Evolution of dendritic patterns during alloy solidification: From the initial instability to the steady state

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ABSTRACT The evolution of the crystal–melt interface was investigated during directional solidification of a dilute binary alloy, starting at the marginal stability time \( t_i \) at which the planar interface first becomes unstable. The time delay between \( t_i \) and the crossover time \( t_0 \) at which the interface modulation becomes observable was determined experimentally. The interface morphology was analyzed as the cellular pattern appeared, and it was followed through the coarsening phase to the final steady-state dendritic pattern. The relevance of the initial instability for steady-state pattern selection was verified experimentally, and some aspects of the coarsening dynamics were measured and compared with theoretical predictions of Warren and Langer [Warren, J. A. & Langer, J. S. (1990) Phys. Rev. A 42, 3518–3525; Warren, J. A. & Langer, J. S. (1993) Phys. Rev. E 47, 2702–2712].

In the previous paper (1), the Warren–Langer (WL) approach (2, 3) to the theory of solidification instabilities was reviewed and was used to analyze the evolution of the crystal–melt interface in the binary alloy succinonitrile/coumarin 152 (SCN/C152). The experiments were described and were analyzed from the initiation of growth up to the marginal stability time \( t_i \) at which the first linear growth coefficient \( a_q(t) \) crosses over from negative to positive. In this paper, we analyze the development of the cellular pattern beyond \( t_i \), determine the time required for the initial instability to develop sufficiently to become visible, and follow the evolution of the interface position and dendrite spacing as the final steady-state morphology is approached.

Onset of the Instability and the Crossover Time

When the pulling speed \( V \) in a directional solidification experiment exceeds the critical value \( V_c \), the planar interface eventually destabilizes and evolves into a cellular or dendritic pattern. The WL analysis (3) shows that the onset of morphological instability occurs at the marginal stability time \( t_i \) while the concentration field is still building up. The WL analysis also determines a crossover time \( t_0 \) when the modulation amplitude becomes comparable to the mean wavelength \( \lambda_0 \), as described in the preceding paper (1) at the end of the Theory section.

To obtain predictions for \( t_i \) and \( \lambda_0 \), the amplitude evolution is calculated for a large range of modes, starting from thermal noise, which is orders of magnitude smaller than an observable amplitude. When the pulling motor is started at \( t = 0 \) the interface initially remains planar until \( t_i \), when the linear growth coefficient \( a_q(t) \) crosses over from negative to positive for some \( q \). Subsequently the interface becomes morphologically unstable for a continuously expanding range of wavelengths. It takes longer, however, for the modulation to grow until it becomes observable (and nonlinear) at \( t = t_0 \).

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Abbreviations: WL, Warren–Langer; SCN, succinonitrile; C152, coumarin 152.

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Fig. 2. Amplitude of the largest Fourier component of the interface modulation during the planar–cellular crossover of a planar interface, with perturbations applied at 600 s and 1,200 s (×, ○), and without perturbations (□). The experimental crossover time \( t_{0,\text{exp}} \), measured over nearly three orders of magnitude in \( V \), is shown in Fig. 3 Lower. It agrees reasonably well with the WL calculation (solid line) but is systematically slightly shorter. This difference could be due to the fact that the initial noise level is larger than the thermal noise Warren and Langer assumed in their calculation, because an increased initial level of noise-induced modulations would decrease \( t_0 \) (see Fig. 1). One possible source for an increased initial noise level at the interface is the contact line between the SCN crystal and the glass capillary, which can lead to a localized mechanical perturbation of the planar interface. Nevertheless, the WL theoretical marginal stability time \( t_i \) (dashed line) is considerably shorter than both the experimental and the theoretical \( t_0 \), except at the lowest velocity.

These experiments show that the WL linear stability analysis is a useful method for calculating the crossover wavelength \( \lambda_{0,\text{exp}} \) and that \( \lambda_{0,\text{exp}} \) is not very sensitive to the amplitude and power distribution of the initial noise at the start of the experiment. The most unstable mode predicted by the steady-state theory deviates significantly from the experimental data, indicating that the introduction of time dependence into the Mullins–Sekerka linear stability analysis by WL is clearly necessary.
newspaper. The other WL refinement of the Mullins–Sekerka analysis—the introduction of an initiation mechanism for the modulation together with the calculation of modulation amplitudes beyond \( t_i \)—is also necessary for the determination of \( \lambda_0 \), because the wavelength of the most unstable mode, at both \( t_i \) and \( t_0 \) (not shown), deviates from the experimental results as well. The marginal stability time \( t_i \) when one mode first becomes unstable provides a lower bound for the experimentally observed planar–cellular crossover time \( t_{0,\text{exp}} \), and the WL calculation of \( t_0 \) provides a good upper bound for the observed \( t_{0,\text{exp}} \) because it assumes the smallest possible level of spontaneous perturbation (thermal noise).

**Coarsening and the Approach to Steady State**

The last stage of the development of a cellular or dendritic array is the coarsening of the initial shallow cellular structure with average wavelength \( \lambda_{0,\text{exp}} \) into a final steady-state pattern. This stage, while crucial for the determination of the microstructure of the fully solidified alloy, is highly nonlinear and not well understood. The initial cellular structure starts with an amplitude comparable to \( \lambda_{0,\text{exp}} \) at the crossover time \( t_0 \) but quickly coarsens as cells overgrow their neighbors; eventually, sidebranches start to appear behind the tips of the fastest growing cells that prevent other cells from growing further (see figure 2A of ref. 1). Even though in the process the interdendritic spacing changes by approximately an order of magnitude, a reproducible selection of the final steady-state dendritic spacing is observed in experiments where the pulling motor is started abruptly and kept at a constant pulling speed \( V \). Recent experiments, however, showed that the steady-state growth conditions alone are not responsible for this selection (5). A range of spacings was observed to be stable under the same steady-state growth conditions, and the reproducible selection of one spacing out of this range of stable spacings was found to depend on the experimental history (i.e., the sequence of pulling speeds preceding the final \( V \)). In experiments where the pulling speed is constant, selection therefore has to occur during the planar–dendritic coarsening.

A linear stability analysis for dendritic arrays by WL showed that a uniform array at a given \( V \) is stable for a range of interdendritic spacings, and that outside of that range the array is most unstable against a doubling of the spacing (2, 3). This predicted spatial period doubling instability has been observed experimentally (6).}
All of the overgrown dendrites have already fallen behind at this time, and eventually stop growing and move back toward smaller $z$ at the pulling speed.

One underlying assumption of the WL theory is that the coarsening dynamics are sensitive to the crossover wavelength and $z$ position. This dependence was studied in a series of experiments where the initial pulling speed was set to different values. Before an instability occurred the pulling speed was switched to the same final value in all experiments and the subsequent initial instabilities and coarsening dynamics to a dendritic array were measured. Different crossover wavelengths and $z$ positions at the same final motor speed were observed, and the coarsening was found to result in different final dendritic spacings. A smaller $z$ position at the crossover time leads to a larger steady-state dendritic spacing, as shown schematically by line 2 in Fig. 4. While different interdendritic spacings can thus be reached during the coarsening under the same steady-state conditions, the dendrite tip radius remains unchanged, dependent only on the steady-state conditions. By selecting a sequence of pulling speeds to exploit the dynamics of the coarsening process, it is therefore possible to select (to a certain degree) these important characteristic length scales of the steady-state dendritic array independently.

Conclusions

We have investigated the evolution of a dendritic pattern in directional solidification experiments with the alloy SCN/C152, starting from a planar solid–liquid interface. Our experiments show that changes in the crossover wavelength $\lambda_0$ can affect the steady-state interdendritic spacing even though the wavelength of the pattern changes by approximately one order of magnitude during the coarsening. The focus of our experiments therefore was a detailed understanding of the initial instability from the initial recoil of the planar front after crystal growth is started until an instability becomes observable.

Initially the planar interface remains morphologically stable until the marginal stability time $t_i$ is reached. Planar interface stability was tested directly through applied spatially periodic UV perturbations that induce sinusoidal modulations of the interface profile. The growth or decay of the modulation amplitude (after the perturbation is switched off) allows for a measurement of positive or negative linear growth coefficients for a large range of wave vectors. This quantitative determination of interface stability shows that the interface becomes continually less stable after crystal growth is started, as the solute concentration builds up ahead of the advancing interface. The measured linear growth coefficients are in good agreement with the WL time-dependent linear stability analysis. At $t_i$ a transition from a negative to a positive linear growth coefficient is observed as perturbation-induced modulations start to grow. In unperturbed experiments, however, no instability is observable at $t_i$, because the instability takes time to evolve far enough to become observable, starting from an unobservably small amplitude of noise-induced modulations.

The evolution of the solute concentration field during the initial planar phase was also measured. The solute profile in the liquid ahead of the interface can be described well by an exponential with a time-dependent characteristic length in reasonable quantitative agreement with the WL calculations.

The measured wavelength at crossover (when instabilities become observable in an unperturbed experiment) is in good agreement with the WL time-dependent linear stability analysis, and the crossover time shows fair agreement with the WL analysis. This indicates that the time dependence of the solute field and a reasonable estimate for the initial noise level must be included in the Mullins–Sekerka linear stability analysis to yield agreement with experimental data, whereas nonlinear terms are not necessary in the stability analysis.

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