THE EFFECT OF CONVECTION ON DENDRITIC GROWTH UNDER MICROGRAVITY CONDITIONS

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The Isothermal Dendritic Growth Experiment (IDGE) is an orbital space flight experiment launched by NASA in March, 1994, as part of the United States Microgravity Payload (USMP-2). The IDGE provided accurate measured dendritic growth rates, tip radii of curvature, and morphological observations of ultra-pure succinonitrile obtained at supercoolings in the range 0.05–2.0 K. Data were received in the form of pairs of digitized binary images telemetered to the ground from orbit in near-real-time, and as 35mm photographic film received 3 months after the flight. The IDGE flight data has now been analyzed, permitting a comprehensive comparison between dendritic growth under terrestrial and microgravity conditions. The measured growth kinetics, in the form of velocity versus supercooling, is markedly different from those observed in terrestrial experiments. Above 0.4 K supercooling in microgravity, the process of dendritic growth is diffusion controlled, i.e., thermal conduction is the rate limiting process. Under terrestrial conditions, dendritic growth of SCN remains dominated by convective transport of heat until a supercooling of ca. 1.7 K is exceeded. Beyond a supercooling of 1.7 K, there is excellent agreement between terrestrial dendritic growth measurements, and a theory with one adjustable parameter determined from microgravity measurements. Surprisingly, however, even under microgravity conditions, dendritic growth of SCN becomes dominated by convective transport at supercoolings of ca. 0.4 K and below. The observations confirm that convection, which depends on a sublinear power of the supercooling, will always dominate at low supercoolings, whereas diffusion, which depends on the superlinear power of the supercooling, will always dominate at high supercoolings.

KEYWORDS Dendrite Microgravity Convection Ivanov

INTRODUCTION

The growth of dendrites is commonly encountered when metals and alloys freeze under low thermal gradients, as occurs in most casting and welding processes. In engineering alloys, the details of dendritic morphology directly relate to important material responses and properties. Of more general interest, dendritic growth is an archetypical problem in morphogenesis, by which a complex pattern evolves from simple starting conditions. Thus, the physical understanding and mathematical description of how dendritic patterns emerge during the solidification process are of interest to scientists and engineers.

In the case of cast alloys that solidify dendritically, computational methods are required to elucidate both microstructure and chemical microsegregation patterns formed by the agglomeration of millions of dendritic grains comprising the macroscopic end product. This requires detailing physical processes over a scale change of at least two orders of magnitude. The mesoscopic dendritic grains, which are strongly influenced by the growth pattern of individual dendrites, are considered the
starting point to the problem of micro-to-macro scale modeling. A full understanding of the behavior of a single, isolated dendrite is an important first step in achieving better understanding and control of the final properties of dendritically solidified materials.

A number of theories of dendritic crystal growth, based on various transport mechanisms, physical assumptions, and mathematical approximations, have been developed over the last forty years to predict dendrite tip velocity, \( V \), and radius of curvature, \( R \), as functions of the supercooling, \( \Delta T \) (see [Glicksman and Marsh (1993)]). The growth of dendrites in pure melts is known to be controlled by the transport of latent heat from the moving crystal-melt interface as it advances into its supercooled melt. Ivantsov (1947) provided the first mathematical solution to the dendritic heat conduction problem and modeled the steady-state dendrite as a paraboloidal body of revolution growing at a constant velocity, \( V \). The resultant thermal-conduction field can be expressed mathematically in paraboloidal coordinates moving with the dendritic tip. This temperature-field solution is known as the Ivantsov, or diffusion-limited, solution. This solution is incomplete insofar as it only specifies the dendritic tip growth Péclet number, \( VR/2a \), (here \( a \) is the thermal diffusivity of the molten phase) as a function of the initial supercooling, and not of the explicit dynamic operating state, \( V \) and \( R \). The Péclet number obtained from the Ivantsov solution for each supercooling yields instead an infinite range of \( V \) and \( R \) that satisfy the diffusion-limited solution at that particular value of \( \Delta T \).

In the early 1970s, succinonitrile (SCN), a BCC organic plastic crystal, was developed [Glicksman, et al. (1976)] as a model metal analog system for studying dendritic growth. SCN solidifies like cubic metals (i.e., with an atomically “rough” solid-liquid interface) yet exhibits convenient properties for solidification experiments, such as a low melting temperature, optical transparency, and accurate characterization of its thermo-physical properties. The use of SCN greatly facilitated dendritic growth studies over the past twenty years so that dendritic tip velocities could be accurately measured and used in critical tests of theory.

Since the mid-1970s, considerable theoretical and experimental efforts have concentrated on the determination of an additional equation or length scale, which when combined with the Ivantsov heat conduction solution, “selects” the observed operating states (see references within [Glicksman and Marsh (1993)] and [Brener and Meñikov (1991)]). Although the underlying physical mechanisms for the various “theories of the second length scale” are quite different, their results are usually expressed through a scaling constant, \( a^* = 2a d_0/VR^2 \), where \( d_0 \) is the capillary length scale, a materials parameter defined from the equilibrium temperature of the crystal-melt interface, the solid-liquid interface energy, and the specific and latent heats. Although some theories predict the value of this scaling constant, in practice, the scaling constant is used as an adjustable parameter to describe the dendritic growth data that is observed in various materials.

The early SCN dendritic growth data showed that natural convection might be modifying the conductive heat transfer process, (i.e., the shape of the dendritic growth chamber and the direction of [100] growth of the primary dendrite with respect to gravity affected the measurements). It became necessary to not only characterize dendritic velocity and radius measurements as functions of the applied
supercooling, but also to parameterize the measurements by growth orientation with respect to gravity.

Unfortunately, more detailed subsequent work showed that gravity-induced convection actually dominates dendritic growth, particularly in the lower supercooling range typical of castings [Huang and Glicksman (1981), Glicksman and Huang (1982)]. There have been some attempts to estimate the effects of natural or forced convection on dendritic growth [Ananth and Gill (1988 and 1991)], but these results are more of a semi-quantitative insight into the process of convective effects on dendritic growth rather than a method to yield a priori predictions of V and R independently. Ananth and Gill were able to determine the extent to which the experimental results of Huang and Glicksman (1981) were consistent with the Navier-Stokes and energy equations, and, as such, provided insight into how to characterize the transport field around a dendrite tip in the presence of convection. However, from the perspective of the search for a priori solutions, these calculations remain coupled to yet-unproven elements of basic dendritic growth theory, and, consequently, can not provide an independent test of the theory.

In the higher supercooling range, where convective influences diminish in comparison to thermal conduction, the morphological scale of dendrites becomes too small to be resolved optically at the high growth speeds encountered. The experimental situation prior to the microgravity experiment reported here was that there appeared to be too narrow a range of supercoolings in any crystal-melt system studied terrestrially that both remained both free of convection effects and permitted an accurate determination of the dendrite tip radius of curvature. As shown here, the microgravity environment broadens this range of supercoolings and permits, for the first time, a critical assessment of both the heat transport theory and interfacial kinetics behavior. Furthermore, with a clearly identified diffusion-limited dendritic growth regime, a detailed quantitative analysis of the effects of convection on dendritic growth becomes possible.

Experimental Apparatus, and Procedures

The Isothermal Dendritic Growth Experiment (IDGE), as one of four experiments comprising the second United States Microgravity Payload mission (USMP-2), was launched on March 4, 1994, to a low-Earth circular orbit of approximately 163 nautical miles, housed in the payload bay of the space shuttle Columbia (STS-62) [Glicksman, Koss, and Wiusa (1994)]. The IDGE was designed to obtain dendritic growth data on better than 99.999% pure SCN at greatly reduced acceleration levels (microgravity). After launch, the IDGE operated for more than nine days at an ambient quasi-static acceleration environment of approximately 0.7 μg₀ and produced 37 independent experimental runs in which dendritic velocity and radius data pairs of acceptable accuracy (± 2% and ± 3% respectively) were measured over a supercooling range of about 0.06 to 1.9 K. The ground-based laboratory techniques for accomplishing simultaneous kinetic and morphological measurements for freely growing SCN dendrites are well established [Huang and Glicksman (1981), Glicksman et al. (1976)]. However, the challenge of designing and building a dendritic growth chamber capable of operating under microgravity conditions was to
integrate these established techniques with the special requirements of an autonomous, or tele-operational, microgravity experiment.

Full details of this development are reported elsewhere [Glicksman et al. (1988 and 1991), Rubinstein et al. (1990)]. In brief, the flight growth chamber and photographic-optic system are central to the flight experiment apparatus (Fig. 1). The stainless steel and glass growth chamber provides an unobstructed view of the growing dendrites through four perpendicularly set windows which permit stereo observation. A thin capillary injector tube, called the stinger, has thermoelectric coolers attached to one end so that dendritic growth can be initiated electronically. The other end of the stinger (where the dendrite emerges) is located near the center of the chamber. The growth chamber itself is mounted inside a temperature-controlled tank, or thermostat, and is connected to a modified shadowgraphic optical system that includes 35mm camera, and Slow Scan Television (SSTV) system used for dendrite detection and near-real-time monitoring system.

For each dendritic growth cycle, the thermostatic bath is heated to 3 K above the SCN, melting temperature of 58.08°C and the bath is maintained at this elevated temperature to ensure that all the SCN in the chamber is melted and no stray crystals are present. The bath temperature is then reduced to provide the desired supercooling in the SCN. After the molten SCN has cooled and stabilized at this prescribed supercooling, thermoelectric coolers initiate solidification at the capped end of the stinger. The solidifying SCN grows down along the inside wall of the stinger until it emerges from the end of the stinger in the center of the growth chamber, which still contains molten and isothermal SCN.

For each dendritic growth cycle the experiment produced a time series of stereopairs of 35mm photographic images; low-resolution binary digitized electronic images; and data streams which characterized the dendrite's growth conditions during

![Diagram of IDGE growth chamber, thermostatic bath, 35mm cameras, and Slow-Scan Television cameras.](image)
the cycle. For each image pair, either 35mm film negative or digitized image, the position of the dendrite tip and the time of the image were recorded. The dendritic velocity was calculated from the slope of the displacement-time curve in the steady-state regime of the growth cycle. The velocity was then adjusted by appropriate stereographic and magnification factors. This procedure yields a set of dendrite velocity measurements accurate to about ± 2%.

The overall experimental task of measuring the size and shape of steady-state dendritic crystals tips is complex. This complexity is evidenced by noting that the typical experimental uncertainty encountered in the best terrestrial laboratory-based tip radii measurements is typically ± 10%. Some of the details of our tip shape analysis methods are described elsewhere [Glicksman et al. (1994), LaCombe et al. (1995)]. In summary, the gray-scale film image is digitized to locate the solid-liquid interface, and reduced to a list of coordinate pairs that define the dendrite edge. The data describing the dendrite edge profile is regressed to an equation for the model of a dendrite, from which the radius of curvature at the tip is calculated. The tip radii data extracted from the IDGE image data files are usually accurate to better than ± 5% for supercoolings less than 1.0 K.

Results from IDGE/USMP-2

The two ground-based dendritic growth velocity data sets used for comparison with the microgravity data, Glicksman/Huang and IDGE terrestrial, are in close agreement over the entire supercooling range (Fig. 2). Thus, negligible differences exist between these data sets arising from the different chamber sizes and shapes, the different SCN specimens employed, and the different eulerian angles (between 0 and 45 degrees) of the [100] growth direction with respect to gravity. However, there are dramatic differences in the observed growth rates between terrestrial and microgravity conditions, particularly at lower supercoolings. At about 1.7 K supercooling there appears to be a slight reduction, about 5%, in the growth velocity under microgravity conditions as compared to terrestrial conditions. This reduction in velocity increases to approximately 15% at 1.3 K supercooling where it had previously been thought that no differences between terrestrial and microgravity conditions would occur. This difference between the two data sets increases to almost a 200% reduction at yet smaller supercoolings. This indicates that substantial convective effects exist at 1g0 for even the highest supercoolings at which solidification microstructures can be resolved during in situ observations. However, both the microgravity data set and the Huang/Glicksman terrestrial data set merge smoothly with the Glicksman, Schaefer & Ayers’ terrestrial data set at approximately 2.0 K supercooling.

By treating the scaling constant, \( \sigma^* \), as an adjustable parameter (which we designate \( \sigma_v \)), we are able to fit the observed microgravity velocity data at supercoolings of 0.47 K and above with the theoretical velocity calculations, using \( \sigma_v = 0.0164 \). The good agreement of the velocity calculations with the microgravity data over the range of supercoolings from 0.47 K to 1.7 K show that this is the supercooling range over which dendritic growth theories employing heat conduction as the rate limiting transport process must be tested. This theory line also matches the terrestrial data at supercoolings above 2.0 K.
The slope of the microgravity data changes from being steeper than terrestrial data at higher supercoolings, to being almost parallel at supercoolings less than ca. 0.4 K (Fig. 3). This may indicate that, even in microgravity, substantial convective effects reappear at lower supercoolings; however, more detailed analysis and evidence are required. The deviations between the microgravity data at lower supercoolings and the theory-line fit to the data at the larger supercoolings, using $\sigma_0$ as the adjustable parameter, provide quantitative evidence that the quasistatic residual microgravity level of approximately 0.7 $\mu g_0$ may have a significant, and eventually dominant, effect on dendritic growth. Thus, at sufficiently low supercoolings, even in microgravity, the dendritic growth velocity is substantially altered from what would be expected by heat conduction alone.
Both data sets exhibit more scatter at lower supercoolings than at higher supercoolings. The scatter in the microgravity data at lower supercoolings is 10%–15%. This is three times the scatter in the comparable terrestrial data sets in the same supercooling range, and up to five times the measurement uncertainty of the microgravity data. This disparity is too large to be caused by measurement uncertainty, and is perhaps due to variations in the acceleration levels during a growth cycle or between different growth cycles.

Such acceleration level variations have been determined from the Orbital Acceleration Research Experiment (OARE), which measured the low frequency, quasi-static acceleration in three perpendicular directions taken once every 25 seconds. The OARE data shows an average quasi-static acceleration of about 0.7 \( \mu g_0 \), with a nearly continuous jitter spectrum of 0.2–0.8 \( \mu g_0 \), and an occasional spike up to 3 \( \mu g_0 \) (Fig. 4). We have attempted to correlate a) the scatter in the microgravity data at the lower supercoolings to b) the differences in the direction of growth of the
measured dendrite with respect to the direction of the local net microgravity acceleration, which vary growth to growth, and the variations in the acceleration environment. To show any correlation between acceleration levels and the variation in velocities at the lower supercoolings two potential effects must be separated: 1) the effect of the amplitude of the acceleration; and 2) the effect of the direction of the acceleration with respect to the direction of growth of the measured dendrite. However, at this time, without a theoretical model, it is not straightforward to decide or determine the range of times of dendrite growth over which acceleration data is relevant, and what, if any, are the effects of deviation from the average acceleration. There are not sufficient data, with either the same average acceleration or the same angle between the growth direction of the measured dendrite and the acceleration vector, to determine a definitive correlation. Thus, although the data suggest qualitatively that the microgravity data below approximately 0.4 K supercooling is strongly effected by convection, there is little definitive quantitative evidence at this time.
The discussion of the radii results (Fig. 5) parallels that already presented on the velocity data, except that here the radius is not as strong a function of supercooling as the velocity, so that the ratios between the terrestrial and microgravity results are smaller. A one-parameter fit to the radii data over the supercooling range from 0.47 K to 1.57 K yields the adjustable scaling parameter \(\sigma_R = 0.0179\), which describes the microgravity radii data at higher supercoolings. The microgravity measured Péclet number data set is clearly separated from the terrestrial data (Fig. 6). Most importantly, the microgravity data vary from that predicted by the Ivantsov theory for paraboloidal dendrites. At lower supercoolings these deviations are ascribed to convection. The approximate agreement achieved between the transport theory and the microgravity data indicates that dendritic growth is indeed most likely governed by the conduction of latent heat from the crystal-melt interface. The remaining deviations are unexplained, but it is clear, even in microgravity, non-diffusive effects are important.

An experimentally determined microgravity scaling constant, \(\sigma^* = 2\pi\eta_0/(VR^2)\), calculated from the microgravity measurements of \(V\) and \(R\) at high supercoolings where convection affects are assumed minimal, yields \(\sigma^* = 0.0194\) (Fig. 7). Curiously, this value of \(\sigma^*\) almost exactly matches the terrestrially measured value of \(\sigma^*\). Thus, despite a six order of magnitude change in the gravitational acceleration environment and dramatic changes in both the measured dendritic \(V\) and \(R\) between those two environments, \(VR^2\) is the same, independent of the acceleration field. However, the direct calculation of \(\sigma^*\) from combined \(V\) and \(R\) measurements is different than the adjustable scaling parameters used to fit either the velocity or radii data. Thus,

![Graph showing dendrite tip radii as a function of supercooling.](image)

**FIGURE 5** Dendrite tip radii as a function of supercooling.
there appears to be no single scaling parameter, whether calculated directly from the combined velocity and radius data or inferred as an adjustable parameter, that when coupled to the Ivantsov heat transfer results, properly describes both the velocity and radius data. If \( \sigma^* \) properly encapsulates the physics of a second length scale, then it is inconsistent to have different values of \( \sigma^* \) generated from the \( V \) or \( R \) data. This strongly suggests that the Ivantsov function combined with a unique scaling constant might not correctly describe steady-state dendritic growth. The inability of Ivantsov’s conduction solution to describe the diffusion-limited growth regime is discussed more fully elsewhere [Glicksman et al. (1995)]. However, it now appears that the deviations between Ivantsov’s conduction theory and our microgravity measured \( V \) and \( R \) data at higher supercoolings is the main source of differences among \( \sigma^*, \sigma_v \), and \( \sigma_R \). Thus, the best way to examine the scaling constant \( \sigma^* \) is through direct measurements and calculation.

The scatter, both within and among the three \( \sigma^* \) data sets, is significant. There appears to be a trend where \( \sigma^* \) increases inversely with supercooling. If \( \sigma^* \) has a dependence on supercooling, that dependence is rather weak compared to the scatter in the new dendritic growth data, especially in the diffusion-limited regime.
established by the microgravity velocity data. Thus, assuming that \( \sigma^* \) remains constant over this limited supercooling range would only introduce a slight error in any quantity that depends on \( \sigma^* \). This finding is in agreement with our earlier observation that the average of \( \sigma^* \) from the diffusion-limited microgravity data is almost the same as the average value of \( \sigma^* \) derived from terrestrial measurements. Most striking is the observation that although the terrestrial and microgravity velocity and radii data are markedly different, the terrestrial and microgravity \( \sigma^* \) data are virtually indistinguishable from each other. It is particularly surprising that \( \sigma^* \), at a particular supercooling, may be insensitive to the differences between the terrestrial or microgravity convective environments, but may instead vary weakly with supercooling.

**Summary and Conclusions**

We measured dendritic growth velocities and tip radii of curvature of succinonitrile in microgravity using the IDGE instrument flown on the USMP-2 platform in the payload bay of the space shuttle Columbia (STS-62). The on-orbit microgravity data, when compared to terrestrial dendritic growth data, demonstrate that: (1) convective effects under terrestrial conditions cause growth speed increases of a factor of 2 at lower supercoolings (\( \Delta T < 0.5 \text{ K} \), and remain significant even up to values as high as \( \Delta T = 1.7 \text{ K} \) supercooling; (2) In the supercooling range above 0.47 K, the data remain virtually free of convective effects, and may be used reliably for examining diffusion-limited dendritic growth theories; (3) Below 0.4 K, dendritic growth is strongly affected by non-diffusion effects, for which we suspect, that even in microgravity, is due to convection. When modeling dendritic growth under terrestrial conditions, which is a goal of dendritic growth research, the effect of convection
must be included. The data provided by the IDGE can then be used to compare quantitatively diffusion-limited and convective-based dendritic growth theories, with dendritic growth observations at two well characterized (0.7 \times 10^{-6} \text{ and } 1 \text{ g}_0) gravity levels.

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REFERENCES


NOMENCLATURE

\begin{itemize}
\item \(d_0\) \text{ Capillary length scale}
\item \(g_0\) \text{ Gravitational acceleration at the earth’s surface, terrestrial gravity}
\item \(V\) \text{ Dendritic tip growth velocity}
\end{itemize}
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<th>Symbol</th>
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<tr>
<td>$R$</td>
<td>Dendritic tip growth radius of curvature at tip, tip radius</td>
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<tr>
<td>$\alpha$</td>
<td>Thermal diffusivity</td>
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<tr>
<td>$\Delta T$</td>
<td>Melt supercooling</td>
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<tr>
<td>$\mu g_0$</td>
<td>One one-millionth of the gravitational acceleration at the earth's surface, one micro-g</td>
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<tr>
<td>$\sigma^*$</td>
<td>Scaling constant, scaling parameter ($2\pi d_0/VR^2$)</td>
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<td>$\sigma_R$</td>
<td>Adjustable scaling constant determined from the measured dendritic growth tip velocity</td>
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<td>$\sigma_v$</td>
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