Nonconstant Tip Velocity in Microgravity Dendritic Growth

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Pivalic acid dendrites grown and observed as part of the USMP-4 isothermal dendritic growth experiment aboard the space shuttle Columbia show an initial transient followed by an accelerating tip growth that may approach a constant growth rate. The mechanism responsible for this behavior is not as yet clear and may be due to experimental factors such as the long-range thermal interactions between the dendrite and its neighbors or container, or it may arise from a fundamental characteristic of isothermal, diffusion-limited, dendritic growth.

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Dendritic microstructures are commonly formed during the solidification of most metals and alloys. It is important to understand the process by which dendrites form because traces of these geometrically complex microstructures persist through subsequent material processing stages, and can affect the properties of the finished product.

Over the past five decades, the scientific community has produced a large body of experimental and theoretical work describing dendritic growth. Glicksman and Marsh [1] discuss this research in their 1993 review article, and Bisang and Bilgram [2] include a detailed review as part of their 1996 article on dendritic growth. As these reviews and others make clear, the dendritic growth rate, or the speed of the advancing tip, is a critical characteristic of dendritic growth. In 1947, Ivantsov [3] introduced two simplifying assumptions in the theory of thermal dendrites growing in a supercooled melt: (1) that the dendrite could be represented as a shape-preserving paraboloidal interface with a tip radius R; (2) that the tip grew at a constant rate V. The notion that real dendrites (as opposed to paraboloidal needle crystals) also grow in a steady-state manner, with constant velocity, is generally considered to be supported by numerous experimental observations of isolated dendrites (see Huang, for example, [4]). Most experimental and simulation studies of dendritic growth attempt to extract a constant velocity measurement as a parametrization of the kinetics, whereas most theoretical studies assume constant velocity behavior.

Recently, our attention was turned towards understanding the interactions between a dendrite and its surroundings, and quantifying what is required for a dendrite to grow in a truly "isolated" manner. Thermal interactions between a dendrite and neighboring tips, container walls [5], or even its own initial structure [6] and trailing side branches [7,8], are considered to be potential sources for an observed velocity that does not match the velocity predicted for an isolated diffusion-limited dendrite with the thermal boundary conditions set at infinity. If the strength of these thermal interactions changes over time, one would expect the velocity of the dendrite to change as well, and one should further expect to see deviations from the conventionally prescribed behavior predicted by an Ivantsov-like solution for an isolated dendrite.

At low supercoolings, the length scale over which the thermal field extends is known to be large, compared to the morphological length scales relevant to the dendritic growth process. With such long thermal diffusion lengths, proximate dendrite arms can mutually interact to generate an operating state that differs from the steady state. For example, Almgren, Dai, and Hakim [9] investigated a similar process in the case of Hele-Shaw flow, and concluded that early in the growth the dendrite tip position advances as $t^{3/5}$ under constant flux conditions. Recent computations from phase-field simulations [10] suggest the existence of an early-time transient in two-dimensional dendritic growth prior to approaching a steady-state growth rate.

The data and conclusions presented here result from two related analyses of dendritic growth data obtained from the isothermal dendritic growth experiment (IDGE). These experiments compiled data on the growth of pivalic acid (PVA) dendrites in microgravity. A more detailed description of the IDGE experiments can be found elsewhere [11–13]. The PVA sample material was contained within a quartz growth chamber located within a temperature-controlled bath. The growth chamber interior volume measured approximately 31 mm square by 50 mm long. Nucleation was achieved through the use of a hollow stinger tube that penetrated the wall of the growth chamber. The exterior end of the stinger tube was closed and surrounded by a thermoelectric cooler. The interior end was open, allowing the PVA sample material in the chamber to also fill the stinger. Each dendritic growth cycle began by completely melting the sample, followed by lowering the melt temperature to the desired supercooling. After the supercooled melt's temperature reached steady state, the thermoelectric cooler was activated to nucleate the crystal, which then propagated down the stinger tube to emerge into the chamber as a dendrite. Once completed, a new growth cycle was initiated by remelting the sample and proceeding as described above. This arrangement, combined with the microgravity conditions, produced dendritic crystals grown under diffusion-limited

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conditions, with the bath temperature controlled to within 0.002 K (spatially and temporally).

During the growth cycles, once a crystal emerged from the stinger, images of the growth were obtained from two perpendicular views using electronic and film cameras. These images constituted the primary data source for the IDGE experiments. The results presented here derive from electronic video cameras. The preliminary analyses were conducted using a lower spatial and temporal resolution data set that was available during the actual flight experiment. This preliminary analysis was performed on dendrites grown at six different supercoolings between 0.13 and 1.25 K. After these data were analyzed, a second and more detailed analysis was performed on one of the same microgravity dendritic growths. The difference was that superior spatially and temporally resolved video (640×480 pixels at 256 gray scale and $\sim$30 frames per sec) became available postflight, providing considerable improvement in measurement resolution. The data presented here show that dendritic growth rates are not constant over the time scale of observation. Specifically, the dendrite tip, after completing its initial transient, continues to experience a small and ever diminishing acceleration. The tip might or might not approach a truly constant velocity.

These results derive from a dendritic growth cycle at a supercooling of 0.41 K. They are qualitatively representative of the entire body of analysis conducted to date. The data consist of measurements of the dendrite tip position measured from video images as a function of time (Fig. 1). Dendrite tip locations were obtained through the use of a subpixel interpolation scheme applied during image processing. Uncertainty in the measured tip positions, at present, is approximately 2 μm, which is considerably less than the pixel size of $\sim$20 μm. For the single growth presented here, there are approximately 240 sec of data, obtained from approximately 7000 image frames. This cycle shows a typical displacement versus time plot of a microgravity-grown dendrite. The initial transient, clearly accelerating, evolves toward a regime from which one can extract the velocity.

Despite the relative linearity of the latter part of the displacement plot, there is a residual curvature. Hence, some acceleration remains here too, although not as much as within the initial transient. We least-square fitted a constant velocity line to the displacement versus time data from the portion of the growth between the hash marks of Fig. 1 (after $\sim$130 sec). Over this range of time, the growth *appears* to be constant. Despite the apparent constant velocity behavior, closer examination of the residuals resulting from the linear regression reveals a systematic deviation from steady-state growth (Fig. 2). A dendrite growing at constant velocity would be expected to exhibit random residuals arising from the uncertainties in the tip displacement measurements. These residuals form a Gaussian distribution centered about zero. Instead, as shown, a distinct nonrandom residual occurs, with monotonic upward curvature in the graph. Additionally, the regression residuals yield information that is useful in determining the uncertainty in the measurements, because their spread is a good measure of the uncertainty resulting from the subpixel interpolation measurement of the tip location. Thus the $\sim$2 μm standard deviation of the residuals is considered to be the limiting resolution of our current image processing and analysis techniques.

A convenient measure of the asymptotic behavior of the tip speed, as evidenced by the nonrandom residual, is the growth rate exponent κ calculated by comparing the data to a power law for the displacement proportional to t^{κ}. An exponent of $\kappa = 1$ indicates linear displacement in time (i.e., constant velocity). Although at first this method seems less intuitive than simply comparing the instantaneous velocity versus time, the slope of which is the acceleration, the power law helps elucidate the variations from, and approach to, constant velocity ($\kappa = 1$) behavior. The

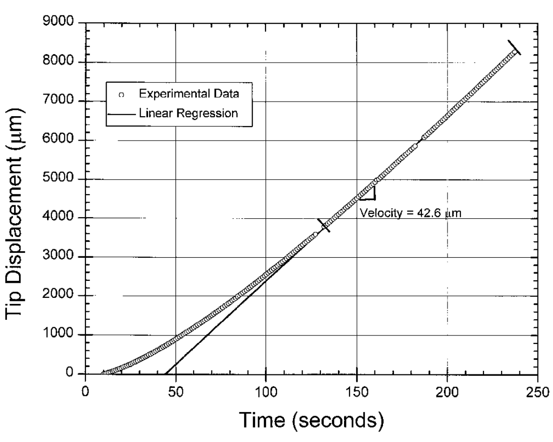

FIG. 1. Tip displacement vs time for USMP-4 cycle 4, grown at 0.41 K supercooling. Linear regression performed using data from the more steady-state portion of the growth (after $\sim$130 sec). The growth rate results from the slope generated by the regression. Note: 1 of every 30 data points plotted.

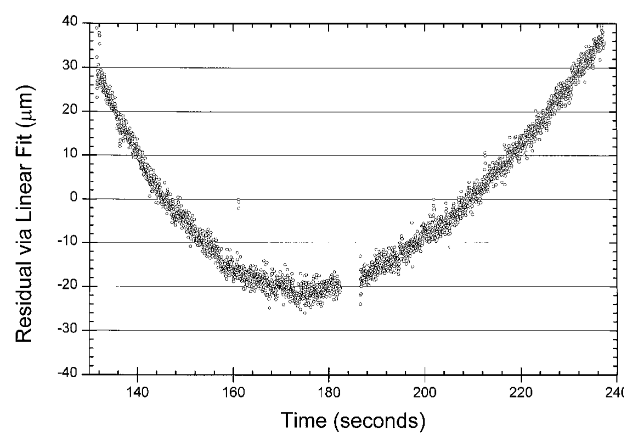

FIG. 2. Residuals resulting from a linear fit to the tip displacement vs time data in Fig. 1 for the latter growth stage (i.e., after $\sim$130 sec). The spread in the residuals is $\sim$2 μm, representing the uncertainty in the tip-position measurements.

measured values of κ (Fig. 3) calculated after first smoothing the displacement data using a two-second moving average reveal that this USMP-4 PVA dendrite was accelerating throughout the microgravity experiments at a rate that was slowly diminishing. This is evidenced by κ approaching unity from above. In addition to the data presented in these figures, the measurements and analysis performed at other supercoolings (the lower resolution *in situ* data) revealed qualitatively similar behavior of κ, although the details of the early transient varied from growth to growth, which will be discussed later.

The non-steady-state velocities observed during these experiments suggest that there appear to be features of this process that are not well described by steady-state dendritic growth theory. Alternatively, within experimental practice, the establishment of *strict* isothermal conditions at the dendritic interface may be illusory. We will discuss briefly several potential sources for non-steady-state behavior. It is beneficial to discuss the time-dependent process as occurring in two distinct periods. The first of these is the initial transient, which is most evident in Fig. 3 prior to approximately 60 sec. The second distinct time-dependent period is beyond ~60 sec, where a more gradually decreasing growth rate exponent is observed.

A plausible explanation for the presence of an initial transient in the displacement data is that during the early phase of growth, the dendrite evolves from its own predendritic morphological structure. The evolving crystal pattern needs time to develop a thermal field that is commensurate with steady-state growth. Ivantsov demonstrated theoretically that isotherms arranged as confocal paraboloids satisfy the requirement for constant-velocity growth. It is proffered that to develop such a steady field from the initially uniform and isothermal melt takes time on the order of what was observed experimentally (~60 sec). A subsequent corollary would be that any dendrite, whose physical size (root to tip) is less than a few thermal lengths has not reached steady state, because it has yet to fully occupy its own thermal domain.

In the nascent stage of a dendrite's development are potential interactions with neighboring dendrites. To investigate this effect, the IDGE experiment [13] obtained data from microgravity where tip velocities for a large number of dendrites grown at an identical supercooling were compared and plotted as a function of the distance to the nearest neighboring dendrite tip. This distance was normalized with respect to the thermal length, which is defined here as the thermal diffusivity of the liquid (7×10^4 μm^2/s) divided by the growth rate (42.6 μm/s). Thus, at 0.41 K supercooling, the characteristic thermal diffusion length for isolated PVA dendrites is ~1640 μm. Note that this measure is dependent upon the growth rate, which is shown here to be non-steady-state, particularly early in the growth. Figure 4 shows that dendrites grown at 0.45 K supercooling, with the closest neighbor farther than approximately three thermal lengths, reach a velocity of ~55 μm/s. Dendrites with neighbors closer than three thermal diffusion lengths have systematically smaller growth rates. This is consistent with the notion that each dendrite is normally surrounded by a thermal boundary layer of a few diffusion lengths thickness. When the dendrites are "close enough" together (three diffusion lengths, for example), their thermal fields begin to overlap, decreasing the temperature gradient (relative to an isolated dendrite), and thus the growth rate as well. This can be visualized most easily with the situation of an equiaxed dendrite. The perpendicular primary arms of an equiaxed dendrite all begin growth close to one another and proceed to grow away from each other. Thus, as their separation increases over time, the degree of thermal interaction diminishes, producing an expected acceleration early in the growth. Such interactions also exist for proximate dendrites growing in the same direction. Eventually, if the interactions are strong enough, one of the dendrites will experience favorable thermal conditions and will "pull away" from the slower dendrite. Tracked over time, such an interaction could account for the observed initial transient phase, prior to ~60 sec. By this time, the dendrite was

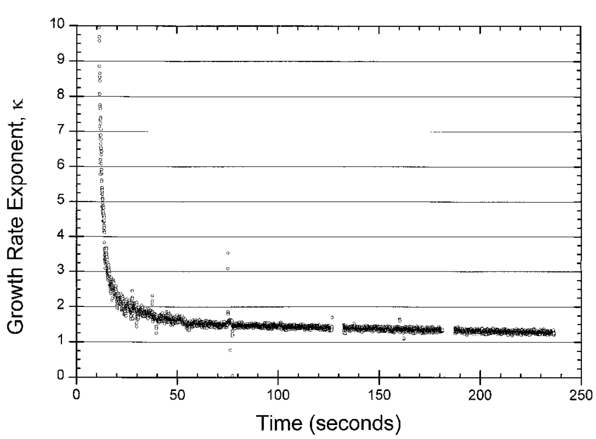

FIG. 3. Growth rate exponent κ as a function of time for the dendrite data in Figs. 1 and 2. A value of $\kappa = 1$ corresponds to steady-state, constant velocity growth.

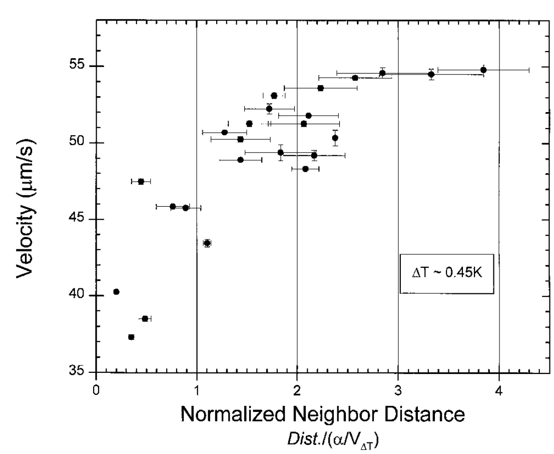

FIG. 4. Dendrite velocity vs distance to nearest neighbor, normalized using the thermal diffusivity α and the nominal growth rate (~55 μm/s) at 0.45 K supercooling, $V_{\Delta T}$.

~0.45 thermal lengths from its closest neighbor, and ~0.8 thermal lengths in overall size. Once the tip "escapes" from this period of strong interactions it is more able to grow as an isolated dendrite, at a more constant velocity.

The experimental (and simulated) observation of an initial transient, preceding steady-state growth, is not new. In fact, it has been generally assumed that this interaction, although easily distinguishable from the steady state that emerges, is less germane and fundamental to the kinetics than the steady state itself. However, as is revealed here, the growth phase following the initial transient might not ever reach a constant velocity. Thus, the more important issue for such investigations is whether the time-dependent state is fundamental to the physics of isothermal dendritic growth, or appears as an artifact of experiments carried out in finite volumes of supercooled melts.

The data showing the effect of neighboring dendrites on growth velocity (Fig. 4) suggest possible long-range thermal interactions with dendrite arms growing in the same general direction as the tip in question. Additionally, these interactions may contribute to a second stage of time-dependent growth seen after the initial transient is completed. However, many of the analyzed dendrites were isolated growths (noting that "isolation" is a strong function of time and supercooling), where the closest neighbor was beyond three thermal lengths. Thus for neighbor interactions to be the mechanism responsible for the nonconstant velocity at the end of the growth cycle, one must firmly establish that the thermal interactions between dendrites separated by more than three thermal lengths are still enough to produce the measured changes in growth velocity.

An additional experimental artifact that may be involved is the finite size of the growth chamber. Pines, Chait, and Zlatkowski [5] showed that a dendrite may interact with the walls of a container as growth proceeds. However, the growths examined here have small thermal lengths compared to the distance between the dendrite tip and the growth chamber wall. Here also, one must investigate whether the closing rate between the dendrite and the wall is commensurate with the measured time variations in velocity. Given these considerations, it is our hypothesis that it is more likely that the second stage of non-steady-state behavior is fundamental to isothermal dendritic growth. Since dendrites are not truly parabolic bodies of revolution [4,14], there is no compelling phenomenological reason that dendrites *should* grow strictly at a constant rate. Insofar as the data set from which these observations and conclusions were drawn is the only one we know of that is both diffusion controlled and measured at the necessary temporal resolution to make the described measurements, we plan additional work in data reduction, analysis, and modeling to explore this hypothesis further.

In summary, a method was developed for evaluating dendritic growth rates that discriminates fine distinctions in the non-steady-state behavior. This method is applied here to PVA dendrites, seen to grow in microgravity at non-steady-state velocities. The mechanism responsible for this behavior may be related to thermal interactions between a dendrite and its surroundings, or it may be intrinsic to the dendritic solidification process. Efforts to further discriminate among the possible causes are currently under way.

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