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The crystallization of sub-cooled water: Measuring the front velocity and mushy zone composition via thermal imaging

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ABSTRACT

Water is one of the most abundant resources on Earth and the study of its phase changes holds many practical implications to both science and technology. In the past, several investigations have researched aspects of the solidification process that occurs when water freezes to ice. One of these facets is its associated linear growth velocity. Traditionally, this has been measured either through free growth or capillary tube techniques. The current study uses a methodology where measuring this velocity is accomplished through the use of infrared imaging. A conventional velocity vs. sub-cooling plot was generated and the data was fitted to a modified Noyes and Whitney growth model. Furthermore, with the use of the thermal images, a novel model was developed to calculate the percentage of water that was frozen after the initial growth step. From this, a conversion vs. sub-cooling plot was produced and the results showed that for the primary growth step, less ice forms under higher sub-coolings.

1. Introduction

Water is one of the most prominent and important naturally occurring compounds found on Earth. It is also the only substance that can be found in large quantities in the solid, liquid, and gaseous states [1]. The transition from liquid water to solid ice is a phenomenon that has been studied at length due to its practical implications. The growth of an ice layer on a lake, the crystallization of water droplets in the atmosphere, and the stability of glaciers are all examples of the phase change's importance to natural scientists [2–4]. Furthermore, industrial applications cover a wide range of interests from the prevention of freezing in pipelines, the de-icing of airplanes, to product preservation [5,6].

The crystallization of ice can occur from either one of two pathways. Either the ice will grow from sub-cooled water or through the direct deposition of water vapor [7]. The research concerned in this article will focus on the former pathway. The formation of ice, as with any other crystallization process, can be separated into three distinct kinetic steps [8,9]. The first step involves the initial propagation of a thin film of the solid phase and a release of the latent heat of formation. The second step includes the thickening of this initial film whose development slowly releases more heat. The final step of the process involves the decrease in system temperature where bulk conversion of the phase is achieved through solid state diffusional adjustments. An approximate timescale for the three kinetic steps is in seconds for the first, minutes for the second, and hours to days for the final phase [8,9]. Much research has been performed on measuring the velocity of the first phase of the crystallization of water at various system sub-coolings. Classically, the measurement of this velocity was performed using one of two general techniques [7]. The first method, often called free dendrite growth, minimizes substrate effects but difficulties arise when one wants to accurately measure the interface temperatures [10–12]. The second technique involves confining the growing ice into a thin tube that is submerged into a cooling bath [13–16]. One of the major disadvantages of this technique includes the increase in substrate effects on the crystal growth. Recent advancements have allowed the initial propagation velocity of the tetrahydrofuran/water hydrate front to be measured via thermal imaging [17]. This non-intrusive, free dendrite-like measurement technique allows for the temperature of the sub-cooled solution to be measured using an infrared camera that monitors the progress of the thermal front propagation associated with the exothermic release of the latent heat of crystallization. In this study, an identical technique was used to measure the velocity of the primary phase of ice crystallization under a range of sub-cooling levels.

Once the first phase of the crystallization process is completed, there exists both ice and a certain amount of unfrozen water. This is sometimes referred to as the "mushy zone" due to the fact that the volume of interest is composed of a mix of the solid and liquid phases [9]. For the crystallization of sub-cooled water, Hallett

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(1964) proposed a simple model that estimates the maximum possible ice conversion after the first crystallization step [11]. This proportion of the masses of water to ice is given as [11]:

$$\frac{\text{Water}}{\text{Ice}} = \int_0^{\Delta T} \frac{C_p dt}{\Delta H_f} \tag{1}$$

where C_p is the specific heat of water at constant pressure, T is the temperature, ΔT is the level of sub-cooling, and ΔH_f is the latent heat of fusion of ice. The results of this theoretical estimation are highly inaccurate as the calculation assumes the phase change occurs in an adiabatic system. In order for a crystallization process to proceed, heat must be lost in order to sustain crystal growth. Furthermore, at the onset of the mushy zone period, the remaining liquid may not reach its equilibrium temperature. This current study proposes a new means to calculate the actual composition of the mushy zone at the completion of the initial front propagation. Using thermal imagining, it is possible to accurately measure the level of system sub-cooling as well as the temperature of the liquid after the first phase of crystallization. With the velocity of the front and the temperature of the heat sink, the amount of heat lost to the system can also be calculated and used in this new model. With these parameters, the initial conversion of sub-cooled water to ice can be accurately calculated.

2. Materials and experimental apparatus

The experimental apparatus comprises of two 12.70 × 30.48 cm insulated Aavid Thermoalloy Hi-Contact Aluminum Cold Plates connected to a Neslab RTE 740 chiller. The cooling fluid running through the cold plates is a 50/50 by volume mixture of ethylene glycol and water. With the use of the chiller, this fluid can be set to any temperature between −40 and 200 °C and can be held constant with an accuracy of 0.01 °C. The cold plates are set on a Newport VH-3600-SG4-325A optical bench that is used to dampen any vibrations. Reverse osmosis water (0.22 μm filter) with a conductivity of 10 μS and a total organic content of <1 ppb is used in all the crystallization experiments. During the experiments, the water is placed in an aluminum dish of 19.60 cm inner diameter and 3.2 mm base thickness.

In order to measure the temperature throughout the crystallization process, a Jenoptik IR-TCM 384 infrared camera was used. It records images of 384 × 288 pixel resolution and can capture temperatures in between −40 and 300 °C. The camera underwent a calibration in order to obtain a measuring accuracy of 1–2% in the temperature range of −20 to 20 °C. Furthermore, it is capable of producing a noise equivalent temperature difference (NETD) of <0.08 °C. The camera is mounted on a stand and placed directly above the water-containing aluminum dish. A black tarp is used to cover the entire apparatus, acting as an optical barrier to prevent any reflections from interfering with the readings. The infrared camera is connected to a computer used for data acquisition via an IEEE 1394 (Firewire) connection. Both VarioCapture and VarioAnalyze software is used in order to record and process the images. Post processing of the data was done using MATLAB®. A schematic diagram of the experimental apparatus can be seen in Fig. 1.

3. Procedure

A 100 g sample of reverse osmosis water was placed inside the aluminum holding dish. At atmospheric pressure, water crystallizes to ice when cooled below 0 °C [7]. The latent heat of crystallization releases 334 kJ/kg [7]. The evolution of this heat is the basis for the measurement of the thermal front velocities as well as the proposed conversion model.

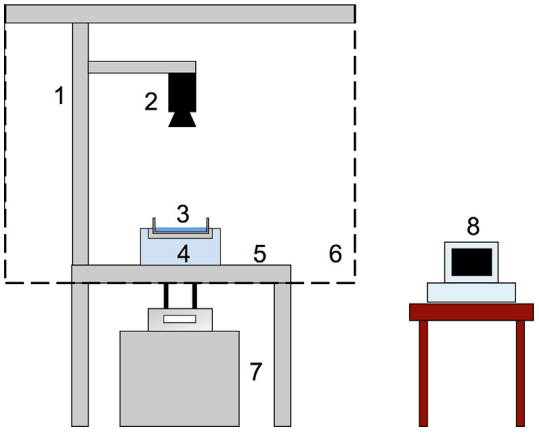

Fig. 1. Experimental apparatus. (1) Camera stand (2) infrared camera (3) aluminum dish with water (4) insulated cooling plates (5) vibration dampening bench (6) optical barrier (7) chiller (8) computer data acquisition system.

Before placing the sample on the cooling plates, the Neslab chiller is turned on and allowed to reach steady state. When the cooling plate's temperature becomes invariant, the water-containing aluminum dish is placed on top of the experimental setup. At this point, the water begins to cool and the VarioCapture software is initiated. Thermal images of the sample are taken at a frequency of 10 Hz. The recording process is terminated after the onset of nucleation once the progression of the thermal front has reached completion. At this point, the VarioAnalyze software is used to convert each image into a matrix of temperatures. The analysis of these matrices are then conducted through scripts written in MATLAB®. The thermal front velocity is measured by tracking the location of the liquid–solid interface in reference to the initial site of nucleation. In each experiment, five velocities are calculated at different angular directions, each starting from the nucleation location. This serves the purpose of producing an average velocity per experiment as well as a validation that the growth behaves isotropically. For these experiments, ice growth is considered to occur in the radial direction primarily. The height of the liquid sample is considered to be negligible. Furthermore, the azimuthal coordinate can be disregarded if growth is isotropic. Additional thermal information required for the proposed conversion model are the cooling plate temperature, the average sub-cooling temperature before nucleation as well as the average sample temperature after the initial crystallization step. This information is read off of the VarioAnalyze software.

4. Results and discussion

4.1. Sub-cooling at nucleation

Due to the stochastic nature of crystal nucleation, the temperature (and by extension, the level of sub-cooling) at which the ice formed was an uncontrollable variable in this study [18]. The longer the water cooled in the holding dish, the greater the system sub-cooling became and the greater the chances of nucleation. As shown by Pasieka et al. (2013), the ability to produce replicates at precisely the same level of sub-cooling in these types of experiments is impossible [17]. In order to overcome this issue, 70 experiments were conducted in order to obtain a spread in the sub-cooling values. An average level of sub-cooling is not reported in this study as the chiller settings ranged from −10 to −13 °C in order to get a good distribution. One of the reasons why the holding dish was fabricated out of aluminum is due to the high thermal conductivity of the material. This both reduces the time for the