



Dynamics of planar interface growth during directional solidification of alloys

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The dynamics of plane front growth during directional solidification is investigated in a well-characterized system of succinonitrile–acetone, and the results show significant deviations from the predictions of existing models. This discrepancy is shown to arise from the assumption of solidification from one end in the theories that ignore the presence of an initial solute boundary layer generally present in experiments. A numerical model that relaxes this assumption is presented that gives excellent agreement with the experimental results.

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During directional solidification of alloys, the solidification dynamics go through an initial transient before a steady state is established. During this initial transient, a solute boundary layer gradually develops next to the interface due to the imbalance between the rate of rejection of solute from the interface and the rate of diffusive transport away from the interface. For solidification conditions that give rise to a diffusion-controlled stable planar interface growth, the solute build-up initially increases during the transient regime, then remains constant when the planar interface reaches the steady-state growth condition, and finally increases as the end of the sample is approached. A quantitative understanding of the initial transient is important for many solidification and crystal growth processes. For example, in order to produce crystals with uniform composition, it is important to determine the length of the sample that is growing under steady-state growth conditions with uniform composition in the solid since the initial and final transient lengths can be significantly large under the low growth rate conditions required for planar front growth. The measurement of the solute profile in the crystal during the transient time, and its comparison with the diffusion-limited model, may indicate any departure from pure diffusion-controlled growth mode and the existence of non-equilibrium effects at the interface or the presence of convection in the liquid. A precise characterization of the initial transient is also

critical in establishing the planar interface instability condition since in real systems the interface becomes unstable before reaching the steady-state condition.

A simplified conceptual model to predict the transient composition profile in solids for a planar interface growth was first proposed by Tiller et al. [1] and a detailed mathematical analysis was subsequently given by Smith et al. [2] for diffusive growth conditions. These models are approximate in that they assume that the solid–liquid interface velocity is precisely equal to the pulling velocity from the very beginning of the solidification (i.e. at $t=0$). This implies that the interface response time to the change in the external velocity is exactly zero. In an actual directional solidification experiment, the interface velocity does not instantaneously rise from zero to the externally imposed velocity and this variation in interface velocity significantly influences the rate of solute build up in the liquid and the resulting composition profile in the solid. The constant-velocity model also has a drawback in that it does not show the effect of temperature gradient on the solute profile in the solid during the transient time. Since the interface moves from near the liquidus to the solidus isotherm during the transient, the imposed temperature gradient must influence the transient dynamics.

A significant insight into the dynamics of solute build up ahead of a transient growth of a planar interface was first obtained from the model of Warren and Langer (WL) [3], which used the integral method for the solution of partial differential equations originally developed by Theodore von Karman [4,5]. Subsequently, Caroli et al. [6] developed

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an analytic al model using the Green function method, and Coriell et al. [7] developed a numerical model for the initial transient. All these models relaxed the assumption of initial velocity to be equal to the imposed velocity, but retained the assumption of no diffusion in the solid, no thermal lag in the system and uniform composition in the liquid at the start of solidification.

Although detailed models of initial transient have been proposed, experimental studies have not been conclusive due to the presence of convection [8] or due to lack of independent evaluation of system parameters [9]. The major aim of this paper is to present the results of benchmark experimental studies that quantitatively investigate the transient behavior in thin rectangular samples of succinonitrile (SCN)–0.23 wt.% acetone in which convection effects are negligible, and all relevant parameters of this system are accurately known and measured independently so that no free parameter is needed to fit the model based on diffusive growth conditions. Experimental results are found to deviate significantly from the predictions of the existing models, and the deviation is observed to increase as the thermal gradient is increased. This deviation is evaluated by comparing the experimental results with a numerical model [10] that allows us to relax the assumptions of the existing models.

The experimental procedure, sample preparation, interface position and interface temperature measurement technique used in this study are identical to those described by Xu et al. [11], and therefore will not be repeated here. The sample was held in the thermal gradient stage for some time to obtain thermal equilibrium and when the interface appeared to remain stationary the sample was directionally solidified at a fixed imposed velocity. The temperature of the interface was characterized as a function of time, including $t = 0$, when the directional solidification run was started. Experiments were carried out for two different velocities and four different thermal gradient values, and the position of the interface and the temperature of the interface as a function of time were measured experimentally for each experiment.

The interface position as a function of time for $V = 2.25 \mu\text{m s}^{-1}$ and $G = 3.8 \text{ K mm}^{-1}$ is shown in Figure 1a along with the predictions of existing theoretical models [3,6,10], and a significant deviation is observed. Similar discrepancies were found at other experimental conditions. To obtain an insight into this deviation, the interface temperature as a function of time was also measured, and the results for an experiment with $V = 1.0 \mu\text{m s}^{-1}$ and $G = 4.0 \text{ K mm}^{-1}$ are shown in Figure 1b; these also show significant disagreement between the theoretical predictions and experimental data. However, an important observation is made in that the initial temperature of the stationary interface is not at the liquidus temperature, as assumed in all theoretical models, which indicates that the initial composition in the liquid is not uniform and a small boundary layer of solute is present in the liquid at the interface. To examine the effect of thermal gradient on the initial interface temperature, experiments were carried out for four different temperature gradient values and the results are presented in Figure 2a. The initial interface temperature was found to decrease with an increase in the thermal gradient value, as shown in Figure 2b, which is in contradiction to the theoretical assumption that the initial interface will be at the liquidus temperature for all temperature gradient values.

We now evaluate the reason for the deviation of experimental results from the predictions of the current theoretical models so that a proper model can be developed to

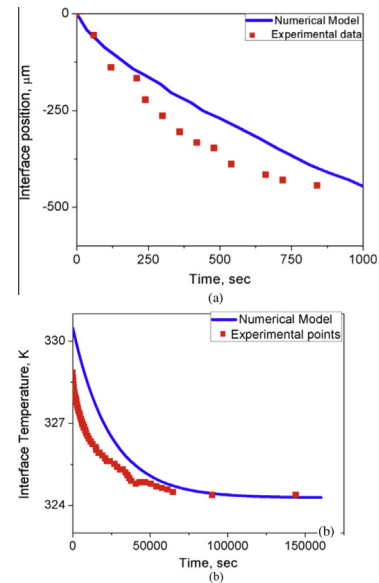


Figure 1. (a) The variation of interface position with time for $V = 2.25 \mu\text{m s}^{-1}$ and $G = 3.8 \text{ K mm}^{-1}$, and (b) the variation of interface temperature with time for $V = 0.75 \mu\text{m s}^{-1}$ and $G = 3.7 \text{ K mm}^{-1}$ in SCN–0.23 wt.% acetone. Experimental results are shown as circles, and the corresponding results of the theoretical model that assumes initial uniform composition C_0 in the liquid are also shown for comparison.

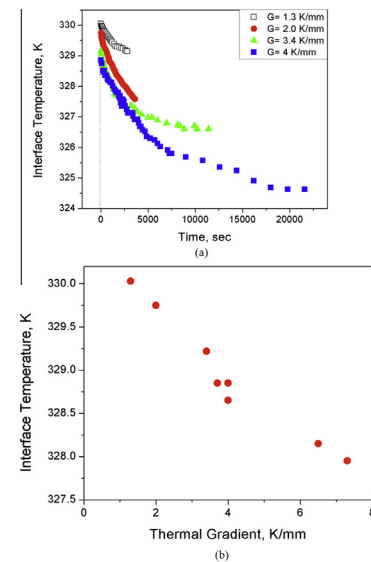


Figure 2. (a) Interface temperature as a function of time in SCN–0.23 wt.% acetone for $V = 1.0 \mu\text{m s}^{-1}$, and $G = 1.3, 2.0, 3.4$ and 4.0 K mm^{-1} . (b) Temperature of the initial stationary interface as a function of imposed thermal gradient.

characterize the initial transient during the directional solidification process. We use a finite-difference model based on a fixed frame of reference [10], which will allow us to relax different assumptions in the existing models. We shall examine three key assumptions of all current models, which are: (i) the diffusion in the solid is negligible; (ii) the isotherms move instantaneously with the pulling speed so that no thermal lag is present in the system; and (iii) the composition in the liquid is uniform initially at the start of solidification and is equal to the initial composition of the alloy or the interface temperature is at the liquidus temperature of the alloy.

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