by first interpolating a straight line between the two points that bound the grey level 127.5. The edge was defined as the x or y position that, for the interpolated line, corresponded to an intensity of 127.5. The uncertainty for the edge location in this work was around 0.2 pixels. The image forming system was calibrated by digitizing a stainless-steel ball with a known diameter of 2.498 ± 0.002 mm. The coordinates of the digitized sphere were processed to calibrate the average length between pixels along a row and along a column. The calibration procedure yielded values of 54.7 μm/pixel horizontally and 54.9 μm/pixel vertically for the side-view camera.

2.2. Materials

The water used was purified by a Millipore water purification system with a specific conductance of 0.056 μS/cm. Two distinctive graphite substrates were prepared following the methods illustrated by Hong et al. [38]. These graphite surface morphologies were characterized by different advancing water contact angles (θ_a): 140 and 160°. In the present work, these surfaces were termed as "G-140" and "G-160", respectively. The advancing contact angle was obtained from the location of the air/solid interface and a theoretical drop profile curve generated from the video-recorded silhouettes of the sessile drop and a best-fitting algorithm based on the Laplace equation, as fully described in Lin et al. [39]. This technique is capable of giving advancing contact angles measurements of ±0.2° precision.

The graphite sheets were purchased from NTC (IGS-743, 99.7%). The G-140 was obtained by peeling off the graphite superficial substrate with an adhesive tape. The Scanning Electron Microscope (SEM) analysis of this substrate revealed the existence of a micro-scaled roughness (Fig. 2 a, b).

The G-160 was obtained via ultrasonication in acetone for about 15 min of G-140. In addition to the micro-scaled structure shown by