Investigating Ice Nucleation and Heat Transfer Dynamics in Supercooled Liquid Water Using Thermography

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Abstract— **This study delves into the intricate interplay of thermography, ice nucleation, and heat transfer during the phase change from supercooled liquid water to crystallized ice. Utilizing high-resolution, high-speed infrared thermography, real-time temperature data is captured during ice nucleation events. By analysing these temperature profiles, valuable information about the dynamics of ice nucleation is revealed and presented. One of the key highlights of this study is the observation of nucleation under supercooled conditions. The evidence of how supercooled liquid water transforms into crystalline ice is provided, which sheds light on the underlying physics and mechanisms involved, like recalescence and phase change. This phase change process is significantly important in the context of cloud formation and freezing rain phenomena. The study may form the basis of developing a mathematical model for defining nucleation phase. These findings have practical implications across multiple industries and can aid in the development of more efficient anti-/de-icing systems, refrigeration systems, improved weather prediction models, and enhanced cryopreservation techniques. The study opens new avenues for further exploration in this field, ultimately advancing our understanding of these critical processes.**

Keywords— ***Ice Nucleation, Thermography, Heat Transfer Dynamics, Latent Heat***

I. INTRODUCTION

Ice nucleation is a key process in understanding ice accretion physics as most of the passive techniques use extending the nucleation time. The phenomena of ice nucleation and subsequent ice growth have paramount importance in various fields, including cold climate engineering, atmospheric science, cryopreservation, and refrigeration systems [1-5]. According to Classical Nucleation Theory (1928) nucleation involves the formation of the initial embryos of tiny stable solids from a supercooled or supersaturated mother phase (vapor or liquid phase). For freezing and ice growth to occur, an ice nucleus must reach a certain size called the critical size (r*) to be thermodynamically stable (Figure 1). Below this critical size, an ice nucleus is unstable and will be destroyed, reverting to the mother phase [1, 6]. Ice nucleation and its growth will happen under a thermodynamic driving force. However, according to modern study, the process of nucleation is dynamic and is also impacted by varying environmental factors [7]. In this regard homogenous and heterogenous nucleation are often studied that are characterized by absence or presence of any external agent to create nucleation sites in water droplets, respectively [1, 8].

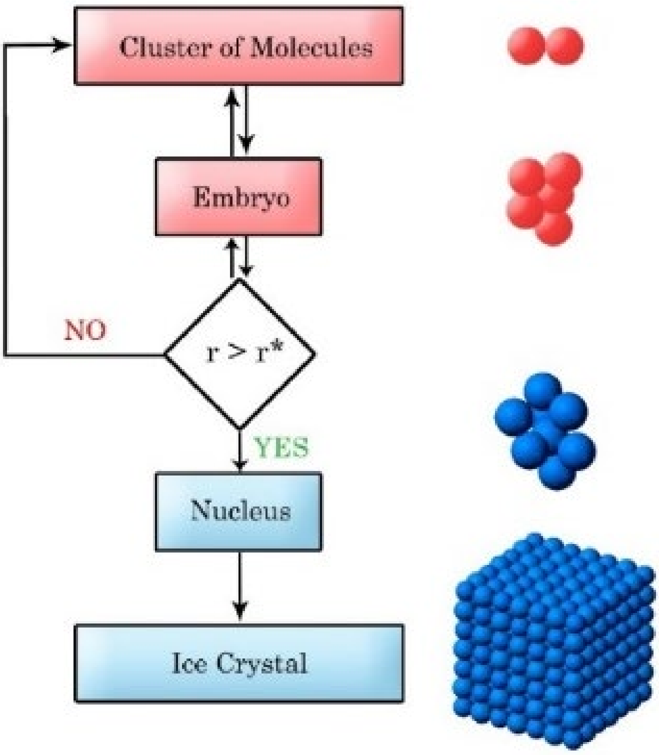


Fig. 1 A graphic representation of ice nucleation phenomenon according to Classical Nucleation Theory (CNT) [9]

Various experimental studies have been conducted to understand the formation of ice from water droplets. The studies focus on monitoring water droplet impact on cold substrates as it experiences freezing. The process concludes in four steps: supercooling, recalescence (nucleation), phase change, and ice cooling (see Figure 2). When droplet impinges a supercooled surface, a liquid-substrate interface is created by which conductive heat transfer takes place and droplet undergoes supercooling. During this stage the morphology of water droplet changes; its contact angle decreases with an increased droplet contact area. Treated hydrophobic surfaces make this contact angle large to create small interface with the solid substrate and hence take longer time for supercooling. Moreover, time duration of supercooling stage is dependent on drop size; smaller droplets comparatively get cooled faster until nucleation initiation [10]. Once a thermal equilibrium is established at this liquid-substrate interface, droplet loses its latent heat and nucleation begins; this stage is termed as recalescence. During this stage an ice shell appears on the circumference of the droplet, rising from the droplet-substrate interface up till the top. It is an instantaneous step and not much literature is available that makes an in-depth analysis for it. After the evolution of latent heat temperature remains constant while phase change takes place, in which the water content inside the frozen shell starts solidifying gradually. A pointed tip is formed at the completion of solidification stage followed by ice cooling [10, 11].

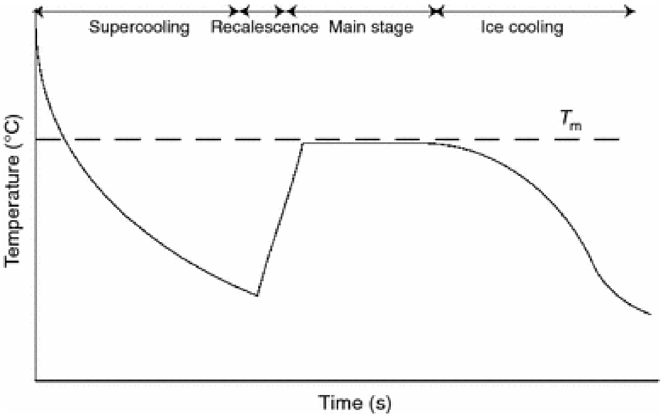


Fig. 2 Freezing process of a supercooled droplet takes place in four stages [7].

High speed infrared thermography can be used to study the phase change and track temperatures changes in water and ice [12-15]. There is also work in progress on the development of ice detection system based on the thermography [16-18]. There are basically two arrangements for IR camera in this context: taking a side view of droplet or taking a top-down view. Side view makes it possible to quantify conduction heat transfer between solid substrate and liquid droplet. Li and Liu [19] recorded top-down view temperature maps during droplet freezing using IR thermography to determine the onset of freezing in pure water droplets versus droplets containing anti-freezing agents. Alizadeh et al. [20] used top-down IR thermography along with highspeed visual imagery to study the impact of droplets impinging on hydrophilic, hydrophobic and superhydrophobic surfaces. Their studies revealed that drop-substrate contact area can delay nucleation by heat transfer as well as by reducing probability of heterogenous nucleation at the interface.

Tavakoli et al. [10] carried out infrared thermography of a droplet freezing experiment on hydrophilic and hydrophobic glass substrate. They used FLIR® A600 thermal camera to get a top-down view of water droplet and observed that this main drop surrounds itself on the top side by condensed micro drops that make an appearance of bumpy straight-line pattern, which is called ‘recalescence front’. This front of condensed micro drops travels down the drop with the release of latent heat as the nucleation and solidification proceed. They postulated that solidification of main drop is triggered by these ‘travelling’ condensed micro drops surrounding it when they reach the main drop, and this way they deduced that nucleation front is activated from point of trijunction (formed by liquid-substrate interface, substrate-gas interface, liquid-gas interface). Castillo et al. [21] performed numerical simulations of solidification stage (phase change stage) using the boundary conditions determined by IR temperature. For that purpose, they employed side-view IR thermography. They revealed that heat transfer rate between drop-substrate is greater than heat transfer between drop-ambient air, and same applies in case of latent heat release. Li et al. [22] used high-speed visual and infrared imaging to observe supercooled large droplets colliding with a hydrophilic and hydrophobic surface, and studied droplets impact, spreading and rebound.

Among all the four steps of droplet freezing, recalescence has not been studied in detail up to the best knowledge of authors, and since it is a split-second process thermocouples are not a reasonable choice for this purpose. High speed imaging is an efficient tool for high resolution studies. The current article highlights recalescence during the impinging water droplets on a cold aluminium substrate using high speed colour camera and a high-resolution long wave thermal infrared camera. The generated data from sequence file is analysed within MATLAB.

II. METHODOLOGY

Experimental design consists of a set of experiments involving ice nucleation which are carried out in cold room laboratory at UQAC – Université du Québec à Chicoutimi, Canada. Individual droplets are dropped using medical syringe with hypodermic needle on aluminium substrate maintained at -13℃ and observed for solidification.

The overall setup includes a small chamber supported by Peltier cooler and thermocouple, cold bath, high speed camera, infrared (IR) camera, aluminium substrate, data acquisition system, temperature and humidity sensors, and a vibration-free table. (Graphic setup of experiment with some actual visuals are shown in Figure 3). A thermally insulated and optically transparent double layer chamber placed on motionless table ensured that the parameters affecting ice nucleation were uniform during experiments in order to increase the accuracy of results and reproducibility of experiments. Its transparency facilitated in imaging the side view of freezing water droplet with Mikrotron MotionBLITZ EoSens Cube7 Mono high-speed camera. The camera offers 525 fps at 3MP resolution with capability to adjust framerate to 200,000 fps at lower resolutions. This feature enabled capturing fast motion sequences such as, water droplet freezing, quite reliably with sufficiently sharp details. LED lights were used to illuminate the scene for high-speed optical camera. Since the chamber walls were opaque for infrared analysis, an IR camera (T1030sc with 50µm close-up lens) was set at height for recording top view of the droplet. The chamber is also supported by temperature and humidity sensors that recorded the surrounding and in-chamber atmospheric parameters before and during the experiments. Prior to experiment the temperature of the aluminium substrate was measured with an IR camera and a thermocouple (K-type) which was then removed during the experiments.

The temperature of thermostatic bath was decreased to a desired value (-15°C) and aluminium substrate was placed over it. Once the temperature became stable water droplets of 10 µL/20 µL volume were dropped onto it with no frost formation. The freezing process onwards was recorded by both high-speed and IR cameras with side and top views, respectively.

By visual high-speed & IR cameras recordings all the four stages of cooling and freezing of water droplets are identified: supercooling – recalescence – solidification – ice cooling. Recalescence, which is an instant stage and happens in a split second can be easily monitored using high-speed camera at 1000 fps.

While working with the thermal camera it needed to be stopped after every 5 minutes due to its limitations. Visual camera, however, could snap a continuous video. For thermal infrared camera, before each recording it was made sure that the target (droplet) was well focused and not blurry, and correct temperature range was selected from camera settings. The sequence file generated by IR camera is analysed by FLIR® ResearchIR software. Circular region of interest (ROI) is sketched over the location of a singular droplet to measure average temperature change over it. Temporal data of this ROI is extracted and then analysed in MATLAB.

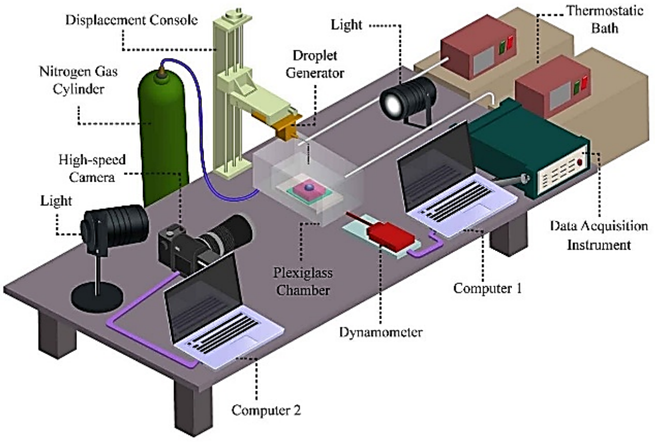
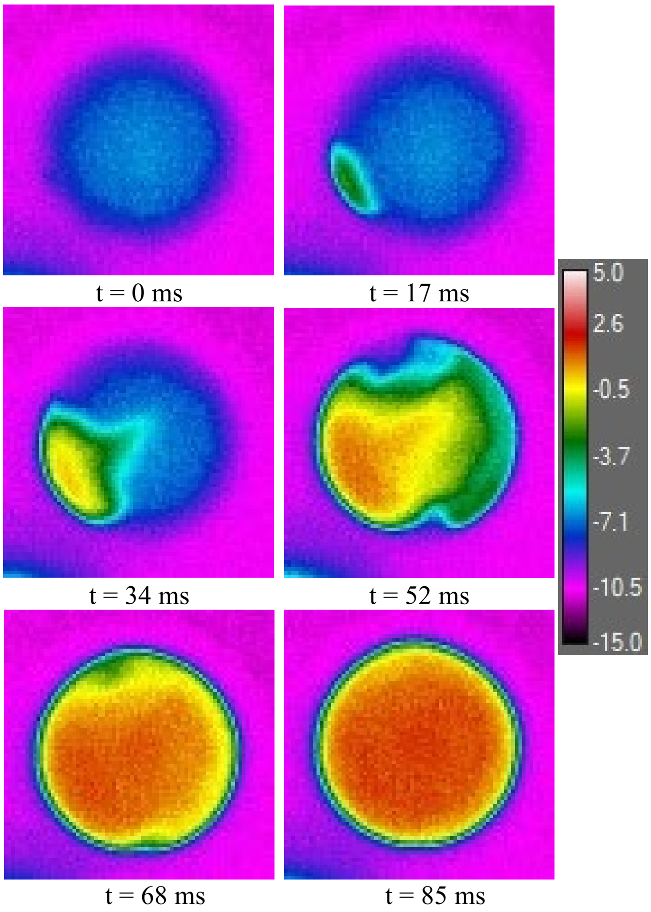


Fig. 3 Graphic representation of experimental setup for ice nucleation droplet test.

III. Results and Discussion

Sequential temperature distribution against elapsed time during recalescence as observed through high-speed infrared imaging is presented below in Figure 4. Evolution of thermal gradient is clearly visible on droplet as it initiates from droplet-substrate-air interface. Immediately prior to recalescence at t = 0, the droplet is supercooled at about -8°C and then after t = 17ms a gradual conductive heat transfer takes place from the substrate and rises across the droplet height. The low temperature regions (blue coloured) remain in supercooled state while the crystallization front rises. This crystallization front is composed of ice-water mixture and covers the whole droplet. At about t = 85ms the crystallization process is completed, and droplet temperature is maintained at approx. 0°C.

IR camera covers the split-second transition at recalescence during which a visible, translucent ice front starts developing from liquid-substrate interface rising towards the tip making a dome shaped outer cover with liquid water inside. Such observations are also recorded by Keshavarzi et al [23] while studying ice nucleation process via high-speed imaging on hydrophobic and superhydrophobic surfaces. Visually this process is apparent through high-speed camera because ice and water have different refractive index (ice: 1.31, water: 1.33). Moreover, the air bubbles get trapped in ice shell giving it a translucent outlook [24]. The transparency of water droplet fully disappears after the solidification (or phase transition event) in which the water content inside the shell solidifies gradually forming a cusp shape.



t = 0 ms t = 17 ms
t = 34 ms t = 52 ms
t = 68 ms t = 85 ms

Fig. 4 Sequential infrared images of recalescence in sessile water droplet

IV. Conclusion

The paper discussed in detail the sequential process of freezing water droplet. utilizing the high-speed infrared thermography to study the freezing droplet on cold substrate. Supercooling can be better depicted in IR imagery whereas phase change is better analysed in visual high-speed camera. Utilizing high-resolution, high-speed infrared thermography, real-time thermal images were captured, and temperature data was analysed during ice nucleation events. By analysing these temperature profiles, we gained valuable information about the dynamics of ice nucleation. Our findings reveal the crucial role of nucleation sites, their distribution, and their impact on the overall heat transfer process. The overall knowledge contributes to a more comprehensive understanding of ice nucleation mechanisms and is a precursor to understanding the microphysics of surfaces undergoing ice accretion. The insights can be found useful in making hydrophobic coatings or treating those surfaces such that duration of supercooling and recalescence stages can be extended.

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