# REVIEW OF THE INFLUENCE OF CONVECTION ON THE MICROSTRUCTURE OF FIBROUS EUTECTICS

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We review experimental research on the influence of gravity on rod eutectic microstructure, including solidification in microgravity and in the centrifuge, as of 1993. Solidification has also been performed with mechanical stirring and while using a magnetic field to reduce convection. Most experiments were performed on the MnBi-Bi eutectic. The rod spacing depended on the growth conditions, with several contradictions between results of different investigators. Proposed mechanisms are discussed and their predictions compared with the experimental results. Vigorous mechanical stirring appears to coarsen the microstructure by altering the concentration field in front of the freezing interface. Gentle convection is believed to alter the microstructure of a fibrous eutectic only when it causes a fluctuating freezing rate with a system for which the kinetics of fiber branching differs from that for fiber termination. These fluctuations may cause the microstructure to coarsen or to become finer, depending on the relative kinetics of these processes.

## 1. INTRODUCTION

During cooperative eutectic solidification, two phases solidify side by side. The growth is coupled by the diffusion field in front of the growth interface. Component B is rejected by the growing $\alpha$ phase, while A is rejected by $\beta$. Consequently A and B must diffuse laterally to the growth interface. Connected to this segregation and lateral diffusion is a concentration field in which the local melt composition deviates from the eutectic. The distance into the melt over which this deviation extends is on the order of the interphase spacing $\lambda$ [1]. Since $\lambda$ is small, on the order of a few $\mu$m, one would not expect gentle convection to influence either the concentration field or $\lambda$. Thus it was surprising in 1976 when Larson reported from his Apollo-Soyuz Test Project experiment that directional solidification in microgravity caused a significant reduction in the fiber spacing of the MnBi-Bi eutectic $\lambda$ [2]. Indeed the first reaction to this result was disbelief, that it was in error, perhaps because of a large difference in freezing rate between earth and microgravity. However, subsequent careful experiments showed that the effect was real and reproducible [3-13].

Since Larson's ASTP experiment on Mn-Bi, a large number of experimental and theoretical studies have been performed to try to understand the influence of convection on eutectic microstructure. With the completion of experiments here on the influence of electric current pulses [14], we now believe we have this understanding. We begin with a summary of the experimental results. Then we compare these results with the predictions of proposed mechanisms.

## 2. EXPERIMENTAL RESULTS

Our basis of comparison here is the microstructure of fibrous eutectics directionally solidified upward on earth at 1 g, i.e. with the melt above the solid. The following rod-forming systems are considered: MnBi-Bi, InSb-NiSb and $Al_3Ni$-Al. The freezing rates reported here yielded rods or fibers of the minority phase. For MnBi-Bi, fibers form above about 1 cm/hr [4,15-17]. As freezing rate is increased, the morphology of the MnBi changes from irregular faceted to triangular to circular cross sections, with increasing regularity in fiber arrangement and decreased scatter in fiber spacing. In agreement with theory [e.g.,1], the average fiber spacing $\lambda$ is inversely proportional to the square root of the freezing rate V, i.e. $\lambda^2 V$ is constant [refs 4,5,15-20 for MnBi; 21,22 for InSb-NiSb; 18,23,24 for $Al_3Ni$-Al]. Similarly as predicted by theory [1], for the MnBi eutectic $\Delta T$ was proportional to V, where $\Delta T$ is the interfacial undercooling and V is the freezing rate [10]. The majority of the research on the influence of convection has been performed on the Mn-Bi system, for which the eutectic composition is $0.72 \pm 0.03$ wt% Mn, or $3.18 \pm 0.09$ vol% MnBi [3,4,25].

The microstructure of eutectics is normally characterized by examination of longitudinal and cross sectional slices. Some authors use a computer algorithm to automatically measure the distance between fibers on the cross sectional slices and take an average to obtain $\lambda$ [e.g. 3-14,16,17,26-29]. Other authors count the number of fibers per unit area and assume $\lambda$ is inversely proportional to the square root of this fiber density [21,30]. We have used both techniques at Clarkson, with no apparent influence on trends.

With fibrous eutectics, little is learned about fiber morphology from longitudinal slices because these intersect only some fibers for a limited distance. The fibers are not perfectly aligned with the plane of the section. Consequently one cannot see fiber orientations, variation in cross section, branching or termination. In order to view these characteristics it is necessary to remove the matrix and expose the fibers. Although this has been successful with some eutectics, no one has yet succeeded in finding a chemical etchant or other treatment that would remove Bi without attacking MnBi. In fact, most etchants preferentially attack MnBi. Chandrasekhar [31] succeeded in exposing MnBi fibers by using a different approach. Eutectic rods were pulled apart mechanically while they were heated by passing a large electric current down them, using a Gleeble. Melting occurred as the rods broke. Typical fracture surfaces are shown in [31,69]. Several conclusions could be drawn:

1. A variety of fiber cross sections occurred. These tended to be faceted at lower freezing rates, and rounded at higher rates.
2. The fibers were not well aligned with one another.
3. The fiber spacing and arrangement were irregular, one might say even random, especially at lower freezing rates.
4. The surfaces of the fibers were generally smooth.
5. There was little evidence of branching.

Chandrasekhar also decanted MnBi-Bi eutectic interfaces during solidification [31]. It was concluded that all of the fibers projected out in front of the interface. Although the distance of projection was usually about 1 diameter at all freezing rates, a few projected out much larger distances.

### *2.1. Microgravity solidification*

Larson and Pirich at Grumman Corporation used NASA's Advanced Directional Solidification System (ADSS) furnace to directionally solidify the MnBi-Bi eutectic on sounding rockets, in the

Shuttle, and on earth under a variety of conditions [3-13,15,26-29]. A 14 cm long heater was used with booster heaters at both ends and a water-cooled copper block at the solidification end. This arrangement produced gradient regions over about 3 cm at both ends and a relatively constant temperature in between. The ampoule was translated through the furnace. The inside diameter of the ampoules was 4 mm. The temperature gradient in the melt at the interface was about $100^{o}C/cm$. Although there were large erratic fluctuations in the local value of the fiber spacing $\lambda$, there were no systematic variations down the ingot or in the cross sectional slices. Temperature measurements inside the ampoules showed that the heater temperature and the axial temperature gradient in the melt were slightly higher in microgravity [7], but the freezing rate was unaltered. The table below summarizes the change in $\lambda$, area %MnBi, and interfacial undercooling caused by solidification in microgravity as compared to solidification on earth with the melt above the solid. It is seen that reducing buoyancy-driven convection in low gravity caused $\lambda$ to decrease, the volume fraction of MnBi to decrease, and the interfacial undercooling to increase (lower interfacial temperature).

**Table 1.** Changes compared to growth upward on earth

| Flight | Freezing rate | Average $\lambda$ | Fraction MnBi | Undercooling | References |
|---|---|---|---|---|---|
| SPAR VI | 30 cm/hr | 35 ± 12% lower | | 7.4% less | 3,6,11 |
| SPAR IX | 49 cm/hr | 47% lower | 8% less | $5.5^{o}C$ larger | 3,7,11 |
| STS-26 | 3 cm/hr | 40% lower | | | 10 |

Smith and Kaya [32] also solidified the MnBi-Bi eutectic in microgravity. The ampoule was translated through a furnace with a relatively constant temperature gradient, producing a value of $\lambda$ ranging from 5 to 8 μm. Contrary to the above results of Larson and Pirich, no change in $\lambda$ was detected between growth in microgravity and solidification upward on earth.

Müller and Kyr [21,22,33,34] directionally solidified the InSb-NiSb eutectic in the TEXUS-10 rocket and in the Shuttle on the Spacelab-1 and D-1 missions. A single ellipsoidal mirror furnace was used on the ground, in D-1 and with TEXUS-10, while a gradient furnace was used on Spacelab-1 and on the ground. The current interface demarcation technique was used to measure freezing rates from 0.6 to 10.8 cm/hr. The value of $\lambda$ was reduced 14% by solidification in microgravity, independent of freezing rate.

Favier and de Goer [18] directionally solidified the $Al_3Ni$-Al eutectic in a TEXUS rocket from 7.9 to 8.4 cm/hr, as estimated from thermocouple readings in the cartridge containing the growth ampoule. The cartridge temperature profile was not changed in low gravity. The value of $\lambda$ was about 15% larger from the microgravity experiments.

## *2.2. Influence of magnetic field*

The MnBi-Bi eutectic was solidified upward in the presence of a 3 kG transverse magnetic field or an 80 kG vertical field using the ADSS furnace described above [10,27-29]. Such a magnetic field should strongly decrease the buoyancy-driven convection, similar to microgravity. The values of $\lambda$ at freezing rates of 30 and 50 cm/hr were the same as given above for the SPAR VI and IX experiments, i.e. reduced from the values without a magnetic field. The interface undercooling was also increased similar to the results in microgravity. The results for a magnetic field down to V = 0.55 cm/hr fell on a $\lambda^2V$ = constant line, which was parallel to and lay below the line for solidification upward without a magnetic field applied. Thus reduction of buoyancy-driven convection by use of microgravity or a magnetic field reduced $\lambda$ by the same amount independent of V.

### *2.3. Influence of temperature gradient*

Experiments at Clarkson and at Grumman showed no measurable influence of temperature gradient on $\lambda$ of MnBi at freezing rates from 3 to 30 cm/hr [5,15,19,20].

### *2.4. Influence of solidification direction*

The ADSS furnace described above also was used to solidify the MnBi-Bi eutectic horizontally and downward (melt below solid) [3,5,15]. These arrangements would be expected to produce significantly more convection than the usual upward solidification. At a freezing rate of 30 cm/hr no change in $\lambda$ was observed, while at 3 cm/hr $\lambda$ was about 67% larger when solidification was downward. Horizontal solidification also seemed to give a slightly larger $\lambda$ at 3 cm/hr.

Smith and Kaya [32] also investigated the influence of ampoule orientation on MnBi using their gradient furnace. With growth down, temperature fluctuations occurred in the melt and severe banding was produced, making any conclusions impossible.

The $\lambda$ for the InSb-NiSb eutectic was increased 9% by solidification downward as compared to solidification upward, and independent of freezing rate [21,22].

The $\lambda$ for $Al_3Ni$-Al was decreased by horizontal solidification as compared to solidification upward, independent of freezing rate [23,24].

### *2.5. Solidification in a centrifuge*

Solidification of InSb-NiSb downward in a centrifuge at 5 to 30 times earth's gravity g caused $\lambda$ to increase 27% compared to solidification downward at 1g, or 38% compared to solidification upward at 1g [21,22]. Centrifugation in this thermally unstable configuration should produce vigorous convection.

### *2.6. Solidification with ACRT*

Eisa [16,17] studied the influence of accelerated crucible rotation (spin-up/spin-down) on the microstructure of the MnBi-Bi eutectic using a vertical Bridgman-Stockbarger furnace. The period of turning on and off the ampoule rotation was varied from 9.6 to 13.6 s. The results were successfully correlated by:

$$\lambda V^{0.5} = 6.26 + 0.000112 (R N^{1.5} / V)^{1.1}$$

where $\lambda$ is in $\mu$m, R is radial position in mm, and N is the rotation rate in RPM.

### 2.7. *Influence of electric current pulses*

Passage of electric current through a solidifying ingot perturbs the freezing rate and can even cause meltback. The Peltier effect causes heat to be liberated or consumed at the freezing interface. The Thomson effect either liberates or consumes heat in the bulk material wherever a temperature gradient is present. The Joule effect liberates heat in the bulk material. When current is turned on, the growth rate is instantly either retarded or increased. Subsequently, the growth rate moves back toward its pre-pulse value. When the current is turned off, the freezing rate instantly changes in the opposite direction.

Single current pulses ranging from 40 to 160 amp/cm$^2$ were passed for 5 to 10s through solidifying MnBi-Bi eutectic in the ADSS described above [26]. Temperature measurements were made in the material during these pulses. For 10s or more of increased growth rate, breakdown of cooperative growth occurred and banding was produced. Some bands were free of MnBi, whereas MnBi was enriched in others.

## 3. THEORY AND DISCUSSION

Several mechanisms have been proposed to explain the influence of convection on eutectic microstructure. We will examine these and compare their predictions with the previous experimental results.

### 3.1. *Influence of convection on the concentration field in the melt during solidification of eutectics*

The starting point for theoretical treatments of eutectic solidification is the classic paper of Jackson and Hunt [1]. They considered the steady state solidification of both lamellar and rod eutectics from an infinitely large, convection-free melt. The total interfacial undercooling $\Delta T$ was taken to be the sum of the undercooling due to curvature and that due to the deviation of the interfacial concentration from the eutectic. In order to estimate the concentration undercooling, the differential equation for diffusion in the melt was solved for a planar interface. It was implicitly assumed that the volumetric properties of all three phases and both constituents are the same. The resulting average concentration undercooling was proportional to the freezing rate V and the lamellar or fiber spacing $\lambda$. By considering the non-planarity of the solid-liquid interface, the average curvature undercooling was estimated to be inversely proportional to $\lambda$. Thus the total $\Delta T$ was a function of $\lambda$ and V. By minimizing $\Delta T$ at constant V, both $\lambda^2 V$ and $\Delta T^2/V$ were predicted to be constant and independent of V. The same result is obtained by maximizing V at constant $\Delta T$. The validity of this extremum assumption was discussed in many subsequent papers.

All of the theoretical work to assess the influence of convection on eutectic microstructure has been aimed at the concentration undercooling term in the Jackson-Hunt treatment. The change in the concentration field was calculated and used to determine the change in average concentration undercooling along the freezing interface.

Verhoeven and Homer [35] made the first attempt to estimate the influence of convection on eutectic lamellar microstructure. A stagnant film model was used. Jackson and Hunt assumed that the melt composition is fixed at an infinite distance from the freezing interface. In Verhoeven-Homer the concentration was fixed at the bulk melt value at distance $\delta$ from the freezing interface. It was concluded that the usual levels of convection utilized in solidification should have no influence on the microstructure of lamellar eutectics. (Although rod eutectics were not addressed, the conclusion would be the same from this model.) On the other hand, the equations show that if convection does influence $\lambda$, the change in $\lambda$ would increase as $\lambda$ increases, i.e. for small V. This predicted trend does not agree with the low g experiments that yielded a change in $\lambda$ that was independent of V. It does agree qualitatively with the ACRT results of Eisa [16,17].

There are fundamental problems with the stagnant film model, which, in the crystal growth literature [e.g.,36,37], is often confused with true boundary layer models. In the stagnant film model, it is assumed that there is no fluid motion inside a thin film of thickness $\lambda$. Outside this film the fluid is taken to be completely mixed, i.e. of uniform composition. In reality, the fluid motion only approaches zero as one approaches the freezing interface. The stagnant film model does not correspond to reality. The value of $\lambda$ must be obtained from experiment or a theoretical computation based on the differential equations of motion and convective transport. In other words, $\lambda$ is defined as the thickness that gives the correct answer! It is not known *a priori*. Furthermore the model predicts that the mass transport rate in various operations is proportional to the diffusion coefficient D, while experiment and exact theory gives a fractional power dependence on D. It is true that the stagnant film model has been reasonably successful at correlating the influence of freezing rate on macroscopic segregation via the Burton-Prim-Slichter equation. However, its applicability to other situations cannot be assumed and must be confirmed by experiment or exact theory for each situation.

We set out several years ago to develop models more soundly based on modern transport phenomena. We noted from the Jackson-Hunt results that, for eutectic melts, the region of perturbed concentration extends only a short distance into the melt, on the order of $\lambda$, which is only a few $\mu$m. Thus we needed to consider the velocity field only near the interface. In this region the fluid flows parallel to the interface at a velocity proportional to the distance from the interface. That is, the velocity gradient at the interface becomes the parameter characterizing the intensity of the convection. This velocity gradient can be calculated by solving the equations of motion for the melt as a whole, as has been done frequently by numerical techniques in recent years.

Thus we used numerical calculations to determine the concentration field in the melt near a freezing interface at steady state. The results were used to calculate the average deviation from the eutectic composition along the interface. Substitution of this in the Jackson-Hunt model allowed us to determine the change in $\lambda$ caused by convection. We did this for lamellar eutectics with a planar interface [17,38-41,57], fibrous eutectics with a planar interface [42], lamellar eutectics with one phase projecting out into the melt [43,44], and with the Soret effect included [45]. Although we predicted changes in $\lambda$ when the stagnant film model of Verhoeven-Homer said there should not be, the changes were much smaller than those observed experimentally by solidification in microgravity or using a magnetic field. Other predictions not in agreement with experiment are:

1. Decreased convection is predicted always to cause $\lambda$ to decrease. Experimentally, an increase was observed in the $Al_3Ni$-Al system when solidification was performed in microgravity [18].
2. Convection is predicted to influence lamellar eutectics only slightly less than fibrous eutectics. Experimentally, the $\lambda$ of lamellar eutectics was not influenced by microgravity [18,68] or by ACRT [46-48]. Only very vigorous convection caused $\lambda$ to increase [49].
3. The percent change in $\lambda$ is predicted to decrease with increasing freezing rate V.

Experimentally, the influence of low gravity [3,6,7,10,11,21,22,33], a magnetic field [27-29], ampoule orientation [21-24] and centrifugation [21,22] was nearly independent of V.

4. $\lambda$ is predicted to increase as the temperature gradient increases because buoyancy-driven convection increases as the temperature gradient is increased. Experimentally, $\lambda$ was independent of the temperature gradient for the Mn-Bi eutectic [5,15,19,20].
5. $\lambda$ is predicted to vary over the cross section of the ingot because the melt velocity gradient at the interface varies spatially. Experimentally, no systematic cross sectional variation in $\lambda$ was observed, except in MnBi solidified with ACRT.

On the other hand, the theoretical predictions did agree quantitatively with the ACRT results of Eisa for MnBi [16,17]. As predicted, the change in $\lambda$ decreased as V increased, increased with radial position R, and increased with increasing stirring.

### *3.2. Off-eutectic solidification*

Although the region of perturbed concentration extends out into the melt only a short distance for eutectics, this is not true for off-eutectic mixtures. When the composition of the bulk melt differs from the eutectic, Jackson and Hunt showed that the concentration changes over a distance on the order of D/V, where D is the diffusion coefficient in the melt and V is the freezing rate [1]. Thus Favier and de Goer [18] suggested that convection would have a much larger influence on $\lambda$ when the melt is off-eutectic, and that this might explain the effect of low gravity on $\lambda$.

When the bulk melt differs from the eutectic composition, cooperative solidification of two phases can occur if the temperature gradient is sufficiently steep to avoid cellular growth or formation of primary dendrites. Although the conditions required to achieve cooperative solidification have been discussed in several papers, that is not the topic of concern here. Let us assume that cooperative solidification does occur. The average composition of the solid must adjust itself to the altered composition of the melt. For example, without convection and at steady state, the average composition of the solid must equal the composition of the bulk melt. As the amount of convection is increased, the average solid composition will move away from the bulk melt composition toward the nearest primary phase. Although the compositions of the two phases may change slightly as the average solid composition deviates from the eutectic, the principle means by which the solid assumes a new average composition is for the relative amount of the two solid phases to change. The ratio of the volumes of the two phases, $\zeta$, appears in the Jackson-Hunt convection-free treatment and results, so that one would expect $\lambda$ to depend on the bulk melt composition.

The ratio $\zeta$ also appears in the Verhoeven-Homer [35] stagnant film treatment of the influence of convection on $\lambda$ through the change in the two-dimensional concentration field. Verhoeven and Homer also estimated the average solid composition using a one-dimensional stagnant film treatment. The interfacial melt composition was assumed to be at the eutectic. If one assumes the compositions of the two solid phases are fixed, one could use this result to estimate $\zeta$, although this was not done by Verhoeven and Homer. In their treatment, Favier and de Goer [18] substituted the one-dimensional estimate for the value of $\zeta$ into the Jackson-Hunt result for lamellar eutectics without convection. They did not calculate the change in the two dimensional concentration field as Verhoeven and Homer had done. Favier and de Goer then proceeded to apply their equations to rod eutectics and concluded that a change in melt concentration on the order of 1% could account for the experimental results on the influence of convection on $\lambda$. Following is a comparison of these predictions with the experimental results:

1. Rod eutectics typically have a small volume fraction of the rod-forming component. Thus a 1% change in eutectic composition is actually enormous. For the MnBi-Bi system, for example, the volume fraction MnBi at the eutectic composition is only 3.18% [25]. Thus a change of 1% is actually a change of almost 1/3! It seems highly unlikely that such a large error could be made in determining the eutectic composition or in weighing out the components. Furthermore cooperative solidification would be difficult to achieve under such conditions.
2. When the feed material is off-eutectic, the average solid composition varies down the length of the ingot [e.g.,4,5,10,35,50-54,66]. With convection, the average solid composition also varies over the cross section [55,56]. Thus these models predict that $\lambda$ would vary systematically down the ingot and over cross sections. Experimentally, such variation was not observed in materials solidified in microgravity or with a magnetic field [3-7,10,11,27-29].
3. The change in $\lambda$ caused by convection is predicted to diminish as the freezing rate V increases. Experimentally, the influence of low gravity [3,6,7,10,11,21,22,33], a magnetic field [27-29], ampoule orientation [21-24] and centrifugation [21,22] on $\lambda$ was nearly independent of V. Only for the ACRT MnBi experiments did the change in $\lambda$ decrease as V was increased [16,17].
4. The value of $\lambda$ is predicted to be a monotonic function of $\zeta$, or, equivalently, of the fraction of the rod phase. Larson and Pirich [4,6] observed a decrease in both %MnBi and $\lambda$ when solidification was carried out in microgravity. Barczy et al. [23,24] observed an increase in $\lambda$ of $Al_3Ni$ rods as the nickel content of the bulk melt was increased. However the $\%Al_3Ni$ in the microstructure was not measured and the structure was always cellular.

### *3.3. Fluctuating freezing rate*

Although the models discussed above can explain the influence of the vigorous forced convection caused by ACRT on the $\lambda$ of MnBi, they cannot explain the influence of microgravity and of a magnetic field on the $\lambda$ of rod eutectics.

In the early 1980's, at Clarkson and at Grumman, it was proposed that a fluctuating freezing rate was causing the $\lambda$ of MnBi to be larger on earth. The hypothesis was that fiber branching occurs less readily than does fiber termination, resulting in a value of $\lambda$ that is larger than when the instantaneous freezing rate is constant and equal to the average value. To test this hypothesis, experiments were performed on the MnBi eutectic in which the ampoule translation rate was suddenly increased or decreased [58-61]. Because of heat transfer limitations, the freezing rate did not immediately equal the translation rate, but rather approached it asymptotically [62-65]. It was found that the microstructure of MnBi always corresponded to the instantaneous freezing rate, i.e. the microstructure adapted more quickly than heat transfer allowed the freezing rate to change.

To adequately test the notion of freezing rate fluctuations causing a change in $\lambda$, a technique is required that causes rapid fluctuations of magnitude below that which would totally disrupt the microstructure by causing all fibers to terminate. Furthermore this must be done in such a way that the convection pattern is not significantly changed. A technique that meets these requirements is electric current pulses. Peltier heat is instantly liberated or consumed at the solid-liquid interface, causing an instantaneous change in freezing rate. As reported above, Cai observed that the MnBi $\lambda$ is increased proportionate to the frequency and the amplitude of the current pulses [14]. This is qualitatively consistent with our hypothesis.

We must also consider the disagreement between the results of Larson-Pirich, who found that solidification of Mn-Bi in microgravity decreased $\lambda$, and those of Smith-Kaya [32], who found no difference in $\lambda$ for MnBi-Bi eutectic solidified in microgravity and on earth. To explain this disagreement, it is necessary to consider the experimental apparatus used by both. Smith-Kaya used a furnace with a nearly constant vertical temperature gradient. Consequently the buoyancy-driven convection should have been very weak and steady on earth, and the freezing rate not fluctuating. On the other hand, Larson-Pirich used a long heater to form the melt, producing a short gradient region at the freezing interface, a long relatively constant temperature region above, and another short gradient region at the end of the heater. Such a temperature profile would be expected to generate moderately strong convection on earth, both due to radial temperature gradients and to an unstable axial gradient in some locations. It would not be surprising if this convection were time-dependent, causing temperature fluctuations and freezing rate fluctuations. Indeed Larson and Pirich reported that they observed low frequency temperature fluctuations of about $3^{o}$C in some of their ground-based experiments [5,7]. Unfortunately their measurement system was not capable of detecting the small, rapid temperature fluctuations that were probably responsible for the increase in $\lambda$ on earth.

To illustrate our hypothesis, let us consider the solidification of MnBi-Bi eutectic with an oscillatory freezing rate. While the freezing rate V is increasing, the system wants the MnBi fiber spacing $\lambda$ to decrease in order to maintain $\lambda^2V$ constant. In fibrous eutectics, this must occur by branching of the existing fibers. Because MnBi is facetted, branching occurs with considerable difficulty. Consequently, the microstructure lags behind the velocity change, until the freezing rate begins to decrease. With a decreasing freezing rate, the system wants $\lambda$ to increase. This is accomplished by the matrix growing around and pinching off fibers. Apparently in the Mn-Bi system, fiber termination occurs more readily than does branching. The net effect of this hysteresis in fiber creation and termination is to yield a $\lambda$ that always exceeds the value expected for the average freezing rate.

For other fibrous eutectics, it may be that fiber branching is easier than fiber termination. Fiber termination would become difficult, for example, if the fibers extend out in the melt a long distance in front of the matrix. As noted earlier, the MnBi fibers project out about one diameter in front of the Bi matrix (31). Apparently this is not sufficient to cause termination to become more difficult than branching for this system. In other systems, it may be, and this would explain why for some fibrous eutectics $\lambda$ is increased when solidification is carried out in microgravity (18).

The above mechanism is less relevant to lamellar eutectics, for which $\lambda$ adjusts by propagation of faults. As noted earlier, solidification in microgravity and use of ACRT had no influence on the $\lambda$ of lamellar eutectics. However Carlberg and Fredriksson did note that the lamellar spacing $\lambda$ depended on the rate of change of the freezing rate [67].

When the freezing rate is fluctuating, the solidification is no longer at steady state. Consequently the volume fractions of the two phases and the interfacial undercooling will depart from their steady state values. Thus the results of Larson-Pirich are not surprising, but cannot be understood using steady state theories. A quantitative theory of oscillatory freezing is needed for comparison with experiment.

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