International Journal of Heat and Mass Transfer 124 (2018) 83–89

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Quantitative measurements of nanoscale thin frost layers using surface plasmon resonance imaging

Chan Ho Jeong [a], Dong Hwan Shin [b], Vinaykumar Konduru [c], Jeffrey S. Allen [c], Chang Kyoung Choi [c,*], Seong Hyuk Lee [a,*]

[a] School of Mechanical Engineering, Chung-Ang University, Seoul 156-756, Republic of Korea
[b] Department of Energy Conversion Systems, Korea Institute of Machinery & Materials, Daejeon 34103, Republic of Korea
[c] Department of Mechanical Engineering-Engineering Mechanics, Michigan Technological University, Houghton, MI 49931, United States

ARTICLE INFO

Article history:
Received 8 January 2018
Received in revised form 16 March 2018
Accepted 16 March 2018

Keywords:
Frosting process
Frost thin layer
Surface plasmon resonance (SPR)
Thickness measurement

ABSTRACT

This study reports the presence of a nanoscale thin frost layer. During the frosting process, the surface plasmon resonance (SPR) imaging method can be used to overcome conventional optical limits and quantify this layer. The research outlined here also provides quantitative thickness measurement of the thin frost layer via a proposed calibration method based on the measured SPR intensity. The SPR system established in this study consists of a 50 nm gold-coated BK7 cover glass, a prism, a light source, a polarizer, a lens and a filter for the collimated light of a 600 ± 5 nm wavelength, and a CCD camera. The SPR angle of the ice phase is 72°, which corresponds to the ice refractive index of 1.307. The gold-glass specimen is cooled from room temperature (23 ± 1 °C) to −4.0 ± 0.8 °C by using a thermoelectric cooler to maintain the relative humidity of 20 ± 3% (at the room temperature). As a result, it is found that the nanoscale thin frost layer between the frozen condensates exists on the surface. Also, the present study yields the spatial distribution of reflectance that is associated with the frost layer thickness, indicating that the local information about thin frost layer thickness can be obtained through this SPR imaging method. It is found that the SPR imaging method enables successful capture of the depthwise spatial variations of the thin frost layer, showing that the frost layer was grown over time as a result of the de-sublimation of water vapor.

1. Introduction

Ice physics has been continuously studied for a long time, and many reports have furthered understanding of the frosting process. However, it remains a significant issue affecting many industrial applications, including as safety of aircraft, refrigeration, air conditioning (in HVAC system), food preservation, power transmission lines, and wind turbine industries [1–8]. Due to the accumulation of ice on a surface, for instance, the thermal resistance and the pressure drop caused by the ice layer increase and subsequently degrade the thermal and mechanical performance of the affected equipment (e.g., heat exchangers or airplane wings [9,10]). Thus, it is necessary to determine detailed information about the frosting mechanism, which is of great importance in understanding the growth of the frost layer on a surface.

In general, the frosting mechanism consists of three different stages, including frost-seed formation and growth, phase change, and frost growth [11–13]. The first stage explains the formation of the supercooled frost-seed on a substrate and its subsequent growth. During this stage, the frost-seed remains in a supercooled state until the onset of freezing in the phase change stage. The onset of freezing occurs through two different ice nucleation modes. The first is homogeneous ice nucleation, which initiates the freezing of the seed within the supercooled condensate away from the substrate. The second is heterogeneous ice nucleation, which starts at the solid-liquid interface [14]. The onset of freezing triggers the phase change stage by the presence of ice bridging [15,16]. Recently, inter-droplet ice bridging phenomena were discovered, indicating one dominant mechanism during the phase change stage. For hydrophobic and mildly hydrophilic surfaces, the frozen seeds make an ice bridge that grows toward neighboring frost seeds, consequently creating an interconnected ice network [17]. Previously, we observed the variation in the propagation speed of the ice bridge according to the condensation mode; we had reported ice bridge propagation, and frost coverage of the whole hydrophilic surface within a minute [18]. In contrast, it took a few minutes for a superhydrophobic surface. In addition, Zhang et al. [19] also reported that the water film made the network between the droplets in the filmwise condensation mode that

* Corresponding authors.
E-mail addresses: cchoi@mtu.edu (C.K. Choi), shlee89@cau.ac.kr (S.H. Lee).

https://doi.org/10.1016/j.ijheatmasstransfer.2018.03.053
0017-9310/

resulted in much quicker freezing and ice-bridging than the dropwise condensation mode. Nath et al. [17] noted that droplets froze all at once on the hydrophilic surface where filmwise condensation occurred.

Therefore, a thin supercooled liquid layer with sub-micron scale thickness might exist, and consequently, it is frozen on the surface together with the supercooled droplet or condensates. This layer is believed to play an essential role in creating the inter-droplet ice bridge between two supercooled condensates during the phase change stage on a hydrophilic surface. Thus, the present study aims to do a quantitative analysis of a frozen thin layer that might be present near the wall. Many scientific works have been conducted for in-situ measurements of the frost layer thickness using the visualization techniques [20–24]. Most of the researchers have used a conventional optical system, such as bright field reflected microscopy (BFRM), with the use of an irradiated light source for collecting the reflected light [20]. For instance, Hermes et al. [20] measured frost thickness in the range of a few millimeters on the side of the frost. This team proposed the growth model of frost layer thickness and the density of the frost layer under a forced convection condition. Fossa and Tanda [21] investigated the frost growth behavior inside the narrow vertical channel under natural convection conditions. Also, Hao et al. [22] developed the prediction model for frost temperature, frost thickness, and heat transfer coefficient by considering the simplified geometry of the frost layer. Finally, Kandula [23,24] proposed a theoretical model of its effective thermal conductivity and assumed that the frost layer could be regarded as the porous packed beds with uniform spherical particles.

Unfortunately, the above-mentioned visualization methods have a critical/intrinsic problem, specifically, diffraction limits that prevent us from seeing the sub-micron thin layer formed on the cooled surface. In particular, a thin frost layer has sub-micron scale order, and its thickness is changed between condensates during the cooling process. Hence, a more promising visualization method with a high spatial resolution is needed to capture the variation of a thin frozen layer. In fact, surface plasmon resonance (SPR) microscopy, widely used for bio-engineering applications, breaks the diffraction limit and theoretically enables infinitely fine resolutions. It also responds very sensitively to the change in refractive index of the test medium near the metal surface (typically made of gold) during the phase change. We have successfully visualized the phase change process (e.g., evaporation and freezing) by using SPR microscopy [25,26]. As a result, we observed the presence of a thin layer near the wall and the growth of the frozen region in freezing droplets over time. Therefore, the present study reports that the nanoscale thin frost layer exists on the surface; the research uses the SPR method and conducts a quantitative analysis to provide the spatial distribution of the thin frozen layer formed between condensates. It is believed that the current study is the first to quantitatively observe the presence of this thin frost layer.

2. Experiment

SPR refers to the oscillation of free electrons that occurs at the metal-insulator interface [27]. The specimen for the SPR microscope consists of four media, namely a prism, a metal layer (gold), a test medium, and air. Based on Maxwell's Equation, the reflectance of the single wavelength light incident on a thin layer can be predicted [28]. The following Eq. (1) can theoretically predict the reflectance of the incident light on a four-layer system;

$$R = \frac{r_1[1+\exp(-2ik_2d_2)] + [r_1r_2+\exp(-2ik_2d_2)]r_3\exp(-2ik_3d_3)}{1+r_1r_2\exp(-2ik_2d_2)+[r_2+r_1\exp(-2ik_2d_2)]r_3\exp(-2ik_3d_3)} \quad (1)$$

where r_i denotes the reflection coefficient between the *i-th* and (*i* + 1)*-th* layers, k denotes the wave vector of the medium, and d_i denotes the thickness of the *i-th* medium layer. Subscripts 1, 2, 3 and 4 indicate glass (BK7), the gold layer (~50 nm), the test medium (water, ice, or air), and the bulk medium, respectively. If the thickness of the test medium is greater than the measurable thickness range of the SPR signal, the medium itself appears to be infinite, and the configuration is reduced to a three-layer system, thereby simplifying Eq. (1). The penetration depth is the distance from the metal-dielectric interface at which the intensity drops to approximately 37% (1/e) of the interface intensity. The theoretical value of the penetration depth of the SPR signals can be obtained from the expressions

$$L_d = \frac{\lambda}{2\pi}\left(\frac{\varepsilon_m+\varepsilon_d}{\varepsilon_d^2}\right)^{1/2} \quad (2)$$

$$L_m = \frac{\lambda}{2\pi}\left(\frac{\varepsilon_m+\varepsilon_d}{\varepsilon_m^2}\right)^{1/2} \quad (3)$$

where λ is the wavelength of incident light, ε_m is the real part of the relative permittivity of the metal layer, and ε_d is the relative permittivity of the dielectric layer [27]. For the incident light of λ = 600 nm on the gold, the SPR penetration depth in gold, water, ice, and air media is 35 nm, 173 nm, 180 nm, and 295 nm, respectively. The relative permittivity (ε_r) of the dielectric layer is expressed as $\varepsilon_r = n^2$ with a refractive index [27]. The refractive index (n) of the test medium (dielectric layer) depends on its phase and/or temperature. For example, n at −4.0 °C corresponds to 1.0027, 1.3330, and 1.3070 for air, supercooled water, and ice, respectively [28]. From Eq. (1), the reflectance changes sensitively with respect to the refractive index of the test medium and the incidence angle of the light.

Fig. 1 shows the SPR imaging system used in the present study. The light from the LED light source (Edmund Optics, SugarCUBE™ LED Illuminators) is converted into a single wavelength light beam through a bandpass filter (600 ± 5 nm, Edmund Optics). A collimation lens array is installed to convert the diverging light beam into a parallel light beam, as depicted in Fig. 1. Finally, p-polarized collimated rays enter the prism. The prism is composed of BK7 glass (Edmund Optics, Dove Prism), and the refractive index corresponds to 1.516 (at λ = 600 nm). The coverslip with a 50 nm gold thin layer (Platypus Tech., Au.0500.CSS) is attached to the prism by using an index matching liquid (Norland, IML150) with the same refractive index of the BK7 prism. The 7.9 × magnification lens is attached to CCD camera (JAI CM-141MCL), and the pixel resolution is 810 nm. The single-wavelength light reflected from the multiple thin layers is captured by the CCD camera at 0.2 frames per second. When the test media consists of air, water, or ice, the reflectance values based on the SPR angle are predicted using Eq. (1), as shown in Fig. 2(a). The most sensitive SPR angle for the refractive index of ice ($n_i \sim$ 1.3070 at −4.0 °C) corresponds to the incident angle of 72°. In Fig. 2(b), the bright, gray and dark regions correspond to air, water, and ice, respectively.

Before cooling the surface, 4 μl of a DI-water droplet was placed on the surface to observe the frost propagation process. The specimen was cooled from room temperature (23 ± 1 °C) to −4.0 ± 0.8 °C using a thermoelectric cooler in which the relative humidity corresponds to 20 ± 3% (at the room temperature). The surface temperature was measured *in-situ* by attaching a thermocouple to the surface of the specimen. It took approximately 3–4 min to cool down to reach −4.0 °C.

For quantitative analyses, a calibration is necessary. Calibration allows calculation of the corresponding reflectance from the pixel intensity of the experimental images. The calibration methodology used in this research is described here. First, a scale factor (S) is