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Supercooled water droplet impact on superhydrophobic surfaces with various roughness and temperature

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ABSTRACT

This paper investigates the impact behaviors between supercooled droplets and various superhydrophobic surfaces. We analyze the thermodynamic reason of the liquid adhesion at low temperature and discuss the influence of the substrate roughness, temperature and wetting property on the impact dynamics of supercooled droplets both experimentally and theoretically. The liquid adhesion results from the ice nucleation rather than the enhanced viscous effect induced by the low temperature. The dramatic increase of the liquid–solid contact area induced by the high-speed collisions between droplets and textured surfaces leads to a larger nucleation rate and a lower critical adhesion temperature. The analytical argument of the liquid–solid contact area fraction at different impact speed throws light on the understanding of the controversy if hierarchical superhydrophobic surfaces are icephobic at low temperature. Valuable suggestions are given that smooth superhydrophobic surfaces with nanoscale roughness near the critical nucleus radius perform better water-repellency against high-speed supercooled droplets.

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1. Introduction

Atmospheric icing on structures has a significant impact in a wide variety of fields, such as the crash of aircrafts, the collapse of transmission lines, the damage of industrial facilities and so on [1–3]. Therefore it is essential to get more knowledge about the ice accumulation process in cold environment and come up with rational design of surfaces with exceptional icephobic property. Various anti-icing and deicing methods have been developed recently. The promotion of the droplet bouncing, the delay of the ice nucleation time before freezing, and the reduction of the adhesion strength after freezing are the three main factors considered in defining icephobicity [4–7]. Inspired by the non-wetting structures in nature [8,9], superhydrophobic water-repellent surfaces which could effectively entrap air cushions in the surface textures, were widely used to prepare anti-icing materials under low temperature, due to their ability to rapidly repel the impacting droplets before freezing occurs [4,5,10–12] and the coalescence induced self-removal of condensed water droplets [13–15]. Besides, when the degree of supercooling of the water droplets is low, the ice nucleation delay time could last several minutes or even several hours [15–17], which provides more possibility to remove the impacting or condensed droplets before freezing occurs. After the ice formation, fabrication of surfaces with low ice adhesion strength is preferential, which allows easier and faster deicing process. Inspirations from the slippery peristome surface of Nepenthes pitcher plants leads to the development of the self-healing, slippery liquid-infused porous surfaces (SLIPS) with exceptional water-repellency, pressure stability and enhanced optical transparency [18–20]. However, the requirements of these surfaces to be scalable, inexpensive, stable under pressure, mechanically durable, self-healing and environmentally friendly limits the development of the superhydrophobic and slippery anti-icing materials and structures.

The ice accretion on structures includes the impacting and freezing process of water droplets on solid surfaces. The solidification of a sessile droplets involves five different stages [21,22]: (1) A pre-cooling or supercooling stage, during which the liquid droplet is cooled from an initial temperature to below the equilibrium freezing temperature, until ice crystal nucleation occurs. (2) An ice nucleation stage. (3) A recalescence stage, during which supercooling drives rapid kinetic crystal growth from the crystal nuclei. This stage is terminated when the droplet reaches its equilibrium freezing temperature. (4) A liquid freezing stage, where crystal growth is governed by the heat transfer rate from the droplet to the point where the droplet liquid is completely frozen. (5) A solid cooling or tempering stage, where the solid droplet temperature is reduced to near the ambient air temperature. The homogeneous nucleation for supercooled free droplets generally occurs at the

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temperature approximately below −40 °C [23]. However, in the presence of impurities or solid substrates, heterogeneous nucleation would first occur at the liquid–solid interfaces at a lower degree of supercooling [24].

Superhydrophobic surfaces were found effective in preventing ice formation instead of fighting its build-up, compared with hydrophilic or hydrophobic surfaces, due to their ability to repel impacting droplets before ice nucleation occurs [25,26]. Many efforts have been made on revealing the nature of the impact behaviors between supercooled water droplets and cold superhydrophobic surfaces. A physics-based modeling framework was developed to predict ice formation on cooled superhydrophobic surfaces, combining the drop impact dynamics, the classical nucleation theory with heat transfer and the wetting dynamics [26]. The reduction of the liquid–solid contact area on superhydrophobic surfaces was found to play an essential role in delaying nucleation by reducing the heat transfer together with the heterogeneous nucleation at the interface [27]. Besides, it was found that the critical Weber number for the drop impalement was independent of the substrate temperature since the meniscus penetration occurred much faster than the heat transfer time scale [28]. However, for droplets with large supercoiling degree (−17 °C), the increased viscous effects were found to significantly influence the impact and impalement dynamics, with a reduction of maximum spreading and an increase of the contact time [29]. The meniscus penetration was demonstrated to occur with full penetration at the center at large supercooling instead of a ring shape at room temperature [29]. The maximum spreading diameter was found to be not influenced by the decrease of temperature [30,31], and the minimum receding diameter and the ice accretion speed increased with the decreasing of wall temperature [31]. Recently, the influence of the nanometer-sized air cushion entrapped on the motion of the three-phase contact line was discussed on hierarchical nanotube surface above and below 0 °C [32].

The application of hierarchical (both micro- and nano-) roughness on materials with low surface energy, is believed to be the key procedure to fabricate superhydrophobic surfaces [8,9] with excellent water-repellency and self-cleaning property. However, in a severe cold environment below the freezing point, whether superhydrophobic surfaces with hierarchical roughness perform better icephobicity is controversial [33–36]. Therefore it is of more importance to study the preparation methods of superhydrophobic surfaces to behave better icephobicity at low temperature. In this work, we investigate the dynamic process of supercooled water droplets with different velocities impacting on various superhydrophobic surfaces with either single-tier or hierarchical roughness. An analytic argument is carried out to explain the transition of outcomes from rebound to adhesion at low-temperature impact, which results more from the various impact behaviors at different speed than from the effect of temperature on the liquid viscosity. An investigation is made to discuss the effects of various impact factors, especially the substrate roughness, temperature and the liquid–solid contact area, on the ice nucleation and the droplet adhesion. Constructive suggestions are proposed for the fabrication of superhydrophobic anti-icing substrates at high-speed impact and under extreme environment.

2. Experimental

2.1. Materials and methods

In this study we fabricated four different superhydrophobic surfaces with various scales of roughness. The geometrical parameters, average roughness and the wetting characteristics of the experimental substrates are shown in Table 1. The average roughness of the substrates was measured using the three-dimensional white light interference surface morphology measurement system (NexView, ZYGO Corporation). The static and dynamic contact angles of a deionized water droplet (8 μL) were measured using a standard contact angle goniometer (JC2000CD1, POWEREACH). Then the substrates were cleaned using high purity nitrogen flow to avoid contamination. The superhydrophobic substrate with single-tier nano-coating, which was applied as the control group in this study, was made by coating flat silicon surface with a commercial nano-coating agent (Glaco Mirror Coat "Zero", Soft 99 Company) [28,29] to modify their hydrophobicity. And the microscopic morphology of the substrate Nano-Si is shown in Fig. 1(a). Superhydrophobic surfaces with hierarchical roughness were prepared as the experimental group, with either regular textures or random roughness. Substrates P4S3 and P100S100 are hierarchical superhydrophobic surfaces with micrometer-scale and submillimeter-scale posts. The posts were first fabricated using photolithography and etching of inductively coupled plasma (ICP) on silicon surfaces. Then they were coated with nanoparticles, with similar roughness to the single-tier surfaces. The microscopic structures of the substrates P4S3 and P100S100 are shown in Fig. 1(b) and (c). The hierarchical superhydrophobic substrate with random roughness SHS-Al was prepared by spraying a superhydrophobic coating composed of hierarchical micro-nanoparticles (Neverwet multi-surface aerosol, Rust-Oleum Corporation) uniformly on an aluminum surface. The average roughness is approximately 15 μm, as shown in Fig. 1(d).

2.2. Experimental setup

Experiments were performed by releasing an ultrapure water droplet ($D_0 = 2.4$ mm ± 0.1 mm) with varied velocity ($U_0 = 1.4$ m/s ~ 2.4 m/s) on different superhydrophobic surfaces placed on a cold plate with a controllable temperature (−30 °C ~ 15 °C). The ultrapure water with electrical conductivity less than 0.1 μS/cm was generated by filtering and deionizing water using an ultrapure water system. The impact experiments were done in a refrigerator in which the temperature and the humidity was controllable. The relative humidity was kept around 25% ± 5% by a constant supply of dry nitrogen to avoid frost formation. The experimental setup is shown in Fig. 2. The experiments were performed by releasing supercooled water droplets ($D_0 = 2.4$ mm ± 0.1 mm, $T_0 = -5$°C ± 0.5 °C) on different cooling superhydrophobic surfaces (T_S ranging from −5°C to −30 °C). The supercooled water was stored in a sterile syringe and supercooled droplets were generated through a capillary tube connected by a disposable sterile tube with the syringe pump. The impact experiments between droplets above the freezing point and various superhydrophobic surfaces were performed at the temperature from 0 °C to 15 °C. The dynamic process of the collision between the drop and the solid was recorded by a high-speed camera (FASTCAM Mini UX100, Photron) at the frame rate of 8000 fps with a shutter speed 1/40,000 s. High purity nitrogen flow was applied to remove the residuals and clean the surface after each experiment. Repeat experiments were performed at least three times at each condition.

3. Results and discussion

3.1. Outcomes: Rebound v.s. Adhesion

Different drop impact behaviors were observed on superhydrophobic surfaces with various roughness and a wide range of temperature from −30 °C to 15 °C. Droplets could bounce off all the experimental superhydrophobic substrates at temperature above the freezing point. However, as the cooling of the substrate,

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