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Research Paper

# Sessile droplet freezing on polished and micro-micro-hierarchical silicon surfaces

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## HIGHLIGHTS

- Droplet freezing on polished and micro-micro-hierarchical silicon surfaces is observed.
- The duration of surface recalescence is 0.2–0.8 s depending on the locations on the droplet.
- Recalescence starts simultaneously but ends earlier as closer to the droplet bottom.
- Freezing rate at the solidification front edge is higher than in the middle.
- The micro-micro-topology determines the dominance of wetting characteristics on ice nucleation.

## ARTICLE INFO

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## ABSTRACT

Droplet freezing on a polished silicon wafer and a micro-micro-hierarchically structured silicon surface, the contact angles of water (at 25.7 °C) on which were 78.77° and 143.82° respectively, were experimentally observed. The surface temperature was maintained at −6.3 °C. Combining high-resolution photography and infrared thermometry, the ice nucleation onset of the droplet was captured. It is found that the hierarchical surface remarkably postpones the ice nucleation compared with the hydrophilic polished silicon because of enhanced free energy barrier for nucleation. The unique micro-micro-scale topology determines the dominance of wetting characteristics on ice nucleation. Besides, the formation of ice shell is initiated randomly at the water-surface-air trijunction base and propagated both circumferentially and vertically upwards during the surface recalescence stage. Subsequently, the internal solidification proceeds from bottom up when the ice shell develops. Moreover, the duration of surface recalescence and internal solidification stages, as well as the shape of solidification front (SF) are evaluated based on the local droplet surface temperatures. The surface recalescence begins simultaneously but ends earlier as closer to the droplet bottom. The temperature at SF edge is higher than that in the middle. Compared with the fully frozen solid cooling stage, the heat transfer rate is much higher during internal solidification.

## 1. Introduction

Surface treatment attracts increasing attention in anti-icing applications. Surface hydrophobicity is crucial on frost nucleation and frost deposition mainly because of the change of surface energy compared with regular un-coated surfaces [1]. Droplet freezing process is of great significance since it corresponds to the frost crystal nucleation period, which is the very first stage of frost deposition.

The study on droplet freezing on different surfaces can be traced back to the 1950s. Carte [2] found minor impact of surface types on the temperature barrier of heterogeneous nucleation of ice. Nevertheless, surface coatings showed detectable influence on the subcooled water droplet freezing temperature in the study of [3]. Encouragingly, the icephobicity of (super)hydrophobic surfaces has been testified in many studies [4–12]. Recently, we experimentally observed the droplet freezing processes under natural convection on surfaces with different topographical textures [13]. A type of two-stage black silicon hydrophobic surface showed excellent icephobicity. Generally, it is believed that the increase of contact angle would substantially rise the potential barrier for ice nucleation [1]. Some also argued that the reduced contact area between the condensed droplets and the surface [5,14,15], and the interfacial air pockets [12] are responsible for droplet freezing retardation.

It was evidenced that the water freezing usually starts from

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heterogeneous nucleation of ice, which could either be initiated by pre-existing nuclei immersed in the water droplet, or in contact with the droplet surface [16]. Nucleation tends to take place at the trijunction where the molecules' structural characteristics have important effects on icing suppression [3]. However, the mechanisms of droplet ice nucleation and freezing on micro-structured surfaces are still not a closed topic.

Experiments were conducted to dig further into the details. The droplet transparency was found to reduce after nucleation and the crystal grew rapidly [17]. Jin et al. [18] captured clear images showing the solid-liquid interface movement after ice nucleation started on a superhydrophobic surface with CA = 152.6°. Different explanations on the transparency change have been reported, including air bubble release, light scattering effect and a solid ice shell formation [19]. Besides, droplet shape gradually changed after ice nucleation till the completion of fully solidification. It is widely accepted that the specific volume difference between ice and water was the reason for droplet deformation and protrusion formation [17,20,21]. Marín et al. [22] also believed that the geometry of freezing front is responsible for the droplet top tip shape.

The temperature of droplet has also been measured using intrusive and non-intrusive methods. A sudden surface temperature rise of the droplet was found when nucleation began, which was attributed to the release of latent heat of fusion [23,24]. Chaudhary and Li [25] measured the temperature distribution inside the droplet with the aid of inserted thermocouples. Uniform temperature was detected across the droplet after recalescence. However, the thermocouples might serve as pre-existing nuclei and affect the ice nucleation. Infrared camera was also applied on top of the droplet by [25] and the transient temperature of the droplet was revealed.

It is also noticed that the surface topographical parameters such as surface roughness and/or microscopic photos were not provided in many studies. Moreover, the existing visualization results were sometimes very blur. Clear observation of the droplet freezing processes in the open literature is very limited. Thermographic approach was barely applied for droplet surface temperature distribution measurement, especially in the lateral view. In the present study, we experimentally observed the droplet freezing on a polished silicon wafer and a micro-micro-hierarchically structured surface using photographic and infrared thermographic methods.

## 2. Description of the experiment

### 2.1. Experimental setup

As shown in Fig. 1, the experimental system includes a CCD camera, an infrared camera, a thermostatic bath and cold base, test samples, a microsyringe for droplet dosing and a data acquisition system. The temperatures set by the thermostatic bath and the corresponding temperatures of the cold base where the test samples lay on are calibrated prior to the experiment. In the present study, the temperature of the thermostatic bath is set to be −10 °C, the corresponding temperature of the cold base is −6.3 °C.

The cold base is covered by a layer of self-addictive tape, which is removed after the cold base reached the desired temperature to prevent pre-accumulated frost prior to the tests. The textured samples are then located right in the middle of the clean cold base after ripping off the tape. The sample temperature rapidly reduces and eventually approaches the cold base temperature. After around 1 min, the surface temperature of the sample reaches and maintains at −6.3 °C. Then, a 10 μL purified water droplet is manually placed on the surface via a microsyringe. The water is processed via a laboratory ultrapure water system and met the National Laboratory Water Standard of China (GB6682-2000).

A CCD camera and a LED illuminator are utilized for visualization of the droplet lateral profile. The camera can also be positioned on top of the droplet for vertical viewing. An infrared camera (NEC® R300SR-H) is placed on the side of the droplet in the direction perpendicular to the CCD camera optical line. Both of the cameras are trigged simultaneously as the dosing is underway in order to capture the very initial moment of the droplet. InfReC Analyzer NS9500 Professional® software is used for infrared image analysis. The maximum set-speed of the infrared and the CCD cameras is 30fps and 8fps respectively.

Tests under the same experimental condition are repeated for at least ten times. The ambient temperature is controlled using an air conditioner. All the tests are conducted under natural convection at atmospheric pressure, an ambient temperature of 24 °C ± 1 °C, and a relative humidity of 50% ± 1%.

### 2.2. Test samples

The micro-micro-hierarchically structured silicon is a new type of surface structure combining periodical micro-pillars and aperiodic black silicon tops. The micro-micro-hierarchical surface is fabricated in two steps. First, the polished silicon wafer is treated via the deep reactive etching method in conjunction with photolithographic

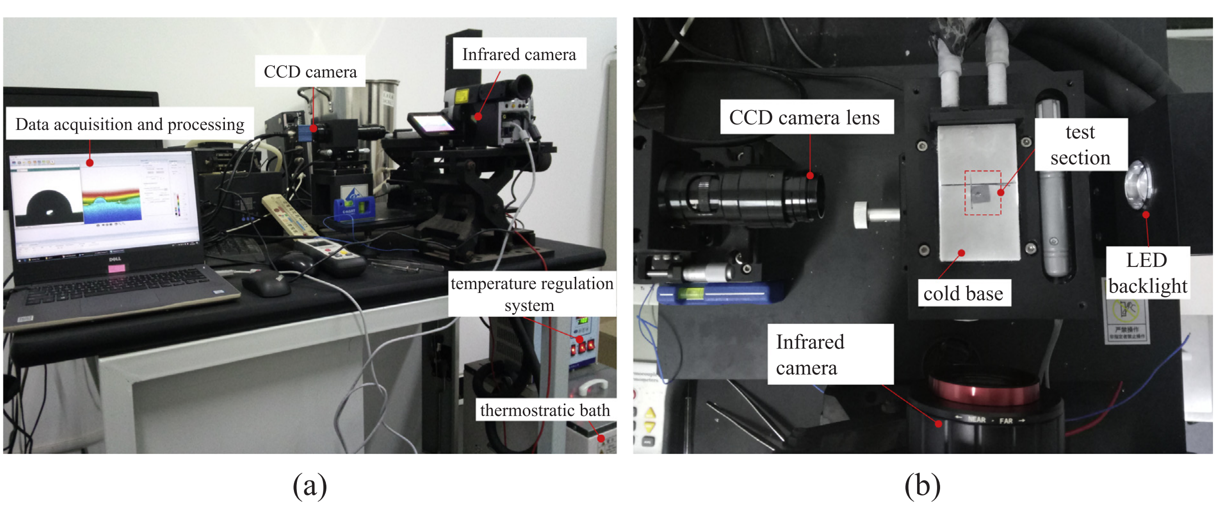


**Fig. 1.** Photos of (a) the entire experimental setups (b) test section.