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# Unified icing theory based on phase transition of supercooled water on a substrate

Weiliang Kong, Hong Liu [*]

*School of Aeronautics and Astronautics, Shanghai JiaoTong University, Shanghai 200240, China*

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ABSTRACT

Lack of knowledge on substrate icing hinders us from predicting the effects of typical substrate properties in icing and anti-icing research. The pattern formation and tip shape of icing on a substrate must be explored to understanding the physics of icing on a substrate. In this paper, the pattern, velocity and tip shape of ice on substrates with different thermal conductivities and surface energies are evaluated in a series of experiments. Experimental results show that as supercooling and thermal conductivity increase, the ice on substrate evolves from a single-needle dendrite to a smooth ice film. In contrast, for free icing the same evolution process can be completed at a higher supercooling than substrate icing. Experimental results on hydrophilic and Plexiglas surfaces demonstrate an abruptly decrease of velocity and Peclet number of ice occurs at approximately 271.6 K, whereas the tip shape of ice does not change obviously; while for free icing this phenomenon is not found. Furthermore, a theoretical analysis on phase transition of substrate icing is performed. It shows that the surface energy of substrates can increase the size of metastable cubic ice and reduce the temperature on the ice tip by 1.5 K compared to free ice, thereby causing the decrease in the velocity and Peclet number of ice. Also the heat conduction of substrates decrease the heat flux in ice growth direction and lead to absolute stability of substrate icing in lower supercooling compared to free icing. In terms of the comparison between free icing and substrate icing, a unified icing theory is proposed to describe the equilibrium icing mode, the non-equilibrium growth state, and the corresponding growth equations. Using this theory, the velocity and pattern of substrate icing with different supercooling, thermal conductivity, and surface energy can be predicted well, as well as these results of free icing.

## 1. Introduction

The solidification of supercooled water on a substrate has attracted much attention in many technical fields, including the icing of aircraft, power systems, or antennae [1]. The role of substrates in icing has become increasingly important over the last decade, especially in consideration of its effect on heat flux [2], ice fraction [3], and icing evolution [4]. While several anti-icing studies have explored the effect of surface properties on icing [5,6], the current icing theories based on free ice growth cannot adequately interpret complex substrate icing phenomena.

Over the past decades, icing has been studied based on the knowledge of free ice crystal growth. Free ice typically has a six-sided dendritic structure and a parabolic-shaped tip. As a result of anisotropy, this structure is transformed into two hollow, six-sided pyramids that are joined at their apices at approximately 271.65 K, and the angle between these pyramids increases along with supercooling [7,8]. Moreover, the ice crystal is not reflective symmetric on the basal plane [9].

The pattern of ice varies along with its growth velocity. As the growth velocity increases from 0, the solidification front will transit from planar to dendritic and eventually transition back to planar [10]. In the former transition, the smooth interface becomes unstable because of amplified perturbations [11]. Meanwhile, the latter transition can be ascribed to absolute stability, above which the planar interface becomes morphologically stable against small perturbations [12]. The experiment shows that free ice crystals turn into platelets in supercooling higher than 14.5 K [13].

Unlike those of free icing, the shape and pattern of substrate icing remain unclear. Lindenmeyer found that icing on brass and glass is faster than free icing [14]. Camp and Barter observed that dendritic ice growth differs from the orientation of the c-axis on aluminum [15]. Qin et al. observed ice growth as thin film on metal in low supercooling [16]. Kong and Liu observed four-edged dendritic ice and ice film on different substrates in supercooling lower than 7 K [4]. Schremb et al. found that icing had a low dependence

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[*] Corresponding author.
*E-mail address:* Hongliu@sjtu.edu.cn (H. Liu).

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## Nomenclature

| | |
|---|---|
| $\Delta H_{h-c}$ | the volume energy difference between Ih and Ic |
| $\Delta \bar{G}_{h-c}$ | the energy difference per unit mass when the control volume turns from Ic to Ih |
| $L_c$ | the latent heat of cubic ice |
| $L_h$ | the latent heat of hexagonal ice |
| $\gamma_c$ | surface energy of cubic ice-water interface |
| $\gamma_h$ | surface energy of hexagonal ice-water interface |
| $\gamma_{iw}$ | surface energy of ice-water interface |
| $\gamma_{si}$ | surface energy of substrate-ice interface |
| $\gamma_{sw}$ | surface energy of substrate-water interface |
| $\rho_i$ | density of ice |
| $\rho_c$ | density of cubic ice (Ic) |
| $\rho_h$ | density of hexagonal ice (Ih) |
| $\theta_c$ | the contact angle of Ic-water-substrate interface on substrate |
| $\theta_h$ | the contact angle of Ih-water-substrate interface on substrate |
| $\theta_i$ | the contact angle of ice-water-substrate interface on substrate |
| $h_{ice0}$ | the initial thickness of ice on substrate |
| $I_0()$ | zero order of modified Bessel function of the first kind |
| $I_1()$ | first order of modified Bessel function of the first kind |

| | |
|---|---|
| $Ic$ | the phase of cubic ice |
| $Ih$ | the phase of hexagonal ice |
| $Iv_{2D}()$ | Ivantsov equation in two-dimension |
| $K_0()$ | zero order of modified Bessel function of the second kind |
| $K_1()$ | first order of modified Bessel function of the second kind |
| $Pe$ | Peclet number of dendritic ice |
| $R$ | the tip radius of dendritic ice |
| $r$ | the radius of control volume on ice tip |
| $r_c$ | the maximum stable size of cubic ice (Ic) |
| $T$ | the ambient temperature |
| $T_m$ | the melting temperature of ice |
| $T_{abs}$ | the temperature of absolute stability |
| $T_{mc}$ | the melting temperature of cubic ice |
| $T_{mh}$ | the melting temperature of hexagonal ice |
| $v$ | the velocity of ice tip |
| $v_x$ | the velocity of horizontal substrate icing |
| $v_y$ | the velocity of vertical substrate icing |
| $v_{abs}$ | the growth velocity of absolute stability |
| $\dot{H}_s$ | heat flux in substrate |
| $\dot{H}_w$ | heat flux in water |

on the properties of substrate in high supercooling [17]. A united understanding on substrate icing has not been formed.

The pattern formation of substrate icing also differs from that of free icing. As the growth velocity increases, the ice perpendicular to the substrate transforms from planar ice film to dendritic ice, while the ice parallel to the substrate transforms from dendritic ice to smooth ice film [4].

Given that the pattern formation depends on the temperature distribution around the ice-water interface, the heat transfer and growth of the substrate ice tip must be investigated to understand its pattern formation.

Ivantsov's theory describes the heat transfer and growth of a parabolic dendritic crystal in supercooled liquid [18]. This theory is extended to include a more general form of elliptical paraboloid-shaped dendrite [19] that is suitable for the anisotropic dendritic ice [20]. However, the solution describes a family of solutions with different tip radii. To obtain a unique solution to the problem, both the capillary effect and anisotropy are considered, and several theories, including marginal stability theory [21], microscopic solvability theory [22], and interfacial wave theory [23], are selected.

The previous explorations on the mechanism of substrate ice growth are very limited compared with those on the mechanism of free icing. Qin et al. assumed a 200 μm initial thickness of substrate ice film based on the results of an experiment conducted in low supercooling [16]. Kong and Liu assumed that the substrate ice tip has a parabolic shape and an initial thickness of 20 μm on a metallic substrate and in supercooling of 1–7 K [4]. Schremb et al. assumed a parabolic-shaped substrate ice tip which radius depends on growth velocity [17]. These studies typically assume that the substrate ice has a parabolic shape, but only few experimental observations of substrate ice tip are available.

The effects of the surface energy of the substrate on icing also need to be investigated. As discussed above, free dendritic ice has an elliptical paraboloid-shape on the tip, while the surface energy of substrate determines the contact angle of ice-water-substrate on the surface, which is independent of the thermal conditions. In this case, the ice would no longer have an elliptical paraboloid shape and its heat transfer differs from that of free dendritic ice. The change in the Gibbs free energy of ice can also affect its phase transition behavior. According to Ostwald's rule of stages [24], the ice will enter a metastable phase during the icing process [25,26] in which its properties will be changed. As the ice on the substrate has a small contact angle, part of its tip may be affected by the surface energy of the substrate.

In summary, substrate icing is much more complex and differs from free icing in terms of pattern formation and growth law. However, the growth and pattern formation mechanisms of substrate ice are often explored based on the knowledge regarding its tip shape.

Given the symmetry of dendritic ice, we can assume that on a thermal insulation substrate, the thermal condition of substrate icing is the same as that of free icing. In previous studies, the scale of the substrate ice tip is too small to be clearly observed and measured. However, the tip radius of ice is inversely proportional to its growth velocity. Therefore, the tip of ice on different substrates can be studied by performing an experiment in low velocity. Given its anisotropy, the ice crystal may have an aspect ratio as high as 30 on its tip [27]. The direction of the basal plane of ice must be identified before conducting observations and measurements on the ice.

The pattern transition of substrate icing must follow the same stability criteria for the solidification front with free icing [11,12] despite the differences in their evolutionary patterns. Therefore, a whole process of evolutionary pattern must be established to distinguish the state of pattern of substrate icing. Based on the observations of the ice tip, the heat transfer of substrate ice can be modeled and the stability criteria of substrate icing can be formulated. The theory of substrate icing can also be formulated based on the above results.

In this paper, the substrate icing with different thermal conductivities and surface energies is explored through an experiment. Substrate icing and free icing are compared in terms of their pattern, velocity, and shape. The effect of the substrate is also examined. The conclusions drawn from the equilibrium and non-equilibrium phase transitions of substrate icing and free icing are also used to formulate a unified theory of substrate icing.