

The Isothermal Dendritic Growth Experiment (IDGE)

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Introduction

Dendrites describe the tree-like crystal morphology commonly assumed in many material systems--particularly in metals and alloys that freeze from supercooled or supersaturated melts. There remains a high level of engineering interest in dendritic solidification because of the role of dendrites in the determination of cast alloy microstructures. Microstructure plays a key role in determining the physical properties of cast or welded products. In addition, dendritic solidification provides an example of non-equilibrium physics and one of the simplest non-trivial examples of dynamic pattern formation, where an amorphous melt, under simple starting conditions, evolves into a complex ramified microstructure [1].

Although it is well-known that dendritic growth is controlled by the transport of latent heat from the moving solid-melt interface as the dendrite advances into a supercooled melt, an accurate, and predictive model has not been developed. Current theories consider: 1) the transfer of heat or solute from the solid-liquid interface into the melt, and 2) the interfacial crystal growth and growth selection physics for the interface. However, the effects of gravity-induced convection on the transfer of heat from the interface prevent either element from being adequately tested solely under terrestrial conditions [2].

The Isothermal Dendritic Growth Experiment (IDGE) constituted a series of three NASA-supported microgravity experiments, all of which flew aboard the space shuttle, *Columbia*. This experimental space flight series was designed and operated to grow and record dendrite solidification in the absence of gravity-induced convective heat transfer,

and thereby produce a wealth of benchmark-quality data for testing solidification scaling laws [3,4].

The first flight of the IDGE flight, on STS-62, took place in March, 1994, on the second United States Microgravity Payload (USMP-2) [5], with a second flight on STS-75, in February/March, 1996, on USMP-3. Both flights used ultra-pure succinonitrile (SCN) as the test material. SCN is an organic crystal that forms dendrites similar to the BCC metals when it solidifies. Thus, SCN provides a nearly ideal physical model for ferrous metals. The third and final IDGE flight (USMP-4) launched on STS-87, in December and March of 1997, employed a different test material. This flight used pivalic acid (PVA)---an FCC organic crystal that solidifies like many non-ferrous metals. PVA, like SCN, has convenient properties for conducting benchmark experiments. However, unlike SCN, PVA exhibits a large anisotropy of its solid-melt interfacial energy, which is a key parameter in the selection of dendritic operating states.

USMP-2 (SCN)

The main conclusions drawn from comparing the on-orbit data to terrestrial dendritic growth data, obtained using the same apparatus and techniques, are that: 1) convective effects under terrestrial conditions cause growth speed increases up to a factor of 2 at the lower supercoolings ($\Delta T < 0.5$ K), and convection effects remain discernible under terrestrial conditions up to supercoolings as high as 1.7 K, far beyond what was thought. 2) In the supercooling range above 0.47 K, microgravity data remain virtually free of convective or chamber-wall effects, and may be used reliably for examining diffusion-limited dendritic growth theories. 3) The diffusion solution to the dendrite problem, combined with a unique scaling constant, σ^* , will not provide accurate prediction of the growth velocity and dendritic tip radii. 4) Growth Péclet numbers calculated from Ivantsov's solution deviate systematically from the IDGE data observed under diffusion-limited conditions. 5) The scaling parameter σ^* does not appear to be a constant, independent of supercooling. Finally, 6), the σ^* measurements from the terrestrial and microgravity data are in good agreement with each other, despite a difference of over six orders of magnitude in the quasi-static acceleration environment of low-earth orbit and terrestrial conditions [6,7].

USMP-3 (SCN)

The second IDGE flight on USMP-3/STS-75 mostly supported the above conclusions. However, at present at a still non-final stage of the analysis, there are some important modifications. With sufficient repeated observations [8], it now appears that the terrestrial and microgravity σ^* are distinguishable, with the microgravity σ^* larger than those measured under terrestrial conditions. However, even with the built up statistic of repeated experiment cycles, the functional dependence of σ^* with supercooling remains ambiguous. Some of the additional data supports the conclusion of USMP-2 that there is

a functional dependence on supercooling, while some of the additional data argues against such a dependence.

The second flight also clarified some issue at the lower supercoolings as to the role of convection, wall proximity or other explanations by showing definitively that it is not convection [9,10] and argued to what extent the low temperature effects are due to wall proximity effects [11]. Finally, the second flight yielded sufficient data to make a three-dimensional reconstruction of the non-parabolic, non-body-of-revolution dendritic tip shape [12,13].

Moving Heat Source Analysis

The method of moving heat sources is applied to the problem of dendritic growth in order to examine how the actual, non-paraboloidal shape of the dendrite tip and its trailing side branch structure affects the transport process. The model describes the diffusive thermal transport processes around a body of revolution (representing the dendrite) advancing into a quiescent melt at a constant rate. The latent heat produced at each of the points along an advancing solidification front is superposed to determine the net change in temperature at an arbitrary point in the surrounding melt. The calculation is performed by specifying the interface shape used in the model to be a shape-preserving body of revolution, which advances into the melt at a constant velocity. Once the shape and the growth velocity are specified, we solve the heat equation (formulated by the moving heat source method) to determine the supercooling experienced by the tip of the interface that is necessary to support the specified growth conditions.

Results of this work indicate that when corrections to Ivantsov's classical, infinite, parabolic tip shape are incorporated, enhanced agreement with experimental data is obtained, relative to the Ivantsov solution. This result is obtained by making modifications to the interface shape in a manner reflecting actual observed SCN dendrite tip shapes. When done, the predicted Pe vs. Δ (Δ is the dimensionless supercooling) relationship is shifted from the Ivantsov result. In general, the model indicates that for interface shapes that are wider than a parabola, Pe will be lower than Ivantsov's result for a given Δ . Conversely, an interface shape narrower than a parabola will raise Pe relative to Ivantsov's result.

The superposition of the latent heat sources (mentioned above) is accomplished by integration starting at the tip, and extending back to the interface areas behind the tip. When done in this manner, it is seen to be unnecessary to integrate back an infinite distance behind the tip. Instead, far from the tip, the contributions to the tip's required supercooling become small. This indicates that there is some "range of influence" which affects the heat transport at the tip of the dendrite, beyond which contributions become negligible. The size of this range is observed to be a strong function of the growth Peclet number, $Pe=VR/2\alpha$, where V is the growth rate, R is the radius of curvature of the tip, and α is the thermal diffusivity of the melt. The important question this raised was whether

or not this range would extend into the region of the dendrite where side branches were present, and the assumed interface shape was not valid.

The model was next applied to the case of the IDGE experiment's range of Peclét numbers and supercoolings. Using the upper end of the experimental Peclét range, $Pe \sim 0.01$, the integration range was limited to the region of the dendrite that is not dominated by the side arms (i.e. the tip region, within $12R$ of the tip). When this is done (Figure 1),

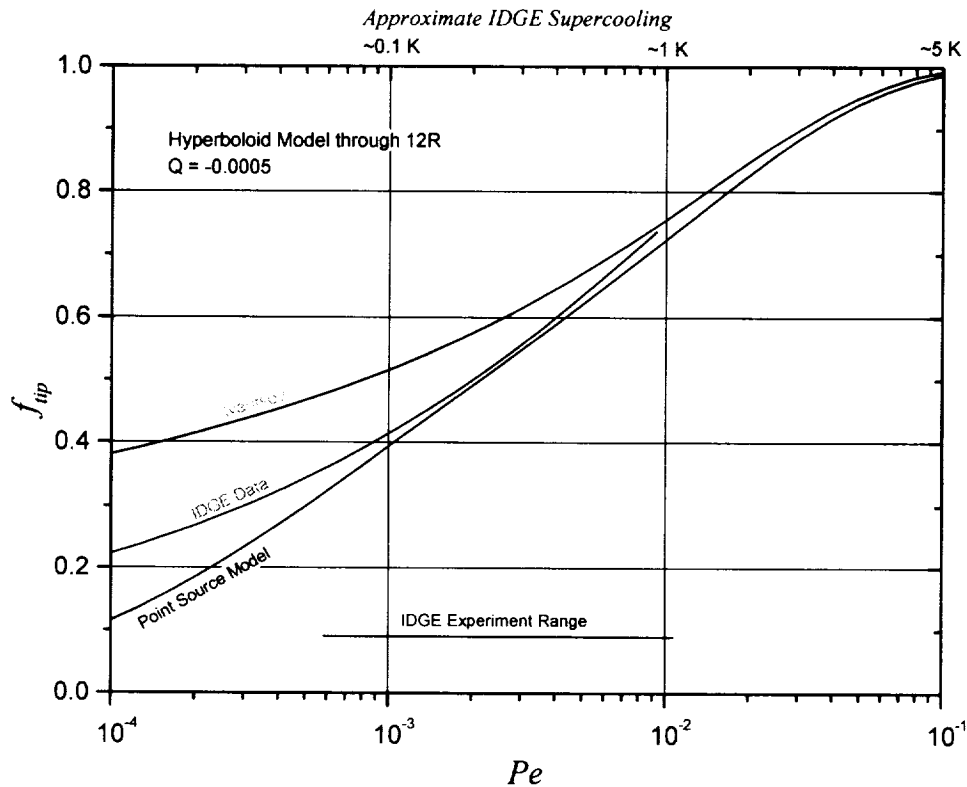


Figure 1. Fractional contribution to Pe from dendrite tip.

at most, only $\sim 75\%$ of the tip supercooling is accounted for. This indicates that at least 25% of the tip's supercooling derives from the side branch region of the dendrite—a region that is clearly not described by any simple function (and clearly, neither is Ivantsov's paraboloid). For the lower Peclét numbers seen in the IDGE experiments, the side branch region contributes up to $\sim 60\%$ of the tip supercooling. This observation joins earlier work by Schaefer [14] in suggesting that under the conditions of the IDGE experiment, the side branch region of a dendrite contributes significantly to the thermal conditions at the tip itself. Furthermore, since there is a considerable stochastic aspect to the side branch structure, it would follow that the scatter in the IDGE data may be Figure

2explained by the variations in the side branch structure, and the corresponding influence that this region has upon the transport processes underway at the tip.

USMP-4

The data and subsequent analysis from the final flight experiment are currently at a preliminary stage, based on images received using telemetry from space. We compared the dendritic growth speed of PVA as a function of the supercooling to both terrestrially measured PVA data, and an estimate scaled from prior SCN microgravity data. The preliminary results of these tests indicate that the PVA data are in good agreement with the SCN data (Figure 2). This implies that dendritic growth in PVA is, like SCN, diffusion-limited, with little, if any, kinetic response. This observation conflicts with the conclusion reached by other investigators that there are large interfacial kinetic effects in PVA. Currently we are extracting more accurate velocity and tip radius, shape, and side-branching measurements from post-flight 35mm film and videos.

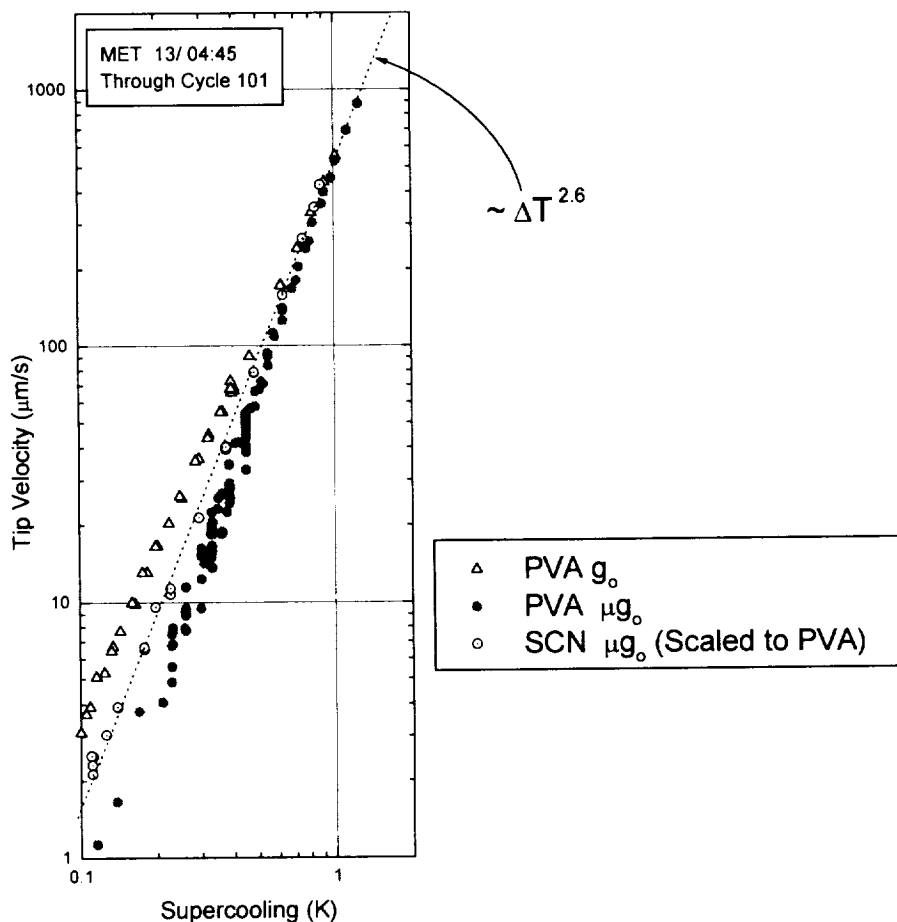


Figure 2. Velocity versus supercooling for PVA.

Figure 3 shows some preliminary data assessing the nature of these boundary layer interactions in PVA. We see that when the nearest neighbor distance exceeds about two thermal diffusion distances, λ , where the diffusion distance $\lambda = \alpha/v$ (α is the diffusivity and v is the tip speed) the velocity levels off at its maximum steady-state rate. When nearest neighbor spacings fall below about 2λ , the velocity is reduced through thermal interactions of the boundary layers. This phenomenon of neighbor interactions has never been observed before, because microgravity conditions are needed to insure growth limited by thermal diffusion from the solid-melt interface.

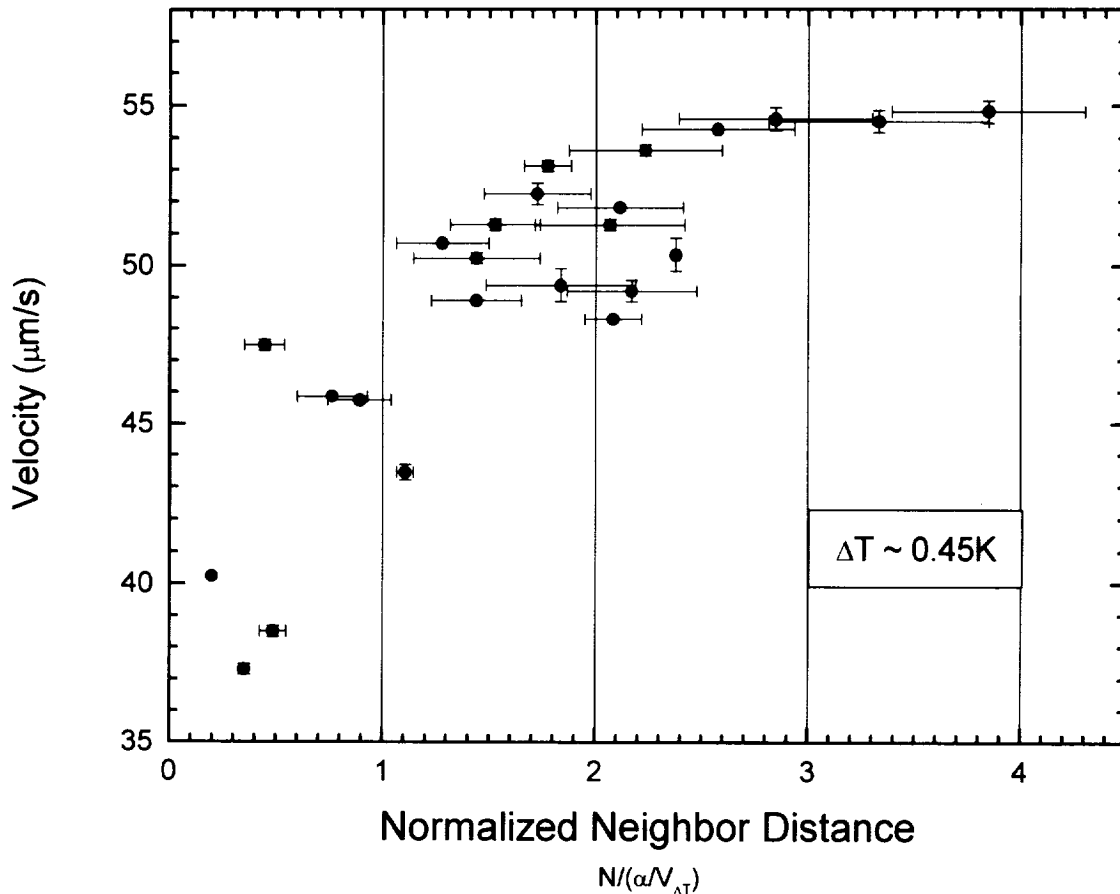


Figure 3. Dendrite velocity versus distance to nearest neighbor.

Telescience

In addition to our investigation of dendritic solidification kinetics and morphology, the IDGE has been part of the development of remote, university-based teleoperations. These teleoperation tests point the way to the future of microgravity science operations on the International Space Station (ISS). NASA headquarters and the Telescience Support Center (TSC) at LeRC, set a goal for developing the experience and expertise to set up

remote, non-NASA locations from which to control space station experiments. Recent IDGE space shuttle flights provide proof-of-concept and tests of remote space flight teleoperations [15].

Summary and Conclusions

The data and analysis performed on the dendritic growth speed and tip size in SCN demonstrates that although the theory yields predictions that are reasonably in agreement with experiment, there are significant discrepancies. However, some of these discrepancies can be explained by accurately describing the diffusion of heat. The key finding involves recognition that the actual three-dimensional shape of dendrites includes time-dependent side-branching and a tip region that is not a paraboloid of revolution. Thus, the role of heat transfer in dendritic growth is validated, with the caveat that a more realistic model of the dendrite than a paraboloid is needed to account for heat flow in an experimentally observed dendrite. We are currently conducting additional analysis to further confirm and demonstrate these conclusions.

The data and analyses for the growth selection physics remain much less definitive. From the first flight, the data indicated that the selection parameter, σ^* , is not exactly a constant, but exhibits a slight dependence on the supercooling. Additional data from the second flight are being examined to investigate the selection of a unique dendrite speed, tip size and shape.

The IDGE flight series is now complete. We are currently completing analyses and moving towards final data archiving. It is gratifying to see that the IDGE published results and archived data sets are being used actively by other scientists and engineers. In addition, we are also pleased to report that the techniques and IDGE hardware system that the authors developed with NASA, are being currently employed on both designated flight experiments, like EDSE, and on flight definition experiments, like TDSE.

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