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Crystallization of supercooled water: A level-set-based modeling of the dendrite tip velocity

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ABSTRACT

'It is well-known that solidification front of a supercooled liquid is unstable; consequently, this instability leads to the appearance of an array of dendrites of sub-micron diameter. The shape and the velocity of the dendrite propagation are determined by the thermodynamic properties of the liquid and solid phases, including interfacial energy as well as the initial temperatures of both. Accordingly, the numerical simulation of solidification process is a rather challenging problem which requires an accurate prediction of high temperature gradients near the moving solidification front. In this study a relevant level set formulation has been developed enabling correct determination of the position and the curvature of the liquid/solid interface. At this interface a Dirichlet boundary condition for the temperature field is imposed by applying a ghost-face method. For the purpose of updating the level set function and optimizing computing time a narrow-band around the interface is introduced. Within this band, whose width is temporally adjusted to the maximum curvature of the interface, the normal-to-interface velocity is appropriately expanded. The computational model is firstly validated along with the analytical solution of stable freezing. The tip velocity of dendritic patterns (pertinent to unstable freezing) is investigated by performing two-dimensional simulations. The computational results exhibit excellent qualitative and quantitative agreement with the marginal stability theory as well as with the available experiments in the heat-diffusion-dominated region.

1. Introduction

Several influencing factors are involved simultaneously in the process of a phase change of pure water: heat transfer, absorption (if a melting process would take place) or release (pertinent to freezing) of latent heat (with possible solute rejection in case of binary substances), surface effects, changes in thermophysical properties with the temperature, etc.

The pure water is supercooled if its temperature becomes lower than the melting point, corresponding to T_m = 273.15 K under atmospheric pressure. This occurs when the energy barrier above which the atoms start to move into the solid lattice structure, required for the formation of a crystal, is not entirely exceeded. Such a state of supercooling is thermodynamically unstable. Correspondingly the liquid is in a metastable state, implying a weak disturbance can initiate a rapid and unstable process of the so-called dendritic solidification. Small perturbations in the initial state can produce significant changes in relation to the time-dependent solid/liquid interface. Behavior of such supercooled water drops is rather important for cloud microphysics and understanding of aircraft icing [1,2].

The phenomenon of dendritic crystal growth attracted considerable attention over the last few years. In nature many cases of spontaneous dendritic pattern formation can be found, e.g., in solidification of metals and crystallization of supercooled solutions. The most evident case of dendritic pattern formation are snowflakes which have various types of complex and fascinating shapes. The underlying physics of crystals has been firstly elaborated by Kepler in 1611, [3]. This famous essay about the form of snowflakes represents the first scientific reference to snow crystals. Over three hundred years later, Nakaya [4] has performed a first systematic study about the snow crystals. In this work the crystal morphology created under different environmental conditions has been described. Over the last decades, a number of theoretical and experimental works [5–10], has been published dealing with the detailed macroscopic dynamics of crystal growth (pertaining to non-equilibrium patterns). The common approach to crystallization problems depends on the grade of the supercooling, ΔT (liquid temperature reduction below its freezing point without solidification), in the

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liquid. At low supercooling ($0.06 \leqslant \Delta T \leqslant 10$ K), the freezing process can be considered as a Stefan problem (corresponding to a diffusion-driven growth) [11]. In the case of very low supercooling ($\Delta T < 0.06$ K), a deviation from the theory can be observed in the experiments. This deviation is assumed to be caused by the effect of natural convection around the crystal promoting its increase in this supercooling range [12]. On the contrary, in the case of increasing supercooling ($\Delta T > 10$ K), there is a transition from the diffusional to the kinetics-limited growth. Correspondingly, the rate of solidification depends on how fast the liquid molecules can be brought in the proper position, orientation and conformation pertinent to the solid phase.

Recently, Shibkov et al. [13–15] have investigated the free growth of an ice crystal in a supercooled pure water film in a wide initial supercooling range corresponding to $0.1 < \Delta T_0 < 30$ K. In [14] various shapes of ice crystal patterns in the range between low ($\Delta T_0 = 0.1$ K) and high ($\Delta T_0 = 14.5$ K) supercooling have been observed. Evidently, the shape and the velocity of the solidification front depends strongly on the initial supercooling in the pure water film. The shape of dendritic front is determined by the balance between surface energy criterion and the efficiency of the interface (region of negligible thickness where solid and liquid phases coexist) in removing heat. Thus, when pure water freezes in the diffusive regime, different morphologies can appear depending on supercooling and crystalline anisotropy. At higher supercooling, e.g., at $\Delta T_0 = 10$ K, transition from diffusional growth to kinetics-limited growth is observed. These experimental results have been compared with the theory of dendritic growth of Oldfield and Langer and Müller-Krumbhaar [16–20] exhibiting an excellent agreement within the diffusion-driven growth region. As expected, in the kinetics-limited growth region a certain deviation from the theory is observed.

Mathematically, the phenomenon of solidification can be modeled by utilizing a moving boundary [11]. The heat equation is solved in each phase separately. The temperature fields are coupled through two boundary conditions at the unknown moving boundary between the solid and the liquid phase. The first boundary condition is the velocity of the interface. It depends on how fast the latent heat of solidification is removed from the interface, hence, the velocity can be derived from a heat balance at the interface. The second boundary condition assumes a constant temperature at the interface corresponding to the melting temperature. Locally, the melting temperature at the interface will be altered by an amount depending on the surface tension between the solid and liquid phases and the local curvature, in line with the Gibbs–Thomson effect. Another effect to be taken into consideration are the density changes due to temperature variation. These changes induce flow resembling the natural convection in the presence of gravity which affects the heat transfer in the liquid.

The direct solution of the time-dependent Stefan problem represents a great challenge. An appropriate front tracking method accounting for the moving solid–liquid interface is required. Over the last decades, the phase-field models have been successfully applied for the simulation of dendritic growth. The basic idea behind phase-field methods is to artificially thicken the interface to an extent which can be resolved by the numerical mesh. This thickening procedure is accomplished by introducing the so-called "order parameter" for each cell, ranging between zero and unity. If the order parameter takes the value of unity the corresponding grid cell is filled entirely with the liquid phase. In contrast, the zero value of the order parameter indicates the grid cell comprising completely the solid phase. The position of the interface coincides with the surface whose order parameter takes the value 0.5. The width of the artificially thickened interface corresponds to the area within which the order parameter varies smoothly from zero to unity. The outcome is a smeared interface over which all governing equations can be solved taking into account appropriately averaged (i.e. weighted) material properties. This allows application of the physical models within such a diffuse interface. By doing so, the computational difficulties pertinent to the tracking of a sharp interface are avoided to a great extent. This contributes to the high popularity of the phase-field methods for simulating dendritic solidification [21–25]. Although the phase-field modelling approach has been shown to be very useful in investigating solidification patterns, there were still some important drawbacks which should be addressed. The principal drawback is the non-physical representation of the diffuse interface. The width of the interface represents an adjustable parameter, which may also lead to unphysical interactions. Unless a field equation solver is developed to enforce the conservation of energy for a control volume positioned strictly at the interface, the interfacial velocity will be inaccurately obtained.

The presently adopted level set method is a computational approach aiming at overcoming the limitations of phase-field models with respect to the interface surface tracking; here the boundary movement is tracked implicitly. This method, firstly introduced by Osher and Sethian [26], describes the spatial distribution of a level set function, Φ, within the entire solution domain. The solid–liquid interface is represented by the zero-value contour of the level set function, which is governed by its own advection-type equation of motion. These equations providing a sharp interface are solved directly and can be handled in a straightforward manner. Although the level set methods still did not reach the popularity the phase-field methods have for studying crystal growth phenomena, they have been increasingly applied to several problems involving moving boundaries [27–29] and crystallization [23,30,31] exhibiting good predictive performance in returning qualitative features of the dendrites.

In this work, an extended level set method using a ghost-face algorithm for solving the temperature field is presented. Unlike the conventional level set method, this extended version converges to exact solution of the Stefan problem for planar solidification. Concerning the tip velocity of dendritic growth within the diffusion-driven growth region the results of two-dimensional simulations show excellent qualitative and quantitative agreement with the experiments of Shibkov [13–15] and the theory of Oldfield [16] and Langer and Müller-Krumbhaar[17–20]. Hence, the computational model is capable of capturing both stages of crystallization: the first rapid, dendritic-like growth (corresponding to the phenomenon of unstable freezing) and the second planar-shaped growth stage (associated with the stable freezing process).

2. Theoretical background

The common approach to a freezing problem consists in its consideration as a two-phase Stefan problem. The term "two-phase" refers here to the phases taking an "active" part in the process. Accordingly, both the liquid and the solid phases are active, i.e. the heat conservation is solved in both sub-domains. Let us consider a square domain, D, of pure water where at every time step and at every numerical node the water is either in the liquid (supercooled) state or in the solid state. Let $T(\boldsymbol{x},t)$ represents the temperature of the water. The region where the water appears as solid is denoted by $\boldsymbol{\Omega}_s$ and the region where the water is a liquid by $\boldsymbol{\Omega}_l$. The interface between the solid phase and the liquid phase is of infinitesimal thickness and is denoted by $\boldsymbol{\Xi}$.

2.1. Governing equations

As the flow in liquid region is not considered presently, the energy equation describing time dependent heat conduction in both regions reduces to: