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# Supercooled Water Droplet freezing and Ice Accretion on Cold Surface

*By Xiaomin Wu*[*], *Jingchun Min*[±], *Xuan Zhang*[•] *& Xin Liu*[○]

*Supercooled water droplet freezing exists in nature and engineering areas. To better understand the water freezing characteristics, we have done a series of research, which involve the supercooled water droplet nucleation and freezing, supercooled water droplet impacting and freezing dynamics, and ice accretion caused by supercooled water droplets in airflow, the present paper introduces some of them, aiming to concentratively show some fundamental aspects of supercooled water droplet freezing and ice accretion. The relevant work and results can be described as follows: the nucleation temperatures of sessile water droplets on a cold plate are experimentally studied, and the result supports that ice nucleation is a random process and a smaller droplet yields a lower nucleation temperature; the freezing process of a supercooled water droplet on a cold plate is investigated, and the droplet freezing rate and final droplet profile are obtained; the impacting-freezing dynamics of a supercooled water droplet on a cold surface is studied numerically, three different morphologies of full rebound, partial rebound and full adhesion are identified; a theoretical model is built to describe the aircraft icing caused by supercooled water droplets in clouds, and the ice accretion process is simulated.*

**Keywords:** *supercooled water droplet, nucleation temperature, freezing characteristics, impacting and freezing dynamics, ice accretion, cold surface*

## Introduction

Supercooled water droplet freezing is observed in nature and engineering fields such as aerospace, power and communication, and cryogenic engineering and refrigeration, so studies on such phenomena are of practical importance. The macroscopic icing in reality is usually a process of a single supercooled water droplet beginning to freeze after interacting with a cold wall and gradually forming an ice layer. To better understand the water freezing characteristics, we have done a series of research, which involve the supercooled water droplet nucleation and freezing [1-5], supercooled water droplet impacting and freezing dynamics [6-11], and ice accretion caused by supercooled water droplets in airflow [12-14], the present paper introduces some of them, seeking to concentratively reveal some fundamental aspects of supercooled water droplet freezing and ice accretion. The paper contains seven sections, Section 1 is the

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introduction, it elaborates the research background and significance, Section 2 is the literature review, it describes the research status, Sections 3-6 present and discuss the supercooled water droplet nucleation and freezing processes, water droplet impacting and freezing dynamics, and ice accretion characteristics, with they each containing their respective methodology, results and discussions, and Section 7 provides a concluding remark.

## Literature Review

Icing nucleation of a supercooled water droplet is the beginning of water droplet freezing and the nucleation temperature determines the initial conditions for the whole droplet freezing process and consequently the freezing rate and time. Jung et al. [15] reported that evaporative cooling of a supercooled liquid by airflow resulted in ice crystallization by homogeneous nucleation at the droplet-air interface as opposed to heterogeneous nucleation at the substrate surface. Chu et al. [16] observed condensed droplet freezing on s superhydrophobic surface and stated that Wenzel droplets with larger contact areas had shorter freezing delay times. Seidler and Seeley [17] proposed a correlation for the nucleation rate of sessile water droplets on a cold plate based on classical nucleation theory. Although many studies have been done, great uncertainties exist with the estimated nucleation temperature.

A supercooled water droplet placed on a cold wall may freeze and eventually form a pointy shape different from that before freezing due to the volume expansion caused by the phase change from water to ice [18]. Anderson et al. [19] compared various simplified models for simulating the shape change of a water droplet during freezing, and built a dynamic growth angle model by assuming a plane freezing front. Marin et al. [20] developed a modified fixed contact line model by assuming a spherical freezing front, but they treated the angle between ice-water and ice-air interfaces as a constant of 90° and analyzed only the tip formation instead of the entire freezing process. Although many works on droplet freezing have been done, most of them either ignored the influence of gravity or failed to consider the effect of supercooling, which can alter the ice-water mixture properties.

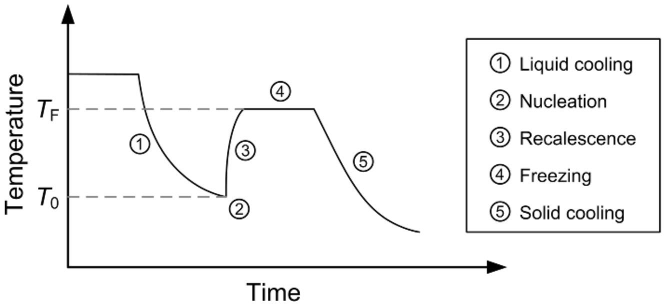
The impacting-freezing phenomenon of a supercooled water droplet involves two processes of impacting and freezing. Jin et al. [21] experimentally investigated the impacting-freezing process of a water droplet on cold cylindrical surface and observed successive freezing processes of water droplet on an ice surface. Yao et al. [22] experimentally and numerically studied the freezing of a room temperature water droplet impacting on a cold surface, the numerical model includes the VOF (volume of fluid) and Solidification/Melting model. Ding et al [23] studied the dynamic behavior of a water droplet impacting on superhydrophobic surfaces with different inclinations and supercooling degrees, they observed that the droplet might undergo full rebound, partial rebound and no rebound with reducing surface temperature. The previous investigations mostly focused on the impacting-freezing process of a room temperature droplet on a cold surface.

Most simulation models for ice accretion start from the classical Messinger icing model [24]. In 1990s, NASA conducted extensive experiments on aircraft icing [25], which provide useful validations for simulations. Many investigators modified the Messinger model that considers only the energy balance between the latent heat release and heat transfer at gas/liquid interface but omits the heat conductions in the ice layer and water film, e.g. Al-Khalil et al. [26], Myers et al. [27] and Du et al. [28] took into account the existence of rime ice as well as the effect of runback water in their models but they simplified the rime ice as property-constant glaze ice and assumed that the rime ice had the same physical properties as the glaze ice at constant.

## Nucleation Temperature of Supercooled Water Droplets on a Cold Plate

Fig. 1 illustrates the temperature transition during the whole freezing process of a water droplet on a gradually cooled cold plate, which involves five distinct stages including: (1) liquid cooling (supercooling), (2) nucleation, (3) recalescence, (4) freezing, and (5) solid cooling [29]. According to the nucleation theory, nucleation takes place when the water temperature drops to a value which is obviously lower than the water freezing point, and the water temperature at this time is called the nucleation temperature. The nucleation/ recalescence stage is the starting point of the freezing process, at which the water turns into an ice-water mixture, which varies the thermophysical properties at the solidification stage.

**Figure 1.** *Temperature Transition during freezing process of a Water Droplet*



Experiments are conducted to investigate the icing nucleation characteristics of water droplets on a cold plate. Fig. 2 shows the experimental system, which consists of a test section, a semiconductor thermoelectric cooler system, a data acquisition system and a photograph acquisition system. Experiments are done on a large number of droplets and the experimental results are statistically analyzed.

**Figure 2.** *Schematic Diagram of the experimental System used for the supercooled Water Droplet Nucleation Tests*

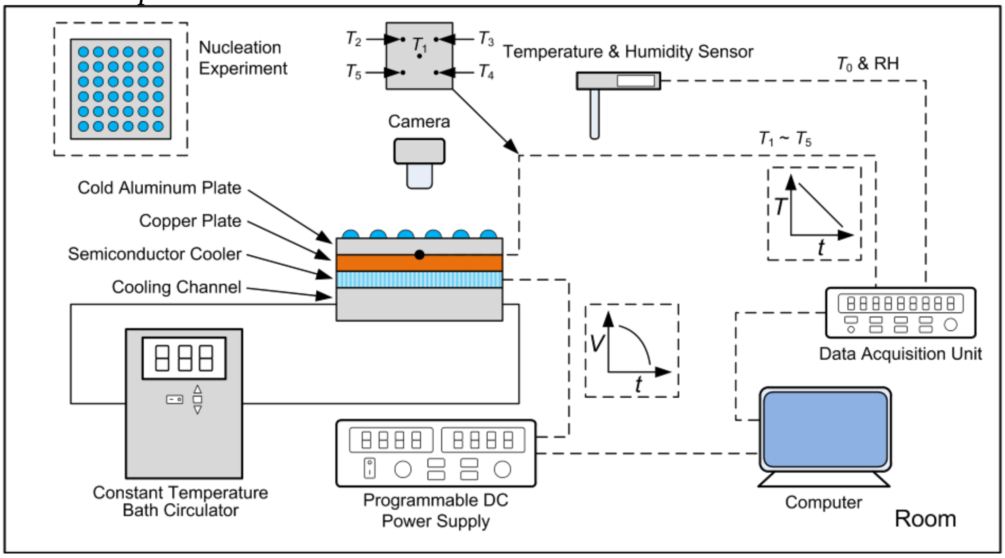
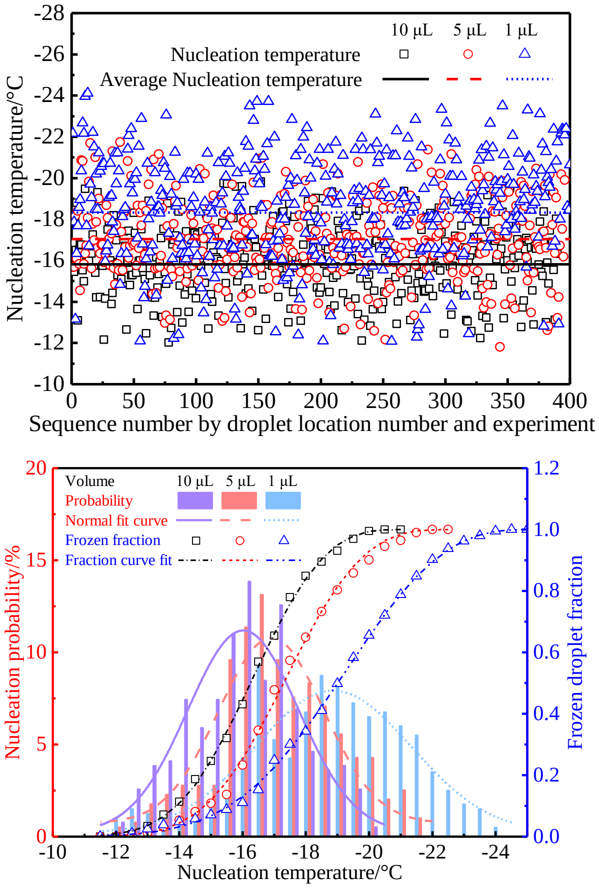


Fig. 3 depicts the nucleation characteristics of water droplets with different volumes on a cold plate. Fig. 3(a) suggests that the ice nucleation in a supercooled droplet is a random process and the nucleation temperature data scatter over a wide range of temperature. As the droplet becomes smaller, the nucleation temperature data scatter more widely, with their average becoming lower. Fig. 3(b) indicates that the variation of the nucleation probability with the nucleation temperature basically satisfies a normal distribution. A smaller droplet tends to have a higher frozen droplet fraction for a given temperature. The average nucleation temperature decreases and the standard deviation increases as the droplet volume decreases. The average nucleation temperatures are -15.8, -17.0 and -18.3°C for droplets having volumes of 10, 5 and 1 μL with standard deviations of 1.7, 1.9 and 2.5°C.

**Figure 3.** *Nucleation characteristics of water droplets with different volumes on a cold aluminum plate: (a) nucleation temperature, (b) nucleation probability and frozen droplet fraction*



More detailed descriptions of this research can be found in our previous work [1].

## Shape Evolution of Water Droplets during Freezing on a Cold Surface

Experiments are carried out to study the water droplet freezing process. The experimental setup used is similar to that as seen in Fig. 2, different sizes of water droplets are placed on cold aluminum plates having different contact angles and temperatures. Fig. 4 presents the freezing process of a 20 μL water droplet on a cold plate whose contact angle is 78° and temperature is -18.4°C, the process includes the supercooling (Fig. 4a), nucleation/recalescence (Fig. 4b) and solidification (Fig. 4c-f) stages as well as the formation of a final pointy shape (Fig. 4f). At the nucleation/recalescence stage, the droplet state changes from water to water-ice mixture with its appearance turning from brightness to darkness; at the solidification stage, the freezing front moves from the droplet bottom to the top.

**Figure 4.** *Freezing process of a 20 μL supercooled water droplet on a cold plate whose contact angle is 78° and temperature is -18.4°C*

(a) $t$ = 0.0 s (b) $t$ = 0.1 s (c) $t$ = 2.0 s (d) $t$ = 6.0 s (e) $t$ = 10.0 s (f) $t$ = 11.7 s

A theoretical model is built to simulate the freezing process of a sessile water droplet, it considers both the supercooling effect on droplet physical properties and the gravity effect on droplet shape and assumes the freezing front (ice-water interface) to be a spherical surface. Fig. 5a depicts the stage of nucleation/recalescence, which occur instantaneously and last only about 0.1 s. At that stage, the water turns into an ice-water mixture, which alters the thermophysical properties at the solidification stage. Fig. 5b describes the solidification stage, during which the upper unfrozen ice-water mixture changes completely into solidified ice.

**Figure 5.** *Freezing process of a supercooled water droplet on a cold surface: (a) nucleation/recalescence stage, (b) solidification stage.*

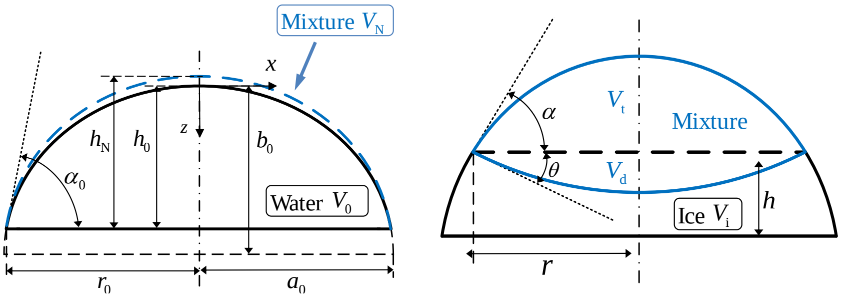


Fig. 6 compares the droplet shapes at different times calculated by the model with those obtained from the experiments. The initial droplet shape (t = 0.0 s), nucleation/recalescence shape (t = 0.1 s) and final frozen shape (t = 11.7 s) given by the model all agree well with the experimental observations. The freezing time obtained from the experiment is 11.7 s while that calculated by the model is 11.9 s, they agree within 2%, supporting the reliability of the model. Fig. 7 shows the evolution of droplet profile obtained from the experiment and that of freezing front calculated by the model. As time goes by, the freezing front moves from the bottom liquid-solid interface up to the topmost point, and a pointy tip eventually forms. The final droplet volume is 21.81 μL, as compared to the initial volume of 20 μL, yielding an expansion rate of 1.091, due to the smaller density of ice relative to water.

**Figure 6.** *Droplet shape comparisons between calculation and experiment*

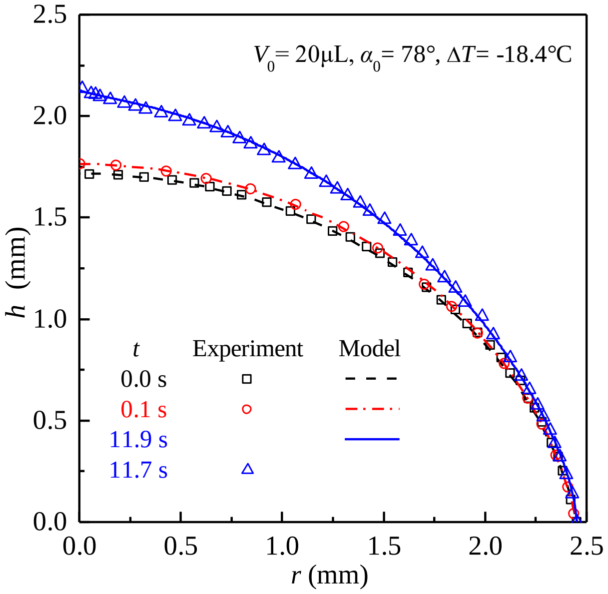
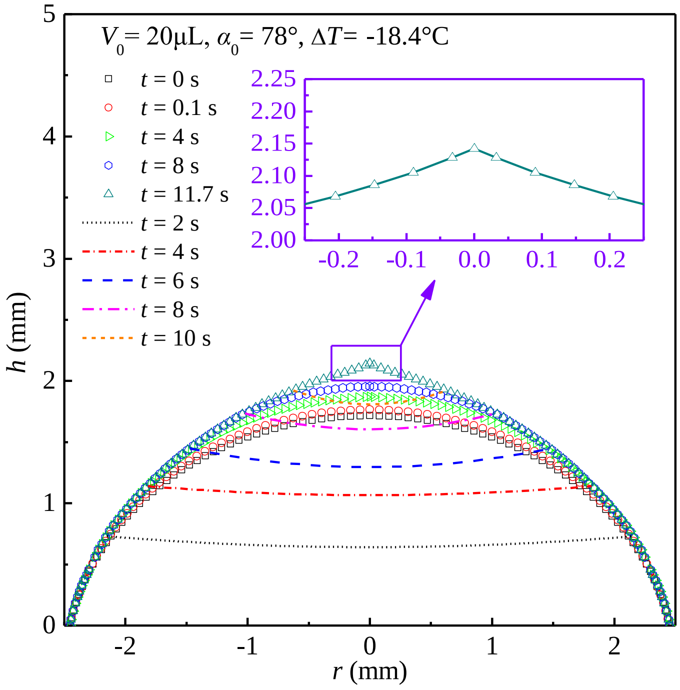


**Figure 7.** *Evolution of droplet profiles obtained from calculation and experiment*

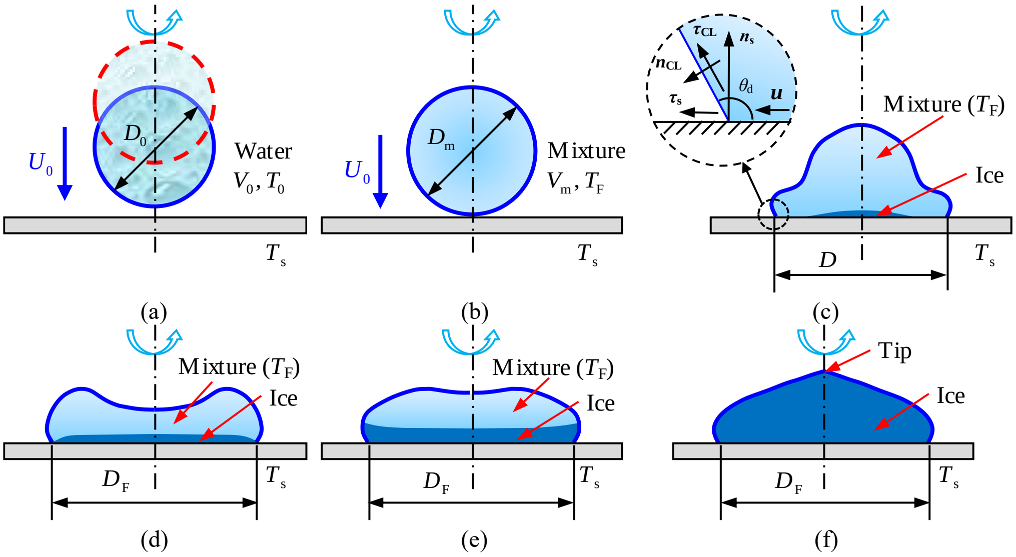


More information about this research can be found in our previous work [2].

## Impacting-Freezing Dynamics of Supercooled Water Droplets on Cold Walls

When a water droplet impacts on a wall, it usually experiences spreading, receding, oscillating and stable stages. When a supercooled water droplet impacts on a cold wall, it generally undergoes the nucleation, recalescence, freezing and solid cooling stages. Heat transfer occurs with phase change during the droplet impacting and freezing processes. Since the impacting can promote ice nucleation, the nucleation occurs upon the droplet touching the cold surface, with the recalescence spreading more rapidly. The supercooled water droplet can therefore be assumed to complete the nucleation/recalescence stage upon touching the cold surface or in a very short time (Figs. 8a and 8b), it changes into a water-ice mixture instantaneously, which has different physical properties from water, with its temperature recovering to the freezing point. After impacting on the cold surface, the droplet deforms and the contact line spreads under the action of the inertia force, surface tension and gravity, accompanied by the propagation of the freezing front along the cold surface inside the droplet (Fig. 8c). The droplet then starts to recede after reaching the maximum spreading state (Fig. 8d). During the spreading or receding stages, the contact line may be frozen and can no longer move. The upper part of the droplet continues oscillating with the freezing front advancing from the bottom to the top (Fig. 8e) until the water-ice mixture becomes completely solidified (Fig. 8f). A conical tip forms at the end of the freezing process.

**Figure 8.** *Schematic of the impacting-freezing process of a supercooled water droplet on a cold surface: (a) before touching the surface (supercooling stage), (b) upon touching the surface (nucleation-recalescence stage), (c) spreading stage (freezing stage), (d) receding stage (freezing stage), (e) oscillating stage (freezing stage), (f) stable stage (completely frozen).*



Experimental and numerical studies are conducted to investigate the supercooled water droplet impacting and freezing behaviors. Experiments are done using an apparatus as seen in Fig. 9 and numerical simulations are implemented using the VOF (Volume of Fluid) method and DCA (Dynamic Contact Angle) model. Fig. 10 compares the droplet temporal profiles at different times during the impacting processes of a room temperature droplet on a room temperature surface and a supercooled water droplet on a cold surface between the experiments and simulations. The simulations agree well with the experiments, supporting reliability of the model.

**Figure 9.** *Schematic diagram of the experimental apparatus used for supercooled water droplet impacting and freezing tests*

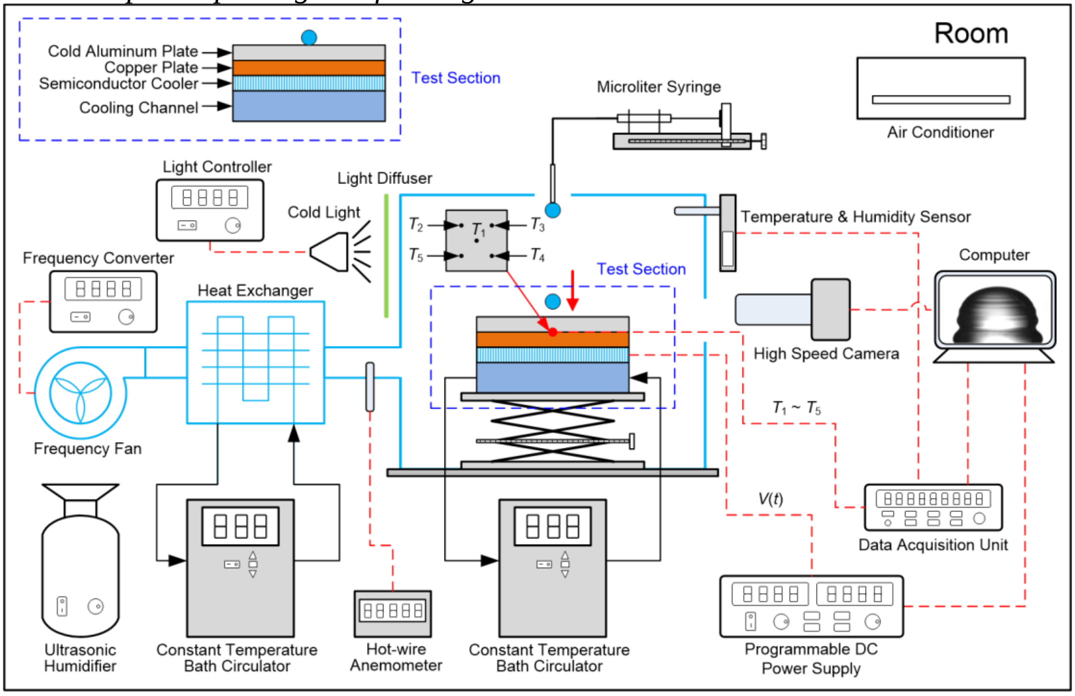
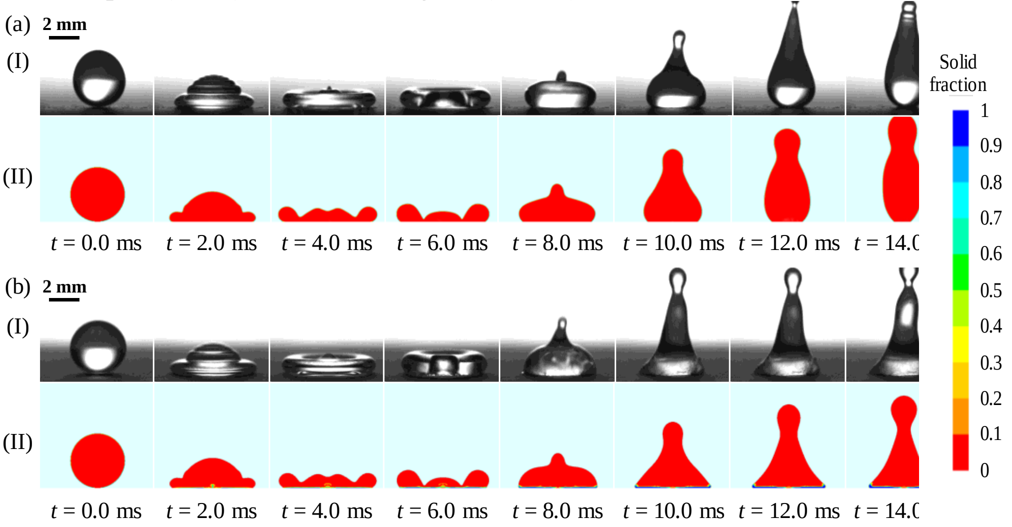
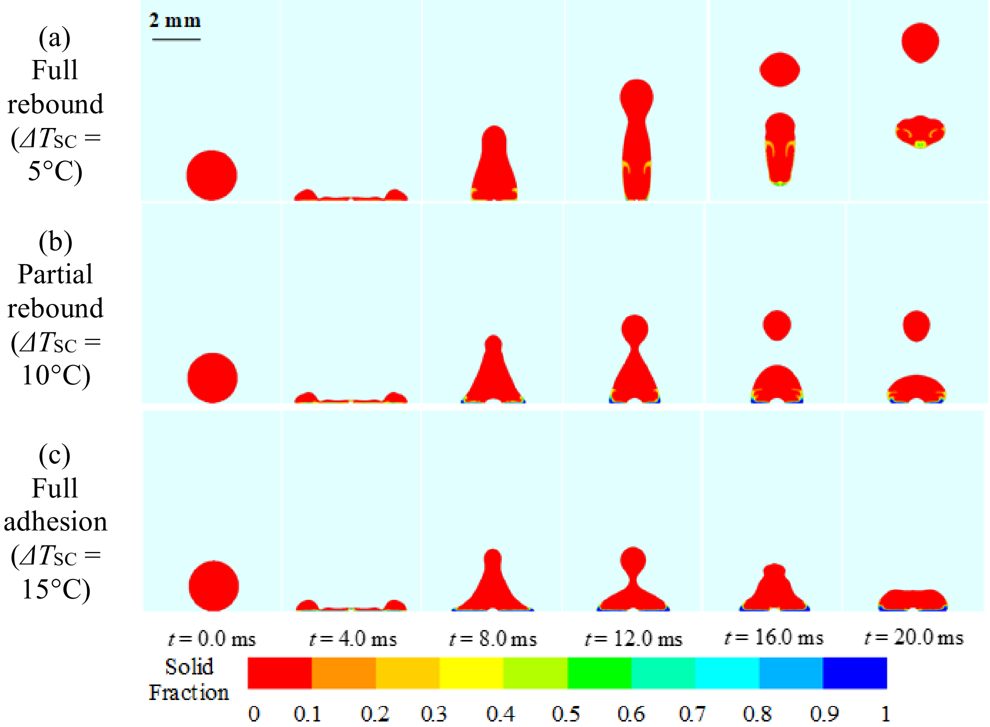


**Figure 10.** *Comparison of the temporal droplet profiles at different times between (I) experiments and (II) simulations in the impacting processes of (a) a room temperature droplet on a room temperature Surface (15°C) and (b) a supercooled water droplet (-5°C) on a cold surface (-30°C)*



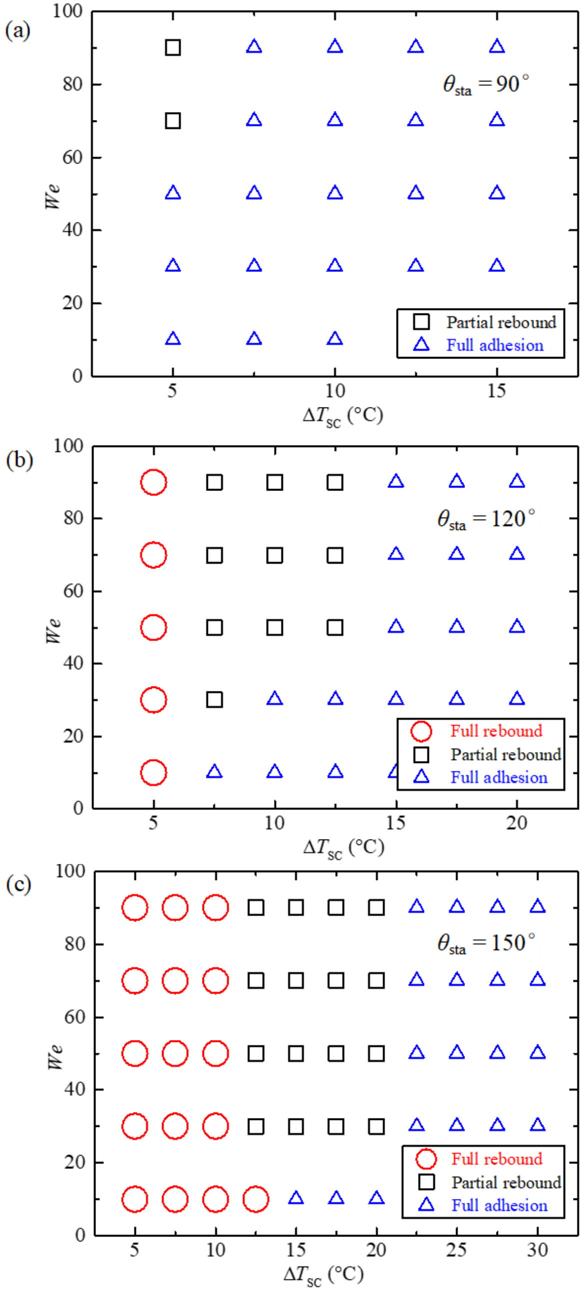
A supercooled water droplet impacting on a cold surface at the same Weber number ($We = \rho U_0^2 D_0 / \sigma$) may exhibit three different morphologies at different supercooling degrees ( $\Delta T_{SC}$ , difference between the freezing point and initial droplet temperature) as shown in Fig. 11, namely the full rebound, partial rebound and full adhesion, indicating a competition between the fluid flow (reflected by the Weber number) and heat transfer (or phase change, characterized by the supercooling degree) near the cold surface. At a smaller supercooling degree (Fig. 11a), the contact line motion in impacting-freezing process is dominated by the fluid flow inside the droplet while little liquid is frozen near the cold surface due to the weak heat transfer and slow freezing speed. The droplet finally bounces off the cold surface. In contrast, a larger supercooling degree (Fig. 11c) enhances the heat transfer and increases the liquid freezing speed. Consequently, the contact line is frozen in impacting-freezing process and the droplet finally adheres to the cold surface. At an intermediate supercooling degree (Fig. 11b), the upper part breaks away from the droplet while the bottom part sticks to the cold surface.

**Figure 11.** *Three morphologies of (a) full rebound ($\Delta T_{SC}$ = 5°C), (b) partial rebound ($\Delta T_{SC}$ = 10°C) and (c) full adhesion ($\Delta T_{SC}$ = 15°C) when a supercooled water droplet impacts on cold hydrophobic surfaces ($\theta_{sta}$ = 120°, We = 50) having different supercooling degrees*



Based on the definitions of the three droplet morphologies in Fig. 11, Fig. 12 is plotted to present the morphology map of the rebound and adhesion when a supercooled water droplet impacts on a cold hydrophobic surface having different supercooling degrees ($\Delta T_{SC}$) and contact angles ($\theta_{sta}$) under different Weber numbers (*We*). When the surface becomes more hydrophobic, the limits for the full rebound and adhesion both shift to the larger supercooling degree. A more hydrophobic surface causes a smaller spreading, weakening the heat transfer and liquid freezing. The droplet is thus easier to bounce off the cold surface at the same Weber number and supercooling degree. So, a greater supercooling degree is needed for the full adhesion of a supercooled droplet on a more hydrophobic surface. Noting that the droplet shows either full rebound or full adhesion while partial rebound is not observed at a low Weber number (*We* = 10) in Fig. 12, the reason is that the inertial force is too weak to break the droplet in the impacting-freezing process compared to the surface tension.

**Figure 12.** *Morphology map of the rebound and adhesion when a supercooled water droplet impacts on a cold hydrophobic surface under different Weber numbers, supercooling degrees and contact angles*



More detailed descriptions of this research can be found in our previous work [6].

## Ice Accretion Caused by Supercooled Droplets Impinging and Sticking on a Cold Surface

Ice accretion on a cold surface exists in nature and engineering processes, different conditions may cause different types of ices, with rapid freezing leading to porous rime ice and slow freezing to dense glaze ice, as illustrated by Fig. 13. Fig. 14 gives a picture taken during icing wind tunnel tests, it includes the variation of gray value along the ice thickness, which can reflect the existences of a sublayer of rime ice and an upper layer of glaze ice.

**Figure 13.** (a) *Slow freezing under a cold temperature leading to the formation of glaze ice, and (b) Rapid freezing under a very cold temperature leading to the formation of rime ice*

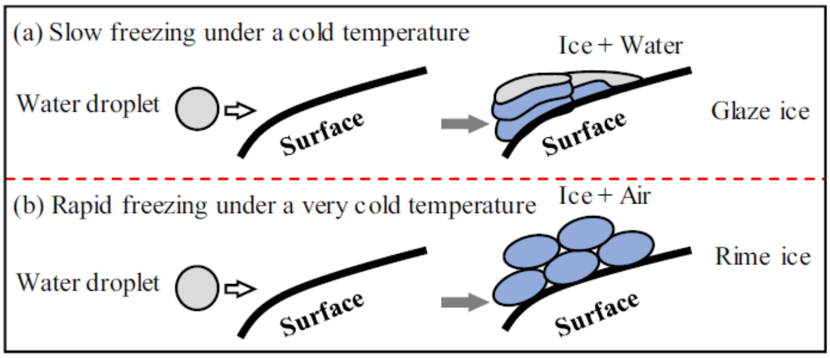
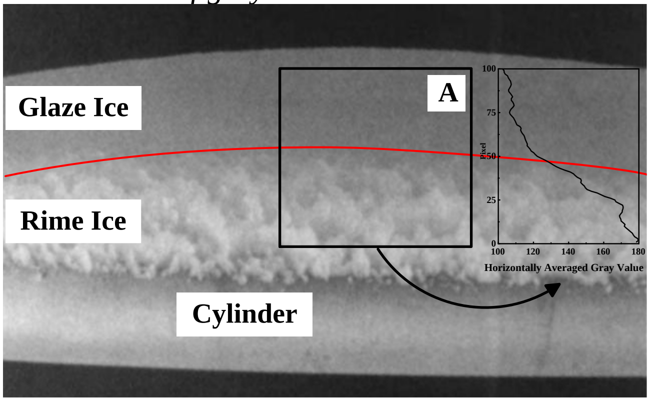
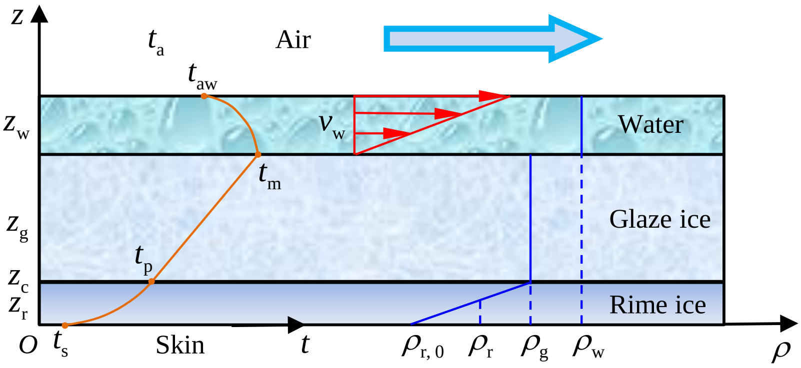


**Figure 14.** *Wind tunnel experimental icing types on a cylindrical surface and the variation of gray value with the ice thickness*



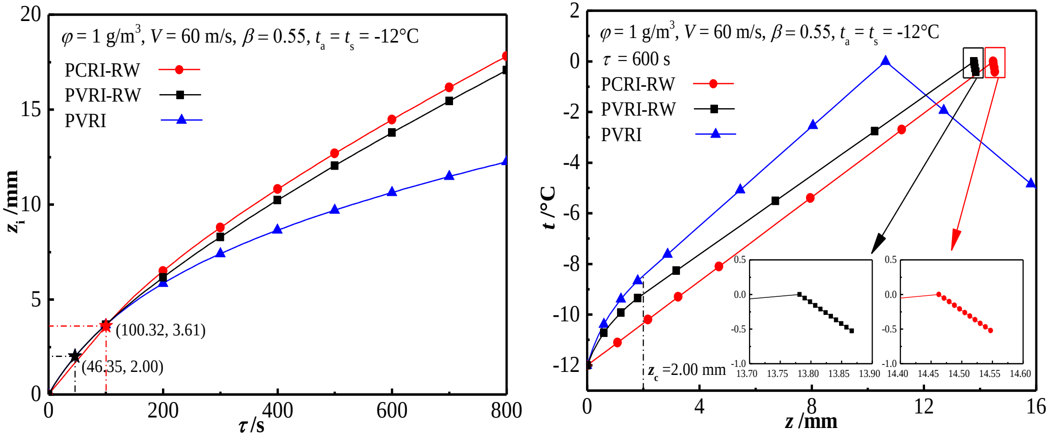
A one-dimensional model is built to simulate the aircraft icing process, which is caused by supercooled droplets impinging and sticking on the outer surfaces of an aircraft flying in clouds. Airflow parameters influence the final icing types including the rime ice and glaze ice. No matter which type of ice forms, a sublayer of rime ice, which has a porous structure and variable physical properties, usually forms due to the high freezing rate of supercooled droplets at the initial stage. At the later stage when glaze ice develops on rime ice, water film may form simultaneously on the glaze ice because only a portion of supercooled droplets can freeze instantaneously. The stage of rime ice formation is generally called the dry mode icing and the stage after water film emergence the wet mode icing. The time when the dry mode transits to the wet mode is referred to as the critical time and the ice thickness at that time as the critical ice thickness. Fig. 15 illustrates the basic physical model that consists of the rime ice layer, glaze ice layer, and water film, density varies linearly along the rime ice thickness but remains constant in the glaze ice layer and water film, Couette flow takes place in the water film, implying that water flows with a linear velocity distribution, through which the runback water effect is taken into account. The model is validated by comparing the calculations with the experiments in Refs. [30-32], the detailed information on the comparison can be found in our previous paper [12].

**Figure 15.** *Physical model including property-variable rime ice and runback water*



Calculations are conducted for $MVD$=20 μm mean volumetric droplet diameter, $V = 60$ m/s airflow velocity, $\varphi = 1$ g/m$^3$ liquid water content, $\beta = 0.55$ collection efficiency, $t_a = t_s = -12$°C air and solid surface temperatures, and $t_m = 0$°C ice melting temperature, these parameters are taken based on the data in Ref. [33]. Fig. 16 shows the results generated by the PCRI-RW, PVRI-RW and PVRI models, where PVRI stands for property-variable rime ice, PCRI for property-constant rime ice, and RW for runback water, so PVRI-RW is the model we recommend, it considers both the rime ice variable property and runback water effects, while the other two are incorporated for the purpose of comparison.

**Figure 16.** *Ice thickness evolutions and temperature profiles at* $\tau = 600\ \mathrm{s}$ *generated by various models (PVRI: property-variable rime ice, PCRI: property-constant rime ice, RW: runback water)*



(a) (b)

In Fig. 16(a), the asterisks express the critical point. The critical time and critical ice thickness given by the PVRI model are the same as those by the PVRI-RW model because they share the same dry mode icing stage. However, the PVRI-RW model yields a shorter critical time and a smaller critical ice thickness than the PCRI-RW model because the PVRI-RW model treats the rime ice as a porous medium that has smaller effective thermal conductivity and density than the glaze ice while the PCRI-RW model treats the rime ice as the glaze ice and assumes that the rime ice has the same properties as the glaze ice as constant. In Fig. 16(b), the rime ice temperature profiles generated by the PVRI and PVRI-RW models are logarithmic curves due to the variations of the rime ice physical properties with the ice thickness while that by the PCRI-RW model is a straight line. The water film temperature profiles given by these three models differ from one another due to different water film thicknesses. The water film temperature profiles given by the PVRI-RW and PCRI-RW models both are parabolic.

More results and further discussion can be found in our previous papers [12, 14].

## Concluding Remarks

A series of research have been done to investigate the supercooled water droplet freezing and ice accretion on cold surface, some of them are concentratively introduced in this article, the relevant works and achievements can be described as below:

(1) The nucleation temperatures of sessile water droplets with different volumes on a cold plate are experimentally studied, and statistical analyses are implemented to obtain the average nucleation temperatures and standard deviations. The results support that supercooled water droplet nucleation is a

random process, with the nucleation temperature scattering over a wide range of temperature that approximates a normal distribution, a smaller droplet yields a lower nucleation temperature and a larger standard deviation.

(2) The freezing process of a supercooled water droplet on a cold plate is studied theoretically and experimentally. A model that considers both the effect of supercooling degree on droplet physical properties and that of the gravity on droplet shape is built to simulate the droplet freezing behaviors. The calculation results suggest that the final droplet profile is less dependent on the supercooling degree, and the freezing rate increases but the freezing time decreases as the plate temperature declines;

(3) The impacting-freezing dynamics of a supercooled water droplet on a cold surface is studied experimentally and numerically. A numerical model that considers both the supercooling effect on droplet physical properties and the dynamic contact angle effect on contact line motion is presented to mimic the droplet impacting-freezing behaviors. Three different morphologies of full rebound, partial rebound and full adhesion are identified in the impacting-freezing process of a supercooled droplet on a cold hydrophobic surface;

(4) A one-dimensional model is presented to describe the ice accretion process on a cold surface, which can be divided into the dry and wet mode icing stages. Rime ice forms on the surface skin at the dry mode icing stage while glaze ice grows on the rime ice and water film develops on the glaze ice at the wet mode icing stage. The calculation results show that the rime ice property variability and runback water effect are affected by the airflow parameters, which influence the heat conductions in the ice layer and water film and consequently the ice accretion characteristics.

Items 1 and 2 are concerned with stationary droplets, they may provide some fundamental understanding of droplet freezing, item 3 involves impact droplets, which are directly related to item 4, i.e. the ice accretion caused by supercooled water droplets in airflow, so item 3 may provide some insights into item 4. Nevertheless, it should be noted that the droplets in item 3 have much larger diameters (a few millimeters) than those in item 4 (0.02 mm), at the same time, the droplets in item 3 have much lower impact velocities (a few meters per second) than those in item 4 (60 m/s), as a result of these, the weber numbers in item 3 are about one order of magnitude smaller than those in item 4. The reason for taking smaller Weber numbers in item 3 (less than 100) is because droplet may break up and splash when it impacts on a solid surface with Weber numbers exceeding 100, making the simulations more difficult to be implemented. Larger Weber numbers should be challenged in future work so that item 3 can contribute more to item 4.

## Acknowledgement

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