ABSTRACT

Effects of the Hydrophobic Coating on the Initiation of Frost Formation Tongxin Zhang, M.S.M.E Mentor: Dennis O'Neal, Ph.D.

The growth and freezing process of water droplets forming on hydrophobic coated surface were investigated by using a digital high-speed microscopy system. The images indicated that hydrophobicity of surfaces can affect the fusion of inter-droplets to form some relative dry areas between droplets and further result in a sparser droplet distribution. These dry gaps can limit the growth rate of ice crystals and retard frost formation on the hydrophobic coated surfaces. Three empirical correlations for dimensionless freezing time of single water droplets with three various static contact angles were developed. The correlations can be used to predict dimensionless freezing time in a wide range of experimental conditions. Dimensionless freezing times among droplets with different shapes were also compared. Through analyzing the coefficients of these correlations, effects of the hydrophobic coated surfaces on the freezing process of intra-droplets are better understood. Effects of the Hydrophobic Coating on the Initiation of Frost Formation

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NOMENCLATURE

T_w	Surface Temperature
T_{∞}	Air Temperature
V	Air Velocity
RH	Relative Humidity
Am	Am Number
Re	Reynolds Number
Ste	Stefan Number
D	Body Size
$ ho_\infty$	Density of Air
μ_∞	Kinematic Viscosity of Air
\mathbf{k}_{∞}	Thermal Conductivity of Air
C _p	Special Heat of Ice
L	Latent Heat of Freezing
τ	Dimensionless Freezing Time
W	Uncertainty
SCA	Static Contact Angle
PDMS	Poly(dimethylsiloxane)
PTFE	Poly(tetrafluoroethylene)
PECVD DACS	Plasma-Enhanced Chemical Vapor Deposition Data Acquisition and Control System

LED	Light-Emitting Diode
fps	Frame Per Second
ROI	Region of Interest
NI	National Instrument
cFP	Compact FieldPoint
RTD	Resistance Temperature Device
RWHCS	The Radius of The Widest Horizontal Cross-Section
Al	Pure Aluminum
Al_Si	Silicon-Based Conformal Polymer Coated Aluminum
Al_HP	Hydrophobic Coated Aluminum

CHAPTER ONE

Introduction

Frost formation is a common phenomenon in nature as well as in engineering systems. It can form on aerospace systems, electrical power lines, and refrigerant-to-air heat exchangers in heat pump and refrigeration applications. When an object is cooled below 0°C and exposed to humid air, ice crystals can eventually form and accumulate on the substrates causing undesired consequences. For example, on the heat exchangers of heat pumps, frost can obstruct airflow between the fins and insulate the fins from the air. This can reduce the heat transfer in the heat exchanger by 50 to 75% and substantially increase the pressure drop of the air through the heat exchanger [1]. Many researchers have investigated the process of frost accretion on surfaces since the 1940s.

A five-stage model for frost formation on a heat exchanger fins was proposed by Gong et al. [2]: water condensation, liquid droplet growth, freezing, crystal growth, and frost layer growth. Many studies have characterized the effect of environmental conditions [3-5] and surface properties [6-8] on frost formation. Ambient air conditions, such as velocity, humidity, temperature, together with the surface conditions, including surface temperature, surface wettability and surface roughness, have been identified as variables affecting frost formation.

Wettability of surface can be quantitatively expressed in terms of static contact angle (SCA). Hydrophilicity is defined by a water SCA less than 90 °. On the contrary, if a surface can obtain a water SCA greater than 90 °, it is defined as a hydrophobic

substrate. With the rapid development of advanced materials, hydrophobic surfaces with larger SCAs for water have become candidates for anti-frosting or icing applications because of their exceptional performance on retarding frost formation and reducing ice adhesion strength [9-11].

Recently, high-performance computing and high-speed cameras have made it possible to explore the mechanisms that affect the initiation of frost formation. Water droplets first form on the surface, then the droplet solidifies from the first ice nucleus. When a droplet freezes, ice crystals or ice bridges then reach outward from the frozen droplet to the nearest unfrozen droplets. Once these ice crystals make contact with an unfrozen droplet, the droplet quickly freezes and becomes a new location from which ice crystals grow. During this process, evaporation of unfrozen droplets near the ice bridges also appear. The evaporated water vapor becomes the mass sources for the growth of the ice crystals and the frozen droplet, in addition to ambient humid air [12, 13].

Few authors discussed the difference of the growth mechanism on the initiation of frost formation between hydrophilic and hydrophobic surfaces. Because the shapes of the water droplets are significantly different on hydrophilic substrates versus hydrophobic surfaces, the processes of droplet growth and freezing may include different phenomena. There is a need to conduct research to compare the freezing performance of inter-droplets and intra-droplets on the surfaces of various wettability.

The main purposes of this study are to:

1. Investigate how hydrophobic coatings influence the mechanism of frost growth.

- 2. Assess the effect of advanced coatings on the period of droplet growth and freezing.
- 3. Develop correlations among flow variables, the properties of water droplets, and other measured parameters for water droplets on hydrophobic surfaces to better understand the underlying mechanism of frost growth.

To meet these goals, two types of experiments were conducted. The first, called Type-I tests, aimed to qualitatively characterize the freezing process of inter-droplets on a pure aluminum (Al) surface and the hydrophobic coated aluminum (Al_HP) surface. The second type of experiments, called Type-II tests, focused on the phase change process within single water droplets. To monitor and record the microscopic and transient freezing events, a high-speed digital microscopy system was applied in both types of tests. The results of this investigation revealed some details about the freezing behavior of early frost formation on both hydrophilic and hydrophobic surfaces. Additionally, this study also provided three empirical correlations corresponding to their different surface wettability. Through the analysis and discussion of these outcomes, the effect of hydrophobic coatings on the initiation of frost formation was comprehensively described and explained with more details.

The thesis consists of five chapters. Chapter 2 presents a review of related literature that included frost formation phenomenon, frost nucleation, defrosting coatings, and phase change process. Chapter 3 describes the experimental setup and operating procedures. Chapter 4 shows the reduced results obtained from both types of experiments. Chapter 5 discusses the results obtained in this study by making comparison with previous research.

CHAPTER TWO

Literature Review

Many researchers have investigated frost and ice formation in engineering systems. The formation of frost often occurs in two different modes: frosting under cryogenic temperature and condensation frosting. When the air temperature is below the triple point of water 0.01°C, frost is directly deposited on the cold surface. However, this phenomenon does not occur in condensation frosting where the air temperature is usually above freezing. Condensation frosting is a problem that occurs in heat pump and refrigeration systems. When the surface temperature is below the ambient dew point temperature, water condenses on the cold surface to form water droplets [14]. The size of the droplets can grow due to continuous condensation or the combination of surrounding droplets. If the surface temperature is below freezing, these droplets eventually freeze to ice and begin the process of crystal growth and densification [15]. In this study, only the mode of condensation frosting was investigated.

In this chapter, a wide range of literature relevant to this study was reviewed and organized into four topical areas: the overall understanding of frost formation, the mechanism of ice nucleation, the wettability of surfaces, and the phase-change process.

Frost Formation Phenomenon

Frost formation has been documented in the literature for over 60 years. The majority of researchers focused on the overall understanding of frost accretion. For heat pump and refrigeration systems, studies indicated that air velocity, air humidity, surface

temperature, and surface finishes had significant effects on the microstructure and properties of frost formed on a cold surface. The effects of these parameters on frost growth, however, could vary by an order of magnitude [3].

Kondepudi [5] investigated the effects of frost growth on finned tube heat exchangers under laminar flow. The fin types included louvered, flat plate, wavy, corrugated, and spine fins. Results showed that under similar conditions, louvered fins had the greatest frost accumulation while the lowest frost growth occurred on flat fins. He indicated that the condensate droplets with large diameters could be retained and spread to form a thin film on surfaces with complex geometry, such as louvered fins. In contrast, small water droplets could be retained on the surface of flat fin. Therefore, the draining property of a surface was an important factor for frost growth.

Tao et al. [16] proposed a two-stage model for frost growth with airflow on a flat plate: a crystal growth period and a fully developed frost layer growth period. First, the ice nucleated and grew from condensate droplets on the surface. After the condensate droplets were completely frozen, the ice crystal layer continued to expand and thicken by direct condensation. Many parameters, including frost height, thermal conductivity, droplets location, humidity ratio, air and wall temperature, Reynolds number, Fourier number, and Nusselt number were used as variables to the frost growth process.

Microscopic imaging has become an important tool in studying the frost growth process. Tao et al. [4] observed and recorded the early stage of frost formation from an above view using a microscopy system with 100X magnification. It was concluded that the average radius of condensate droplets varied with different surface temperatures. Gong et al. [2] also used a microscopy system to study the growth of ice crystals. They

observed the process from the lateral view of the test section. The results showed that ice crystals grew along the edge of the contact region between the condensate droplet and the cold surface, to form a cover of ice on the surface. This phenomenon indicated that the heat transfer around the wall-droplet contact area was much greater than that at the top of the droplet. The ice film formed created an insulating layer which could reduce the heat transfer between the surface and ambient air.

Na and Webb [6] revealed that the surface energy could affect the supersaturation degree of air at the cold surface where frost nucleation occurred. The surface energy was represented in terms of the water SCA. Qualitatively, higher supersaturation degree was required for frost nucleation on a lower energy surface. Shin [7] studied the effects of surface energy on frost formation. The surfaces had an SCA with water droplets at either 23 °, 55 °, or 88 °. Results showed that frost density and thermal conductivity increased over time, and the higher frost density and thermal conductivity appeared on the surface that had a lower SCA for water. Additionally, empirical correlations were also established for frost thickness, density and thermal conductivity as the functions of time and surface energy.

Water condensate droplets could form different shapes on the surfaces with various SCAs. The droplet shapes would influence the frost structures and further affect the frost properties. Hoke et al. [8] studied the effect of substrate wettability on frost properties. They observed the frost deposition on hydrophilic and hydrophobic surfaces using microscopy system and indicated that the frost layer growth on the hydrophilic surfaces was faster than that on the hydrophobic surfaces during the fully developed growth. This was due to two reasons, 1) the surfaces wettability affected the distribution

of condensate droplets, which was the initial condition for frost growth; and 2) the frost layer formed on the hydrophilic surface obtained the higher thermal conductivity.

With the development of advanced materials, hydrophobic surfaces with large SCAs have been evaluated for their potential to affect frost formation. Liu et al. [9] observed the progress of frost deposition on a cold hydrophobic substrate where SCA with water droplets was 162 °. The observation process was divided into four parts: the condensation of water droplets, the freezing process, the initial frost crystal formation, and the structure of frost layer. Compared to the conventional copper surface, the hydrophobic surface showed a strong ability to retard frost formation. The frost layer structure formed on the hydrophobic surface was thinner and looser, and easy to be removed.

Cai et al. [10] applied hydrophobic coatings and hygroscopic coatings on regular copper surfaces and compared their abilities of disturbing the frost formation process. The results demonstrated an evident effect of these coatings on the initial stage of frost growth. The frost formed on these coated surfaces was delayed and appeared to reduce frost height and provide a sparser distribution of crystals. In light of these findings, there is a promising future for the applications of these kinds of coatings to heat exchangers used in heat and refrigeration equipment.

Development of high-speed cameras has provided investigator with a tool to study the mechanism of frost growth on solid substrates. Dooley [12] studied ice propagation on four samples, whose water SCAs ranged from 25 ° to 85 °. He indicated that the freezing velocity between droplets was independent of substrate temperature and but slightly dependent on the humidity and surface energy. However, the freezing velocity

within a single droplet was determined strongly by substrate temperature and weakly by surface energy. The air humidity had no effect on the intra-droplet freezing velocity. Dooley also pointed out that ice crystals grew outward from the frozen droplet towards the nearest unfrozen droplet. During the process, evaporation of surrounding unfrozen droplets was associated as mass sources that served for the growth of ice crystals. Petit et al. [13] observed a very similar phenomenon when he studied frost growth on a substrate with 124 °SCA. They confirmed that the observed behavior of frost growth was the well-known Wegener-Bergeron-Findeisen effect, which usually occurred in atmospheric phenomena.

Frost Nucleation

Nucleation is an important process during the early stage of frost formation. The supersaturated air and the condensate droplets both need to exceed the change in Gibbs free energy, also known as the change of free enthalpy. Water molecules need to form large enough embryos and pass the barrier of nucleation to induce the phase change process [17].

Turnbull and Vonnegut [18] summarized the nucleation theory for analyzing the liquid nuclei formation between a surface and supersaturated vapor. The change in Gibbs free energy ΔG^* between a liquid nucleus of critical size and the supersaturated vapor was described as:

$$\Delta G^* = \frac{16 \pi \sigma^3}{3(\Delta G_V)^2}$$
(2.1)

where σ is the interfacial energy per area between liquid and vapor, and ΔG_V is the Gibbs free energy difference per unit volume between vapor and liquid phases. Further, the free

energy of formation ΔG_c^* of a liquid nucleus of critical size on the catalyst surface was given as:

$$\Delta G_c^* = \Delta G^* f(\theta) \tag{2.2}$$

where $f(\theta) = (2 + \cos\theta)(1 - \cos\theta)^2/4$ is a function of the SCA between the liquid and the surface of the solid catalytic body. Equation 2.2 showed a proportional relationship between the critical Gibbs free energy difference and the SCA function. Turnbull and Vonnegut also reviewed a classical expression for estimating the rate of embryo formation of heterogeneous nucleation:

$$I = A \exp\left(-\frac{\Delta G_c^*}{kT}\right)$$
(2.3)

where A is a kinetic constant and k is the Boltzmann's constant. According to Equations 2.2 and 2.3, the value of $f(\theta)$ decreases with an increasing SCA, which leads to a declining critical Gibbs free energy difference ΔG_c^* . When other parameters remain unchanged, the heterogeneous nucleation rate has a decreasing tendency with a reducing critical Gibbs free energy difference, which means that a relatively small embryo formation rate would exist for a high SCA. The tendency of SCA versus heterogeneous nucleation rate is shown in Figure 2.1.

An simple heterogeneous nucleation model was developed by Fletcher [17] and described in depth by Na [6]. The model was shown in Figure 2.2. The parameter, m, the cosine of the SCA, was defined as a function of the interfacial energies:

$$m \equiv \cos \theta = \frac{\sigma_{js} - \sigma_{is}}{\sigma_{ij}}$$
(2.4)



Figure 2.1. Static contact angle versus heterogeneous nucleation rate (Based on Equations 2.2 and 2.3).



Figure 2.2. Contact angle and the Fletcher's heterogeneous nucleation model [6].

The term σ denoted the surface energy of the interface. It was used to quantify the disruption of intermolecular bonds that occur when a surface was created. The subscripts i, j, and s represented parent phase, new phase and the substrate, respectively. The change in Gibbs free energy of heterogeneous nucleation at the critical embryo size r_c was expressed as:

$$\Delta G_c^* = \frac{16 \pi \sigma_{ij}^3}{3 \Delta \bar{g}_V^2} \frac{(2+m)(1-m)^2}{4}$$
(2.5)

$$r_c = -\frac{2\sigma_{ij}}{\Delta \bar{g}_V} \tag{2.6}$$

where $\Delta \bar{g}_V$ is the Gibbs energy change per unit volume between the parent phase and the new phase during the phase change. A greater critical Gibbs free energy change of

heterogeneous nucleation is needed when the substrate has a larger SCA for water. The tendency is the same as shown in Figure 2.1.

In addition to the theoretical analysis, experimental efforts have been made to study the environmental parameters that could potentially affect the nucleation process. Na and Webb [6] revealed that a higher water vapor supersaturation degree was required for frost nucleation on a low energy surface. In the investigation of frost nucleation on flat surfaces by Piucco [19], a basic physical criterion was developed to predict the appearance of frost nucleation as a function of the operating conditions and surface properties. The experimental results showed a good agreement with the mathematical model based on Fletcher's heterogeneous nucleation theory.

Defrosting Coatings

Investigations on frost formation demonstrate that surface wettability is a major factor influencing frost formation because wettability determines the properties of frost. In the past twenty years, interests in defrosting materials have risen as a result of the advanced material science and technology [20]. Generally, there exists mainly three types of defrosting or deicing coatings: low surface energy coatings, heterogeneous/composite coatings, and porous/ hydrophobic coatings [11].

Low Surface Energy Coatings

A low surface energy coating is fabricated by depositing a material of low surface energy on a solid surface. Commercially available poly(dimethylsiloxane) (PDMS or silicone) and poly(tetrafluoroethylene) (PTFE) are both great candidate materials. The

reduced ice adhesion on these polysiloxane-based polymers is mainly an effect of the dissimilar rheological-mechanical properties between them [11].

Croutch et al. [21] studied the properties of polymers with $-CH_3$ and $-CF_3$ groups and they reported that the maximum water repellency could be obtained if $-CH_3$ and - CF_3 groups were oriented outward from the coating surface. Deposition of self-assembled monolayers was a valid method to fabricate such layers of polymers on metal or nonmetal oxides surfaces.

Plasma-enhanced chemical vapor deposition (PECVD) can be employed to create low free energy coatings. Ji et al. [22] produced highly reactive radical species such as CF_2 • or CF_3 • during the PECVD process and subsequently deposited on aluminum surfaces to form hydrophobic fluorinated carbon coatings. The coatings produced SCAs with water as large as 130°. During fabrication, surface morphology of the coatings was tunable by adjusting vapor pressure ranging from 1 to 67 Pa. For convenience, the author classified that the range of low pressure was from 1.3 to 6.0 Pa and high pressure varied from 10.6 to 66.5 Pa. Generally, high vapor pressure often yielded rougher surface when low vapor pressure with smoother surface. The surface morphology and surface chemistry, together, determine the wettability of the coatings. The produced coatings using the above-described method could be adhered to aluminum surfaces with high affinity, yet delivering purely elastic behavior with a hardness >1.8 Gpa. The formed coatings never resulted in a degradation of substrate mechanical performance, such as delamination or brittle fracture produced by scratching, due to the qualified mechanical properties of coatings.

Heterogeneous and Composite Coatings

The main purpose of applying heterogeneous polymer coatings on a surface is to lower the ice adhesion strengths that would otherwise occur on homogeneous coatings. Murase et al. [23, 24] investigated three heterogeneous polymer coatings: organopolysiloxane grafted with fluoro polymer (A), polyperfluoroalkyl(meth)acrylate combined with hydrophobic silicon dioxide (B), and an organopolysiloxane modified with a lithium compound (LiMO₃) (C). The authors found that the water molecule showed a slight repulsion against a siloxane group, but a strong attraction to a fluorocarbon group. Furthermore, the water molecule displayed completely different orientations at the surfaces of these two groups. Due to the existence of differences in energy bond and the water molecule orientation, the ice-substrate interface was weakened with the probable creation of a wide range of dislocations and slips in the structure of the liquid-like layer.

Similar to heterogeneous coatings, composite coatings also have unique properties that their original components do not. Der Ger et al. [25] and Zhao et al. [26] used electroless deposition techniques to produce Ni-P-PTFE composition coatings. It was shown from that study that the incorporation of surfactant or PTFE particles into Ni–P matrixes had a significant influence on the surface free energy of the coatings, resulting in water SCA up to 110 °. Wang et al. [27] produced nickel/ultra-dispersed PTFE composite films by applying electrochemical deposition techniques. The PTFE particles were suspended using a cationic fluorocarbon surfactant in a Watt's nickel-plating bath and distributed homogeneously within all the composite films. The PTFE particle

concentration strongly influenced the water-repellency of Ni-PTFE composite films. The SCAs of water droplets on the surface of the composite films could reach 154.9 °.

Porous Hydrophobic Materials

When water droplets are in contact with lotus leaves, they tend to present in pearls-like shape with a large SCA. Barthlott [28], a botanist at the University of Bonn, studied natural lotus leaves and found the interdependence between surface roughness, reduced particle adhesion, and water repellency was the keystone in the self-cleaning mechanism of many biological surfaces. This phenomenon was called the Lotus-Effect. Since then, a bionic hydrophobic surface technology was developed to manufacture surfaces simulating the lotus self-cleaning mechanism. So far, the common idea underlying all these approaches was to modify a rough, patterned or porous surface, which has methyl or fluorine terminal groups [11].

Numerous hydrophobic materials have been developed [29] by either etching a given substrate, chemically depositing oxides nanoparticles, spin-coating or spraying polymers, using nanolithography. The primary strategy is to create a porous structure. These methods were motivated by the analysis of wettability of porous materials from Wenzel [30] and Cassie [31]. In their theories, a substrate was denoted as textured surface if its topography was altered by chemical or mechanical modification. Wenzel found that the true and observed SCAs on a textured surface were different due to the existence of surface roughness. He proposed that:

$$\cos\theta_{\rm c} = R\cos\theta \tag{2.7}$$

where θ_c is the SCA on a perfectly smooth surface, θ is the observed SCA, and R is the roughness factor defined as the ratio between the actual and geometric surface area. Since then, Cassie further modified Wenzel's equation for textured porous surfaces. The formation of liquid-air and liquid-solid interfaces at the pores of these surfaces led to wider apparent SCAs. Cassie proposed the following alternative expression:

$$\cos\theta_{\rm c} = f\left(1 + \cos\theta\right) - 1 \tag{2.8}$$

where f is the area fraction of solid-liquid interface. Both Equations 2.7 and 2.8 show that the surface roughness factor has a great impact on the apparent SCA.

Many porous hydrophobic materials have been assessed under frosting conditions and have shown acceptable icephobic behavior. Momen et al. [32] spray-coated a silicone rubber/stearic acid mixture on a flat surface. The stearic acid concentration governed the surface morphology and further determined the wettability of the coatings. The water SCA could reach 160° in their study. Moreover, the hydrophobic coating was able to retard the freezing process and showed good icephobic behavior at the supercooled temperature. Jafari et al. [33] created a micro-nanostructured aluminum oxide underlayer on the alloy substrate and coated the rough surface with RF-sputtered polytetrafluoroethylene (PTFE). A "bird's nest"-like structure was shown under scanning electron microscope. The coating exhibited a high SCA of 165 °. The authors also observed the high quantities of CF3 and CF2 groups by using X-ray photoelectron spectroscopy. Techniques utilized by these groups further enhanced the hydrophobic behavior of the coatings. Under atmospheric icing conditions, the surfaces showed great icephobic performance. The ice adhesion strength was 3.5 times lower than that on the polished aluminum substrate.

The mechanism of delayed frost growth on hydrophobic surfaces was investigated by Hao et al. [34]. Their findings supported that only hydrophobic surfaces with jumping condensates were able to retard frost formation. If the structure gaps on surfaces are narrow enough (scale of submicrometer or nanometer), the water condensed within these nanogaps could automatically ascend to the top of the structures and form droplets on the surface. This condensate is called jumping condensate. These droplets can form a Cassie state, in which air was trapped in the nanogaps between surface features to form a composite solid-air hydrophobic surface. In light of these results, a key approach in delaying frost growth was to retard the initial ice crystal formation.

Phase Change Process

The phase change process of condensate droplets is an important stage in frost formation. Investigators aim to evaluate the parameters involved in the phenomenon and their relationship to develop models for predicting the freezing process.

Usually, the liquid-solid phase change process occurs at a constant temperature. However, when the droplet is contacted with a cold substrate (below 0 $^{\circ}$), the transition might take place over a small temperature interval (supercooling). During the transition process, the droplet is separated into two phases with different thermophysical properties with energy released at the liquid-solid interface. Generally, the phase change interface could be regarded as an isothermal or quasi-isothermal boundary.

The freezing of condensate droplets could be classified as the Stefan problem where the temperature distribution in a homogeneous medium undergoes a phase change. The liquid droplet and the substrate were initially at a constant temperature above the freezing point, followed by a sudden decrease in plate temperature to a value below the

freezing temperature. After a period of undercooling, the liquid-solid phase change of water droplet occurred. The Stefan number, a dimensionless number from heat transfer, was employed to analyze the process quantitatively.

$$Ste = \frac{C_p(T_\infty - T_w)}{L}$$
(2.9)

where C_p is the specific heat of liquid and L is the latent heat of freezing. The Stefan number is a key number in solving the problem of heat transfer during solidification or melting [35].

Some researchers [36] hypothesized that internal circulation inside a droplet might affect the temperature field of the droplet and further influence the phase change process. The circulation could be caused by either buoyancy force or surface tension force. Sparrow et al. [36] studied the transition from natural convection controlled freezing to conduction controlled freezing. At the early period, when the liquid was at a temperature above freezing, the temperature differences through the liquid induced a natural convection motion, which could retard the freezing process. Later, as the differences decreased over time, the freezing rate tended to be controlled by heat conduction. Experimental results showed that the frozen samples for the early freezing time were smooth-surfaced and tapered, while the surfaces of those for the later time were overlaid with a thicket of large discrete crystals. These characteristics validated the authors' analysis about the roles of natural convection and heat conduction in phase change process.

In contrast, Lorenz et al. [37] postulated the importance of natural convection to droplets temperature distribution during phase change process was negligible. He studied

a hemispherical water droplet on a flat isothermal surface. He indicated that the heat flux through the water droplet from the cold surface was much larger than the heat capacity of the water droplets, so the transient effect of natural convection was insignificant on the temperature distribution. Based on his calculations, he concluded that the increase of heat transfer in the model considering internal circulation was less than a few percent higher over the one controlled by heat conduction.

Gong [38] experimentally explored heat transfer during the freezing of condensate droplets. The hemispherical droplets were initially at a steady state on a cold surface when microscopy observation was applied to monitor and record the freezing process of the droplets. Through analyzing all the parameters involved in the process of phase change, Gong developed an empirical correlation with four dimensionless numbers to predict the freezing time of water droplets.

Summary

Several conclusions could be drawn from the previous review of literature:

1. Frost formation has been well studied experimentally and theoretically. Researchers have evaluated factors including the environmental parameters and the substrate properties, and their specific effects in different stages of frost formation. Some investigations also tended to explore both the mechanisms of frost growth and the initiation of frost formation from a micro perspective. Recently, a growing number of researchers focused on using advanced coatings to reduce frost formation and improve defrosting performance. However, few of them discussed how these coatings might retard frost growth or explain how these coatings influence the mechanism of frost growth.

2. Many advanced coatings have been fabricated and used for deicing. These coatings either reduce the ice adhesion strength or decrease the ice nucleation. However, frost formation on these coatings has been extensively evaluated only in their influence on frost growth, but not how they affect the transition from droplet growth to freezing. Thus, there is an opportunity to assess the effect of advanced coatings on this period of droplet growth and freezing.

3. The process of phase change in a hemispherical water droplet previously investigated by Gong [38] provided correlations among flow variables, the properties of water droplets, and other measured parameters. These results were helpful to understand the underlying mechanism of frost growth, yet similar analysis for other shape droplets, with different SCAs, was missing.

CHAPTER THREE

Experimental Facility

To investigate the effect of hydrophobic coatings on the initiation of frost formation, experimental work was needed. The experimental facility used in this study was originally designed by previous graduate students at the Energy Systems Laboratory of Texas A&M University and recently rebuilt at Baylor University. As shown in Figure 3.1, the facility consisted of four main components: A) a closed-loop psychrometric wind tunnel, B) a test section, C) a digital microscopy system and D) a data acquisition and control system (DACS).



Figure 3.1. Overall view of the experimental setup.

Closed-loop Psychrometric Wind Tunnel

The wind tunnel was made of several modular sections, including an adjustablespeed blower, two cooling coils, a duct heater, a humidification system and a dry gas injection system. Figure 3.2 gives the schematic drawing of the wind tunnel loop system with relative locations of these components. The function of the wind tunnel was to provide continuous, conditioned airflow over an interchangeable test surface mounted to the bottom side of the test section.



Figure 3.2. Schematic overview of the wind tunnel loop system.

The test loop was constructed of 2.54 cm thick aluminum coated fiberglass duct board. Each duct module was secured with weather stripping and C-clamps to minimize the loss of heat and air leaks. A VWR recirculating chiller (model 13271-212) was used to provide 50/50 propylene glycol-water coolant for the primary and secondary cooling coils. The piping system was designed to connect the chiller and cooling coils. Two pairs of rotameters and globe valves were set in the return line of each cooling coil to control and measure the coolant flow rate. A 120VAC blower adjusted with a silicon controlled rectifier was applied downstream of the primary cooling coil to provide the air circulation in the wind tunnel as high as 15 m/s. The precise control of the air humidity could be realized by simultaneously using a steam injection nozzle and a gas injection nozzle. In addition, a 120VAC, 500 W electric duct heater, powered by a programmable Red Lion Model TCU controller, was used as the final air temperature control module. The whole wind tunnel system allowed the air temperature to vary from 1 $^{\circ}$ to 55 $^{\circ}$ and the air humidity to change from 20% to 95%.

Test Section

Acrylic Test Stage

The acrylic test stage was located at the top of the bypass duct, the central component of the wind tunnel loop. The detailed schematic of test stage is shown in Figure 3.3.



Figure 3.3. Detailed schematic of test section module.

The test stage was made of machined optical transparency acrylic glass that had a low thermal conductivity ($0.2 \text{ W/m}\cdot\text{K}$). A 0.17 mm thick single pane of optical quality glass on the acrylic panel of the test stage allows the in-situ, top-down viewing in the

microscopy system, without disruption to the experimental process. On the lateral sides of the test stage, two similar viewports provided a horizontal view of the test surface. At the upstream of the test section, a group of 1.5 mm diameter, 20 mm long, cylindrical, plastic flow straighteners were applied at the inlet of the inner annulus to generate a laminar flow. A relative humidity (RH) sensor and thermocouple arrays, mounted upstream from the test surface, were used for the measurement of the inlet air temperature and the relative humidity, respectively. Furthermore, there was a type-T thermocouple and a hot wire anemometer downstream from the test surface for the measurement of the outlet air temperature and the air velocity.

Phase-change Module

A phase-change module mounted at the bottom of the test section was the central component of the experimental system used to control the test surface temperature. The main components consisted of a Peltier module, a copper heat sink with a 30 mm × 30 mm surface plate, and two type-T thermocouples. Figure 3.4 gives the detailed view of the thermal stage. The Peltier module, powered by a Kepco Inc. 100 W analog-controlled, DC regulated, power supply (model ATE-15-6M), was a Marlow Industries 36 W thermoelectric cooler (model DT12-4-01LS). It was located right below the test surface. An electromechanical switch was applied to alter the working condition of the Peltier module in cooling or heating mode. The copper heat sink with an internal circuit could conduct the heat from the Peltier module to the coolant (50/50 propylene glycol-water solution), which was provided by a Cole-Parmer (model 12108-10) benchtop circulating chiller. The two type-T thermocouples were for the measurement of the surface temperature and the heat sink temperature. All the components in the phase-

change module were bonded by a thermally conductive material (Berquist Co. Hi-Flow® 565 UT) with 0.254 mm thickness and the 52 °C phase-change temperature. The phase-change module was capable of controlling the test surface temperature from -20°C to 120°C.



Figure 3.4. Exploded view of thermal stage.

Digital Microscopy System

A digital microscopy system was used to monitor the test surface through a customized high speed imaging system associated with a compound microscope. It allowed for a real-time observation of all experiments at the different magnitudes, providing more detailed information about the fast and micro freezing process. Figure 3.5 gives the overall view of the digital microscopy system.

Compound Microscope

An upright style Nikon stereoscopic microscope (model LV150) was mounted above the test section to observe the experimental process vertically or horizontally. Two long-working-distance objective lenses, 5X magnification and 20X magnification, were
used in this study and mounted on a four-objective capacity rotating nosepiece turret. A customized light-emitting diode (LED) illuminator was applied to replace the original Phillips LUXEON (model K2-TFFC) LED aiming to minimize the effect of heat generated from the light source. A machined copper heat sink with serval cylindrical fins was used to drive the heat away from the customized LED illuminator. When needed, a modified OEM halogen lamp could be used to cooperate with the LED illuminator if needed.



Figure 3.5. Overall view of the digital microscopy system.

A two-degree-of freedom telescoping boom stand supported the whole microscope system to observe the experimental process right above the viewport of the test section. For a horizontal view of the test surface, a machined metal extension rod was connected between the focusing mounting bracket of the telescoping stand and the optical microscope. In addition, to reduce the vibration from the system components, such as chillers and blower, the telescoping stand was put on a Newport RS 2000TM active pneumatic vibration isolation table.

High-speed Camera

To provide a real-time visualization of the freezing processes on the test surface, an IDT-Redlake M3 high-speed digital camera was attached to the upper viewport of the Nikon microscope and stabilized with a C-clamp. The camera operated with a 15.4 x 12.3 mm proprietary complimentary metal-oxide-semiconductor photodiode array. Under the maximum resolution, 1280 by 1024 pixels, the optimal operating frame rate could reach 520 frames per second (fps). The high speed camera was connected with a 32 GB RAM workstation through a BitFlow-Karbon CL frame grabber card, which was directly installed on the north-bridge slot of the PCI Express bus on the motherboard of the workstation. In this configuration, recorded images were able to be stored in the RAM directly during the experiment and then quickly transferred to a 4 TB USB 3.0 external hard drive when the test was completed.

IDT's proprietary Motion Studio software was installed to handle the camera during the image acquisition. This software could adjust the image recording rate from 10 to 520 fps at a full frame resolution, 1280 by 1024 pixels. To optimize the recording of the entire freezing process, the camera operated in a circular recording mode, meaning that the images were stored in a pre-allocated memory area in a first in, first out style. The real-time monitoring images of the freezing process were displayed on the screen through the interface of Motion Studio software. When the experimental process was completed, an external event trigger would be activated to finish the image acquisition and record images to memory. In the study of scale relationship between pixel and length for each objective lens, an original equipment manufacturer Nikon stage micrometer slide with 0.01 mm scale marks was secured to a 3.0 by 3.0 cm unpolished, low reflective aluminum surface. After taking a series of images, a single calibration image for each objective lens was extracted and post-processed using Adobe Photoshop 64 Professional. By counting the pixels between length markings on the stage micrometer, the pixel to length relationship could be calibrated. Table 3.1 shows the calibration data for each objective lens and the geometric specifications of the region of interest (ROI) under each objective lens.

Table 3.1. Camera image calibration data for each objective lens.

Objective Lens	Calibration value	ROI length	ROI width	ROI area
	μm /pixel	μm	μm	μm^2
5X	2.38	3048	2438	$7.43x10^{6}$
20X	0.59	753	602	$4.54x10^5$

Data Acquisition and Control System

The DACS was used to measure and control multiple analog input/output signals from various instruments simultaneously. A National Instruments (NI) Compact FieldPoint (cFP) 1804 controller was connected to the workstation and operated by a NI LabVIEW program. The controller was used to handle an 8 channel cFP-TC-120 thermocouple module for measuring the temperature and an 8 channel cFP-AIO-610 analog input/output module for dealing with the measurement of air velocity, air relative humidity and the adjustment of the Peltier Module voltage.

Temperature Measurement

Serval type-T thermocouples and thermocouple arrays were engaged in the measurement of multiple temperatures in the experimental setup. Table 3.2 gives the list

of measured variables with the instrumentation used for each measurement. All these thermocouples needed to be calibrated before initiating the experiments. A circulating 50/50 propylene glycol-water bath and a NIST calibrated Traceable platinum resistance temperature device (RTD) (model 23609-228) were employed during the calibration process. The calibration ranged from -20 °C to 30 °C, which within the expected temperature range during the experiments. For the 0 °C calibration, an ice bath of high purity water was used. The max error was ± 0.75 °C.

Channel ID	Measurement variables	Instrumentations
TC0	Chiller tank temperature	Type-T thermocouple
TC1	Cooling coil supply temperature	Type-T thermocouple
TC2	Peltier heat sink temperature	Type-T thermocouple
TC3	Test surface temperature	Type-T thermocouple
TC4	Test section traverse temperature	Type-T thermocouple
TC5	Test section air inlet temperature	Type-T thermocouple array
TC6	Test section air outlet temperature	Type-T thermocouple array
AI0	Airstream bulk temperature	RH transmitter
AI1	Airstream bulk RH	RH transmitter
AI3	Test section air velocity	A constant temperature anemometer

Table 3.2. List of measured variables with instrumentations.

Velocity Measurement

The air velocity was measured by a TSI 1750 constant temperature hot-wire anemometer (model 1201 sensor probe). The anemometer was calibrated in a custom designed calibration facility at Baylor Subsonic Wind Tunnel Laboratory. The calibration range was from 0 m/s to 12 m/s.

Relative Humidity

The air relative humidity and the bulk air temperature at the inlet of the test section was measured by a Vaisala HMT-333 relative humidity sensor. For a more accurate relative humidity data during experimental process, the sensor needed to be calibrated by the saturated salt solution method [39, 40], which was easy to use, accurate and cost-effective. The calibration salts, MgCl₂, NaCl, and K₂SO₄, were purchased from VAISALA and used to prepare the non-volatile and chemically stable saturated solution following the manufacture's instruction. When calibrating, the solution was contained in a 50 mL glass beaker covered with a modified rubber stopper. The instrument probe and a secondary reference thermocouple were secured inside the glass beaker by the stopper. The glass beaker was subsequently put into a standard household refrigerator (4 $^{\circ}$). The temperature and relative humidity of the salt solution were monitored with the multimeter thermocouple and the instrument control panel, respectively. For each salt solution, the process took 45 to 60 minutes. The calibration range was from 33.6% to 98.5% with the max error $\pm 1.7\%$.

Images Acquisition System

The Images acquisition system was described in the section of digital microscopy system. During the experiments, the pictures of the freezing process were taken at 10 fps, 20 fps or 100 fps for different purposes. An imaging software called ImageJ was utilized for determining droplet size and SCA during single water droplet test. Furthermore, a Matlab code developed by Dooley [12] was used to analyze the freezing front of frost formation process.

CHAPTER FOUR

Experimental Procedure

The experiments for this study consisted of two main types tests, with the objectives of observing the inter-droplet freezing (Type-I) and intra–droplet phasechange process (Type-II) during frost formation. In Type-I experiments, the samples were placed under simulated frosting air and surface conditions typically experienced by heat exchanger fins in heat pumps. Water vapor would condense on the sample surface and eventually solidify to form a frost layer. These experiments were designed to qualitatively study the differences of the early stage frost formation between hydrophilic surfaces and hydrophobic surfaces. For Type-II experiments, a single water droplet was implanted on the test surface using a syringe or a pipette. The high speed camera was applied to monitor the freezing process from the lateral viewport of the test section. The aim of these tests was to quantitatively investigate the impacts of surfaces with different wettability on freezing time of a single water droplet. The two types of tests, collectively, should expand the understanding of the mechanism of frost formation on diverse surfaces.

Test Surface Preparation

All samples used in this study were aluminum based and well-polished before employing in the tests. Table 4.1 shows the characteristics of these four specimens.

Surface ID	Coating	Size (cm ²)
Al_01	None	3×3
Al_HP_01	Lotus Shield hydrophobic coating	3×3
Al_02	None	3×3
Al_HP_02	Lotus Shield hydrophobic coating/half coated	3×3
Al_Si_02	Silicon-based conformal polymer coating/half coated	3×3

Table 4.1. Coating and size of test surfaces.

Sampling for Type-I Experiments

Two 3 cm by 3 cm aluminum surfaces was prepared for making the test samples of Type-I experiments. In the sampling stage, both surfaces were thoroughly cleaned before usage to avoid any contamination that might affect the frost nucleation during the experiments. The entire cleaning process consisted of regular dish soap wash, sonication in acetone and drying. The samples were first placed in a glass beaker with the cleaning solution and manually shaken orbitally for 5 minutes until there was no visible contamination. Samples were then transferred into another beaker filling with deionized water. Second, the samples were moved to a JT Baker low-water acetone containing glass beaker, and subjected to sonication for 15 to 30 min. The samples were then immediately removed into JT reagent grade methanol before drying. Meanwhile, an electric hotplate was preheated to around 40 °C to evaporate the methanol retained on samples. Then the Al sample was prepared. During the entire cleaning process, great care was taken when handling the samples to avoid any scratch on the surfaces.

After cleaning, the other aluminum surfaces were placed in a spin-coater with the hydrophobic coating material from Lotus Shield to make the Al_HP surface. A computer cooling fan was modified and used as the spin coater. First, the sample was fixed in the spin-coater plate, and generally covered with the hydrophobic coating using a disposable transfer pipette. Spinning velocity was next increased by adjusting the supplying voltage

of the fan and droplets were slowly deposited on the coating at the rate of one per 1 to 2 seconds for around 10 seconds. Lastly, the sample was dried on 40 $^{\circ}$ C electric hotplate for 15 minutes. Once the test surface preparation was completed, the samples were kept in petri dishes in a desiccator.

Sampling for Type-II Experiments

To study the effects of different surfaces on the freezing time of a single water droplet, three specimens were employed, a polished Al sample, a silicon-based conformal polymer coated aluminum (Al_Si) sample, and an Al_HP sample. The same cleaning steps as Type-I experiments were applied to prepare these samples. In the process of chemical coating, the Al_HP sample was made by using the same method mentioned before. The only difference was that only half of the area of the sample was coated. The other was covered with tape during the coating process (the tape would be removed once the sampling process was finished). If a single droplet was placed on a fully-coated surface, it tended to absorb water vapor from the ambient air and grow then combine with some condensing water droplets which could reduce its SCA and result in a longer waiting time before freezing begins. All these influences could reduce the accuracy of the measurements. Since the freezing process occurred transiently on the pure aluminum surface (the uncoated half), the single water droplet began to freeze sooner than if on a fully coated surface. The Al_Si sample utilized the same "half-coated-region" strategy. The specimen was covered with the coating sprayed on the surface and then immediately spin-coated for about 10 seconds. The Al_Si sample was then dried at 200 °C at least 1 hour and then kept in a desiccator for 24 hours.

Measurement of Static Contact Angle

The images used to measure the SCAs on each specimen were captured by the digital camera system. The samples were horizontally mounted in the test section. The microscopy system was set in the horizontal working mode. The height of the microscope was then carefully adjusted to gain an appropriate view position. A Signature SeriesTM VWR single channel, variable volume, electronic pipetter (model 516) was employed to implant water droplets with various volumes from 1.0 μ L to 5.0 μ L. By calculating the average value, the effect of droplet volume on the SCA was minimized. Two LED lamps provided the light source from the backside of the droplet to enhance the contrast degree of the droplet contour. Software *ImageJ* was employed to measure these SCAs manually from the images. An example was shown in Figure 4.1. The same measurements (5 times per sample) were repeated on different specimens. The results were shown in Figure 4.2 and Table 4.2.



Figure 4.1. Measurement of the SCA of a droplet with a volume of 3.0 μ L.



Figure 4.2. SCA versus droplet volume for all test surfaces.

Surface type	Averaged SCA (degree)	Standard Deviation(degree)
Al	74.6	2.1
Al_Si	86.5	1.1
Al_HP	127.1	1.9

Table 4.2. Volume-averaged SCAs for all test surfaces.

Experimental Process

As mentioned previously, the experiments in this study consisted of two main type tests: observing the overall frost formation process (Type-I) and investigating the single water droplet freezing process (Type-II). Five experimental procedures were involved in either type of the experiments, and they included system warmup, droplet formation onto the test surface (condensation in Type-I tests or implantation in Type-II tests), measurement and recording of the experimental variables, acquisition of the images during the freezing process, and data analysis.

Overall Frost Formation Process (Type-I)

The wind tunnel connection, the coolant leaks, and all power wires connection, were first inspected before operating any system. The test section chiller and the wind tunnel cooling coil chiller were then powered on and were set at $0 \,^{\circ}$ and $-5 \,^{\circ}$, respectively. The power source for DACS were then turned on. Finally, the workstation with the LabVIEW control interface was started to monitor all these experimental variables.

During Type-I experiments, four parameters had to be controlled and adjusted. These included the test surface temperature (T_w) , the test section inlet air temperature (T_{∞}) , the relative humidity (RH) of air, and the test section air velocity (V). To simulate the nominal refrigeration and heat pump operating conditions, the air temperature (T_{∞}) and velocity (V) were set at constant values of 5°C and 5 m/s, respectively. After the target air temperature was reached, the steam boiler was activated (if needed) and manually adjusted to provide the target RH value 65%. If the RH was too high for the desired conditions, dry nitrogen gas was injected into the air stream through the dry air nozzle. The Peltier module was kept in heating mode to avoid any condensate droplets formation on the test surface. The optical microscopy system was configured to use magnifications of 5X and 20X, corresponding to recording the images at 10 fps and 100 fps, respectively. Once the steady environmental conditions (T_{∞} =5°C, V = 5m/s and RH = 65%) were reached, the phase-change module was switched to the cooling mode to decrease the surface temperature down to -10°C.

The high-speed camera operated in the continuous recording mode to record the water droplets condensation, growth, and solidification process. The test matrix for Type-I experiments was shown in Table 4.3.

Test	Test	Magnification	Image	Environmental
Surface	Number	Magnification	Rate	Condition
Al	1	5X	10 fps	$T_w = -10^{\circ}C$
	2	20X	100 fps	$T_{\infty} = 5^{\circ}C$
Al_HP	3	5X	10 fps	V = 5 m/s
	4	20X	100 fps	RH = 65%

Table 4.3. Test matrix for Type-I experiments.

When the first sign of freezing appreared in the ROI, the camera trigger was switched on to record the freezing process until the condensed water droplets in the monitored region were completed frozen. The data file was marked simultaneously with the activation of the trigger to synchronize the recorded images and the time series data. In the end, the images from both types of samples were compared and analyzed, for an evaluation of the surfaces on the initiation of frost formation. Repeated experiments were conducted to capture enough details in the frost formation and to improve data accuracy and confidence.

Freezing Process of a Single Water Droplet (Type-II)

The Type-II experiments followed a similar system warm-up operations to the Type-I tests, such as system inspections and equipment initiation. However, Type-II experiments were designed to be conducted in different conditions to investigate the effect of environmental variables on the droplets' freezing time on various surfaces. The experiments differed in that the air temperature was varied from 4°C to 24°C, the surface

temperature from -5° C to -13° C, the radius of the droplet from 0.5 mm to 1.5 mm, and the air velocity from 0.20 m/s to 1.85 m/s.

When the test channel reached steady state, a single water droplet was implanted on the dry test surface by using a HAMILTON 5 μ L syringe and a Signature SeriesTM VWR single channel, variable volume, electronic pipettor (model 516). To minimize the impact on the evaporation and condensation at the interface between the ambient air and droplet, the test surface temperature was maintained at a value slightly greater than the dew point of the ambient air. Once the droplet was on the surface, the phase-change module was immediately switched to the cooling mode. The microscopy system was set at 5X magnification and 20 fps image rate. Two LED lights were applied to highlight the freezing front of the droplet.

After the entire freezing process was recorded, the *ImageJ* software was employed to gain the geometric information and the freezing time of the droplet. The same type of experiments were conducted on three different surfaces, a polished Al sample, an Al_Si sample and an Al_HP sample.

CHAPTER FIVE

Experimental Results and Data Analysis

The experimental work was conducted to explore the effects of hydrophobic coatings on the initiation of frost formation. Data consisted of a series of images captured by the digital microscopy system and the environmental parameters recorded by the data acquisition system during the freezing events. In this chapter, the experimental data and its analysis are discussed.

As mentioned previously, the overall experiment was divided into two categories: Type-I and Type-II. The results from both groups revealed the effects of the microbehavior of the freezing droplets from the test substrates. Type-I experiments provide a qualitative analysis from the standpoint of inter-droplets performance while Type-II experiments concentrated on the effects from surfaces on a single droplet phase change process.

Type-I Experiments

In this group of experiments, a series of images were recorded above the test surfaces by the high-speed camera in 5X and 20X magnifications during the condensation and freezing events. Through data acquisition and control system, all the environmental parameters were maintained in the same conditions: air velocity 5 m/s, air relative humidity 65%, wall temperature -10°C, and air temperature 5°C. The observation with 5X magnification and 10 fps showed the behavior of the bulk freezing front of the droplets on Al and Al_HP surfaces. The method used was the optical phase tracking technique developed by Dooley. [12]. This technique is based on the phenomenon that opacity of ice is higher relative to water. Through contrasting the intensity of major visible droplets, the frozen ones can be readily identified by their darker appearance. To further study the freezing performance, the observation with 20X magnification and 100 fps was conducted.

Observation with 5X Magnification

The bulk freezing front is of considerable interest because it defines local regions on the surface where the freezing occurs. Figures 5.1 and 5.2 present the digital image sequence recorded from the Al and Al_HP tests, respectively. The images were processed by the optical phase tracking technique mentioned previously. Within the region of interest (ROI), the black area was the region occupied by the frozen droplets while the white dots indicated the unfrozen ones.



Figure 5.1. Images of freezing front position on the Al surface at (a) t=0 s (b) t=10 s (c) t=20 s (d) t=30 s.



Figure 5.2. Images of freezing front position on the Al_HP surface at (a) t=0 s (b) t=50 s (c) t=100 s (d) t=150 s.

The freezing process occurred on the ROI of Al surface with a SCA of 74.6° lasted around 40 sec. It was easy to identify the boundary between frozen and unfrozen droplets and observe the motion of the bulk freezing front. However, the freezing events on the Al_HP surface (127.1° SCA) presented huge differences compared to that on the Al surface. The freezing front line was hard to be recognized. It seemed like the water droplets had frozen randomly on the ROI. The entire freezing process lasted about 200 sec, five-fold longer than that measured on the Al surface. In other words, the freezing process on the Al_HP surface was significantly retarded. The reason will be presented in detail in next section, the observation with 20X magnification

Further observations on the 5X raw images of freezing at t = 0 sec (shown in Figure 5.3) revealed several important details about the features of the Al and Al_HP surfaces undergoing dropwise condensation. The initial time t=0 sec was define as the

moment right before the first frozen droplet appeared on the ROI. Before freezing occurred, some tiny droplets uniformly condensed on the Al surface. With the increasing time, these droplets then gradually grew and coalesced with surrounding droplets. They distributed evenly on the Al surface, with relatively uniformed sizes. There were about 300 visible droplets per mm^2 on the ROI at t=0 sec. In contrast, during the condensation process on the Al_HP surface, although some tiny droplets uniformly condensed with similar sizes at the beginning period, the changes of these liquid droplet sizes varied a lot at the rest of this period. These droplets presented very circularly spherical shapes, which didn't perform on the Al surface. The distribution of these droplets was apparently sparser than those on the Al surface. There were about 100 visible droplets per mm^2 on the ROI at t=0 sec. Moreover, quite large droplets (marked by ellipses in Figure 5.3 b) were noted, and appeared randomly on the ROI of the Al_HP surface. Interestingly, there appear to be dry areas around the large droplets, with barely any water on the surface. The diameter of the greatest droplets on the Al_HP surface was about 0.18 mm, but this value for the Al surface was around 0.11 mm.



Figure 5.3. 5X Raw images of freezing at t = 0.0 s (a) on test surface Al (b) on test surface Al_HP (representative large droplets referred in red circles).

Another raw image recorded at t = 35.0 second is Figure 5.4. It shows the freezing pattern on the Al_HP surface. The greater height in these large droplets (exemplified in red circle) made them more susceptible to the disturbance by the ambient air flow, resulting in an inevitable longer waiting time before freezing than those small ones in the yellow subregions. Additionally, some area without water condensate appeared along the edges of the frozen region, as indicated by yellow circles in the raw image of Figure 5.4.



Figure 5.4. 5X Raw image of freezing at t = 35.0 s on test surface Al_HP. (Red circle showed the location of a representative large droplet. Yellow circles showed the location where freezing had occurred.).

To gain more details about the differences in freezing behaviors of both substrates, the microscope was increased to 20X magnification and the frame rate of the camera was increased to 100 fps.

Observation with 20X Magnification

The higher magnification and imaging frequency could offer more detailed information about the freezing process. The raw images of Al test at the start and end of freezing times are presented in Figure 5.5. On the Al surface, the locations of the primary droplets, as well as the total number of the main droplets were almost unchanged during the freezing process. On the Al_HP surface (shown in Figure 5.6), however, the positions of the primary droplets in some regions shifted significantly throughout the test. The population of the main droplets had a noticeable decline from around 40 droplets down to less than 30 droplets while some droplets' sizes were visibly increased. The main reason of these changes is the different behaviors of droplet coalescences on the Al and Al_HP surfaces, which will be described in details later.



Figure 5.5. 20X freezing images on the Al surface at (a) t = 0.00 s and (b) t = 6.64 s.



Figure 5.6. 20X freezing raw images on the Al_HP surface at (a) t = 7.52 s and (b) t = 128.03 s. (The ellipses marked the main region that droplet coalescences happened.)

Through further observation, examples of droplet coalescence on the Al surface and the Al_HP surface were captured and shown in Figure 5.7 and Figure 5.8, respectively. The coalescences of droplets had different performances on these two surfaces. On the Al surface (shown in Figure 5.7), the new formed droplet (marked in the circle in Figure 5.7 (c)) still possessed the regions of its parent droplets (marked in the circle in Figure 5.7 (b)). On the contrary, the new formed droplet (marked in the circle in Figure 5.8 (c)) on the Al_HP surface evidently had a smaller covered area than its parent droplets (marked in the circle in Figure 5.8 (b)) and created a relative dry boundary. The different performances of droplet coalescences on the two surfaces were due to the variances of the SCAs. The lower surface tension in droplets with lower SCAs tended to spread the droplet on the surface. Conversely, droplets with a higher SCA had a tendency to maintain the shape close to a sphere and reduce contact area as a result of higher surface tension. The motion of the droplet coalescence would usually sweep past the subregion occupied by the former droplets and leave a "dry area".



Figure 5.7. The fusion of droplets on the Al surface: (a) the entire ROI at t=6.07 s (b) the magnified subregion at t=6.07 s (c) the magnified subregion at t=6.08 s.



Figure 5.8. The fusion of droplets on the Al_HP surface: (a) the entire ROI at t=7.52 s (b) the magnified subregion at t=7.52 s (c) the magnified subregion at t=7.53 s.

During the freezing process on the Al_HP surface, droplet fusion randomly occurred in the form of either liquid droplet growth or droplet solidification, as shown in Figures 5.9 and 5.10. The relatively dry regions formed during the freezing period were marked by the ellipses. In these zones, ice crystals, formed from frozen droplets to liquid droplets, grew slowly due to limited mass resource besides the ambient air flow. Consequently, these subregions became the gaps to retard the freezing front from directly crossing the frozen-unfrozen boundary. The last two images in Figure 5.10 showed that the ice crystal (marked in a square frame) failed to bridge the gap at about 50 sec. If the gap (e.g. the area that the dash arrow pass in Figure 5.10) was too broad for ice crystals to cross, an alternative freezing route would be chosen by the frosting front (e.g. the solid arrow in Figure 5.10), regardless of time consumed. This phenomenon was an important approach on how the hydrophobic surface could retard the freezing process of condensate droplets.



t = 12.94 s

t= 12.95 s



t = 17.53 s

t = 17.55 s



t = 26.28 s

t = 26.29 s

Figure 5.9. Image sequence of droplets fusions during the freezing process on the Al_HP surface. (The ellipses marked the dry areas and the circles pointed out coalescences of droplets. The curve presents the freezing front line.)



t = 27.95 s

t = 27.96 s



t = 43.84 s

t = 43.85 s



t = 51.97 s

t = 100.00 s

Figure 5.10. Image sequence of "dry area" formation during the freezing process on the Al_HP surface. (The ellipses marked the dry areas and the circles pointed out coalescences of droplets. The curve presents the freezing front line. The dash arrows showed the ideal freezing direction and the solid arrows gave the real freezing direction.)

Type-II Experiments

Unlike the freezing behavior among the condensate droplets explored in Type-I experiment, the Type-II test series focused on the phase change process in a single water droplet. The freezing process was recorded by the digital microscopy system under a range of test conditions. By analyzing the consecutive images of the phase change process, the droplet freezing time was obtained.

In the test series, the range of the air velocities were from 0.20 m/s to 1.85 m/s with \pm 2% variation. The surface temperature controlled by the Peltier module was ranged from -5°C to -13°C and the variation during a test was within 1°C. The range of the ambient air temperature was from 4°C to 24°C with the \pm 1°C variation. The dimension of the water droplets was described by the radius of their widest horizontal cross-section (RWHCS). For the AL_Si and Al_HP samples, RWHCS was the radius of the droplet. However, for the Al sample, it was the radius of the circle formed by the contact area between the droplet and the cold substrate. The maximum RWHCS was set at 1.5 mm according to the width of the observed region, while the minimum was set at 0.4 mm due to the volume limitation from the electronic pipetter and the hypodermic syringes. Additionally, the relative humidity of air was controlled from 24% to 54%, which was within the range of the environmental conditions relevant to heat pumps.

The experimental results of the Type-II tests were put in tabular form (see Appendix A) and in graphic representation. The uncertainty values were at a 95% confidence level. The correlation equations predicting the droplet dimensionless freezing time for three different surfaces were developed from the experimental data.

Dimensional Analysis

Dimensional analysis is commonly used to identify the physical relevant variables of an individual process. To study the freezing of hemispherical droplets, Gong [38] applied the method to find some relevant dimensionless groups that could be directly used for the interpretation of the droplets freezing process on a cold substrate. The technique has the advantage to extend the results of a few tests run to a wide range of experimental conditions. In our study, the technique was modified to explore the phase change process of droplets on three different substrates: the Al surface, the Al_Si surface, and the Al_HP surface.

Based on Gong's analysis [38], the Buckingham Pi theorem, the key theorem in dimensional analysis, was used in this study. All the variables related to the droplet freezing process are listed as below: freezing time *t*, ambient air temperature T_{∞} , surface temperature T_w , air velocity *V*, body size *D*, air density ρ_{∞} , air viscosity μ_{∞} , air thermal conductivity k_{∞} , ice special heat C_p , and water latent heat of freezing *L*. The body size *D* is defined as the volume-based particle size:

$$D = 2\sqrt[3]{\frac{3 * volume}{4\pi}}$$
(5.1)

According to the Buckingham Pi theorem, the freezing time was firstly written as a function of these eight variables mentioned above:

$$t = f\left(T_{\infty} - T_{W}, V, D, \rho_{\infty}, \mu_{\infty}, k_{\infty}, C_{p}, L\right)$$
(5.2)

There are four primary dimensions: time, temperature, length, and mass. Then through the analysis of the theorem, the Equation 5.2 can be rewritten as a function of several dimensionless Π groups:

$$\frac{V}{D}t = f\left[\frac{k_{\infty}(T_{\infty} - T_{w})}{\rho_{\infty}V^{3}D}, \frac{\rho_{\infty}VD}{\mu_{\infty}}, \frac{C_{p}(T_{f} - T_{w})}{L}\right]$$
(5.3)

or

$$\tau = f \left(Am_{\infty}, Re_{\infty}, Ste_L \right) \tag{5.4}$$

where τ is the dimensionless freezing time, defined as the ratio of the distance covered by the ambient air during time *t* to the droplet body size *D*. It provides a possibility to compare the freezing times of different droplets frozen at different conditions. The greater the τ is, the longer the freezing takes. T_f is the water freezing temperature, 0°C. The parameter, Am_{∞} , is defined as the ratio of heat flux through the droplet by conduction from the cold substrate to the kinetic energy flux of the ambient air. The influence of this parameter on the droplet phase change process will be discussed later. The Reynolds number Re_{∞} could reflect the effect of ambient air flow on the dimensionless freezing time. The Stefan number Ste_L represented the importance of the heat capacity of the solid relative to the latent heat during the phase change process.

The experimental results of the freezing of the droplets with three different SCAs were presented in the next section. Both dimensional and dimensionless results in a tabular form were listed in Appendix A while the dimensionless graphic presentations were showed later in this chapter.

The Freezing Time

The freezing problem was expressed by four parameters presented in Equation 5.4 through the dimensional analysis. When the experimental data were plotted on a log-log scale, shown in Figures 5.14, 5.15, and 5.16, the dimensionless freezing time displayed nearly linear relationships with the variables Am_{∞} , Re_{∞} , and Ste_L , respectively.



Figure 5.11. An example of water droplet formed on Al_Si surface.



Figure 5.12. An example of water droplet formed on Al_HP surface.



Figure 5.13. An example of water droplet formed on Al surface.



Figure 5.14. The experimental results on the Al_Si surface: (a) effect of Am_{∞} on freezing time, (b) effect of flow field on freezing time, and (c) effect of heat transfer on freezing time.



Figure 5.15. The experimental results on the Al_HP surface: (a) effect of Am_{∞} on freezing time, (b) effect of flow field on freezing time, and (c) effect of heat transfer on freezing time.



Figure 5.16. The experimental results on the Al surface: (a) effect of Am_{∞} on freezing time, (b) effect of flow field on freezing time, and (c) effect of heat transfer on freezing time.

Take the results of the Al_Si test as an example. Figure 5.14 (a) showed the effect of Am_{∞} on the dimensionless freezing time. The discrete experimental data presented a strong relationship between the two variables. The dimensionless freezing time τ linearly decreased with increasing Am_{∞} . For a small Am_{∞} , the kinetic energy flux of the ambient flow was sufficiently large relative to the heat flux through the body by conduction. Hence, the surface temperature of the droplet tended to be close to the ambient temperature to extend the freezing process. With increasing Am_{∞} , the kinetic energy effect gradually became less important compared to conduction effects, and the temperature of the droplet began to approach the plate temperature. The dimensionless freezing time was thus shortened.

In Figure 5.14 (b), τ increased along with Re_{∞} . The Re_{∞} number increases with rising body size of the droplet or air velocity. For large Re_{∞} , the heat transfer between the droplet and ambient air was great, which means the droplet temperature was more easily raised by the ambient air. As a result, the dimensionless freezing time of water droplet was prolonged.

The relationship between τ and Ste_L , shown in Figure 5.14 (c), was similar to the correlation between τ and Am_{∞} . With increasing Stefan number, the enhanced temperature difference accelerated the cooling process of the droplet and decreased the dimensionless freezing time.

The correlations among these parameters were developed for all the three test series, and had the functional form:

$$\tau = Intercept + C_1 Am_{\infty} + C_2 Re_{\infty} + C_3 Ste_L$$
(5.5)

where C_1 , C_2 , and C_3 were the coefficients determined from a regression analysis. For the calculations, the properties of air flow (density, kinematic viscosity, and thermal conductivity) were evaluated at the film temperature $T_{film} = \frac{T_{\infty} + T_W}{2}$. An example of freezing of a droplet on the Al_HP surface is shown in Figure 5.17.



Figure 5.17. An example of freezing of a droplet on the Al_HP surface (The straight line presents the freezing front.).

Table 5.1 lists the values of these coefficients corresponding to their droplets SCAs. Among the three groups, the maximum values of coefficient C_1 appeared at 127.1 ° correlated equation as well as the largest absolute value of coefficient C_3 . Since both Am_{∞} and Ste_L were dependent on the surface temperature, the freezing process on the Al_HP surface was the most prone to be influenced by the surface temperature difference. Besides, the geometry of the droplet with 127.1 °SCA could reduce the water-surface contact area to decrease the conduction effect. The geometry also could separate the air flow to bring more upper warm air to the lower region to increase the temperature inside the droplet, as shown in Figure 5.17. All these influences resulted in the largest interception and a relatively large coefficient C_2 (Table 5.1) for this correlated equation. The 74.6 ° correlated equation also carried a relatively great coefficient C_2 , but the reason was different. For the droplet with 74.6 °SCA, the ratio of water-surface contact area to body volume was the highest compared to other two types of droplets. Consequently, its dimensionless freezing time was more likely affected by the heat transfer between the droplet and the ambient air. Additionally, for the 86.5 ° correlated equation, the coefficient C_2 and absolute value of coefficient C_3 were the smallest, suggesting lowest sensitivity of the Al_Si surface to fluid flow and surface temperature. The ranges of the dimensionless parameters are shown in Table 5.2.

Table 5.1. Constant values of the correlated equations.

SCA	74.6 Degree	86.5 Degree	127.1 Degree
Intercept	4888.38	6031.18	16426.38
C_1	0.0853	0.0754	0.3676
C_2	107.91	57.47	134.40
C_3	-132872	-115672	-352615
R Square	0.9584	0.9712	0.9562

SCA	74.6 Degree	86.5 Degree	127.1 Degree
Am_{∞}	20~38300	25~16200	20~18000
Re_{∞}	20~230	30~200	40~240
Ste _L	0.035~0.077	0.036~0.073	0.029~0.073

Table 5.2. Ranges of the dimensionless parameters: Am_{∞} , Re_{∞} , and Ste_L .



Figure 5.18. Separation of air flow passing by the droplets with 127.1 °SCA.

The results of regression statistics were summarized in Table 5.1, for an illustration of the correlation between the correlated regression lines and the experimental data. All the values of R-square, also known as the coefficient of determination, were above 0.90. These results indicated that the correlated equation can explain over 90 percent of all the variability of the experimental data around its mean. The graphical representations of the correlated results and the experimental data for the three different test series were displayed in Figures 5.18, 5.19, and 5.20, respectively. The correlated equations provided the feasibility to extend the results of a few experiments to a wide variety of other conditions.



Figure 5.19. The comparison of experimental data and correlation predictions for the Al_Si tests.



Figure 5.20. The comparison of experimental data and correlation predictions for the Al_HP tests.



Figure 5.21. The comparison of experimental data and correlation predictions for the Al tests.

Summary

In this chapter, the Type-I and Type-II experimental results were shown. The freezing phenomenon of the inter-droplets was revealed, in particular the formation of the "dry area". Furthermore, the dimensionless freezing time of a single droplet was also described. Lastly, three empirical correlations corresponding to three different SCAs were developed for the study of the heat transfer during the phase change process of droplets.
CHAPTER SIX

Discussion

The effect of the hydrophobic coating on the initiation of frost formation has been investigated in this study. A comparison of observed phenomena occurring on surfaces of different wettability was provided and should help provide some insights in the stages of frost formation. In this chapter, we further discussed the results obtained in the experiments. Two sections were included corresponding to the two types of tests we conducted previously.

The Freezing Mechanism of Inter-droplets in Type-I Experiments

Thanks to the rapid development of computers and high-speed cameras, the micro and fast freezing phenomenon has been explored to a great extent to reveal the undergoing mechanism, especially about the differences of freezing on the hydrophilic and hydrophobic surfaces. Previous investigations [8-10, 13, 34, 41] indicated that the hydrophobic surfaces have a strong ability to retard frost formation, mainly because of the potential barrier of nucleation for these surfaces being much larger than that for hydrophilic substrates. In this experiments, similar results were gained after comparing the freezing behavior happened on the Al and Al_HP surfaces. However, we observed some interesting phenomena that were not reported before, which could aid in describing the freezing event in a more comprehensive manner.

Water Droplets Distribution

Water condensation is considered as the first step of frost formation at the air and surface temperature considered here. At the end of the condensation period, the size, number, and pattern of water droplets depend on the surface. Hoke [8] studied the effect of substrate wettability on frost properties and indicated that the distribution of water on the surface was dependent on the SCA of the surface. The distribution of droplets could determine the frost structure and further significantly affect the conductivity of the frost layer which directly regulates the growth rate of frost at the later stage of frost formation. Moreover, the number and distribution of water droplets on the substrate could impact the density of the initial frost layer. So the exploration of the droplets distribution on the hydrophilic and hydrophobic surfaces in this study is quite meaningful.

With the help of the high-speed camera operating at 100 fps and microscope with 20X magnification, these images provided some unreported information about the distribution of water droplets. First, the vapor water started condensing on the cold surfaces and formed many liquid droplets. Due to the differences of SCAs, the droplets on the Al and Al_HP substrates showed different shapes; flatter on the Al substrate, and closer to a sphere on Al_HP. The various geometries lead to the different behaviors during the coalescence and growth of droplets. For the Al surface, the new coalesced droplet occupied the previous regions of parent and daughter droplets. This phenomenon mainly happened before freezing. Conversely, the coalescence and growth on the Al_HP surface occurred during both the condensation and freezing processes. The form of this kind of coalescence could be liquid-liquid droplets or liquid-solid droplets. The new droplet was pulled in to form a sphere-like droplet with a smaller occupancy area

compared to the parent and daughter droplets. Once the freezing started on the surfaces, the distribution of water droplets on the Al surface changed slightly as well as their sizes and amount. However, on the Al_HP substrate, the coalescences constantly altered the distribution pattern on the surfaces. The new coalesced droplet would sweep past the surrounding subregion to merge more droplets into it and further change the distribution of the condensate (see Figure 6.1). As the results of the performances mentioned above, the droplets on the Al_HP surface had a sparser distribution, larger droplet sizes, and fewer droplets.



t = 17.53 s t = 17.54 s t = 17.55 s Figure 6.1. An example to show the motion induced by the fusion of droplets.

Similar observations about the droplets distribution were reported previously by Huang et al. [41], Hoke et al. [8], and Cai et al. [10]. However, they focused on the effects of the distribution on frost properties, such as the thermal conductivity, density, and growth rate of the frost layer, without any further exploration into the mechanism for its formation. Instead, these experimental results offered an integrated description about the influence of substrate wettability towards the distribution of droplets on the initiation of frost formation.

Growth of Ice Bridges and Formation of Dry Area

In characterizing frost growth, the same descriptions of the condensate freezing process were reported by other investigations [12, 13, 34]. A droplet starts solidification from the first ice nucleus and then forms several ice crystals or ice bridges outward the nearest unfrozen droplets. Once these ice crystals contact the target droplet, the latter immediately freezes and becomes a new ice point from which ice crystals grow. The chain reaction is the most primary mechanisms of frost freezing. Additionally, evaporation of unfrozen droplets near the ice crystals and the frozen droplets was also an important phenomenon during the process of the chain reaction.

In these experiments, this freezing pattern was also observed on the Al and Al_HP surfaces. Interestingly, there were some obvious differences between the freezing performances on these two surfaces. On the hydrophilic surface, the freezing front was continuous and flowing, and the overall freezing process on the ROI was relatively fast. In contrast, for the hydrophobic surface, the freezing front was stuttered owning to the existence of dry areas on the surface and incoherent growth of ice crystals. The freezing front had to progress slowly droplet by droplet in discrete steps across the hydrophobic substrate. Evidently, the dry gaps among the droplets had a significant effect on the speed of frost growth. For the formation of these dry zones surrounding the frozen droplets, Hao et al. [34] came up with another possible explanation. When studying the mechanism of delayed frost growth on the hydrophobic surface with jumping condensates, it was observed that the region near the ice crystals was generally dry. Since the ice crystals yet appeared and grew, Hao et al. proposed that the mass sources for the growing ice crystals and frozen droplets were the humid air and the evaporating condensate droplets, and the

growth of mass was through the vapor-ice deposition. Similarly, the frozen droplets could also obtain the mass for growth from both the ambient air and the evaporated water. However, our experiments revealed that the fusion of droplets tremendously contributed to the formation of dry gaps among droplets on the Al_HP surface, which has never been reported before. Above all, we concluded that the evaporation of unfrozen droplets and the fusion of droplets held induce the formation of the dry areas on the Al_HP surface caused by the growth of ice crystals and frozen droplets.

Retardation of the Frost Formation

About the reasons for the retardation of frost formation on the substrates with low surface energy, many investigations have given their explanations [8, 41]. The effects of surfaces are considered to impact at every stage of frost formation, including the period of water condensation, the process of droplets freezing, and the growth of frost layer.

First, frost crystals nucleation on a hydrophobic surface requires more activation energy to form an ice nucleus than those on a hydrophilic substrate. Thus, the ice nucleation rate on the former surface is lower due to the heterogeneous nucleation theory [8]. Additionally, we believe that the taller droplet with larger SCA is closer to the upper warm air. Because of the above two factors, the period of droplets condensation on the substrate with low surface energy tends to be longer.

For the freezing process of droplets, Huang et al. [41] indicated that the large SCA on the hydrophobic surface allowed the droplet to take on a smaller contact area between the condensate and the substrate. Both the heat transfer capability and droplets freezing rate were reduced resulting in longer freezing process compared to that on the hydrophilic surface. Furthermore, in our experiments, we noticed that the special dry

areas among droplets on the Al_HP surface could be another factor prolonging the freezing process. The growth rate of ice crystals was stalled in the dry gaps between droplets. When the gaps were wide enough, the freezing front would choose another path which took a longer time. Therefore, both the freezing velocities of the inter droplets and inner droplets on the low energy surface are less than those on the hydrophilic substrate.

Last, the stage of later frost growth was also influenced by the surface wettability. As previously mentioned, the surface wettability could affect the droplet distribution as well as the conductivity of the frost layer. On the hydrophobic surface, the thermal conductivity of frost layer is lower. As a result, its frost layer is thinner than that on the hydrophilic surface [8]. Even though these experiments were not related to this period of frost formation, we analyzed the formation of droplets distribution on the surfaces with different wettability in the first section.

The above description mapped out the entire process of how the surface wettability retardation of the frost growth influenced by surface wettability, which would facilitate an overall comprehension over the behaviors of frost formation.

The Analysis of the Dimensionless Freezing Time in Type-II Experiments

The technique used in this type-II experiments was modified from Gong's method [38] in the study of the freezing of hemispherical droplets. In his analysis, the freezing time was regarded as a function of many system parameters: body size D, surface temperature T_w , ambient air temperature T_∞ , surrounding air velocity V, and fluid properties. One thing needs to be noted is that the body size D was defined as the diameter of the droplet by Gong. When there was only one type of body shape (hemisphere) involved in the study, the diameter was an acceptable parameter to

represent a characteristic of the water droplet. The change of the body size refers to the variance of the droplet volume. However, in this study, there were three different geometries of water droplets required to be investigated, including the 74.6°, the 86.5°, and the 127.1 °SCA droplets. When three different SCA droplets had the same diameter, their volumes were significantly different. Hence, another parameter needed to be introduced to represent the body size for comparing dimensions of the droplets with different non-spherical shapes. The volume-based particle size given in Equation 6.1 (the same as Equation 5.1) was chosen as the body size D in this study. For the droplets on the different surfaces, if their volume-based particle sizes as well as all environmental conditions were equal, the differences in their freezing time were generated by the variation of substrates. For condensates on the same surface, the change of the volumebased particle size indicated the alteration of the droplets volume. Based on the above analysis, it is believed that the volume-based particle size was capable of not only representing the geometric information of the water droplets in this phase-change problem, but also comparing the droplet freezing times tested on the different surfaces in a more feasible and reasonable manner.

$$D = 2\sqrt[3]{\frac{3 * \text{volume}}{4\pi}}$$
(6.1)

To verify the viability of this parameter, we employed both methods to analyze the experimental data collected during the tests on the Al_Si surface. Two groups of reduced data were presented in tabular forms (see Appendix A) and in graphic form (see Figures 6.2, 6.3, and 6.4). The uncertainty values of the experimental results were at a 95% confidence level.



Figure 6.2. The comparison of experimental data and correlation predictions for the Al_Si tests using (a) the diameter of droplet as body size and (b) the volume-based particle size as body size



Figure 6.3. The experimental results on the Al_Si surface: (a) effect of Am_{∞} on freezing time, (b) effect of flow field on freezing time, and (c) effect of heat transfer on freezing time. .(The body size is the volume-based particle size.)



Figure 6.4. The experimental results on the Al_Si surface: (a) effect of Am_{∞} on freezing time, (b) effect of flow field on freezing time, and (c) effect of heat transfer on freezing time.(The body size is the diameter of droplet.)

As shown in Figures 6.3 and 6.4, the dimensionless freezing time displayed good linear relationships with Am_{∞} , Re_{∞} , and Ste_L number when the data were plotted on a loglog scale. They even had the same pattern of data distribution. The only difference was the scale of the variables. From the analysis of the dimensionless parameter equations, the Am_{∞} , Re_{∞} , and dimensionless freezing time τ were linear functions of body size D, but the Ste_L number was not dependent on this variable. As a result, the Am_{∞} , Re_{∞} , and dimensionless freezing time τ would be scaled down at a certain rate if we modified the body size from Gong's method to that used in these experiments. The rate for the tests on the Al_Si surface was around 0.83 which was the ratio of the diameter of droplet to the volume-based particle size. For the Ste_L number, its value did not change along with the alternative.

Figures 6.2 illustrated the correlation relationships obtained from both methods. The pattern of data distribution was found similarly. Same as the above dimensionless groups, the only variance of two graphs was in the scale. The magnification rate was around 1.32. Furthermore, we acquired decent R-square values from both methods, 0.9712 for the volume-based particle size and 0.9686 for the diameter of droplet. It implied that both approaches were qualified for the study of the dimensionless freezing time of droplets on a chosen surface.

Since there were three different substrates investigated in this study, the volumebased particle size was a more appropriate parameter in representing the droplet characteristic related to the phase-change process and making the comparison of dimensionless freezing different SCAs more meaningful.

Summary

In this chapter, we discussed the experimental results with other related research in serval aspects, including the formation of droplet distribution, the importance of dry area on the hydrophobic surfaces, and the qualification of this correlation method.

1. The droplets distribution had a great impact on the frost properties which could further regulate the growth of frost layer. For the low energy surfaces, the water droplets presented in the manner of sparse distribution in small amount, which was caused by the fusion of droplets during the process of condensate growth and freezing. Moreover, the motion induced by the fusions could allow the newly formed droplet to sweep past the nearby area and further reduce the number of droplets.

2. The dry gaps appeared on the low energy surface were formed by the fusion of condensate and the evaporation of those small mass droplets near the growing ice bridges. The growth rate of ice crystal was quite slow. If the gaps were too wide , the freezing front would continue in an alternative path although it takes more time. This phenomenon was considered as one of the main retarding factors of the frost growth.

3. Compared to the diameter of droplet, the volume-based particle size was a better parameter to represent the droplet geometric properties related to the phase-change process. Applying this variable offered more reasonable comparison between the dimensionless freezing time of droplets and different irregular shapes. Through analyzing the coefficients of these dimensionless parameters in the correlation equations, the potential effects of the different surfaces on the freezing process of water droplets were revealed.

APPENDICES

APPENDIX A

Type-II Experimental Data

Table A.1.	Type-II	experimental	data for t	he Al test	series.

	V(m/s)	$T_{\infty}(^{\circ}C)$	$T_w(^{\circ}C)$	a(mm)	h(mm)	D(mm)	t(s)	Am_{∞}	Re_{∞}	Ste_{L}	τ	$\tau_predict$	W_{τ}	W _{Am}	W _{Re}	W _{Ste}
1	0.28	23.5	-12.1	1.3340	1.0210	1.8677	9.00	16278.77	38.17	0.073	1362.46	759.961	27.34	1401.03	0.80	0.011
2	0.39	23.9	-11.5	1.4685	1.1424	2.0709	14.30	5494.53	58.49	0.069	2707.05	2510.505	54.32	472.89	1.23	0.011
3	0.37	21.6	-10.0	1.4351	0.9948	1.9248	14.95	6269.22	51.43	0.060	2873.74	3009.297	57.66	539.56	1.08	0.009
4	0.20	21.7	-12.3	1.4197	1.1138	2.0097	9.50	38208.21	29.82	0.074	964.80	1570.353	19.36	3288.39	0.63	0.012
5	0.50	21.5	-11.3	1.4197	1.0686	1.9731	12.20	2627.90	70.93	0.068	3064.25	3767.608	61.49	226.17	1.49	0.011
6	0.49	21.5	-10.0	1.3756	1.0377	1.9137	13.65	2777.20	67.19	0.060	3466.13	4412.033	69.55	239.02	1.41	0.009
7	0.48	21.5	-10.0	1.3114	1.0139	1.8443	10.85	2908.66	64.56	0.060	2850.35	4139.45	57.19	250.33	1.36	0.009
8	0.27	21.2	-10.1	1.1150	0.8711	1.5754	7.55	19079.51	30.98	0.061	1302.86	1815.735	26.14	1642.08	0.65	0.010
9	0.37	20.4	-10.0	1.1912	0.9068	1.6638	9.70	7042.89	44.44	0.060	2147.65	2321.022	43.09	606.15	0.93	0.009
10	0.39	20.2	-10.0	0.7925	0.6045	1.1079	4.15	8544.17	31.73	0.060	1478.49	1077.584	29.67	735.35	0.67	0.009
11	0.42	19.8	-10.4	0.6593	0.4689	0.8940	2.55	8837.32	27.23	0.062	1194.19	298.4653	23.96	760.58	0.57	0.010
12	0.39	20.0	-9.2	0.5391	0.4189	0.7598	1.95	12843.62	21.26	0.055	991.56	951.6422	19.90	1105.39	0.45	0.009
13	0.49	19.1	-9.1	0.4665	0.3475	0.6454	1.60	7189.05	22.92	0.055	1212.33	728.0411	24.33	618.73	0.48	0.009
14	0.53	18.8	-9.0	0.6962	0.5188	0.9633	3.70	3639.25	37.39	0.054	2052.42	2066.283	41.18	313.21	0.78	0.008
15	0.56	16.6	-8.9	0.8520	0.6664	1.2045	7.90	2331.10	49.13	0.053	3659.15	3301.16	73.42	200.63	1.03	0.008
16	0.57	16.6	-12.9	0.4355	0.3641	0.6346	1.10	4775.56	26.69	0.077	984.48	-2097.18	19.75	411.01	0.56	0.012
17	0.82	14.6	-8.3	0.9282	0.8425	1.4054	14.40	566.20	84.52	0.050	8388.94	7447.266	168.33	48.73	1.77	0.008
18	0.78	13.6	-7.2	0.9925	0.7830	1.4084	13.15	587.47	80.83	0.043	7296.88	7926.893	146.42	50.56	1.70	0.007
19	0.79	12.8	-12.5	0.6355	0.5188	0.9159	2.30	1061.93	53.87	0.075	1972.09	837.4939	39.57	91.39	1.13	0.012
20	0.77	12.1	-11.5	0.7211	0.6021	1.0500	2.75	929.54	60.23	0.069	2007.76	2308.848	40.29	80.00	1.26	0.011

21	0.77	12.0	-6.4	0.5736	0.4617	0.8214	6.50	917.08	46.73	0.038	6116.46	4912.45	122.73	78.93	0.98	0.006
22	0.82	11.5	-6.1	0.4736	0.4094	0.7007	4.55	847.71	42.52	0.037	5348.05	4691.148	107.31	72.96	0.89	0.006
23	0.94	10.6	-9.2	0.8437	0.6807	1.2094	8.25	363.43	85.17	0.055	6436.65	6783.388	129.15	31.28	1.79	0.009
24	0.95	10.1	-7.9	0.8473	0.6950	1.2237	10.45	319.50	86.73	0.047	8130.68	7983.239	163.15	27.50	1.82	0.007
25	1.00	9.6	-8.9	0.7664	0.6759	1.1446	6.65	298.67	85.80	0.053	5822.56	7084.752	116.83	25.70	1.80	0.008
26	1.23	8.2	-7.2	0.9139	0.7259	1.3009	10.05	118.57	119.61	0.043	9508.38	12071.55	190.79	10.20	2.51	0.007
27	1.22	8.0	-7.5	0.9496	0.8401	1.4204	11.60	111.62	129.70	0.045	9963.68	12920.83	199.93	9.61	2.72	0.007
28	1.16	8.2	-5.9	1.1876	0.8758	1.6359	25.35	103.64	141.09	0.035	17958.17	15423.39	360.34	8.92	2.96	0.006
29	1.31	7.8	-8.2	0.4022	0.3475	0.5950	2.00	222.79	58.36	0.049	4392.68	4674.747	88.14	19.17	1.23	0.008
30	1.40	7.4	-8.0	0.4974	0.4617	0.7613	3.10	137.64	79.82	0.048	5685.35	7142.449	114.08	11.85	1.68	0.008
31	1.39	6.5	-7.6	0.8116	0.6688	1.1746	7.15	82.92	122.94	0.046	8468.03	12109.29	169.92	7.14	2.58	0.007
32	1.50	5.2	-6.9	1.4232	1.1186	2.0163	31.80	32.86	227.80	0.041	23634.28	23977.65	474.23	2.83	4.78	0.006
33	1.50	5.0	-5.9	1.1614	0.8806	1.6194	20.55	36.83	183.24	0.035	19097.98	19965.99	383.21	3.17	3.85	0.006
34	1.77	4.9	-5.9	0.8152	0.7259	1.2231	13.70	29.87	162.55	0.035	19793.35	17732.79	397.16	2.57	3.41	0.006
35	1.69	4.9	-6.0	1.1400	0.9853	1.6867	25.45	24.54	214.95	0.036	25559.65	23307.05	512.87	2.11	4.51	0.006
36	1.63	4.9	-6.0	0.9591	0.8616	1.4450	18.85	31.90	177.79	0.036	21337.81	19297.84	428.15	2.75	3.73	0.006

	V(m/s)	$T_{\infty}(^{\circ}C)$	$T_w(^{\circ}C)$	a(mm)	h(mm)	D(mm)	t(s)	Am_{∞}	Re_{∞}	Ste_L	τ	$\tau_predict$	\mathbf{W}_{τ}	W_{Am}	W_{Re}	W _{Ste}
1	1.10	7.2	-10.0	0.8116	0.7283	1.2221	5.20	197.09	101.35	0.060	4663.63	4937.92	93.49	17.36	2.13	0.009
2	1.12	7.3	-10.0	0.8723	0.7902	1.3194	6.40	172.20	111.75	0.060	5431.91	5533.73	108.90	15.17	2.34	0.009
3	0.83	10.2	-10.0	1.1971	1.0805	1.8077	9.80	357.45	113.02	0.060	4523.46	5620.68	90.68	31.48	2.37	0.009
4	0.66	6.9	-10.0	1.1281	1.0186	1.7038	8.80	624.86	85.74	0.060	3431.66	4073.05	68.80	55.04	1.80	0.009
5	0.66	6.7	-10.0	1.1186	1.0020	1.6830	8.40	645.65	83.76	0.060	3278.15	3960.83	65.72	56.87	1.76	0.009
6	0.52	7.6	-10.0	1.0448	0.9472	1.5810	7.00	1476.31	61.86	0.060	2292.32	2764.84	45.96	130.03	1.30	0.009
7	0.52	7.3	-10.0	1.0305	0.9639	1.5830	8.30	1425.20	62.37	0.060	2731.44	2790.30	54.76	125.53	1.31	0.009
8	0.52	7.8	-10.0	0.9389	0.8330	1.4063	6.10	1652.59	55.38	0.060	2262.56	2405.72	45.36	145.56	1.16	0.009
9	0.55	8.3	-10.0	0.9199	0.8973	1.4423	6.60	1405.17	59.93	0.060	2522.47	2648.57	50.57	123.77	1.26	0.009
10	0.34	22.1	-12.0	1.4780	1.3447	2.2403	14.25	7647.72	54.80	0.072	2144.00	1437.69	42.98	673.60	1.15	0.011
11	0.33	22.4	-11.0	1.4447	1.2900	2.1703	14.25	8249.95	51.82	0.066	2169.20	2005.09	43.49	726.65	1.09	0.010
12	0.29	21.5	-10.0	1.4280	1.2757	2.1458	15.30	11061.54	45.72	0.060	2102.92	2559.70	42.16	974.29	0.96	0.009
13	0.38	21.0	-12.0	1.2923	1.1519	1.9399	9.90	6061.69	53.32	0.072	1925.87	1233.10	38.61	533.91	1.12	0.011
14	0.32	20.3	-10.0	1.4340	1.2519	2.1315	17.00	8047.79	50.10	0.060	2585.17	2584.28	51.83	708.84	1.05	0.009
15	0.64	20.0	-10.0	0.6331	0.5831	0.9653	4.15	2242.89	45.07	0.060	2765.12	1857.70	55.43	197.55	0.95	0.009
16	0.43	20.3	-10.0	0.8758	0.8211	1.3468	7.10	5433.66	42.05	0.060	2269.81	1924.62	45.50	478.59	0.88	0.009
17	0.43	20.1	-9.0	0.6783	0.6212	1.0314	4.50	6632.89	32.43	0.054	1896.22	2155.42	38.01	584.22	0.68	0.008
18	0.48	19.7	-9.0	0.7211	0.6616	1.0975	5.70	4625.93	37.98	0.054	2480.93	2323.11	49.74	407.45	0.80	0.008
19	0.50	17.5	-12.1	0.6033	0.5950	0.9511	2.80	4760.33	34.92	0.073	1466.80	8.25	29.41	419.29	0.73	0.011
20	0.65	13.7	-8.1	1.1436	1.0710	1.7577	14.00	865.40	83.73	0.049	5154.21	5292.88	103.33	76.22	1.76	0.008
21	0.69	12.0	-7.0	1.0758	0.9948	1.6434	15.00	651.45	84.25	0.042	6342.49	6069.23	127.15	57.38	1.77	0.007
22	0.71	11.5	-11.0	0.9948	0.9163	1.5168	6.30	759.74	81.05	0.066	2964.51	3120.41	59.43	66.92	1.70	0.010
23	0.65	11.3	-11.2	1.1448	0.9449	1.6581	7.15	927.44	80.54	0.067	2794.18	2965.09	56.02	81.69	1.69	0.011
24	0.72	10.3	-6.0	0.9092	0.8616	1.4054	12.80	584.85	75.00	0.036	6573.44	6225.89	131.78	51.51	1.57	0.006
25	0.82	9.6	-6.0	0.8378	0.7997	1.2996	9.80	405.67	79.21	0.036	6201.83	6454.33	124.33	35.73	1.66	0.006
26	1.10	9.8	-9.0	1.0103	0.9187	1.5310	9.45	171.96	126.08	0.054	6798.23	7050.52	136.29	15.15	2.65	0.008
27	1.38	10.0	-8.1	1.0722	0.9925	1.6387	13.45	78.62	168.36	0.049	11318.58	10097.26	226.91	6.92	3.53	0.008

Table A.2. Type-II experimental data for the Al_Si test series.

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28	1.36	10.2	-9.0	1.0746	0.9996	1.6462	10.55	85.68	167.59	0.054	8735.34	9429.59	175.12	7.55	3.52	0.008
29	1.29	8.6	-7.0	1.1103	1.0424	1.7086	15.10	79.72	164.51	0.042	11409.47	10638.68	228.73	7.02	3.45	0.007
30	1.77	6.8	-8.0	0.7735	0.7616	1.2184	6.30	40.80	162.07	0.048	9136.79	9802.25	183.17	3.59	3.40	0.008
31	1.77	6.9	-8.0	0.7152	0.7140	1.1343	5.40	44.31	150.86	0.048	8414.14	9158.27	168.68	3.90	3.17	0.008
32	1.79	6.6	-7.0	0.7402	0.7354	1.1712	6.80	37.38	157.77	0.042	10417.84	10248.14	208.85	3.29	3.31	0.007
33	1.83	5.7	-7.3	0.9211	0.8782	1.4281	10.35	27.62	196.50	0.044	13231.57	12265.23	265.26	2.43	4.12	0.007
34	1.69	4.9	-6.0	0.8782	0.8497	1.3714	9.75	30.35	174.80	0.036	12040.54	11919.59	241.38	2.67	3.67	0.006
35	0.28	21.3	-10.0	1.3102	1.1900	1.9843	13.20	13310.28	40.75	0.060	1889.66	2443.56	37.88	1172.36	0.85	0.009
36	0.27	21.5	-11.0	1.3138	1.1710	1.9720	10.80	16134.98	38.58	0.066	1478.28	1838.47	29.64	1421.16	0.81	0.010

	V(m/s)	$T_{\infty}(^{\circ}C)$	$T_w(^{\circ}C)$	a(mm)	h(mm)	D(mm)	t(s)	Am_{∞}	Re∞	SteL	τ	$\tau_predict$	Wτ	WAm	WRe	WSte
1	0.25	23.9	-12.2	1.1674	1.8612	2.4134	27.40	17925.26	44.04	0.073	2869.95	3150.87	57.49	1542.60	0.92	0.011
2	0.26	23.9	-11.0	1.1472	1.7707	2.3234	29.70	17041.57	43.07	0.066	3293.70	5231.73	65.98	1466.56	0.90	0.010
3	0.37	21.7	-10.0	0.9913	1.7041	2.1524	34.10	5454.60	58.11	0.060	5925.17	5107.47	118.70	469.41	1.22	0.009
4	0.28	21.1	-12.0	0.9056	1.5946	1.9982	22.70	14430.74	40.76	0.072	3182.51	1848.26	63.76	1241.87	0.85	0.011
5	0.32	20.3	-11.0	0.8390	1.4946	1.8658	21.45	9586.57	43.79	0.066	3707.84	2588.30	74.28	825.00	0.92	0.010
6	0.46	20.4	-9.9	0.8152	1.3423	1.7207	21.15	3450.30	57.54	0.059	5671.13	4505.49	113.61	296.92	1.21	0.009
7	0.45	20.4	-9.9	0.6724	1.2590	1.5471	19.20	4107.92	50.62	0.059	5602.30	3817.18	112.23	353.52	1.06	0.009
8	0.45	20.6	-10.1	0.6497	1.1662	1.4523	14.15	4579.71	46.99	0.061	4348.76	3080.06	87.12	394.12	0.98	0.010
9	0.42	19.6	-9.0	1.2614	1.8873	2.5057	54.50	2931.58	76.57	0.054	9171.20	8774.43	183.73	252.28	1.60	0.008
10	0.39	18.6	-10.0	1.1472	1.7921	2.3411	34.70	3881.42	66.94	0.060	5810.76	5715.95	116.41	334.03	1.40	0.009
11	0.42	18.1	-9.0	1.1055	1.7255	2.2548	35.10	3169.44	68.56	0.054	6502.05	7785.34	130.26	272.75	1.44	0.008
12	0.38	17.9	-8.1	0.9353	1.5898	2.0158	36.09	4578.25	55.35	0.049	6766.60	8429.81	135.56	393.99	1.16	0.008
13	0.72	16.7	-8.0	0.8520	1.4946	1.8752	32.20	682.98	98.07	0.048	12317.74	12950.83	246.76	58.78	2.06	0.008
14	0.70	15.9	-12.0	0.7580	1.2566	1.6071	10.95	952.57	83.34	0.072	4773.60	2616.79	95.63	81.98	1.75	0.011
15	0.76	14.2	-8.0	0.7342	1.1852	1.5300	18.80	637.60	85.04	0.048	9292.71	11182.95	186.16	54.87	1.78	0.008
16	0.74	13.6	-7.1	0.5403	0.9187	1.1647	12.95	844.57	62.96	0.043	8189.47	10193.57	164.06	72.68	1.32	0.007
17	0.73	13.2	-11.1	1.1960	1.8754	2.4465	30.60	477.41	132.95	0.067	9129.09	11011.61	182.88	41.08	2.79	0.010
18	0.77	12.8	-11.1	1.1103	1.8183	2.3353	27.60	425.35	133.41	0.067	9057.40	11054.30	181.45	36.60	2.80	0.010
19	1.13	10.5	-6.0	1.0972	1.6684	2.2015	56.00	97.77	183.79	0.036	28741.55	28482.99	575.78	8.41	3.85	0.006
20	1.13	9.7	-9.0	0.8056	1.5184	1.8622	26.05	129.20	157.82	0.054	15850.41	18664.15	317.53	11.12	3.31	0.008
21	1.12	9.2	-8.0	0.6236	1.3066	1.5543	23.65	145.67	130.47	0.048	17099.29	17107.80	342.55	12.54	2.73	0.008
22	1.34	8.1	-8.0	0.6164	1.2614	1.5103	22.35	82.38	151.88	0.048	19858.23	19961.98	397.82	7.09	3.18	0.008
23	1.22	7.5	-7.1	0.5355	1.0496	1.2722	16.05	117.73	116.04	0.043	15367.31	17060.21	307.85	10.13	2.43	0.007
24	1.29	7.3	-4.8	1.0912	1.9373	2.4211	67.75	43.70	231.92	0.029	36043.74	37467.69	722.06	3.76	4.86	0.005
25	1.41	6.5	-8.1	0.9877	1.7469	2.1858	52.55	44.41	231.57	0.049	33797.99	30446.80	677.07	3.82	4.85	0.008
26	1.43	5.7	-7.2	0.8723	1.5137	1.9059	39.20	43.15	204.84	0.043	29343.66	28755.93	587.84	3.71	4.29	0.007
27	1.85	5.1	-7.1	0.7687	1.2495	1.6091	29.55	22.18	224.53	0.043	33964.24	31605.84	680.40	1.91	4.71	0.007

Table A.3. Type-II experimental data for the Al_HP test series.

28	1.78	5.4	-7.1	0.6866	1.1948	1.5030	26.10	27.35	201.52	0.043	30890.59	28515.26	618.83	2.35	4.22	0.007
29	1.70	5.7	-6.0	0.5902	1.0139	1.2810	24.65	35.05	162.97	0.036	32621.60	25661.78	653.51	3.02	3.42	0.006
30	1.32	7.2	-7.0	1.1757	1.8683	2.4255	50.50	47.23	240.30	0.042	27530.14	33945.83	551.51	4.06	5.04	0.007

	V(m/s)	$T_{\infty}(^{\circ}C)$	$T_w(^{\circ}C)$	a(mm)	h(mm)	D(mm)	t(s)	Am∞	Re∞	Ste _L	τ	$\tau_predict$
1	1.10	7.2	-10.0	0.8116	0.7283	1.2221	5.20	148.39	134.60	0.060	3511.38	3833.88
2	1.12	7.3	-10.0	0.8723	0.7902	1.3194	6.40	130.23	147.76	0.060	4108.22	4287.99
3	0.83	10.2	-10.0	1.1971	1.0805	1.8077	9.80	269.87	149.70	0.060	3415.24	4367.34
4	0.66	6.9	-10.0	1.1281	1.0186	1.7038	8.80	471.87	113.53	0.060	2591.43	3132.48
5	0.66	6.7	-10.0	1.1186	1.0020	1.6830	8.40	485.71	111.35	0.060	2466.11	3058.20
6	0.52	7.6	-10.0	1.0448	0.9472	1.5810	7.00	1116.98	81.76	0.060	1734.39	2088.60
7	0.52	7.3	-10.0	1.0305	0.9639	1.5830	8.30	1094.59	81.21	0.060	2097.82	2067.60
8	0.52	7.8	-10.0	0.9389	0.8330	1.4063	6.10	1237.60	73.96	0.060	1694.39	1829.02
9	0.55	8.3	-10.0	0.9199	0.8973	1.4423	6.60	1101.58	76.45	0.060	1977.48	1903.39
10	0.34	22.1	-12.0	1.4780	1.3447	2.2403	14.25	5796.11	72.31	0.072	1624.91	1008.49
11	0.33	22.4	-11.0	1.4447	1.2900	2.1703	14.25	6197.02	68.98	0.066	1629.42	1508.46
12	0.29	21.5	-10.0	1.4280	1.2757	2.1458	15.30	8310.81	60.86	0.060	1579.97	1991.85
13	0.38	21.0	-12.0	1.2923	1.1519	1.9399	9.90	4549.50	71.04	0.072	1445.43	855.87
14	0.32	20.3	-10.0	1.4340	1.2519	2.1315	17.00	5981.39	67.41	0.060	1921.39	2015.65
15	0.64	20.0	-10.0	0.6331	0.5831	0.9653	4.15	1709.87	59.12	0.060	2108.00	1356.31
16	0.43	20.3	-10.0	0.8758	0.8211	1.3468	7.10	4177.88	54.69	0.060	1745.23	1418.01
17	0.43	20.1	-9.0	0.6783	0.6212	1.0314	4.50	5042.72	42.65	0.054	1441.62	1656.81
18	0.48	19.7	-9.0	0.7211	0.6616	1.0975	5.70	3520.04	49.91	0.054	1887.83	1775.49
19	0.50	17.5	-12.1	0.6033	0.5950	0.9511	2.80	3752.19	44.30	0.073	1156.17	-197.58
20	0.65	13.7	-8.1	1.1436	1.0710	1.7577	14.00	665.06	108.96	0.049	3961.02	4093.71
21	0.69	12.0	-7.0	1.0758	0.9948	1.6434	15.00	497.60	110.30	0.042	4844.61	4763.89
22	0.71	11.5	-11.0	0.9948	0.9163	1.5168	6.30	579.19	106.32	0.066	2260.01	2311.83
23	0.65	11.3	-11.2	1.1448	0.9449	1.6581	7.15	671.64	111.22	0.067	2023.50	2373.50
24	0.72	10.3	-6.0	0.9092	0.8616	1.4054	12.80	452.04	97.03	0.036	5080.73	4880.76
25	0.82	9.6	-6.0	0.8378	0.7997	1.2996	9.80	314.66	102.12	0.036	4810.50	5045.04
26	1.10	9.8	-9.0	1.0103	0.9187	1.5310	9.45	130.29	166.40	0.054	5150.89	5513.79
27	1.38	10.0	-8.1	1.0722	0.9925	1.6387	13.45	60.08	220.32	0.049	8649.38	7897.07

Table A.4. Reduced type-II experimental data of the Al_Si tests by Gong's method.

28	1.36	10.2	-9.0	1.0746	0.9996	1.6462	10.55	65.63	218.78	0.054	6691.28	7321.93
29	1.29	8.6	-7.0	1.1103	1.0424	1.7086	15.10	61.34	213.80	0.042	8778.99	8309.79
30	1.77	6.8	-8.0	0.7735	0.7616	1.2184	6.30	32.13	205.78	0.048	7196.09	7449.19
31	1.77	6.9	-8.0	0.7152	0.7140	1.1343	5.40	35.14	190.23	0.048	6672.75	6911.00
32	1.79	6.6	-7.0	0.7402	0.7354	1.1712	6.80	29.58	199.43	0.042	8242.06	7809.43
33	1.83	5.7	-7.3	0.9211	0.8782	1.4281	10.35	21.41	253.47	0.044	10257.63	9505.87
34	1.69	4.9	-6.0	0.8782	0.8497	1.3714	9.75	23.70	223.88	0.036	9401.18	9235.89
35	0.28	21.3	-10.0	1.3102	1.1900	1.9843	13.20	10079.45	53.81	0.060	1430.98	1901.87
36	0.27	21.5	-11.0	1.3138	1.1710	1.9720	10.80	12109.58	51.40	0.066	1109.48	1415.02

APPENDIX B

Uncertainty Analysis of Type-II Experiments

Let the experimental result 'R' be a function of 'n' independent variables V_n and define this equation the data reduction equation.

$$R = R(V_1, V_2, \dots, V_n)$$
(B.1)

The uncertainties in the individual variables affect the final result through a data reduction equation. If all the variables are normally distributed, then the uncertainty in the result is given by

$$W_R = \left[\left(\frac{\partial R}{\partial V_1} W_1 \right)^2 + \left(\frac{\partial R}{\partial V_2} W_2 \right)^2 + \dots + \left(\frac{\partial R}{\partial V_n} W_n \right)^2 \right]^{1/2}$$
(B.2)

where the W_n are the uncertainties of the independent variables V_n .

In this study, the data reduction equation has the general form

$$R = aV_1^b V_2^c V_3^d \tag{B.3}$$

where a is a constant and the exponents b, c, and d may be positive or negative constants, the general uncertainty analysis equation becomes

$$\frac{W_R}{R} = \left[\left(b \frac{W_1}{V_1} \right)^2 + \left(c \frac{W_2}{V_2} \right)^2 + \left(d \frac{W_3}{V_3} \right)^2 \right]^{1/2} \tag{B.4}$$

In this investigation, the Type-II experiments were performed to measure the freezing time of water droplets during phase-change process on three different substrates. The uncertainties of the direct measured variables (Temperature, length, time, and velocity) were listed below:

$$W_T = \pm 1^{\circ} C (\pm 1 \text{ K})$$

 $W_l = 0.0024 \text{ mm}$
 $W_t = 0.001 \text{ s}$
 $W_V = 0.02$

The average values of these direct measured variables (air temperature, surface temperature, droplet height, diameter of covered area of droplet, freezing time) were listed below:

SCA	74.55 Degree	86.51 Degree	127.14 Degree
$T_{\infty}(\mathbf{K})$	287.15	286.15	287.15
$T_w(\mathbf{K})$	264.15	264.15	264.15
<i>h</i> (mm)	0.7422	0.9361	1.5022
<i>d</i> (mm)	1.8641	2.0375	1.7786
<i>t</i> (s)	10.091	9.382	30.591

Table B.1. Average values of the direct measured variables.

The uncertainty analysis was performed on the fluid properties, body size D, dimensionless freezing time τ , Am_{∞} , Re_{∞} , and Ste_L .

Fluid Properties

The fluid properties in this experiments were not directly measured. All of the thermophysical properties were estimated through the empirical equations obtained from the reference source, National Bureau of Standards Circular 564(1955) [42]. At the condition of the ideal gas model, the thermal conductivity k_{∞} , density ρ_{∞} , and kinematic viscosity v_{∞} depend only on temperature, which means all the uncertainties in these parameters are brought by the measured temperature.

$$k_{\infty}(T) = 0.024061794 + 7.5939994 * 10^{-5} * (T - 273.15)$$
(B.5)

$$\rho_{\infty}(T) = 0.63370723 + 0.65832423 * \exp\left(\frac{T - 273.15}{139.31335}\right) \tag{B.6}$$

$$v_{\infty}(T) = -2.2938017 * 10^{-5} + 3.6276317 * 10^{-5} * \exp\left(\frac{T - 273.15}{417.58729}\right)$$
 (B.7)

The general uncertainty analysis equations of these fluid properties are

$$W_k = \left(\frac{\partial k}{\partial T}\right) W_T = 7.5939994 * 10^{-5} * W_T$$
 (B.8)

$$W_{\rho} = \left(\frac{\partial\rho}{\partial T}\right) W_{T} = \frac{0.65832423}{139.31335} * \exp\left(\frac{T - 273.15}{139.31335}\right) * W_{T}$$
(B.9)

$$W_{\nu} = \left(\frac{\partial \nu}{\partial T}\right) W_T = \frac{3.6276317 * 10^{-5}}{417.58729} * \exp\left(\frac{T - 273.15}{417.58729}\right) * W_T \tag{B.10}$$

For our experiments, $W_T = 1^{\circ}C$ (1K). Table B2 gives the uncertainties of fluid properties for three different test series.

Table B.2. Uncertainties of fluid properties for the three different test series.

SCA	74.55 Degree	86.51 Degree	127.14 Degree
Air Temperature (K)	287.15	286.15	287.15
$W_k (W/m \cdot K)$	$7.594*10^{-5}$	$7.594*10^{-5}$	$7.594*10^{-5}$
W_{ρ} (kg/m ³)	$5.225*10^{-3}$	$5.188*10^{-3}$	$5.225*10^{-3}$
W_{v} (m ² /s)	$8.983*10^{-8}$	$8.962*10^{-8}$	$8.983*10^{-8}$

Body Size

We assume the droplet is not affected by the gravity and have a shape of spherical cab. The volume is a function of the droplet height h and the diameter of droplet covered area d.

$$volume = \frac{\pi h}{6} \left(\frac{3}{4} d^2 + h^2 \right)$$
 (B.11)

$$\left(\frac{W_{volume}}{volume}\right)^2 = \left(\frac{6d}{3d^2 + 4h^2}W_d\right)^2 + \left(\frac{3(d^2 + 4h^2)}{h(3d^2 + 4h^2)}W_h\right)^2 \tag{B.12}$$

The data reduction equation for D is

$$D = 2\sqrt[3]{\frac{3 * volume}{4\pi}} = 2\left(\frac{3 volume}{4\pi}\right)^{1/3}$$
(B.13)

and the general uncertainty analysis equation is

$$\left(\frac{W_D}{D}\right)^2 = \left(\frac{1}{3}\frac{W_{volume}}{volume}\right)^2 \tag{B.14}$$

The values for the variables and the results of the uncertainties in the droplet volume and body size are listed in Table B3.

Table B.3. Uncertainties of the droplet volume and body size for the three different tests.

SCA	74.55 Degree	86.51 Degree	127.14 Degree
W_1 (mm)	0.0024	0.0024	0.0024
h (mm)	0.7422	0.9361	1.5022
d (mm)	1.8641	2.0375	1.7786
W _{volume} /volume	0.00485	0.00412	0.00345
W_D/D	0.00162	0.00137	0.00115

Dimensionless Freezing time

The data reduction equation for τ is

$$\tau = \frac{Vt}{D} \tag{B.15}$$

and the general uncertainty analysis expression is

$$\left(\frac{W_t}{\tau}\right)^2 = \left(\frac{W_V}{V}\right)^2 + \left(\frac{W_t}{t}\right)^2 + \left(\frac{W_D}{D}\right)^2 \tag{B.16}$$

The values for the variables and the results of the uncertainties in the dimensionless freezing time are presented in Table B4.

SCA	74.55 Degree	86.51 Degree	127.14 Degree
W _V /V	0.02	0.02	0.02
W_t/t	0.7422/0.001	0.9361/0.001	1.5022/0.001
W_D/D	0.00162	0.00137	0.00115
$W_{ au}/ au$	0.0201	0.0200	0.0200

Table B.4. Uncertainties of the dimensionless freezing time for the three different tests.

Am_{∞} Number

The data reduction equation for Am number is

$$Am_{\infty} = \frac{k(T_{\infty} - T_{w})}{\rho V^{3}D}$$
(B.17)

and the general uncertainty analysis expression is

$$\left(\frac{W_{Am}}{Am_{\infty}}\right)^2 = \left(\frac{W_k}{k}\right)^2 + \left(\frac{W_{T_{\infty}}}{T_{\infty} - T_w}\right)^2 + \left(\frac{W_{T_w}}{T_{\infty} - T_w}\right)^2 + \left(\frac{W_{\rho}}{\rho}\right)^2 + \left(3\frac{W_V}{V}\right)^2 + \left(\frac{W_D}{D}\right)^2 \quad (B.18)$$

The values for the variables and the results of the uncertainties in the Am_{∞} number are given in Table B5.

Table B.5. Uncertainties of the Am_{∞} number for the three different tests.

SCA	74.55 Degree	86.51 Degree	127.14 Degree
W _k /k	7.594*10 ⁻⁵ /0.0251	7.594*10 ⁻⁵ /0.0250	7.594*10 ⁻⁵ /0.0251
$W_T/(T_\infty - T_w)$	1/(287.15-264.15)	1/(286.15-264.15)	1/(287.15-264.15)
$W_{ ho}/ ho$	5.225*10 ⁻³ /1.23	5.118*10 ⁻³ /1.23	5.225*10 ⁻³ /1.23
W_V/V	0.02	0.02	0.02
W_D/D	0.00162	0.00137	0.00115
W _{Am} /Am	0.0860	0.0880	0.0860

Reynolds Number

The data reduction equation is

$$Re_{\infty} = \frac{VD}{v} \tag{B.19}$$

and the general uncertainty analysis expression is

$$\left(\frac{W_{Re}}{Re_{\infty}}\right)^2 = \left(\frac{W_V}{V}\right)^2 + \left(\frac{W_D}{D}\right)^2 + \left(\frac{W_V}{V}\right)^2 \tag{B.20}$$

The values for the variables and the results of the uncertainties in the Reynolds number are listed in Table B6.

Table B.6. Uncertainties of the Reynolds number for the three different tests.

SCA	74.55 Degree	86.51 Degree	127.14 Degree
W_V/V	0.02	0.02	0.02
W_D/D	0.00162	0.00137	0.00115
W_{ν}/ν	$8.983*10^{-8}/1.458*10^{-5}$	$8.962*10^{-8}/1.458*10^{-5}$	$8.983*10^{-8}/1.458*10^{-5}$
W_{Re}/Re_{∞}	0.0210	0.0210	0.0210

Stefan Number

The data reduction equation for Stefan number is

$$Ste_{\rm L} = \frac{C_p(T_f - T_w)}{L} \tag{B.21}$$

and the general uncertainty analysis expression is

$$\left(\frac{W_{Ste}}{Ste_{\rm L}}\right)^2 = \left(\frac{W_{C_p}}{C_p}\right)^2 + \left(\frac{W_{T_f}}{T_f - T_w}\right)^2 + \left(\frac{W_{T_w}}{T_f - T_w}\right)^2 + \left(\frac{W_L}{L}\right)^2 \tag{B.22}$$

The heat capacity and the latent heat of freezing for water droplets are considered as constants, $C_p = 2.0$ kJ/kg K and L=333.7 KJ/Kg. The water freezing temperature T_f is 273.15 K. Other values for the variables and the results of the uncertainties in the Stefan number are shown in Table B7.

Table B.7. Uncertainties of the Stefan number for the three different tests.

SCA	74.55 Degree	86.51 Degree	127.14 Degree
$W_T/(T_f - T_w)$	1/(273.15-264.15)	1/(273.15-264.15)	1/(273.15-264.15)
W _{Ste} /Ste	0.157	0.157	0.157

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