Abstract

Dendrites are a commonly observed mode of crystallization exhibited by metals and alloys during casting and welding. The development of dendritic patterns depends on the transport of heat and solute away from the growing crystal-melt interface. The kinetics of dendritic crystal growth were studied recently under microgravity conditions in two related space flight experiments called the Isothermal Dendritic Growth Experiment (IDGE). Steady-state, dynamic tip shapes, observed under terrestrial and microgravity conditions, were analyzed using a quartic (4th-order) shape regression. This process yielded the parabolic tip radius, \( R \), and tip speed, \( V \), which each are growth parameters already shown to be sensitive to the presence of gravitationally induced convection, whereas their combined scaling law, \( VR^2 = 2\alpha d_0/\sigma^* \), verified by these experiments, appears to be remarkably indifferent to the gravity level. Here \( \alpha \) and \( d_0 \) are the thermal diffusivity and capillary length, respectively, and \( \sigma^* \) is the scaling constant derived from theory. The quartic shape coefficients, presented here for two supercoolings, are demonstrated to be insensitive to both the gravity level and the supercooling, similar to behavior of the scaling constant itself.

Background

Dendritic microstructures are commonly encountered during the solidification of many engineering materials. It is important to understand how dendrites form because traces of these geometrically complex microstructures persist through subsequent material processing stages and can affect the properties of the finished product. Dendritic solidification, a diffusion controlled process, is commonly observed during casting and welding, where the latent heat released at the solid/melt interface diffuses into the melt ahead of the growing dendrite. A mathematical solution to this heat flow problem was first developed by Ivantsov [1], and may be expressed as

\[
St = Pe \cdot e^{-Pe/\alpha} \int_0^\infty \frac{e^{-u}}{u} \, du ,
\]

where \( St \equiv \Delta T/(L/C_p) \) defines the Stefan number (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 of the melt under constant pressure. \( V \) is the steady-state
dendrite tip velocity, \( \alpha \) is the thermal diffusivity of the melt, and \( R \) is a length scale, taken here to be the radius of curvature at the dendrite tip, *approximated as a paraboloid of revolution*.

In addition to the fundamental transport solution of Ivantsov, various theories (see the review by Glicksman and Marsh [2]), suggest a second useful relationship having the form which is independent of \( \Delta T \),

\[
\sigma^* = \frac{2 \alpha d_0}{VR^2}, \tag{2}
\]

where \( \sigma^* \) is usually referred to as the stability, selection, or scaling constant and \( d_0 \) is the capillary length scale (a materials constant).

When Ivantsov’s diffusion solution, Eq. (1), is combined with the selection rule expressed in Eq. (2), it then becomes possible to define the operating state of the dendritic growth process in terms of the tip velocity, \( V \), a length scale, \( R \), and a single process parameter, \( \Delta T \). Recently, Glicksman et al. have shown [3,4] that the combination of Eqs. (1) and (2) does not adequately describe the dendritic solidification process. Furthermore, although Ivantsov’s transport equation itself agrees approximately with data from microgravity experiments, it still tends to overestimate \( \text{Pe} \) as a function of supercooling, as evidenced by the experimental data in Figures 1a and 1b.

Several limiting assumptions were made in the original Ivantsov analysis, including the condition that a dendrite tip is well represented by a paraboloidal body of revolution growing

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**Figure 1 (a, b)**

a) Ivantsov Diffusion theory is in approximate agreement with the Péclet number measured under microgravity conditions, while terrestrial-based measurements differ significantly at lower supercoolings where the process is not diffusion-limited.

b) Microgravity Péclet number as a function of supercooling shows that despite the approximate agreement in Figure 1a, the Ivantsov Diffusion theory (dashed line) tends to overestimate \( \text{Pe} \).
at a constant velocity. The validity of these assumptions was examined by LaCombe, Koss, Fradkov, and Glicksman [5] through careful measurement of the three-dimensional anisotropy in the shape of dendrite tips at a single supercooling. This paper extends that work by presenting measurements of this shape anisotropy under the reduced melt convection conditions of microgravity.

The notion that dendrites are not paraboloids of revolution was clearly demonstrated by the data of Huang and Glicksman [6] (Figure 2), which shows two profiles of the same succinonitrile dendrite superimposed upon one another. Here, one tip ($\phi = 0^\circ$) viewed along a (100) direction appears wider than a parabola that best fits the tip region (dotted line), and a second image ($\phi = 45^\circ$) obtained from a profile viewed from the (110) direction is seen to be narrower than the same parabola. Note that both views do closely approximate the parabola near the tip. This early observation has often been used to justify the use of a parabolic (2nd-order polynomial) fitting form when choosing the length scale of a dendrite tip.

The profile of a dendrite’s solid/melt interface in Cartesian coordinates is usually represented with the 2nd-order polynomial equation

$$y = y_0 + \frac{1}{2R}(x-x_0)^2,$$

where $x$ and $y$ are the coordinates of the profile, $R$ is the radius of curvature at the tip, and the point $(x_0, y_0)$ represents the location of the tip. Profile data from micrographs of growing dendrites can be fitted to the parabola in Eq. (3) to yield $x_0$, $y_0$, and $R$. This procedure, long considered as a standard method to measure dendrite tip radii, is, of course, limited by the assumption of a paraboloidal shape. When this mathematical form is applied to objects that are not parabolic bodies of revolution, the calculated tip radius becomes less accurate when data further back from the tip is used. Departures from a parabolic profile often occur before side branching becomes prominent, as shown in Figure 2.

A fundamental difficulty in quantifying dendritic tip shapes is the need to include information that comes from regions of the solid/melt interface away from the tip in order to obtain the radius of curvature at the tip. For a variety of reasons discussed in LaCombe et al. [5], the uncertainties of the optically measured solid/melt interface position near the tip are larger than those from regions further from the tip. This leads to the use of data from regions of the dendrite profile that may not be parabolic to fit the shape of the tip region (see Figure 2).

Several researchers have suggested the use of a higher-order polynomial to characterize more realistically the shape of dendrite tips [5,7-12]. A fourth-order equation describing a dendritic profile is

$$y = y_0 + \frac{1}{2R}(x-x_0)^2 + \frac{1}{6R^3}(x-x_0)^4.$$

Figure 2

SCN dendrite tip viewed from two different orientations [6]. This figure illustrates the difference in shape of two superimposed views of the same dendrite, as compared with a “best-fit” parabola indicated by the dotted line.
where $q$ has units of length$^{-3}$ and represents the amplitude of the quartic deviation from the second-order paraboloidal basis shape. This relationship can be written in the non-dimensional form

$$Y = \frac{X^2}{2} + Q(\phi)X^4,$$

where $X = x/R$, $Y = y/R$, and $Q(\phi)$ is equal to $qR^3$ and varies with the projection angle, $\phi$. $Q(\phi)$ for cubic crystals like succinonitrile is assumed to have the functional form $Q(\phi) \propto \cos(4\phi)$ [7-11]. However, recent analysis of dendrite profiles in BCC succinonitrile [5,12] shows that this form is not valid. In the present analysis, $Q$ is treated as a general function of the orientation, $\phi$.

The use of an additional parameter in fitting the experimental data improves the quality of the tip-profile’s fit merely by adding a degree of freedom to the regression. There are, however, more significant advantages. Extending the 2$\text{nd}$-order polynomial through the addition of a 4$\text{th}$-order term results in a second-order coefficient, $(2R)^1$, that is less sensitive to the angle, $\phi$, from which the measurement is made. In coordination with this, the shape anisotropy subsequently appears strongly in the 4$\text{th}$-order term. These observations give credence to the use of a paraboloidal basis shape along with 4-fold deviations as one moves away from the tip.

**Experiment Description**

Experiments were conducted in the microgravity environment aboard space shuttle Columbia’s STS-75 mission. Ultra-pure succinonitrile (SCN) in a stainless steel and borosilicate glass growth chamber was placed in a temperature-controlled bath exhibiting a temperature stability of $\pm 2\text{mK}$. The dendrites were grown and measured at supercoolings of

![Figure 3](image_url)

Schematic representation of the growth chamber within the temperature-controlled bath. Dendrites are grown within the growth chamber and recorded by both 35mm film and CCD camera. The 35mm film constitutes the primary shape information data source.
The experimental apparatus depicted schematically in Figure 3 consists of a growth chamber containing the SCN immersed in a thermostatically controlled bath. Dendritic crystals are grown by first melting the SCN and then lowering the temperature to the desired supercooling. Once the temperature of the SCN has stabilized at the desired supercooling, a thermoelectric cooler is activated causing the stinger (see Fig. 3) to cool further. The molten SCN within the tubular stinger nucleates and freezes, propagating a solid film along the stinger’s inside wall until it emerges into the large volume of the growth chamber. In this unconfined state, the solid grows into the undercooled melt as an equiaxed dendrite. Photographic images are then recorded at regular time intervals along two perpendicular optical axes. These photographs are the primary data source for the measurements of the tip shapes. A more thorough description of the apparatus and the associated measurement process is given by LaCombe et al. [5].

Results

Tip shape information was extracted for dendrites grown at two supercoolings (0.26 K and 0.48 K). Each crystal grows at a random orientation relative to the photographic system. The 35mm negatives were prepared and analyzed using computer-based image processing software to produce digitized images of the dendrite tips. Each image was processed to produce a set of coordinate pairs ($x,y$) that describes the profile shape of the dendrite as viewed from each camera orientation. This set of $x$-$y$ coordinates describing the edge was then inserted into a regression routine based on Equation 4, producing fitted values of $R$ and $q$ (or $Q$) using a

![Graph](https://via.placeholder.com/150)

**Figure 4**

Fourth-order constant $Q$ as a function of orientation at 0.26 K. The four-fold symmetry of the underlying cubic structure is evident in this experimentally measured parameter.
Fourth-order constant $Q$ as a function of orientation at 0.48 K. Comparison with Figure 4 reveals a uniform relation between $Q$ and $\phi$ for different supercoolings.

method described in LaCombe et al. [5]. The resulting values of $Q$ are plotted as a function of the view angle (orientation) in Figures 4 and 5.

The data shown in Figures 4 and 5 reveal the assumed four-fold rotational symmetry of the underlying crystallographic structure. $Q$, representing the deviation from parabolicity, varies with observation angle $\phi$. Comparison of data in Figures 4 and 5 reveals that there is not a significant difference in the function $Q(\phi)$ as supercooling changes. These observations agree with the earlier work of LaCombe et. al. [12]. Each of these figures also shows that there is not a significant difference between $Q(\phi)$ as measured under terrestrial or microgravity conditions, suggesting that melt convection does not noticeably affect the shape of a dendrite tip in this range of supercoolings.

Conclusions

Contrary to assumptions made in many dendritic growth theories, dendrite tips are not parabolic bodies of revolution—especially as regions beyond the tip are considered. The degree to which this deviation from parabolicity is present, does not appear to be significantly affected by convective heat transport within the range of detection of our experiment, and within the range of supercoolings that were examined.
In contrast to $Q$'s indifference to the convective environment, Figure 1a shows that the presence of melt convection dramatically affects other parameters of the process such as $Pe$. It is shown elsewhere[13] that the growth rate ($V$) and scale ($R$) of a dendrite are also affected significantly. However, it is also noted that similarly to $Q$, $\sigma^*$ is also independent of the convective environment (shown in Figure 6). The theoretical basis for $\sigma^*$ incorporates the surface energy anisotropy, and thus is inherently linked with $Q$, which serves to characterize the dynamic shape anisotropy of a dendrite tip. [14]

We feel that this work constitutes a step towards a better understanding of the impact of melt convection upon various aspects of dendritic growth. It has become apparent that although some parameters ($V$, $R$, $Pe$) are influenced by convective transport, others, particularly those relating to crystal/melt interfacial energy and shape, do not appear to be affected. Future efforts to understand this observation will be aimed in this direction.

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