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Quantitative analysis of anti-freezing characteristics of superhydrophobic surfaces according to initial ice nuclei formation time and freezing propagation velocity

Junghan Kim, Jaehyeon Jeon, Dong Rip Kim, Kwan-Soo Lee *

School of Mechanical Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul 04763, Republic of Korea

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ABSTRACT

In order to quantitatively analyze the anti-freezing characteristics of superhydrophobic and bare surfaces, the freezing delay properties of the surfaces were experimentally investigated under various operating conditions by placing sessile droplets on their surface. The freezing delay time was calculated using the experimental results and analyzed by employing a stochastic method. The formation time of initial ice nuclei and freezing propagation velocity at a macroscopic level were proposed as measures of surface anti-freezing characteristics. The anti-freezing properties of the bare and superhydrophobic surfaces were analyzed using the proposed quantitative measures. Consequently, the tendency of quantitative results was consistent with that of the qualitative ones according to the changes of the operating conditions (air inlet velocity, relative humidity, and surface temperature). Moreover, the superior anti-freezing performance of the superhydrophobic surface was quantitatively confirmed by the initial ice nuclei formation time, which was delayed by 22–92%, and the freezing propagation velocity, which decreased by 17–30%.

1. Introduction

Frost and ice formation on the heat exchangers of air conditioning and cooling equipment [1,2] and wind turbines [3,4] causes various problems, such as reduced device performance and potential operation interruptions. Meanwhile, superhydrophobic surfaces, which have a higher contact angle than normal surfaces because of their low surface energies, exhibit anti-freezing characteristics as a result of the jumping droplet phenomenon [5] and high Gibbs energy barrier. These surfaces can alleviate the problems caused by frost and ice formation without consuming additional energy. Hence, numerous studies have been conducted on the freezing delay of superhydrophobic surfaces.

In previous research, the freezing delay time was used as a measure of the anti-freezing performance of developed superhydrophobic surfaces. The experimental methods and definitions of the freezing delay time, which was used to verify the surface anti-freezing properties, were different depending on the study. In studies using sessile droplets, a single droplet [6–11] or numerous sessile droplets [12] were placed on the surface to observe the freezing process. In these cases, the freezing delay time was defined from the start time of the droplet cooling on the surface at room temperature [6–8] or at a temperature below freezing point [9–11], to the onset of the droplet freezing. In studies that did not use sessile droplets, freezing propagation on the surface was observed directly [13–17]. Among them, when the entire surface was observed, the freezing delay time was mainly measured from the onset of surface cooling, and the surface freezing propagation process was observed continuously over time [13–15]. On the other hand, when the small part of the entire surface was observed, the freezing delay time of the observation area was defined as the period from the freezing time of the first water droplet to that of the final water droplet [16,17].

However, the majority of previous research studies on the anti-freezing properties of superhydrophobic surfaces using freezing delay time exhibit certain limitations. First, it is difficult to conduct a strict comparison using the previous results, because a variety of experimental methods and definitions exist for the freezing delay time and there is a lack of quantitative performance measures. Second, it is difficult to analyze the anti-freezing characteristics accurately using only the freezing delay time. Anti-freezing characteristics can be regarded as the retarded formation of initial ice nuclei and delayed propagation of ice nuclei. The freezing delay time can describe the formation of initial ice nuclei according to

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* Corresponding author.
E-mail address: ksleehy@hanyang.ac.kr (K.-S. Lee).

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Nomenclature

A	area
f	function
ΔG^*	Gibbs energy barrier, J
n	number
N	total number
r	radius, nm
RH	relative humidity, %
T	temperature, °C
t	time, s
u	air inlet velocity, m/s
v	freezing propagation velocity, cm^2/s
w	absolute humidity, g/kg_{DA}

Subscripts

a	air
s	surface
sat	saturation
st	static

Greek symbols

Φ	cumulative distribution function
θ	contact angle, °

isolated droplet freezing, but this is not sufficient for describing the freezing propagation process, which is characterized by inter-droplet freezing [18]. Therefore, it is necessary to characterize the freezing propagation process by performing an analysis using the rate or velocity. In order to achieve this, certain researchers have made use of freezing propagation velocity. Boreyko and Collier [18] observed the jumping droplet phenomena and analyzed the inter-droplet freezing process. Chen et al. [19] and Zhao et al. [20,21] analyzed the effects of surface microstructure on freezing propagation behavior. The existing freezing propagation velocity was suitable only for analyzing the microscopic behavior of freezing propagation because the velocity was calculated by observing part of the substrate with a microscope. However, this method exhibits limitations in representing the macroscopic behavior of freezing propagation [22]. Another limitation is that employing a stochastic approach is necessary to analyze the freezing characteristics more accurately. Previously, it was demonstrated that the results of repeated experiments on freezing delay time were expressed as an arithmetic mean, suggesting that freezing was always delayed on the superhydrophobic surface relative to the bare surface. Recently, Kim et al. [23] analyzed the freeze delay time in a stochastic manner in order to consider the randomness of freezing process and found that water droplets on a bare surface occasionally freeze later than on a superhydrophobic surface. This suggests that the arithmetical mean of the freezing delay time may not be a suitable measure for the surface anti-freezing properties, and a stochastic approach is required to evaluate the anti-freezing properties more accurately. However, this study also exhibited the limitation that anti-freezing characteristics were expressed only by freezing delay time. Therefore, in order to analyze the freezing characteristics accurately, it is necessary to improve previous research methods by proposing quantitative performance measures, using macroscopic freezing propagation velocity, and performing a stochastic analysis of anti-freezing characteristics.

In this study, we propose quantitative analyses of anti-freezing characteristics using a stochastic method to compensate for the limitations of previous studies. Freezing delay experiments on bare and superhydrophobic surfaces are performed using sessile droplets while varying the operating conditions, and the experimental results are analyzed by means of the stochastic method. In order to analyze the anti-freezing performance quantitatively, new definitions of the initial ice nuclei formation time and freezing propagation velocity are proposed, which are appropriate for characterizing macroscopic freezing behavior, and the anti-freezing characteristics of the bare and superhydrophobic surfaces are quantitatively analyzed.

2. Experiments

2.1. Experimental apparatus and test conditions

The experiments were conducted using an experimental setup constructed in the same manner as in previous studies [24,25]. Fig. 1 illustrates the test section (300 × 300 × 500 mm) used in these experiments. A high-resolution camera (Nikon D810 with micro 105 mm ED lens) was installed at the top of the test section to observe the freezing propagation across the surface. A thermoelectric cooling module was horizontally installed at the bottom of the test section. As indicated in Fig. 1(b), an aluminum plate for uniform heat transfer and a heat sink for heat dissipation were attached above and below the thermoelectric element, respectively. The test surfaces were placed on the aluminum plate, and thermal grease was applied between these in order to minimize the contact thermal resistance. The surface temperature was measured by type-T thermocouples implanted in the aluminum plate and controlled using a proportional–integral–derivative (PID) controller (Temcoline, T34). Table 1 displays the accuracy of the measurements.

The bare and superhydrophobic surfaces were made of Al 6061 with a size of 5 × 5 cm, and the superhydrophobic surface was fabricated according to the following procedure. The Al 6061 surface was washed with deionized water, acetone, ethanol, and isopropanol, and then immersed in a 1 M NaOH solution (Samchun Chemical) for 2 min to remove the surface oxide layer. Thereafter, the Al surface was etched for 20 min in a mixed solution of deionized water and a 35 vol% hydrochloric acid solution (Daejung Chemicals and Metals) with a volume ratio of 4:1, and dried in a vacuum oven at 190 °C for 1 h. The surface was immersed in a mixed solution of hexane and 1H, 1H, 2H, 2H-perfluorodecyltri chlorosilane (Alfa Aesar) with a volume ratio of 1000:1 for 10 min and dried in a convection oven at 100 °C for 1 h to complete the superhydrophobic surface fabrication. Fig. 2 illustrates the static contact angles and scanning electron microscope (SEM) images of the untreated and superhydrophobic surface microstructures. The test surface with sessile droplets (~8 μl) was photographed at room temperature, and the surface contact angle was measured using axisymmetric drop shape analysis (ADSA) [26]. The droplet shape, calculated by ADSA and represented by the blue curve in Fig. 2, was overlaid on the image of a water droplet. The static contact angles of the untreated and superhydrophobic surfaces were 85 ± 1° and 156 ± 2°, respectively. The sliding angle of the superhydrophobic surface was less than 10°. A smooth bare surface and superhydrophobic surface with a fine microstructure can be observed in the SEM images of the surfaces.