Dendritic Growth Velocities in Microgravity

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(Received 23 March 1994)

We measured dendritic tip velocities in pure succinonitrile (SCN) in microgravity, using a sequence of telemetered binary images sent to Earth from the space shuttle Columbia (STS-62). Growth velocities were measured as a function of the supercooling over the range 0.05–1.5 K. Microgravity observations show that buoyancy-induced convection alters the growth kinetics of SCN dendrites at supercoolings as high as 1.3 K. Also, the dendrite velocity data measured under microgravity agree well with the Ivantsov paraboloidal diffusion solution when coupled to a scaling constant of $\sigma^* = 0.0157$.

PACS numbers: 68.70.+w, 47.54.+r, 81.10.Fq, 81.10.Mx

Dendritic growth is the ubiquitous form of crystal growth encountered when metals, alloys, and many other materials solidify under low thermal gradients, a situation which typically occurs in most industrial solidification processes. A clear elucidation of dendritic growth kinetics under well-characterized diffusion-controlled conditions is crucial in order to achieve a rigorous test of nonlinear dynamical pattern formation theories, and, ultimately, is needed to predict and achieve desired microstructures and physical properties in a variety of solidification processes.

The growth of dendrites in pure liquids is generally acknowledged to be controlled by the diffusive transport of latent heat from the moving crystal-melt interface as it advances into its supercooled melt. A number of theories of dendritic crystal growth, based on various transport mechanisms, physical assumptions, and mathematical approximations, have been investigated over the last forty years, as described in the recent review by one of the authors [1].

A mathematical solution to the dendritic heat flow problem was first analyzed by Ivantsov [2], who modeled the dendrite as a paraboloidal body of revolution, growing at a constant velocity V. The resultant diffusion field can be solved exactly in paraboloidal coordinates moving with the dendritic tip. The solution to this formulation is

$$\Delta\vartheta = \mathrm{Pe}\left(e^{\mathrm{Pe}} \int_{\mathrm{Pe}}^{\infty} \frac{e^{-u}}{u}\, du\right), \tag{1}$$

where $\Delta\vartheta$ is the nondimensional scaled supercooling,

$$\Delta\vartheta = \frac{\Delta T}{L/C_p}, \tag{2}$$

and Pe is the growth Péclet number,

$$\mathrm{Pe} = \frac{VR}{2\alpha}. \tag{3}$$

Here, ΔT is the supercooling, L is the molar latent heat, and C_p is the constant pressure molar heat capacity. In Eq. (3), V is the steady-state dendrite tip velocity, R is the paraboloidal radius of curvature at the tip, and α is the thermal diffusivity of the melt phase.

This diffusion solution is, however, incomplete, insofar as a unique dendritic dynamic operating state (V, R) is not explicitly specified. As one can see from the definition of the Péclet number in Eq. (3), there exists for a specified supercooling an infinite manifold of (V, R) ordered pairs that satisfy the solution set given by Eq. (1). In reality, unique, steady operating states $(V_{\mathrm{exp}}, R_{\mathrm{exp}})$ are observed experimentally for dendrites at each specified supercooling ΔT. Considerable theoretical efforts within the physics community have been directed to answering the question as to whether and under what conditions a second equation or length scale exists, which, when combined with the Ivantsov diffusion solution, selects the unique (i.e., observed) dynamic operating state (see Refs. [1,3–5]). This effort has been motivated in large part by the earlier observation that for several pure materials undergoing unconstrained dendritic growth, VR^2 is a constant, or a weakly varying function of the supercooling, specific to each material (see Ref. [1]). This selection criteria may be based on several different dynamical considerations, such as morphological stability, microscopic solvability, shape anisotropy, noise amplification, traveling waves, etc. Although the underlying physics for these "theories of the second length scale" might in fact be quite different, their results are often encapsulated as

$$\sigma^* = \frac{2\alpha d_0}{VR^2}. \tag{4}$$

Here, σ^* is usually referred to as the stability, selection, or scaling constant, and d_0 is the capillary length scale, a materials parameter defined as

$$d_0 = \frac{T_m \gamma C_p}{L^2}, \tag{5}$$

where T_m is the equilibrium temperature of the crystal-melt interface, and γ is the crystal-melt interfacial energy.

0031-9007/94/73(4)/573(4)$06.00

A great deal of experimental work has been performed using succinonitrile [SCN, the chemical formula of which is $NC-(CH_2)_2-CN$] as a model dendritic growth system, because of its conveniently low melting temperature, optical transparency, and well-characterized thermophysical properties [6,7]. To date, the large body of experimental evidence gathered on SCN and other suitable test materials does not confirm, deny, or differentiate among the different pattern formation theories. The experimental situation, simply stated, is that there appears to be too narrow a range of supercoolings in any given crystal-melt system studied terrestrially that remains both free of convection effects and permits the accurate determination of the dendrite tip radii of curvature. Thus, pure diffusion solutions, and various dynamical selection rules coupled to those solutions, cannot be tested adequately by relying exclusively on terrestrial experiments, where substantial amounts of natural convection are usually present in the crucial range of small supercoolings. A low-gravity dendritic growth experiment was deemed necessary to provide a fundamental and definitive test of the predictions of dendrite growth theory [8]. Such a test requires that setting a single, clearly defined experimental parameter, viz., the supercooling, fully determines the resulting steady-state dendritic growth velocity V and tip size R, without inducing significant convective motions of the melt.

The Isothermal Dendritic Growth Experiment (IDGE) was launched on March 4, 1994, to a low-Earth circular orbit of approximately 163 nautical miles in the payload bay of the space shuttle Columbia (STS-62). The IDGE, one of four experiments comprising the second United States Microgravity Payload Mission (USMP-2), was designed, built, and operated to satisfy these exacting scientific requirements by measuring the growth kinetics and morphology of SCN dendrites growing under microgravity conditions, with its concomitantly reduced natural convection. Complete details of the IDGE instrument and its operational capabilities are available in Ref. [9].

Although the complete analysis of the on-orbit scientific results will have to await the return of the more than 400 photomicrographs taken as 35 mm negatives (that are, as of this writing, still orbiting the Earth), the authors have already analyzed many of the binary, digital images taken during each of some 60 dendritic growth cycles and then immediately telemetered to the Payload Operations and Control Center (POCC), at the Marshall Space Flight Center. These binary images were then interpreted by the IDGE science teams assembled at the POCC, in Huntsville, AL, and at Rensselaer Polytechnic Institute, in Troy, NY. Analysis of the telemetered binary images was limited to dendrite growth velocities only, as extraction of accurate dendritic tip radii measurements demands a spatial resolution that is available in the 35 mm film, but not in the binary images. The velocity measurements were performed in near real time, by measuring the slopes of the dendrites' displacement-time curves assembled from sequential telemetered binary images. These data yielded velocity results with sufficient accuracy and precision (cf. Table I) such that some definitive conclusions can be reached.

Table I lists for 11 supercoolings between 0.182 and 1.312 K dendrite velocity measurements observed on orbit, and their associated errors. The optical system that produced the binary images might have introduced an additional systematic error, which will be checked and, if needed, corrected postflight. Such an error, if it even exists, would be less than 5%, which is small enough not to alter our major conclusions. Figure 1 contains a plot of three sets of dendritic growth data: (1) IDGE velocities measured on Earth with the flight instrument with the same SCN sample that was flown; (2) the laboratory data set of Huang and Glicksman [7]; and (3) a subset of the on-orbit velocity data that we obtained from the binary images. The terrestrial dendritic growth velocity data include only dendrites with the [100] growth axis within $\pm 45^\circ$ of the local g vector. In addition, we show as the dotted curve a theoretical velocity calculation generated with the diffusion theory [Eq. (1)], combined with one fitting parameter σ^*.

Figure 1 shows a dramatic difference in the dendritic growth kinetics, particularly at the lower supercoolings, between terrestrial and microgravity conditions. Surprisingly, even at 1.572 K supercooling, where we anticipated only slight influences of convection on the dendritic growth process, there still appears to be more than an 8% reduction in the growth velocity under microgravity as compared to terrestrial conditions. Accepting the largest possible systematic and random errors likely to be in these data could increase the microgravity growth rate to as much as 5% higher than the terrestrial data set, or, perhaps, decrease it to as much as 19% lower. The reduction in growth velocity appears much more conclusively at 1.3 K, where the dendritic velocity measured in microgravity is 27% lower than the terrestrial data. Again,

TABLE I. Microgravity dendritic growth velocity measurements calculated from telemetered binary images from the space shuttle Columbia (STS-62).

$\Delta T \pm 0.002$ (K)	Velocity (microns/s)	Velocity Error ($\pm$microns/s)
0.182	3.4	0.3
0.202	4.01	0.04
0.232	4.62	0.08
0.292	8.44	0.07
0.372	14.73	0.04
0.472	22.4	0.7
0.612	44.5	0.5
0.782	84.3	1.5
1.012	172.1	4.9
1.312	320	14
1.572	534	44

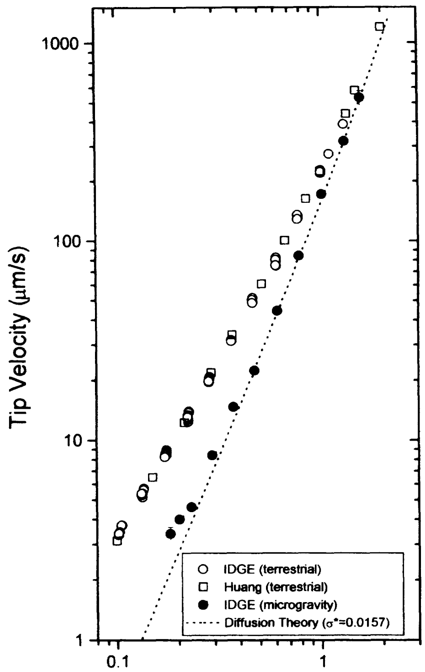

FIG. 1. Steady-state dendritic growth velocity versus supercooling. Dendritic growth velocities of succinonitrile observed in microgravity differ substantially from those measured terrestrially. The convergence of microgravity and terrestrial growth velocities occurs at a supercooling of 1.5 K or above. Moreover, the self-consistent scaling constant, σ^*_{sc}, inferred from the microgravity growth velocities, and represented by the dotted line, is approximately 20% lower than the value derived from terrestrial dendritic velocity and radii data.

accepting the largest possible errors, the measured velocity might range from 21% to 34% lower than that measured on Earth. Clearly, the results presented here show that it is incorrect to assess the validity of pure diffusion dendritic growth theories based on terrestrial dendritic growth measurements of SCN even at supercoolings as large as 1.3 K. Some theorists, unfortunately, have assumed that convective effects in SCN dendrites are unimportant at such elevated supercoolings, but indeed they are not. Furthermore, any calculation of a scaling constant σ^* based on terrestrially observed SCN dendritic growth data, even within the upper range of supercooling, would be in serious error.

At this time, lacking the 35 mm IDGE photonegatives still aboard Columbia, and the subsequent radii extraction measurements we plan to perform, it is not yet possible to calculate σ^* directly from the microgravity dendritic growth data. Rather, we define a self-consistent scaling constant

$$\sigma^*_{sc} = \frac{V_{\exp} d_0}{2\alpha \mathrm{Pe}^2}, \tag{6}$$

calculated from the theoretical Péclet number Pe, using pure diffusion theory [Eq. (1)], the definition of σ^* [Eq. (4)], and the measured velocity under microgravity or terrestrial conditions $V_{\exp}$. The self-consistent scaling yields a value of σ^* that, when combined with pure diffusion theory, produces the observed velocity in microgravity. The results of this calculation, as applied to the sets of the IDGE terrestrial and microgravity velocity data, appear plotted in Fig. 2. If diffusion theory with a scaling constant σ^*_{sc} were correct, then the data would plot as a horizontal line across the entire supercooling range. The results seem to approximate this situation, at least for the range of microgravity velocity data for supercoolings greater than 0.4 K. The average value of the self-consistent scaling constant over this supercooling range is $\sigma^*_{sc} = 0.0157 \pm 3\%$. One should note, as seen in Fig. 1, that using one adjustable parameter, viz., σ^*_{sc}, with the diffusion theory [Eq. (1)] yields velocity calculations in good agreement with the microgravity data over a range of supercoolings above 0.4 K.

The forced agreement here between the dendritic velocity data in microgravity and our calculations with a one-parameter fit merely provides a necessary but not sufficient condition for demonstrating the validity of pure diffusion theory combined with a selection rule of the form shown in Eq. (4). For complete and compelling evidence in support of this type of theory, the dendritic tip radii $R_{\exp}$ must also be consistent with the equation

$$R = \frac{d_0}{\sigma^* \mathrm{Pe}}, \tag{7}$$

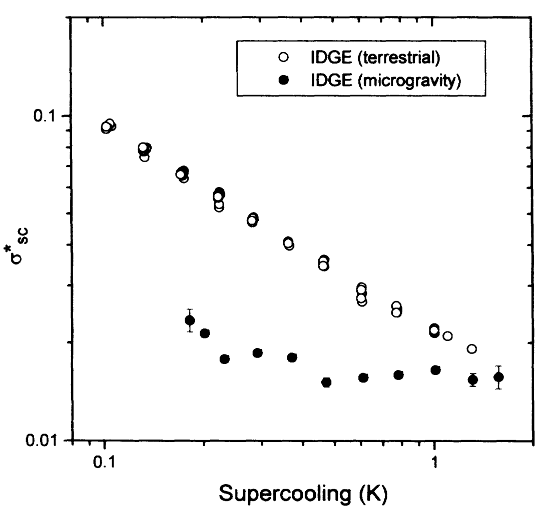

FIG. 2. Self-consistent scaling constant σ^*_{sc}, derived from dendritic growth velocities under microgravity conditions, versus supcooling. σ^*_{sc} is a construct developed to permit an estimate of the scaling constant using only velocities, and assuming the pure diffusion solution. σ^* is frequently assumed to be constant for pure diffusional dendritic growth, and σ^*_{sc} remains nearly constant at the higher supercoolings investigated. The steady increase of σ^*_{sc} observed with decreasing supercooling suggests the increasing importance of residual convective effects at low supercoolings, even in microgravity.

derived consequently from Eqs. (1) and (4), and where σ^* itself has been determined *independently* from the velocity data. Radii measurements will, of course, be reported at a later time, when the IDGE photomicrographs have been analyzed. We can, however, already infer from the observed agreement between the microgravity growth velocity data and the one-parameter fit to those data that the pure diffusion solution indeed provides the proper functional dependence on supercooling to describe dendritic velocity. Furthermore, the inferred value of $\sigma^* = 0.0157$ obtained in microgravity represents approximately a 20% reduction from the best terrestrial estimates ($\sigma^* = 0.0195$ with SCN as the test substance). Finally, we speculate that the growing deviation of the velocity data from the theory at supercoolings less than 0.4 K arises from the increasing importance of the differences from the state of zero gravity, which is required by diffusion theory, and the actual quasistatic acceleration environment achieved in orbit at the position of the IDGE growth chamber relative to the center of mass of the space shuttle.

We measured the dendritic growth velocity of succinonitrile in microgravity using the IDGE instrument mounted in the payload bay of the space shuttle Columbia (STS-62). Data telemetered to the ground were analyzed, and when compared to terrestrial dendritic growth data, demonstrate that (1) terrestrial convective effects remain significant in dendritic growth kinetics at supercoolings as large as 1.3 K, and, perhaps, above; (2) a pure diffusion solution to the dendrite problem combined with a scaling constant proportional to $1/VR^2$ is consistent with the observed dependence on supercooling of the growth velocities in microgravity; and (3) the tentatively revised, *self-consistent* value of the dendritic scaling constant under microgravity conditions in succinonitrile is $\sigma^* = 0.0157$.

Finally, we emphasize that these results could only be produced under precisely controlled, long-duration microgravity conditions of the type just achieved on USMP-2.

The requirements for designing, building, testing, and operating a complex, highly interactive scientific experiment aboard the space shuttle are beyond the skills and resources of any one organization. Diverse teams of dedicated professionals are required. Over the last ten years the following individuals contributed significantly to the success of this experiment: L. T. Bushnell, R. C. Hahn, B. Herbach, P. Kobryn, J. C. LaCombe, T. A. Lagrasso, E. R. Rubinstein, N. B. Singh, M. E. Selleck, S. H. Tirmizi, and A. Velosa, at Rensselaer Polytechnic Institute, Troy, NY; and R. Abramczyk, C. Blachly-Olah, D. Burns, A. Fedak, R. Grant, A. Kawecki, F. Kohl, G. Kraft, M. Lehto, L. Levinson, D. Malarik, J. McDade, D. Miller, A. Patel, D. Priebe, J. Robertson, A. Sobota, D. Vachon, G. Weeks, and R. Whittlesey, at or associated with NASA's Lewis Research Center, Cleveland, OH. Special thanks are due to the crew of Columbia, STS-62, and for the continued understanding, interest, and financial support provided by the National Aeronautics and Space Administration, Life and Microgravity Sciences and Application Division (Code U), Washington, DC, under Contract No. NAS3-25368, with liaison provided through the Microgravity Experiments Branch, Space Experiments Division, Lewis Research Center, Cleveland, OH.

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