



# Freezing of water droplets on solid surfaces: An experimental and numerical study



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## ABSTRACT

We present an experimental and numerical study on the freezing of static water droplets on surfaces with different wettability when the surfaces are subject to rapid cooling. Temperature evolution of the droplets is recorded using both intrusive and non-intrusive methods to identify the processes involved in the cooling and phase change of the droplets. It is found the time taken for a droplet to freeze depends on the droplet temperature at the pre-recalescence instant as well as the surface wettability. To provide insight into the heat transfer during the freezing process, thermal simulation is carried out by numerically solving the enthalpy-based heat conduction equation. To determine the initial and boundary conditions for the simulation of freezing, the thermal history of the droplet prior to the occurrence of freezing is numerically analyzed by solving single phase heat conduction driven by rapid cooling. The numerical results of droplet freezing are compared to the experimental data, showing close agreement on the freezing time.

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## 1. Introduction

Ice formation on surfaces plays an important role in many processes and presents a great challenge to safety, efficiency and performance [1,2]. Many efforts have been made for its prevention on aircraft surfaces, ships, wind turbine blades, heat exchangers etc. [3–5]. Many of anti-icing and de-icing techniques have been developed by researchers to overcome these challenges. The development of surface coatings and textures that limit ice formation is a subject of considerable attention [6,7]. Freezing test of water droplets on these engineered surfaces has become a common method for assessing the anti-icing capability of these surfaces. Therefore, it is necessary to develop a good understanding of the physics involved in the freezing of water droplets on solid surfaces.

Freezing of suspended or free water droplets has been studied extensively over past few decades [8–12]. Efforts have been put on developing theoretical models for droplet freezing and applying new techniques for experimental validation. Strub et al. [8] experimentally and numerically studied the crystallization of a water droplet in a cold humid airflow, which is essential for understanding the behavior of water droplets sprayed out of a snow gun. Hindmarsh et al. [9] experimentally measured the temperature

transition of freezing droplets suspended from a thermocouple junction in a cold air stream. A simple heat balance model was developed to predict the freezing time of droplets. The freezing of free liquid droplets was theoretically studied by Feuillebois et al. [10]. Three different conditions were considered: (1) the center of droplet freezes first; (2) the outer shell freezes first; (3) and ice nuclei are uniformly distributed in the droplet. The problem was then solved using two methods, numerical and perturbation, both giving identical results.

Recently driven by the research and development of ice-phobic surfaces based on super-hydrophobic technologies, the impingement of water droplets on surfaces under supercooled conditions has received rising attention [13–16]. Although most studies on this subject have taken into account heat transfer, both experimental data and understanding of heat transfer and phase change inside water droplets are very limited. This limitation restricts the development of analytical models for water droplet impact under supercooled conditions. This is obvious if comparison is made to the thermal coating research [17–19], for which, based on well-established theories of heat transfer and solidification, a few important analytical models have been developed for the deposition of molten metal droplets onto cold substrates.

To obtain fundamental understanding of water droplets interacting with solid surfaces under supercooled conditions, a few studies have been reported on the freezing of static water droplets on supercooled surfaces. Suzuki et al. [20] used a high speed

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## Nomenclature

$C$	specific heat
$f$	fraction of ice
$h$	heat transfer coefficient
$k$	thermal conductivity
$L$	latent heat of fusion
$T_{\infty}$	temperature of liquid nitrogen flow
$T_e$	equilibrium phase change temperature
$T_{0,r}$	droplet topmost temperature at pre-recalence instant
$T_{s,r}$	droplet-substrate interface temperature at pre-recalence instant
$T_r$	droplet average temperature at pre-recalence instant

$T_{ref}$	arbitrary reference temperature
$\Delta t_f$	freezing time

## Greek symbols

$\Omega$	specific internal energy
$\rho$	density

## Subscripts

$w$	water
$i$	ice

camera to observe the freezing of water droplets on chemically treated silicon surfaces, and used differential scanning calorimetry to investigate the dependence of the freezing temperature on those surfaces' characteristics. With only the high speed images available for the discussion of droplet freezing, some process and events occurring during freezing are still in vague and hence demand a deeper insight. Tabakova and Feuillebois [21] reported a theoretical study on the freezing of liquid droplets on solid surfaces. Based on their numerical results of solving one-phase Stefan problem, they proposed analytical functions to estimate the time taken for a water droplet to freeze. However, no experimental work was conducted to validate the results and functions.

The objective of the present work is to investigate the freezing of static water droplets lying on supercooled surfaces both experimentally and numerically. Temperature histories recorded using both intrusive and non-intrusive methods are used to identify the processes involved in the cooling and phase change of the droplets. Numerical analysis is conducted to provide insight into the heat transfer involved in the freezing process.

## 2. Methodologies

In the present work, the freezing of individual water droplets on solid surfaces was studied. Fig. 1 schematically shows the experimental setup used to conduct the experiments. Water droplets with a relatively large volume of 21  $\mu\text{L}$  and a small volume of 7.2  $\mu\text{L}$  were dispensed from glass pipettes connected to a micro-pump, and were gently deposited on smooth surfaces with different

wettability. The water had been micro-filtered and deionized. Two types of surfaces were used, a hydrophilic surface with contact angle of 45° and a hydrophobic surface with contact angle of 110°. The two surfaces were prepared by coating silicon wafers with two different self-assembled monolayers (SAM) [20]. The roughness of the SAM-coated surfaces was measured to be lower than 10 nm, and its effect on heat transfer is negligible.

As shown in Fig. 1, the Si wafer was kept on a cold plate cooled by liquid nitrogen flowing through internal channels of the plate. The pump was controlled by a temperature controller such that the cold plate can operate at a preset cooling rate (constant change of temperature per unit time). For each test reported in the present work, the cold plate temperature continued to decrease at a constant rate during the entire test process. The cold plate temperature was measured using a thermocouple embedded in the plate. The temperature was different from the surface temperature of the SAM-coated wafer in both steady and transient states. This is not only because of the finite thickness (0.375 mm) of the wafer, but also due to the unknown thermal contact resistance between the wafer and the cold plate.

The tests were conducted in a room where the temperature and relative humidity were maintained within  $20\text{ }^{\circ}\text{C} \pm 1\text{ }^{\circ}\text{C}$  and  $20\% \pm 3\%$ , respectively. The pressure was one atmosphere. As a result, the dew point was approximately less than  $0\text{ }^{\circ}\text{C}$ . The purpose of having this relatively dry environment was to avoid condensation on surfaces at the early stage of cooling. However, long exposure to this condition would cause significant evaporation of the droplet before icing. To minimize evaporation and also to ensure that all droplets land on dry surfaces rather than condensate, during deposition the cold plate was maintained at  $5\text{ }^{\circ}\text{C}$ , which was close to but higher than the dew point.

Temperature at the topmost point of the droplet, referred to as topmost temperature hereinafter, was measured using an infrared camera (FLIR 660 with spectral range 7.5–13  $\mu\text{m}$ ) with a frame rate of 30 frames per second. Using IR camera provides a non-intrusive method for monitoring temperature evolution with high spatial and temporal resolutions [22]. Based on the absorption coefficients of water and ice within the spectral range [23], the IR camera measures the temperature of  $\sim 10\text{ }\mu\text{m}$  surface layer of the droplet, and this thickness is much less than the center height of water droplets in the present work. The IR camera was kept normal to the droplet top to avoid the angular variation of infrared emissivity [24,25]. Emissivity calibration was conducted for water and ice for temperature ranging from  $-20\text{ }^{\circ}\text{C}$  to  $20\text{ }^{\circ}\text{C}$ , and was found to vary between 0.92 and 0.98. A constant emissivity 0.95 was used for IR data processing. The overall uncertainty for temperature measurement associated with the emissivity value and the IR camera is less than  $1.25\text{ }^{\circ}\text{C}$ , which accounts for less than 10% of the range of temperature change studied the present work. No temperature

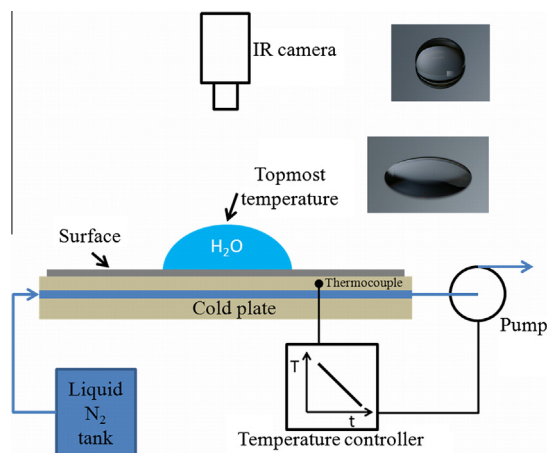


Fig. 1. Schematic of experimental setup. The two inset images are water droplets on the hydrophobic and hydrophilic surfaces, respectively.

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