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Bubbles entrapment for drops impinging on polymer surfaces: The roughness effect

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ABSTRACT

A study on the influence of the substrate roughness on the occurrence and the formation mechanisms of air bubbles entrapped into water drops impinging on polymeric surfaces, typified by distinctive superficial grinding ($15 \leqslant R_q \leqslant 1159$ nm), was conducted by digitizing silhouettes of the impacting droplets. The images were processed to determine the drop outer profile, the morphology of an air cavity formed at the center of the droplet during the droplet deformations, and the shape of the enclosed bubble. The fluid dynamic processes leading to the entrapment of the bubble were meticulously analyzed, and three new formation mechanisms were observed and detailed. The investigation was conducted for different droplet sizes ($2.13 \leqslant D_0 \leqslant 3.87$ mm), and at a wide range of impact velocities ($0.14 \leqslant V_i \leqslant 0.59$ m/s). The entrapped bubbles were more likely found into drops falling on more rugged surfaces, while were rare or totally absent into drops impinging on less rough substrates. However, considering just impacts on more rugged surfaces, the bubble occurrence appeared to be minimally influenced by the substrate roughness. At fixed roughness, the bubbles were caught into droplets impinging at lower velocities with increasing the drop size. For fixed drop size and substrate roughness, the range of the bubble existence was often split in two distinctive intervals of impact velocities, phenomenon especially noted for drops impacting on more rugged surfaces. Finally, we suggested a relation between the peculiar shapes that the liquid assumed in the course of the impact and the occurrence/absence of the entrapped bubble.

1. Introduction

The formation of air bubbles into the liquid during drop impinging processes has drawn tremendous attention since the early 1990s [1–41]. Not considering cavitation cases [1], two very distinctive types of bubble enclosed into a droplet impacting on a solid surface have been reported in literature. The first kind, more precisely named [14,40] *impact bubble*, forms during the initial instants of the *spreading phase* due to the enclosure of a thin air layer caught under the droplet in the course of the impaction. This phenomenon has been deeply investigated using experimental, theoretical and simulation approaches [2,4,6–29,38]. The second kind of bubble, labeled as *entrapped bubble* [39,40], is formed, instead, during the *retraction phase* as consequence of the closure of the top liquid layer of the droplet above an air cavity formed at the center of the drop [1,30,32–41].

This latter bubble formation was first reported in 1972 by Elliott and Ford [30], so that a brief description was included in the 1993 review by Rein [1]. In 2003, using modern high-speed visualization technologies, Renardy, et al. [31] recorded the formation of the air cavity at the center of the drop, though afterward collapsing with the ejection of a liquid jet without entrapping any bubble. Similar air cavities were noted by Khatavkar, et al. [33] for microdrops of initial diameter $D_0 = 157$ μm, simulated using a diffuse-interface model (DIM), falling at impact velocity $V_i = 0.83$ m/s on smooth, flat, and chemically homogeneous solid surfaces (equilibrium contact angle $90° < \theta_{eq} \leqslant 120°$). However the formed bubbles were not stable in their computation, so that they were not visible into the simulated recoiling droplets.

In 2006, entrapped air bubbles were for the first time photographed by Bartolo et al. [32]. In their study, these bubbles were caught into water droplets with $D_0 = 2$ mm falling at impact velocities between 0.55 and 0.65 m/s on superhydrophobic substrates ($\theta_{eq} \sim 160°$). Subsequently, Tsai et al. [34] observed, for $0.28 \leqslant V_i < 0.35$ m/s, the formation of air cavities and enclosed bubbles into water drops ($D_0 = 2$ mm) impinging on different rough surfaces of carbon nanofiber jungles ($\theta_{eq} = 152 \pm 3°, 163 \pm 3°$). Chen et al. [35,36] reported the occurrence of entrapped bubbles into water drops with $D_0 = 2.4$ mm falling at $0.47 \leqslant V_i < 0.5$ m/s on soft polydimethyl-siloxane (PDMS) elastomers (apparent contact angle $\theta_{ap} = 13.4°, 40.74°, 102.8°$, and $111.2°$). Interestingly, they also

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observed that, in specific conditions, the entrapped bubble could coalesce with the air film enclosed under the drop during the impingement. Afterwards, the same group [37] noted the presence of bubbles into water drops with $D_0 = 2.76$ mm impacting at $0.31 \leqslant V_i < 1.13$ m/s on surfaces made depositing carbon nanotubes on silicon substrates patterned with micropost arrays (advancing water contact angle, $\theta_a = 167°$), and at $0.31 \leqslant V_i < 0.83$ on lotus leafs ($\theta_a = 163°$). In 2011, Huang et al. [38] simulated the formation of entrapped bubbles by using a phase-field lattice Boltzmann model (LBM). The bubbles (not only *air* bubbles) were detected for different simulation conditions (drop size, impact velocity, liquid surface tension and viscosity, and substrate hydrophobicity).

In two recent works, Wang et al. [39] and Hung et al. [40] detailed the formation of several entrapped bubbles by analyzing top and side view images of water drops ($1.28 \leqslant D_0 \leqslant 5.86$ mm) falling at different impact velocities on parafilm substrates ($\theta_a \sim 110°$). Particularly, Hung et al. [40] illustrated two general mechanisms for the formation of the bubble, depending on whether or not the central air cavity touched the substrate surface in the course of the impact. The description of these formation mechanisms was expanded by Pittoni et al. [41], in the study of the occurrence of entrapped bubbles into water drops ($1.91 \leqslant D_0 \leqslant 4.87$ mm) impinging on four graphite substrates, characterized by different hydrophobicities ($\theta_a \sim 90°$, 120°, 140°, 160°). Specifically, five unlike mechanisms leading to the bubble formation were reported for definite operative conditions. The substrate hydrophobicity was found to greatly influence the occurrence of the entrapped bubbles, with few or no bubbles noted into drop impacting on the less hydrophobic surfaces.

However, in all the above mentioned analyses, the influence of the substrate roughness on the occurrence and formation mechanisms of the bubble entrapment was never systematically considered. Therefore, for clearly investigating this roughness effect, in this study experiments were conducted on pure water droplets falling on eight different polycarbonate (PC) substrates, typified by distinctive superficial grinding. The analysis was carried out digitizing the silhouettes of the impacting droplets. Then, the images were processed to determine the drop edge coordinates, identifying the morphology of the air cavity and the shape of the enclosed bubble. The impingement experiments were performed for different impact velocities and droplet volumes, delineating the system conditions (V_i, D_0 and R_q) for the occurrence and the formation mechanisms of the entrapped bubble.

2. Experimental

2.1. Materials

The water used was purified by a Barnstead NANOpure water purification system with a specific conductance of less than 0.057 μS/cm. The polycarbonate, PC (CAS #24936-68-3), used in this work was purchased from Sun-Fung Co., Ltd., Taiwan. Some measurements were performed on original PC surfaces, employed *as-it-is*. Other PC substrates underwent a grinding process for 15 min. The average grit sizes used (ISO 6344 grit designation) were 5, 8, 14, 22, 35, 76, and 125 μm.

An atomic force microscopy (AFM) analysis was conducted for investigating the unlike polymers topographies (Fig. 1) and measuring the surface roughness. Areas of 100 × 100 μm size were scanned in contact mode and the AFM images were analyzed for calculating the root-mean-square roughness, R_q. The R_q was found to be 15 nm for the original PC substrate, and 32, 114, 261, 358, 609, 1092 and 1159 nm for the other grinded surfaces. The different substrates were named in this study as "R15", "R32", "R114", "R261", "R358", "R609", "R1092" and "R1159" respectively, based on their roughness.

R15 substrates (Fig. 1a) exhibited smoother and more homogeneous surfaces, even if random little irregularities were detected by the AFM examination. After the grinding processes, the PC substrates presented an increasing number of defects and inhomogeneities: R32, R114 and R261 (Fig.1b–d) were characterized by striped-like topographies, while R358, R609, R1092 and R1159 (Fig.1e–h) showed more rugged surfaces. An analysis of the values of the water advancing contact angle (θ_a) for these grinded PC surfaces has been recently detailed in Pittoni et al. [42]. Specifically, in regards to the hydrophobicity of the substrates, a strong increment of θ_a values with increasing R_q was noted: from ∼85° for R15 to ∼120° for R1159.

2.2. Apparatus and methods

An apparatus similar to the system detailed in Wang et al. [43] was used in this study for recording and analyzing the droplets impingement. The video image system (Optronis CR3000X2 and Mikrotron GmbH mini2) digitized the pictures in 400 lines × 400 pixels. The rate of image acquisition was 6770 images per second. 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 55.3 μm/pixel horizontally and 55.1 μm/pixel vertically for the side-view camera. The uncertainty for the edge location in this work was around 0.2 pixels for the drop profile and 2 pixel for the air cavity/bubble profile [41]. All experiments were undertaken at 25 ± 0.5 °C.

Falling drops were generated from stainless steel needles with impact heights above the substrates ranging between 10 and 17.9 mm. The positions of the top or bottom points of the falling drop were fitted by the equation of a free falling body to determine the time of droplet contact and the impact velocity [43]. In the present work, the impact velocity ranged between 0.14 and 0.59 m/s, while the droplet sizes were 2.13, 2.53, 3.14 and 3.87 mm. The drop size was evaluated from the average drop mass of 100 impinging droplets.

After the droplet fell down and impacted on the solid substrate, sequential digital images of the drop were taken. At the completion of impinging and wetting phases, the images were processed to determine the drop edge coordinates [40,41]. Therefore the droplet outer profile, the morphology of the air cavity formed at the center of the droplet, and the shape of entrapped bubble were delineated.

3. Results and discussion

Fig. 2 illustrates the morphologies of two water drops of $D_0 = 3.14$ mm impinging at $V_i = 0.29$ m/s on two different PC substrates: R1092 (Fig. 2a) and R1159 (Fig. 2b). The drop impacts were recorded from an approximately 45° top view. During the initial phase (images −10 to 33), dominated by the inertia of the impact, both of the drops assumed at first a spherical shape (images −10, 5), and then a multiple layers configuration (images 15–33). Note that the morphology of the drop just before the impact may be influenced by the oscillations that the drop undergoes after its release from the needle [43], so that, with modifying the impact height, the drop shape may assume more oblong or spherical configurations. In the cases illustrated in Fig. 2, the impact heights were similar for the two drops, so that their shapes before the impact were not significantly different (images −10).

At the time of image 37 a conical protuberance stretched out on the upper part of the drop impinging on R1092 (image 37, Fig. 2a). A more blunt bulge was observed on the top of the drop impacting on R1159 (image 37, Fig. 2b). However, due to the oscillations