

CONVECTIVE FLOW DURING DENDRITIC GROWTH

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ABSTRACT

A review is presented of the major experimental findings obtained from recent ground-based research conducted under the SPAR program. Measurements of dendritic growth at small supercoolings indicate that below approximately 1.5 K a transition occurs from diffusive control to convective control in succinonitrile, a model system chosen for this study. The key theoretical ideas concerning diffusive and convective heat transport during dendritic growth are discussed, and it is shown that a transition in the transport control should occur when the characteristic length for diffusion becomes larger than the characteristic length for convection. The experimental findings and the theoretical ideas discussed suggest that the Fluid Experiment System could provide appropriate experimental diagnostics for flow field visualization and quantification of the fluid dynamical effects presented here.

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INTRODUCTION

Accompanying thermal dendritic growth, the latent heat is dissipated from the moving solid-liquid interface through the surrounding supercooled melt. This heat transfer gives rise to a thermal field around the growing dendrite, whereby latent heat flows from solid to liquid along the thermal gradient. The presence of such a gradient alone is responsible for diffusive heat flow. Under terrestrial conditions, however, a pressure gradient develops from the thermal gradient and its associated density gradient. This gradient produces a fluid flow field which changes the thermal distribution near the dendrite, thereby modifying the amount of heat flowing from the interface. The heat flow may be increased or decreased depending on the relative direction between the diffusive flow and the convective flow.

As described above, terrestrial dendritic growth experiments always involve both diffusion and convection. However, the diffusive heat transport process increases rapidly and non-linearly with increased supercooling, whereas the convective heat transport process increases more linearly with supercooling. Thus, at relatively large supercoolings, the diffusive component tends to dominate the heat transfer processes, whereas at small supercoolings, convection must eventually dominate. The diffusion-convection transition in succinonitrile—a material used in a study of diffusion-controlled dendritic growth [1]—occurred at about 1°C. Dendritic growth in succinonitrile at supercoolings smaller than 1°C will thus be controlled

by convection.

In the series of experiments examined in this paper, convection-controlled dendritic growth was the prime subject of study. Experiments reported here include the influence of spatial orientation (from 0-180° relative orientation between the growth direction and the gravity vector) on the dendritic growth of succinonitrile. The description and qualitative explanation of the experimental results were presented in a previous paper [2].

The spatial orientation effect measurements were repeated at several levels of supercooling below 2°C to yield the dependence of the dendritic growth rate on supercooling, in the range where the growth kinetics are controlled by natural convective heat transfer. These results were presented in Refs. [2] and [3]. The discussion of orientation effects on convectively controlled dendritic growth was based originally on a theory proposed by Doherty, Cantor, and Fairs [4]. This theory considers only the case of dendrites growing under counterflow conditions, i.e., where the dendrite tip propagates in opposition to the convective fluid velocity. Moreover, this theory estimates the far-field flow velocity, U_{∞} induced by natural convection, by employing a formula developed by Szekely and Themelis [5]. Finally, the convective heat transfer from the dendrite tip can be calculated using a fluid mechanical model originally proposed for forced convection [6]. The supercooling dependence of dendritic growth velocity predicted by this theory is, however, inconsistent with our experimental results [3].

The failure of that theory may be ascribed to the theoretical assumption that the tip region of a growing dendrite is the sole source of heat in the system. Consequently, the convection length scale is linked to the tip dimensions. Actually, in our method of studying solidification, a

dendritic mass consisting of five or six dendrites emerges from the capillary aperture in the bulb (C) - c.f. Fig. 1. Although each dendrite is growing independently, the whole freezing complex acts as a large-scale heat source. The convective current present in our experimental system thus can be expected to flow more rapidly than a fluid current induced by a single dendrite tip acting alone. Hence, the convection length scale should be associated with the multidendrite freezing complex.

In this report, we will first describe a few details of our experimental method and some salient experimental results. We will then present a model intended to explain the supercooling dependence of dendritic growth kinetics under the influence of convective heat transport. Major emphasis is placed on predicting from theory the supercooling level at which the transition from diffusion-controlled to convection-controlled dendritic growth occurs. It should be noted here that preliminary to any analysis of dendritic growth kinetics when under the control of convective heat transport, one must obtain a description of the kinetics when under the control of thermal diffusion alone. Fortunately, a new, and relatively complete, theory of diffusion-controlled dendritic growth was published recently [7]. Furthermore, this theory has been verified in two experiments [8,9] to be correct to at least $\pm 5\%$. Thereafter in this report, the supercooling dependence of dendritic growth velocity predicted by this new dendritic growth theory will be used to predict the baseline (diffusional) kinetics to analyze the influence of convection on dendritic growth.

EXPERIMENTAL

The present series of experiments was designed to define critically the precise experimental conditions for free dendritic growth with pure

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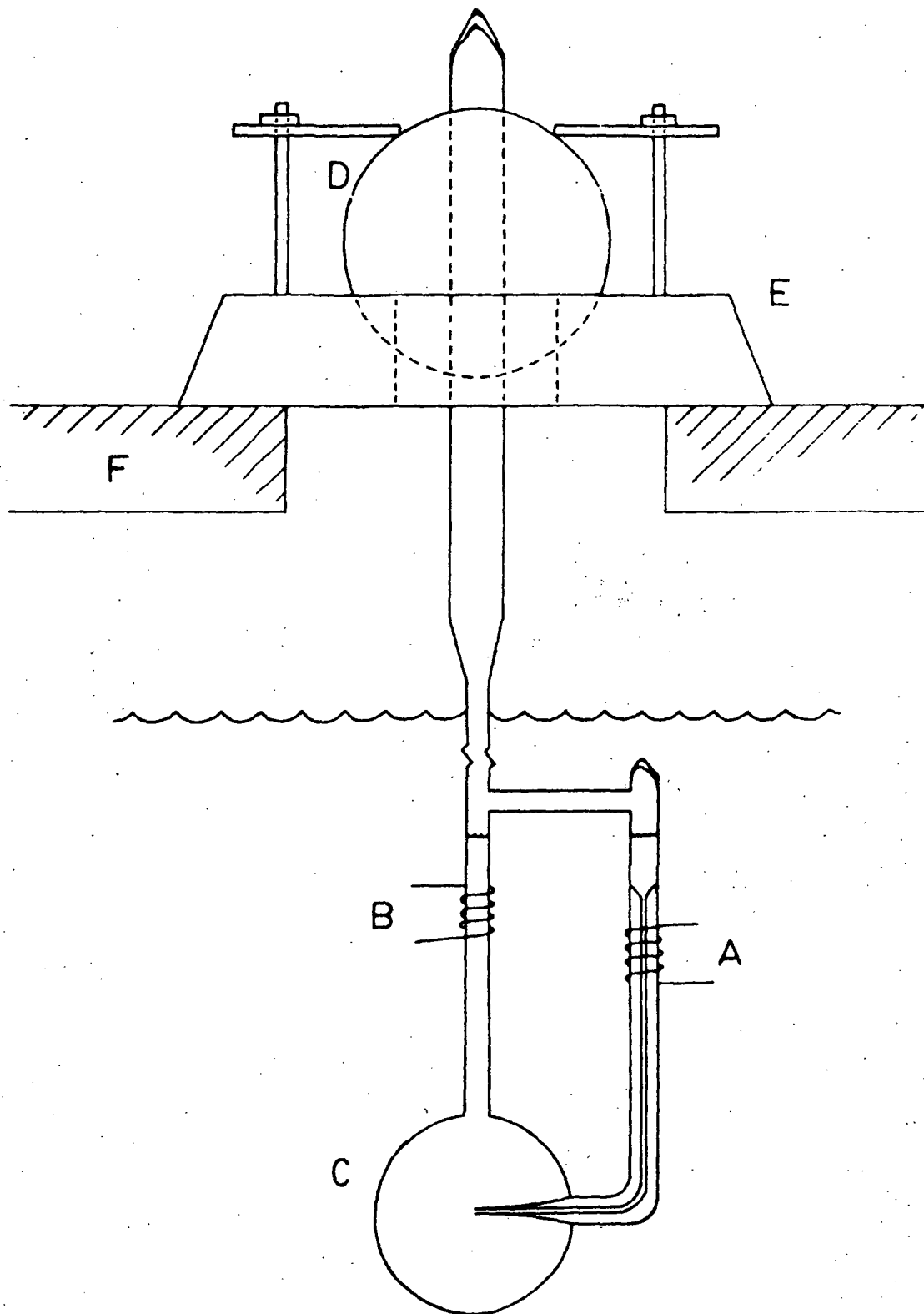


FIGURE 1. Schematic drawing of specimen configuration and support stage. A and B are control heaters; C is the crystal growth chamber; D is the tilting and secondary rotating device; E is the primary rotation and X-Y translation stage; F is the tank cover.

heat transfer (i.e., no solute diffusion). These experiments were designed to permit measurement of pertinent experimental variables, namely: dendritic growth velocity, V ; dendritic orientation angle with respect to gravity, θ ; and supercooling, $\Delta T \equiv T - T_m$, where T is the pre-set temperature of the molten succinonitrile, and T_m is the equilibrium melting temperature of pure succinonitrile.

Details of specimen purification, specimen characterization, temperature measurements, and temperature control were provided in an earlier paper [2]. The accuracies and measuremental resolution of these factors are compiled in Table 1. As may be shown from Table 1, the uncertainty in measuring the initial supercooling, ΔT , is less than $\pm 0.001^\circ\text{C}$. Furthermore, our preparing a specimen of succinonitrile with better than 6-9's purity ensures attainment of growth kinetics controlled solely by the flow of latent heat. Solute effects may be safely ignored.

Table 1. Resolution, Control Accuracy, and Purity of Experimental System

Temperature Measurement Resolution	Temperature Measurement Accuracy	Temperature Control Stability	Uncertainty in Melting Temperature	Purity Level in Specimen
0.0004°C	$\pm 0.002^\circ\text{C}$	$\pm 0.0004^\circ\text{C}$	$\pm 0.0004^\circ\text{C}$	> 99.99995%

Dendritic growth studies were carried out in the specimen chamber detailed schematically in Figure 1. Two control heaters, A and B, prevented stray crystals from growing into the chamber C. The succinonitrile could thus be kept liquid in the chamber at any pre-selected temperature established in the thermostatted observation tank. Normally it took about 50 minutes for the entire specimen to achieve a uniform temperature. At that

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point, control heater A was switched off, and the seed crystal above location A was permitted to propagate into the chamber C through the capillary. The dendrites growing within the chamber were then free of any extraneous interaction with the glass chamber walls until they touched the walls at the end of each run. Furthermore, the outward growing dendrites tended not to interact with one another through overlap of their surrounding thermal fields. Achievement of this unconstrained or free dendritic growth condition is essential to the present kinetics study.

As shown in Fig. 1, the specimen was supported by a special stage which allowed full rotation, a tilt of $\pm 6^\circ$, and a two-axis translation of ± 2.5 cm. The ability to maneuver the growing dendrites into a desired spatial orientation with respect to gravity and the axis of observation was essential to the present study. The growing crystals were observed with a Wild M5A stereomicroscope, equipped with a trinocular assembly and a Wild MKal camera. Photographs were taken through an orange filter using fine-grain Polaroid 105 film (3 1/4" x 4 1/4", ASA 75) with direct, diffused, electronic flash. To minimize optical distortion from the spherical specimen chamber, the observation tank was filled with a mixture of ethylene glycol-17 vol % H₂O, selected to match the index of refraction of succinonitrile as well as act as the heat transfer medium.

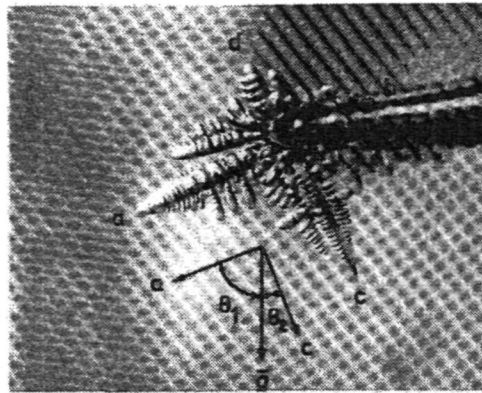
Free dendrites emerged from the tip of the capillary and grew into the spherical chamber in the expected $\langle 100 \rangle$ cube-edge directions. Although such dendrites were either perpendicular or parallel to each other, they grew in random directions with respect to the direction of observation. To determine the true growth velocity and the true growth orientation with respect to the gravity vector, \bar{g} , the dendrites were photographed

from two different directions. Since the microscope was fixed, this required rotation of the specimen chamber.

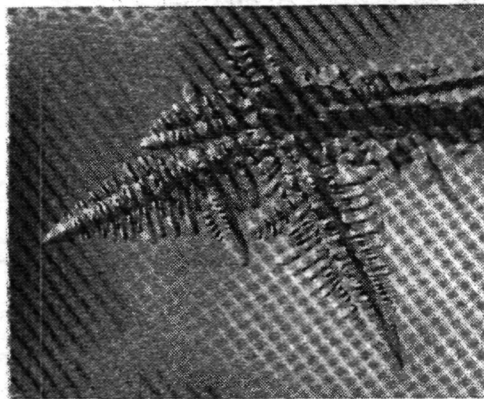
The specimen rotation procedure required to measure true dendritic growth velocities and growth orientations consisted of the following steps:

1) As soon as free dendrites started growing, the specimen chamber was rotated to position a colinear pair of dendrites (e.g., dendrites with axes along $[100]$ and $[\bar{1}00]$ in the plane of observation. Figure 2(a) is a photograph taken after completion of this rotation operation. Note that the images of those branches of dendrite c, which were growing perpendicular to the focal plane, appear as a "string of beads". Also, the tips of dendrites a and b were simultaneously rotated into the focal plane. Under this special circumstance, the relative orientation angle of dendrite a from \bar{g} is expressed by θ_1 , defined in Fig. 2(a), and that of dendrite b by $(180-\theta_1)$. Also, the growth velocity of dendrites a and b can be calculated directly from the tip displacements measured on a series of photographs taken at known time intervals, such as shown in Figs. 2(a) and 2(b).

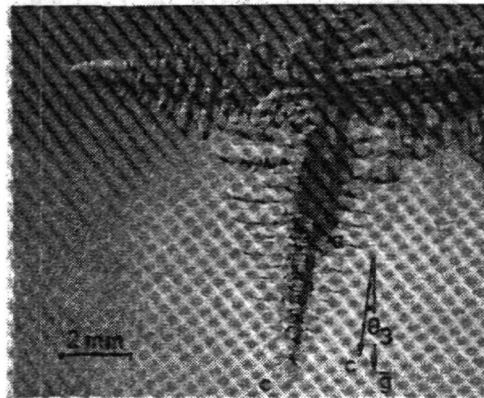
2) The angle θ_2 , shown in Fig. 2(a), however, might not represent the true deviation angle of dendrite c from \bar{g} , because dendrite c might be growing at some angle θ_3 out of the plane of Fig. 2(a). The angle θ_3 was measured on a photograph taken after the specimen chamber had been rotated $+90^\circ$ or -90° about \bar{g} . This is shown in Fig. 2(c). We note again that the perpendicular branching sheet of dendrite c in Fig. 2(c), was previously observed as the side-branches of dendrite c in Fig. 2(a). Accordingly, the true deviation angle of dendrite c from \bar{g} should be given by $\theta_4 = \tan^{-1}[(\tan^2\theta_2 + \tan^2\theta_3)^{1/2}]$ and that of dendrite d by $(180-\theta_4)$. Also, the apparent growth velocity of dendrite c, as well as that of dendrite d, must be multiplied by the factor $(\cos\theta_2 \cdot \sec\theta_4)$, which accounts for the stereographic corrections discussed above.



(a)



(b)



(c)

FIGURE 2. (a) Dendrite a and b rotated to lie in the focal plane. (b) 4.7 minutes after (a). (c) A sideview of the growing dendrite complex seen in (a) and (b), accomplished by a 90° rotation of the specimen chamber about g.

RESULTS AND DISCUSSION

Dendritic growth velocities, V , were measured as a function of growth orientation, θ , at seventeen supercoolings ranging from 0.043°C to 2°C . Five typical experimental curves of V versus θ are shown in Fig. 3. As may be noted in Fig. 3, a downward growing succinonitrile dendrite (propagating against the natural convective fluid current) tends to grow faster than an upward-growing dendrite. Detailed discussion of this orientation effect can be found in Ref. [2] and [3]. Also observable in Fig. 3 is that the dependence of the dendritic growth velocity on spatial orientation increases in degree at small supercoolings. The supercooling dependence of growth kinetics for dendrites growing parallel to gravity is summarized in Fig. 4. Also included in Fig. 4 is the theoretical curve of V_d versus ΔT predicted for diffusion-controlled dendritic growth [7-9]. By comparison, the convective flow tends to enhance the growth of downward-growing dendrites below a certain level of supercooling. Fig. 5 is obtained when the measured growth velocities, V , are normalized to the theoretical diffusive dendritic growth velocities, V_d . Fig. 5 shows clearly that the diffusion-convection transition occurs rather suddenly at a supercooling of about 1.5°C .

The remainder of this discussion concerns the development of a theory to predict the critical supercooling at which the diffusion-convection transition occurs. To account realistically for the heat transport attendant to the crystal growth method used in this study, we will consider the whole dendritic complex (see again Fig. 2) as the heat emitting source which drives the natural convective fluid flow, Fig. 6. As such, the reference length of the convective flow field, l , must be chosen as the radius of the dendritic complex ($l \sim 1 \text{ cm}$). This reference length is rela-

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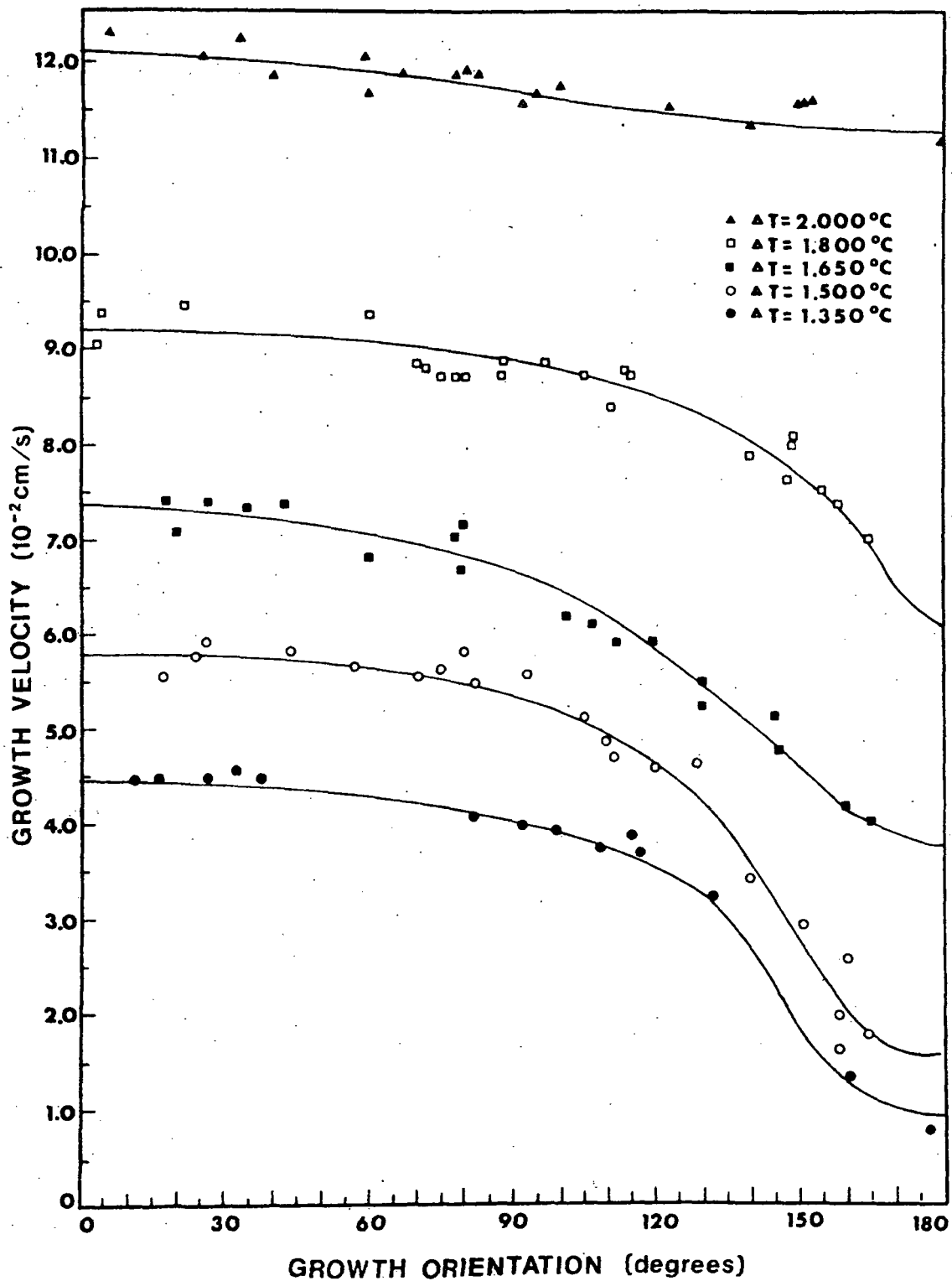


FIGURE 3. Dendritic growth velocity versus growth orientation with respect to gravity at five levels of supercooling.

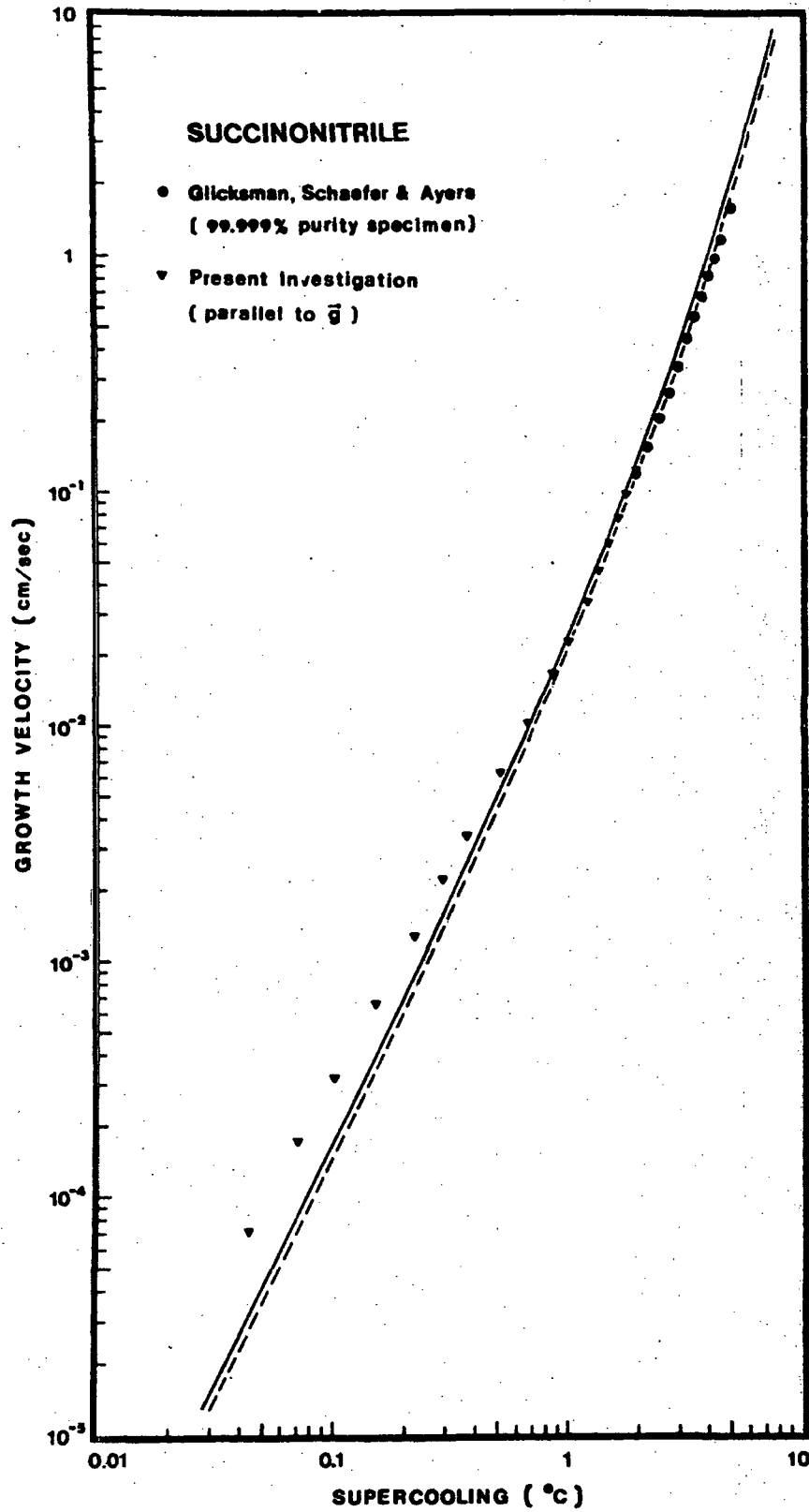


FIGURE 4. Dendritic growth velocity versus supercooling for dendrites growing parallel to gravity.

tively large compared to the dendrite tip radius (10^{-4} to 10^{-3} cm) which was used as the reference length in the model of Doherty et al. [4]. The velocity U_{∞} of the convective flow induced by such a dendritic mass is given by [10]

$$U_{\infty} = A_1 Gr^{1/2} \frac{\nu}{\ell}, \quad (1)$$

where A_1 is a constant approximately equal to unity, ν is the kinematic viscosity, and Gr is the Grashof number defined as

$$Gr = \frac{g\beta\Delta T\ell^3}{\nu^2},$$

where g is the gravitational acceleration, and β is the volume expansion coefficient. The presence of fluid flow modifies the thermal field surrounding a dendrite, which would be governed by the diffusion of heat were the liquid phase in a quiescent state. The characteristics of heat transfer within a thermal field can be conveniently described by the thickness of the "thermal boundary layer". For the case of convective heat transfer, the thermal boundary layer thickness, δ , is given by [11]

$$\delta_T/\ell = A_2 Re^{-1/2} Pr^{-1/3}, \quad (2)$$

where A_2 is a correlation constant approximately equal to 0.5; Re is the Reynolds number defined as

$$Re = \frac{U_{\infty} \ell}{\nu}; \quad (3)$$

and Pr is the Prandtl number defined as

$$Pr = \frac{\nu}{\alpha}, \quad (4)$$

where α is the thermal diffusivity. By combining eq. (1) and (2), the thermal boundary layer thickness can be expressed as

$$\delta_T = A_1^{-1/2} A_2 (g\beta \Delta T)^{-1/4} l^{1/4} \alpha^{1/2} Pr^{1/6} \quad (5)$$

For the case of thermal diffusion at a dendrite tip, δ is given by the Stefan boundary layer thickness defined as

$$\delta_s = \frac{2\alpha}{V_d} \quad (6)$$

where V_d is the diffusion-controlled dendritic growth velocity, which may be described by the power law [1].

$$V_d = \frac{A_3 \alpha \Delta S}{\gamma} \left(\frac{C_p}{L} \right)^{1.5} (\Delta T)^{2.5} \quad (7)$$

Here, A_3 is a constant equal to 0.018, ΔS is the entropy of fusion per unit volume, γ is the solid-liquid interfacial energy, C_p is the heat capacity of the liquid, and L is the latent heat of fusion. Therefore, the Stefan boundary layer for the pure diffusion case can be expressed as

$$\delta_s = \frac{2\gamma(L/C_p)^{1.5}}{A_3 \cdot \Delta S (\Delta T)^{2.5}} \quad (8)$$

When $\delta_T = \delta_s$, a "crossover" can occur in the dominant heat transport mechanism. The "crossover" condition is obtained when the right-hand sides of eq. (5) and (8) are set equal. This procedure yields a critical transition supercooling ΔT^* , which when expressed in a dimensionless form $\Delta\theta^* = \Delta T^* / (L/C_p)$, is given by

$$\Delta\theta^* = \left[\left(\frac{g\beta}{l} \right) \left(\frac{A_1}{\alpha} \right)^2 \left(\frac{C_p}{L} \right)^3 \left(\frac{\gamma}{A_2 A_3 \Delta S} \right)^4 \right]^{1/9} Pr^{-2/27} \quad (9)$$

Inserting the pertinent materials parameters (see Ref. [1] and [2]), into eq. 9 yields $\Delta T^* = 1.23^\circ\text{C}$ for succinonitrile. Comparison of this result to that measured from Fig. 5, indicates that eq. 9 is predictive to within

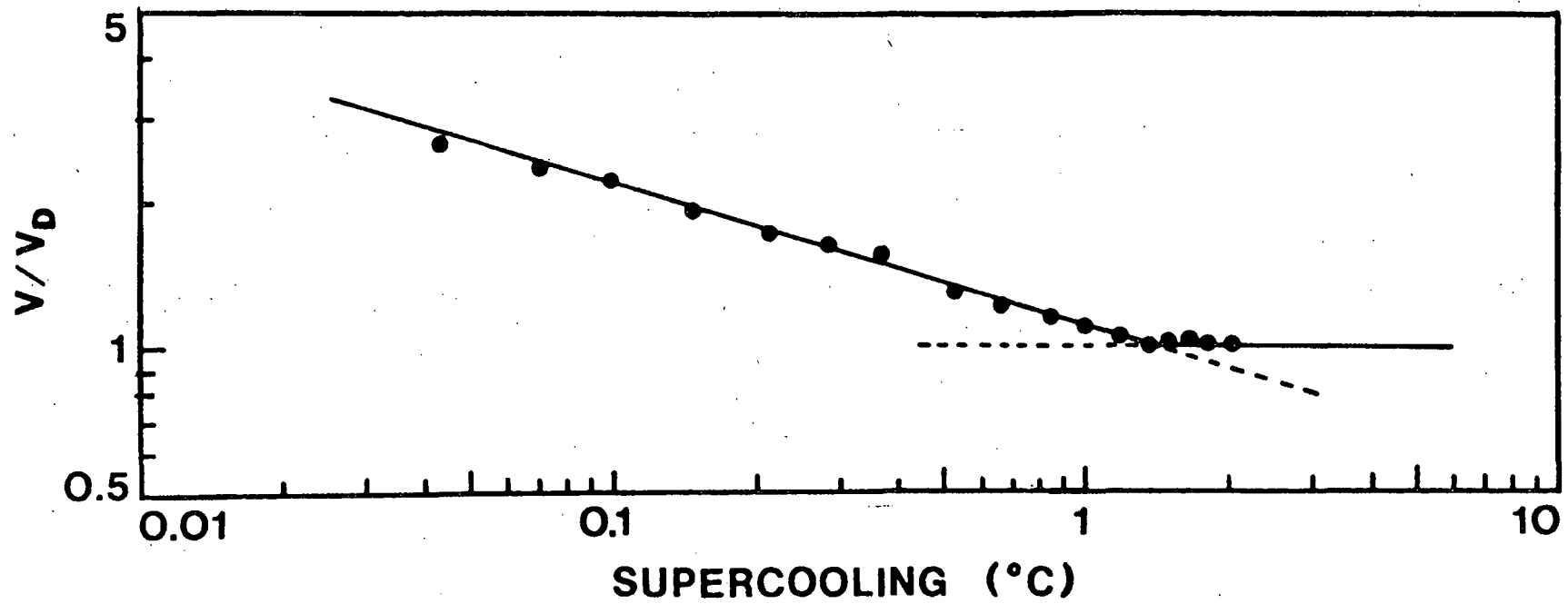


FIGURE 5. Normalized dendritic growth velocity versus supercooling for dendrites growing parallel to gravity.

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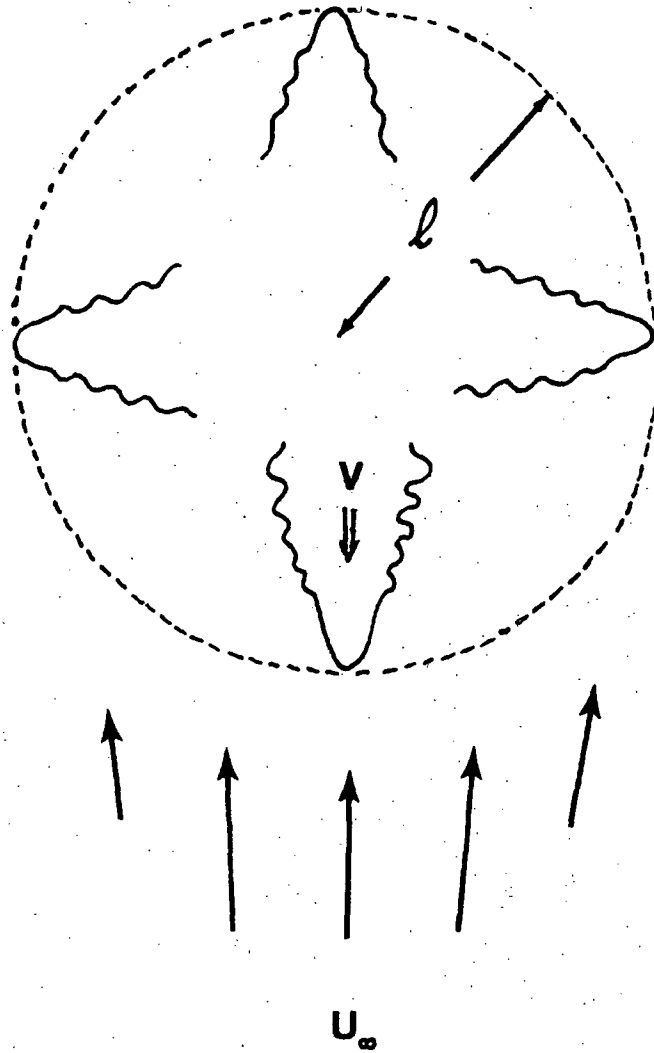


FIGURE 6. Schematic showing the natural convective fluid flow in front of a downward-growing dendrite. The convective flow is induced by heat released by the solidifying dendritic mass.

about 10%, which is the combined level of uncertainty of the parameters.

Normally, viscosity and expansion coefficient are considered important materials parameters in determining the relative ease of natural convection. Equation 9, however, indicates that $\Delta\theta^*$ varies with β to the 1/9 power, and with ν to the -2/27 power. Furthermore, since $\Delta\theta^*$ is proportional to $\ell^{-1/9}$, the manner of choosing the reference scale has relatively little effect on the predicted value of the "crossover" supercooling. The crossover point shows a similarly weak dependence on the gravitational level g .

SUMMARY

1) The kinetics of dendritic growth in pure materials is controlled by the release of latent heat, which is removed from the solid-liquid interface by diffusive and/or convective flow.

2) The diffusion of heat from a dendrite increases rapidly and non-linearly with increasing supercooling, whereas the convection of heat varies in a more linear manner.

3) Significant convection effects in succinonitrile, manifested by the orientation dependence of the growth rate, occur when the supercooling is less than 1.5°C.

4) The crossover between diffusive and convective transport depends on the relative thickness of the Stefan or diffusion length compared with the thermal boundary layer. These lengths become equal at a supercooling which may be calculated from diffusion theory and fluid mechanics.

5) The theoretical expression for the "crossover" supercooling shows that this quantity varies weakly with such factors as the gravitational acceleration, the melt viscosity, and the volumetric expansion coefficient.

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Ground based experiments have been carried out to measure the influence of melt convection on the growth kinetics of succinonitrile - a model solidification system, which simulates the freezing of metals. Growth velocity measurements will be discussed, with supercooling and spatial orientation with respect to the gravity vector as the two major experimental variables. A distinct transition has been observed near 1.5°C supercooling, where the heat transport mechanism changes from diffusive to convective. The desirability to determine, at least semiquantitatively, the nature of the melt flows will be discussed, along with the requirements which might be imposed by such measurements on the F.E.S.

FUTURE DIRECTIONS AND SIGNIFICANCE OF THE FLUID EXPERIMENT SYSTEM

As described in this paper, convective transport can play a major role in dendritic solidification, especially at small supercoolings. A boundary layer analysis developed here suggests, at least in pure systems, that the dominant transport mechanism changes at the "crossover" of the boundary layer thickness. A detailed analysis which characterizes the flow fields surrounding a dendrite has not yet been developed, nor have experiments been performed to elucidate the behavior of these flows. The Fluid Experiment System, (FES), now being developed by NASA for inclusion on Space Lab III, will provide a variety of fluid flow diagnostic techniques. The convective flows during dendritic solidification are generally slow, laminar flows, of three-dimensional character. The use of schlieren, shadowgraphic, or holographic flow visualization techniques could be explored as possible methods to measure the qualitative nature of these flows. More quantitative approaches such as laser doppler or speckle interferometry could be explored for limited, detailed measurements of fluid flow velocities. If the characteristics of the melt convection could be convincingly established at terrestrial gravitational levels, then the effect of reduced gravity under space flight conditions would be justified. Indeed, the elucidation of how convection modifies the kinetics of dendritic growth in different spatial orientations with respect to the gravity vector remains only partially understood. A more quantitative understanding of this complex phenomenon will contribute to better solidification process design—both on earth and in space—and to achieving better materials with controlled chemical distributions and reduced defects. To this end, the FES represents a potentially important opportunity to explore melt convection in far greater detail than has heretofore been possible.

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