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Water drop impingement on graphite substrates with random dilute defects

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ABSTRACT

Droplet impingement experiments for a wide range of Weber numbers were conducted by digitizing silhouettes of impacting water drops onto a tailored grinded graphite substrate, typified by randomly distributed cavities on a generally smooth surface. The aim was to investigate if the anchoring of the triple line, due to friction forces generated by dilute superficial defects, could be observed for drop impingement experiments. During the early inertially dominated spreading phase, the drops showed similar behaviors independently of the initial impact velocities. However, just after this phase, droplets impacting at low Weber numbers indeed exhibited this peculiar friction-induced pinned configuration.

1. Introduction

Impingement of liquid droplets onto dry surface substrates is a complex process that encompasses fluid dynamics, physics, and interfacial chemistry. Sundry parameters, such as drop size, impact velocity, liquid viscosity, surface tension, addiction of surfactants, and substrate morphology have been found to influence intensely the droplet impact and spreading processes [1–8].

After detaching from the tip of the needle, the droplet follows a free falling soft body dynamics. Immediately after impact, a thin film forms on the solid surface and the liquid expands horizontally; this is generally termed as the spreading phase [1,2]. The spreading phase ends as the drop achieves its maximum spreading diameter [6,9]. Generally, a droplet retraction or recoil starts right after this phase [1,2]. Spreading and recoil processes may also repeat and, for a droplet that has gained adequate energy before the impact, splash or rebound phenomena may occur [7]. However, the presence or the extent of a retraction phase and the subsequent three phases contact line movement depend on the initial impact velocity and specific characteristics of the substrate morphology [10–19].

Specifically, for drop impingement experiments, partial or total absence of recoil has been observed for two diverse reasons: drop pinning due to the imbibition of a rough substrate by the liquid [10–14]; or drop pinning generated by a strong adhesion between the water and a smooth substrate due to high superficial hydrophilicity or molecular reorientation [15–19]. The first pinning case has been reported especially for drops impacting onto superhydrophobic substrates typified by extremely rough surfaces, characterized by micro- or/and nano-roughnesses [12–14] or textured microholes or pillars [10–14]. For drops impinging onto these morphologies, the pinning has been recorded as caused by the droplet transition from Cassie (in which the liquid sits on top of the substrate roughness with air remaining trapped in the ditches and troughs under the droplet) to Wenzel (in which the interface between the drop and the substrate is homogeneous, and no air is trapped beneath the droplet) or partial Wenzel states. The contact area increase due to Wenzel or partial Wenzel states significantly raised the liquid–solid adhesion and forced the drop to be locally stuck on the substrate. This transition has been noted as enhanced with increasing the impact velocity [11,13,14]. For this pinning case, however, the drop has been observed as anchored to the substrate after an initial recoil phase.

For the second pinning case, instead, it has been reported a total absence of the retraction phase: the drop pinned just after the end of the spreading process. This case has been detected for drops impacting onto highly hydrophilic surfaces, as polished metals, clean glasses and quartz [15–19] or peculiar substrates in which exposed molecular groups were flexible enough to allow a reorientation when in contact with water [19]. For drops impinging onto these surfaces, the pinning has been observed as caused by the strong liquid–solid molecular interactions, which greatly increases the drop-substrate adhesion, preventing the triple line to recede.

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These pinning phenomena have been widely discussed in describing drop behaviors during slow wetting or dewetting processes [20–26]. Particularly, during evaporative and forced dewetting studies, in addition to the two mentioned above, it has been found a further possible cause for the drop to stick on the substrate: the three phases contact line was anchored to random dilute defects present on the substrate [21–25]. Specifically, the pinning was observed when the friction forces due to single defects, which fixed the triple line, overcame the capillary forces, which tended to bring the triple line to the equilibrium position. Up to a definite concentration of random defects, the pinning phase was more and more promoted. However, when the number of defects was sufficiently high, their collective effect along the triple line favored the depinning of the drop [24,25].

The purpose of this study was to investigate if on the latter pinning case, due to friction forces generated by dilute superficial defects, it could be observed during drop impingement experiments. We thus prepared a tailored graphite substrate, of which morphology was typified by randomly distributed deeper cavities on a generally smooth surface. Moreover, the chosen substrate was not highly hydrophilic, but characterized by an equilibrium contact angle around 90°. We analyzed impacting, spreading, retracting, and rebounding behaviors of water drops impinging onto this graphite surface. The investigation was conducted on a wide range of impact velocities, in order to bring out the correlations between inertial, viscous, friction, and capillarity forces. The impact dynamics were measured in terms of the variation of wetting diameter, contact angle, drop height and shape, and a special attention was given to the dynamics of the triple line.

2. Experimental

The water used was purified by a Millipore water purification system with a specific conductance of 0.056 μS/cm. The graphite substrate was prepared following the method illustrated by Hong et al. [27]. The graphite sheets were purchased from NTC (IGS-743, 99.7%) and, after a rinse in acetone, the equilibrium contact angle was measured [28] and determined as about θ_{eq} = 120°. The sheet was then ground by a 4000-grit sandpaper on a spinner. Subsequent to this grinding process, the θ_{eq} lowered to 90°. A scanning electron microscope (SEM) analysis was conducted on the sample and the substrate exhibited a predominantly smooth surface typified by the presence of random deeper cavities (Fig. 1).

A similar apparatus of the droplet impingement imaging system detailed in Wang et al. [17,18] was used in this work. Falling drops were generated from a stainless steel needle (gauge No. 31, ID = 0.13 mm) at 5.4, 13.1, 18.3, 26.0, 32.0 and 53.1 ± 0.2 mm above the substrates (impact heights, H_f). The positions of the top or bottom points were fitted by the equation of a free falling body to determine the time of droplet contact and the impact velocity [29]. The droplet size (D_0) was evaluated from the average drop mass of 20 droplets. The average droplet size was 2.14 ± 0.02 mm for all the measurement except for the run with H_f = 13.1 mm, in which D_0 = 2.09 mm. The calculated impact velocities were V_i = 0.326, 0.508, 0.599, 0.710, 0.789 and 1.020 m/s, respectively. The Weber numbers, a measure of the relative importance of the liquid inertia compared to its surface tension, defined as

$$W_e = \frac{\rho V_i^2 D_0}{\sigma} \tag{1}$$

where V_i and D_0 are the above defined impact velocity and initial drop diameter, while ρ and σ are the density and the surface tension of the liquid, were 3.16, 7.67, 10.67, 14.99, 18.51, and 30.94, respectively. All experiments were carried out at 25 ± 0.5 °C.

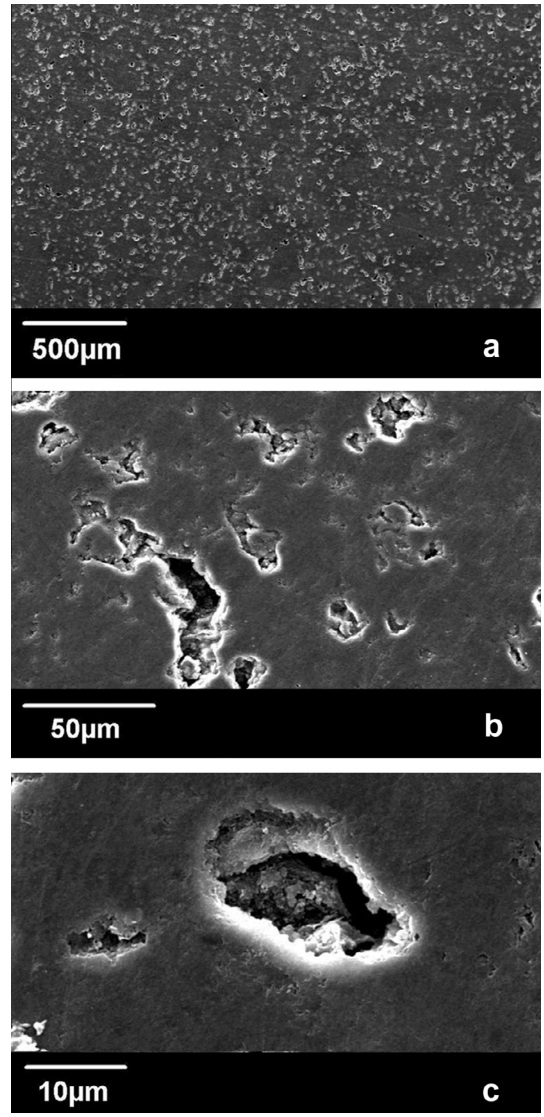

Fig. 1. SEM images of the graphite surfaces for different magnifications: 30× (a), 400× (b) and 1500× (c).

After the impinging and wetting behaviors were complete, the images were processed to determine the drop edge coordinates, i.e., the whole profiles of impacting droplets, the contact angle, the wetting diameter and the height of the drops [18]. The uncertainty for the edge locations in this work was approximately 0.011 mm (∼0.2 pixels). All the measurements were conducted for several runs, of which two randomly chosen ones were initially fully analyzed, checking if their results were in general agreement. However, since the *intrinsic* randomness of the system described (the presence of random defects on the substrate always induced slight differences in the results) and, thus, the main focus on *qualitative* drop behaviors, the reported data were chosen to be referred to a single *example-run*, actually more consistent than showing, instead, the average value of all the analyzed runs.

3. Results and discussion

Fig. 2 shows the wetting behavior for pure water droplets impacting onto grinded graphite substrates. The analysis was initially performed by evaluating quantitatively the time dependence of spreading factor, contact angle, and dimensionless drop height.