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Ice-dependent liquid-phase convective cells during the melting of frozen sessile droplets containing water and multiwall carbon nanotubes

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ABSTRACT

The melting of frozen water droplets is a fundamental and ubiquitous process and the study of the transport processes occurring within the system during phase change is essential to understanding the forces that govern it. Multiwall carbon nanotubes (MWCNTs) can be added to liquid systems prior to crystallization in order to modify the properties of the phase change process and of the liquid or solid systems themselves. In this study, the melting behavior of frozen sessile droplets composed of water and 50 ppm of either functionalized or non-functionalized aqueous MWCNTs is investigated. Droplets are thawed from their base on a hydrophobic substrate set to temperatures between 1 and 30 °C. Tracking of MWCNT clusters during melting shows convective fluid motion occurring within the liquid melt at temperatures above 5 °C. This circulation is contingent on the presence of the ice phase above. The internal fluid dynamics are attributed predominantly to thermocapillary effects as a result of temperature-induced surface tension gradients along the air/liquid interface in the melt. Further, the melting times of MWCNT-containing systems were longer than pure water samples. These results highlight new and important mechanisms driving the melting processes within water droplet systems.

1. Introduction

The existence of stationary water droplets on surfaces is a universal phenomenon in nature and industry, and it plays a significant role in countless physical processes. Tiny beads may linger on a surface after contact with a liquid, yet the widespread and continuous transition of water between its three phases is also a steady source of droplet creation. Liquid drops may arise from the condensation of water vapor or through the melting of ice, and solid drops result from the deposition of water vapor or the crystallization of water. While natural systems exhibit a certain resiliency to these processes, droplet formation in industrial or man-made settings is generally problematic with regard to equipment and machinery. For example, continuous droplet impingement on steam turbine blades causes erosion and wear [1], while impact freezing of supercooled droplets on wind turbines and aircraft components leads to measurement errors, power losses and mechanical failures [2,3]. In these contexts, droplets usually evolve from a phase change on a cold substrate, and significant research is directed toward understanding the vapor condensation, deposition or liquid freezing processes in order to mitigate formation on the material surface. Most substrates tested exhibit varying degrees of hydrophobicity since anti-wetting or ice-shedding characteristics are desirable for a wide variety of applications, especially those wishing to prevent water or ice accumulation [4,5]. Interestingly, relatively less attention has been focused on the melting of a frozen droplet on such surfaces, which is especially relevant considering that active heating is a common anti-icing technique.

The general melting behavior of water is studied for a multitude of purposes that range from common scenarios such as an ice mass immersed in water [6], to phase change materials (PCM) for latent heat thermal energy storage systems, to geophysical processes occurring in river, lake and ocean systems [7]. Under certain conditions within melting systems, convective motion develops in the liquid melt, which significantly augments fluid transport. These instabilities were first documented in detail by Bénard [8] in thin liquid layers and were linked to the presence of thermal gradients within the system. The non-uniform temperature fields were then mathematically proven by later researchers to generate surface tension forces [9] and/or buoyancy forces [10] that set the fluid into motion. Surface tension-driven convection, also known as thermocapillary convection, arises from the stronger cohesive forces between colder molecules along an interface exerting a contractile force on warmer molecules. Momentum is then

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transmitted to the bulk interior by viscous forces, establishing circulatory fluid motion [11]. Density-driven convection, also known as thermogravitational convection, results from the expansion and rise of heated fluid, and the sinking of colder fluid. Whether a system is under surface tension or density control – or a combination of the two – is greatly dependent on the geometry of the system, the applied heat flux, the properties of the material substrate, and the thermophysical properties of the fluid [12,13]. In the past century, a large number of experimental and numerical analyses have been conducted that examine several combinations of the above-mentioned parameters in order to characterize these instabilities. The unidirectional melting of water has been predominantly studied in horizontal layers, such as rectangular and cylindrical containers [14–16], or in spheres melted radially from the exterior [6,17–19]. To the best of our knowledge, the case of a bottom-heated frozen water droplet has not been investigated.

Many technologies are also exploring the benefits of introducing nano-sized solid-phase additives to the process fluids in order to enhance or introduce new properties to the system. If no agglomeration takes place, the nanoparticles (at least one dimension <100 nm) can remain in suspension indefinitely since Brownian agitation predominates over a settling force such as gravity [20,21]. One type of dispersed nanoparticle–host system that garners significant attention is the multiwall carbon nanotube (MWCNT) nanofluid [22,23]. The hydrophobic nature of MWCNTs makes them predisposed to agglomeration in a polar solvent. Two main approaches are employed to preserve particle separation in the liquid. A simple method is the use of surfactants that coat the nanotubes and stabilize them in water. Alternatively, the outer surfaces of the MWCNTs can be functionalized with covalently-bound polar groups to intrinsically promote interaction with water [24–26]. This second approach is more robust and reduces the chemical complexity of the system, as well as any undesirable secondary effects due to the additives [27]. MWCNTs offer tremendous strength-to-weight ratios and exceptional thermal and chemical resistance, but they are probably most renown for enhancements in broadband light absorption, and thermal and electrical conduction [21,27]. MWCNT nanofluids are thus regularly evaluated for improved energy collection and heat transport properties in liquid media. Recently, testing has begun to assess the behavior of MWCNT nanofluids for phase change applications. Regarding enhanced boiling scenarios, there is a wide disagreement surrounding improvements in heat transfer characteristics, however MWCNT nanofluids consistently increase the critical heat fluxes [27,28]. Hordy et al. [29] also demonstrated that surface-treated MWCNTs remained stable in solutions undergoing boiling conditions. In solidification processes, MWCNTs have been reported to enhance the rate of nucleation, the speed of solidification and the total conversion of the base fluid, each of which increases the efficiency of the phase change process [30–35]. MWCNT nanofluids have thus shown great potential to modify the freezing and boiling processes of the base fluid, however their effects on the melting process have been relatively less described.

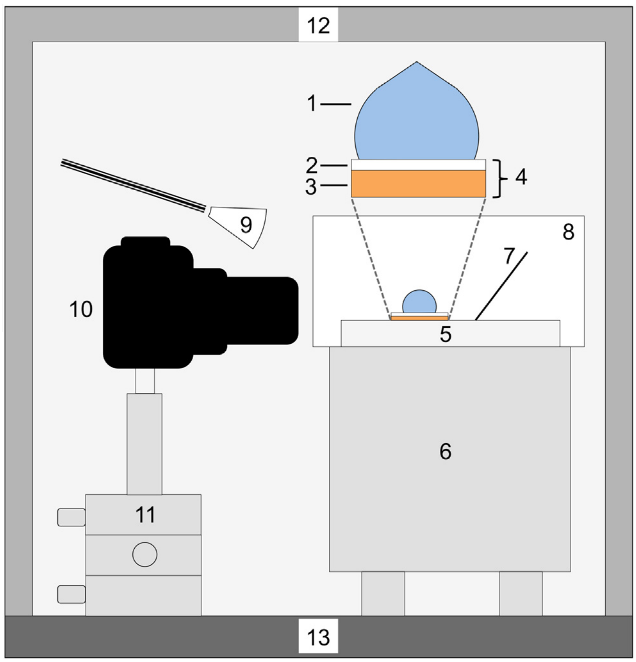

Fig. 1. Schematic of experimental setup. (1) Sample droplet; (2) PTFE strip; (3) copper plate; (4) sample plate; (5) TE warm plate surface; (6) TE warm plate body; (7) reflective sheet; (8) insulated enclosure; (9) light source; (10) camera and lens; (11) *x*, *y*, *z* multi-axis positioner; (12) plastic barrier; (13) optical table.

This study sets out to examine the morphological behavior of a bottom-heated melting sessile ice droplet composed of water, 50 ppm functionalized or 50 ppm non-functionalized MWCNT nanofluid across a range of warm plate temperatures on a hydrophobic substrate. As an essential constituent of the phase transformation scheme, the melting process merits equal study in order to understand the fundamental transport processes governing the interconversion of water between liquid and solid states.

2. Materials and methods

2.1. Experimental setup and materials

The experimental setup is analogous to the one used in a previous report and can be seen in Fig. 1 [36]. The sample plate consists of a thin strip of polytetrafluoroethylene (PTFE, 0.064–0.089 mm thickness) set on top of a copper plate (1.5 mm thickness). Droplet samples are frozen on an Aavid Thermalloy aluminum Hi-Contact liquid cold plate connected to a Thermo Scientific Neslab RTE 740 refrigerated bath/circulator. The process fluid is composed of a 50/50 by volume mixture of ethylene glycol and water. The temperature of the coolant within the bath can be programmed between −40 and 200 °C, and the temperature of the externally coupled cold plate surface is monitored by a 0.25 mm fine-diameter thermocouple probe (Omega, operating range −210 to 2100 °C, accuracy ±1.1 °C). Droplet samples are melted on a thermoelectric (TE) or Peltier plate with a maximum temperature rating of 70 °C. The temperature of the warm surface is controlled by a National Instruments LabVIEW VI and measured by a general-purpose thermistor (operating range −20 to 100 °C, sensor tolerance ±1.0 °C between 0 and 70 °C). The droplet sample is visualized by a Canon EOS 60D DSLR camera (18.0 Megapixel CMOS sensor) fitted with a Canon MP-E 65 mm f/2.8 1–5× macro lens. The camera is placed at an angle parallel to the warm plate surface and the focal plane is adjusted with an *x*, *y*, *z* multi-axis manual positioner stage in order to capture particle motion midway within the droplet. Videos are recorded at 30 fps. Diffuse illumination is provided by a fiber optic LED light source that is set to a low setting (10% of maximum brightness) along with a white reflective sheet placed behind the sample. In order to minimize air circulation onto the sample and to maximize heating from below, the sample plate and warm plate surface are enclosed in a polystyrene insulation chamber, and together with the remaining components are surrounded by a clear poly(vinyl chloride) cover. The entire apparatus rests on an optical table. Room temperature is read by a Fisher Traceable Jumbo thermo-humidity meter with a −5 to 50 °C measurement range to an accuracy of ±1.0 °C.