Dendritic solidification and the influence of interface morphology

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Abstract

The three-dimensional tip-shape of succinonitrile dendrites suggests a possible mechanism responsible for certain features of microgravity dendritic solidification data which do not conform to the theory of Ivantsov. These data constitute part of the Isothermal Dendritic Growth Experiment (IDGE), and were obtained during three different missions of the space shuttle Columbia. Under the conditions of reduced melt motion and convective heat transfer, the experimental data agree reasonably well with the basic transport theory, though discrepancies still exist. These have become the focus of further investigations.

The method of moving heat sources is applied to the problem of dendritic growth of a pure material in a quiescent, isothermal melt. By specifying various steady-state solid-liquid interface shapes in the model formulation, a more clear understanding of the influence of the tip shape and the side-arm cruciform is obtained. The results of this study indicate that for interfaces which take the form of infinite bodies of revolution, the transport solution is sensitive to the shape of the interface.
1 Introduction and Background

During the solidification of metals and alloys, dendritic microstructures are commonly formed. It is important to understand the process by which dendrites form because traces of these geometrically complex microstructures persist through subsequent material processing stages and can affect the properties of the finished product.

Over the past 5 decades, a large body of experimental and theoretical works describing dendritic growth has been produced. This background was discussed in the review of Glicksman and Marsh [1], and also by Bisang and Bilgram [2]. We will focus here on the process in its simplest form, namely as a diffusion controlled process in a pure material, where the latent heat released at the solid/melt interface diffuses into the melt ahead of the advancing interface. A mathematical solution to this heat flow problem was first developed by Ivantsov [3], and may be expressed as

\[
\Delta = Pe \left[ e^{Pe} \int_{0}^{\infty} e^{-u} du \right].
\]  

(1)

Here, \( \Delta \equiv \Delta T/(L/C_p) \) defines the dimensionless supercooling, \( Pe \equiv VR/2\alpha \) defines the growth Péclet number, \( \Delta T \) is the supercooling, \( L \) is the molar latent heat, and \( C_p \) is the molar specific heat under constant pressure. \( V \) is the steady-state dendrite growth rate, \( \alpha \) is the thermal diffusivity of the liquid phase, and \( R \) is a length scale, taken here to be the radius of curvature at the dendrite tip. Ivantsov’s solution applies to the case of an interface in the form of a paraboloid of revolution. While it is generally accepted that more sophisticated approaches to studying dendritic growth are presently available, we will nonetheless, focus here on the transport in an analogous manner to Ivantsov’s result of Eq. (1). With this relatively straightforward analysis, it will be shown that a dendrite’s interface shape can measurably influence the dendritic growth process.

1.1 Succinonitrile Dendrite Interface Shape

The first suggestion that the crystal-melt interface shape may affect the dendritic growth process came about when two sets of experimental measurements were evaluated. The first observation was that although the Ivantsov solution described microgravity dendritic growth data with reasonable success, there were several issues pertaining to the Péclet number data [4,5] that could not be explained by either Ivantsov’s result or other models that included additional influences such as convection or container wall interactions [6-8]. These will be
discussed in more detail later. The second observation derived from the experimental measurement of succinonitrile dendrite tip shapes described in [9, 10]. In this study, it was quantified how dendrite tips are neither parabolic in profile, nor bodies of revolution as is commonly assumed in theory. Succinonitrile dendrites viewed in profile can be characterized in the region close to the tip (prior to side-arms appearing at ~12\(R\)) using the dimensionless 4\(^{th}\)-order polynomial shown in Eq. (2),

\[ Z = \frac{\rho^2}{2} + Q\rho^4. \]  

Here, \(Z\) and \(\rho\) are the cylindrical coordinates, normalized using \(R\), with \(Z \equiv z/R\) and \(\rho \equiv r/R\). \(Q\) is a dimensionless parameter describing the azimuthal variation of the interface shape. For SCN dendrites, \(Q\) exhibits 4-fold rotational symmetry as illustrated in Figure (1), where the experimentally measured \(Q\) is plotted as a function of the azimuthal direction from which the image was obtained (measured in degrees from the nearest [100] side-arm direction). With such a quantified measurement of how a typical SCN dendrite differs from the paraboloidal shape assumed by Ivantsov, it was natural to ask if this feature can explain the details of the Péclet number data.

2 Analysis

In order to evaluate how the interface shape affects the transport of heat, the steady-state conduction equation was solved using the method of moving heat sources, which offers more flexibility with arbitrary interface shapes than the analytical solution form of Ivantsov. This solution scheme, based on a Green’s function approach, was first introduced in its general form by Rosenthal [11] in 1946. While Ivantsov made an early attempt to apply it to the subject of dendritic growth [12], it was not until 1978 when Schaefer presented the first detailed application [13] of the method to this subject matter.

The method is straightforward in principle: each point on the crystal-melt interface is treated as a source of latent heat on an interface growing into a quiescent melt. The contribution of each point to the surrounding temperature field is determined by superposing (integrating) their influences via the conduction equation. This way, it becomes possible to determine the rise in temperature at any point in the thermal field, due to the release of latent heat along the interface. The solution takes the general form of Eq. (3). Here, \(dT\) represents the change in temperature at any point due to a point source of strength \(dq\), traveling at the velocity \(V\), and located a distance \(H\) from the point in question. The equation is written as,
Figure 1: Typical SCN dendrite tip interface shape anisotropy as a function of azimuthal direction measured from the [100] side-arm direction. When characterized using the 4th-order polynomial of Eq. (2), the azimuthal average is $Q \approx -0.001$. Note that these measurements were made on the region of the interface prior to side branching, namely, 0-12R behind the tip.

$$dT = \frac{dq}{4\pi k} \left[ \frac{\exp\left(-\frac{V}{2\alpha}(\xi + H)\right)}{H} \right].$$

(3)

Where, $k$ represents the thermal conductivity, and $\xi$ is the component of $H$ in the direction of the dendritic growth axis.

The strength of this approach to investigating the transport process is in its flexibility. Where analytical solutions such as Ivantsov’s are tied to particular interfacial geometries such as the paraboloid and its relatives [14], Equation (3) allows the use of any arbitrary distribution of latent heat sources (i.e. the interface shape). Once the interface shape is specified, $\xi$ and $H$ become
functions of position and the entire relationship can be integrated over the
interface to determine the net change in temperature at the location in question.

In this study, the interface shape will be specified as either functions of
polynomial form (Eq. (2)), or as hyperboloids of revolution. These particular
functions are selected only because they allow representations of interface
shapes that are somewhat wider and somewhat narrower than a paraboloid and
can be numerically integrated to infinity without difficulty. If the hyperboloid is
first written as a Taylor series expansion, it can be cast in terms of the parameters
used to characterize the 4th-order polynomial,

\[ Z = \frac{1 - \sqrt{1 - 8Q\rho^2}}{8Q} \]  

(4)

Where \( e = \sqrt{-8Q + 1} \) relates the hyperboloid’s eccentricity, \( e \), to the quartic
shape parameter, \( Q \), and \( a = -\frac{R}{8Q} \) connects \( a \) (½ the hyperboloid’s major axis
length) to both the shape \( (Q) \) and size scale \( (R) \) of the interface.

The solution of Equation (3) is found for a point of interest located at the tip
of the dendrite. After substituting the appropriate interface function into Eq. (3)
and some manipulation, the equation reduces to the convenient form of Equation
(5), shown for the case of the 4th-order polynomial (the quartoid):

\[ \frac{d\Delta}{d\rho} = 2Pe \exp \left[ -\frac{Pe}{2} \rho \left( \rho + 2Q\rho^3 + \sqrt{\rho^2 + 4\rho^4Q + 4\rho^6Q^2 + 4} \right) \right] \]  

\[ \sqrt{\rho^2 + 4\rho^4Q + 4\rho^6Q^2 + 4} \]  

(5)

Where, \( d\Delta \) is the differential form of the contribution to the dimensionless
temperature change at the tip, and its variation over the interface \( (d\Delta) \) is solely a
function of the Péclet number and the shape of the interface, \( Q \). References to
the parameters \( V \) and \( R \) have conveniently been encapsulated into the Péclet
number.

The net influence upon the thermal field at the tip of the interface is
determined by separating and integrating Equation (5) (or its analog for the
hyperboloid) over the entire interface.
3 Results

Using the bodies of revolution represented in Equations (2) and (4), the Péclet numbers are calculated via integration of Equation (5) (or similar) for interfaces of varying shapes. The results are plotted along with the experimental data in Figure (2). The functions used to describe the interfaces were chosen so as to be able to effectively integrate back to “infinity”. “Infinity” was realized by progressively increasing the integration range, until the calculated dimensionless supercooling no longer exhibited detectable changes. Interface shapes that were wider than a parabola, were modeled with the hyperbolic formulation, whereas interface shapes that were narrower than a parabola were modeled as 4th-order polynomials (quartoids) with positive values of $Q$.

In representing the interface shape, the range of $Q$ values investigated in this study was selected from the approximate range of values that have been experimentally observed (Figure (1)). The azimuthal average value of $Q$ derived from Figure (1) is approximately $-0.001 \pm 0.0003$, however we must interpret this with care. It is important to note that this shape information was obtained from the region of the dendrite that extends to approximately 10-12$R$ behind the tip—a range that does not display significant side branching. However, the numerical results in Figure (2), were produced by integration to infinity. This is appropriate when making comparisons with the Ivantsov result, but not for making comparisons with experimental data. Nonetheless, Figure (2) reveals how a modest change from a parabolic interface shape can influence the transport in a measurable manner.

Note that the low supercooling behavior of the model in Figure (2) (i.e. the “drop-off”) results for interfaces wider than a parabola. This feature of the model results from the integration to infinity. For such interface shapes, far behind the tip, the hyperboloid approaches its asymptotes and is markedly different from a paraboloid. This influence is more noticeable at lower supercoolings, where thermal lengths are longer.

The most prominent discrepant feature of the experimental data in Figure (2) is seen at the lower supercoolings. In this range, there is a systematic “liftoff” of the experimental data above the Ivantsov transport line. Reasonable explanations of this behavior have been offered by Pines, et. al. and Sekerka et. al. [6, 8], who showed that a dendrite interacting with the container walls, is expected to exhibit this effect.

4 Conclusions

The application of the method of moving heat sources, originally introduced by Schaefer, is demonstrated here to have considerable utility in developing a quantitative understanding of how the interface shape affects the dendritic growth process. Figure (2) illustrated how an interface-shape that is wider than
a paraboloid induces a downward shift in $Pe$ from Ivantsov’s result. Conversely, shapes that are more narrow, will shift $Pe$ upward.

Current efforts are aimed elucidating how the various regions of the dendrite contribute to the transport process, particularly when the interface is treated more like a natural dendrite, which is not an infinite, paraboloidal, body of revolution.

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References


