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Initial dynamics of a solid–liquid interface within a thermal gradient

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In directional solidification experiments an alloy is placed in the thermal gradient assembly and kept stationary to achieve a steady-state thermal profile. During this time an interface motion occurs that is experimentally characterized and shown to generate a solute boundary layer at the interface whose thickness depends on the time the sample is kept stationary before the external velocity is imposed. This boundary layer must be included in the theoretical description of the initial transient during a planar front growth.

Keywords: Directional solidification; Interface dynamics; Bridgman technique; Crystal growth; Boundary layer

The dynamics of solid–liquid interfaces plays a crucial role in the evolution of microstructures during directional solidification of alloys. The most basic process is the motion of a planar interface in a binary alloy system that is controlled by the development of a solute boundary layer with time until a steady-state condition is reached. Theoretical models have been proposed by using approximate analytical treatments or numerical simulations [1–6]. In all the models proposed so far, the sample is considered to solidify from one end so that the initial composition in the liquid is uniform. In contrast, in directional solidification experiments, a solid alloy of uniform composition is placed in a thermal gradient stage. The sample is generally held stationary until a steady-state thermal profile is established and a planar interface forms [7]. The sample is then moved at a fixed rate to achieve directional solidification.

When the sample is placed in the thermal gradient stage (Fig. 1a) the liquid and solid are at the initial alloy composition far from the interface, but the liquid and solid compositions at the interface must be different for local equilibrium to be present. Therefore, solute boundary layers develop, as schematized in Figure 1b, which in turn influence the concentration field when the sample is directionally solidified, and must be taken into account in the description of the initial transient during the planar front growth.

Nguyen Thi et al. [8] first characterized the presence of an initial solute boundary layer in an Al–1.5 wt.% Ni alloy. They discussed the homogenization process by temperature gradient zone melting process in the mushy zone, and concluded that when liquid diffusion is the mode of transport, the limiting condition of a homogeneous liquid may not be fulfilled due to the very long time required.

The present study examines the dynamics of the planar interface that forms quickly in a dilute alloy of a transparent system in the presence of a thermal gradient but no externally imposed velocity. We seek to determine the position or temperature of the interface when the interface stabilizes and remains stationary within the temperature gradient. For a pure material the interface will be at the melting point isotherm. However, in a binary system the interface temperature can have any value between

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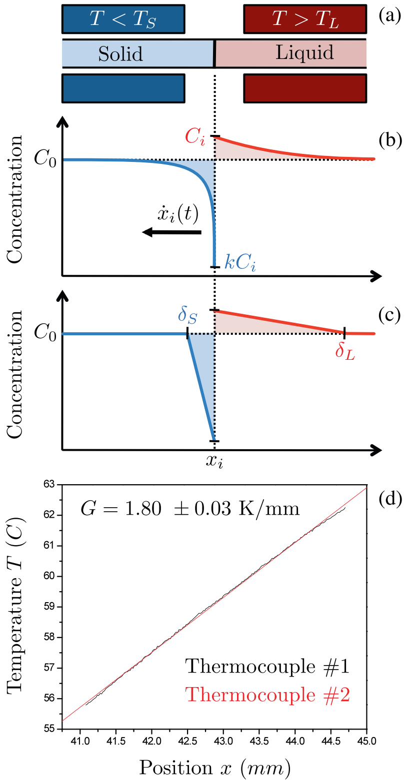

Figure 1. (a) Experimental setup in which a solid of initial uniform composition, C_0, is placed in the thermal gradient stage with zero imposed velocity. Composition profiles develop in the solid and liquid to maintain local equilibrium at the interface while the interface recoils, as illustrated in the schematic snapshot in (b) with boundary layer equivalent profiles in (c). The temperature profiles obtained from two thermocouples, shown in (d), overlap when relatively displaced by the distance between the thermocouples, hence confirming a homogeneous thermal gradient $G = 1.8$ °C mm^{-1}.

the liquidus and the solidus temperatures, and the interface must stabilize between these two temperatures.

Experiments were carried out with succinonitrile (SCN)–0.25 wt.% camphor. As-received SCN was first purified by distillation and a zone-refining process. Camphor with initial purity of 98 wt.% was sublimated twice at a temperature of 353 K under vacuum of 45 Pa. High-purity SCN was then mixed with a camphor in a glovebox filled with an inert environment of nitrogen to obtain the SCN–0.25 wt.% camphor mixture. The liquid alloy was introduced into the sample cell and cooled quickly to obtain a solid sample of fine microstructure that consisted of equiaxed dendrites and eutectic. A rectangular sample 100 μm thick was used to minimize convection in the liquid. Two additional cells, identical to the sample cell, were also prepared, and calibrated thermocouples were placed in these two cells. The cells with thermocouples were placed on the two sides of the sample cell, and the three-cell assembly was placed in the temperature gradient stage. The hot and cold baths were maintained at 73 and 48 °C, respectively, yielding a nominal thermal gradient of 1.8 °C mm^{-1}.

The thermocouples in the outer cells were placed at a fixed distance from each other in the growth direction, such that the solid–liquid interface in the center sample was present between these two thermocouples. First the sample assembly was directionally solidified at a fixed growth rate, and the temperature profiles given by the two thermocouples were characterized. When displaced by the distance between the thermocouples, the temperature profiles from the two thermocouples overlapped, as shown in Figure 1d, hence confirming the presence of steady-state thermal field. The temperature gradient measured in the region between the two thermocouples was 1.8 °C mm^{-1}, identical to a direct calculation from the measured temperature at the two thermocouples divided by the distance between them, hence confirming a linear temperature profile in the sample near the interface.

The temperature profiles are linear and continuous through the solid–liquid interface since the thermal conductivities of solid and liquid SCN are nearly equal, at 0.225 and 0.223 W K^{-1} m^{-1}, respectively [9]. Thus, the interface temperature can be calculated at any time from the thermal gradient value and the location of the interface.

A fresh sample cell was prepared and placed in the thermal gradient for quantitative measurements of interface position with time with no imposed velocity. Once the sample was placed in the thermal gradient stage, melting in the hot zone occurred rapidly above the liquidus temperature and a planar solid–liquid interface formed. Since the sample is held stationary the thermocouple positions remain fixed and only the interface position changes with time. The evolution of the interface location was measured from a fixed reference position, which was taken as the sharp edge of the cold chamber. The initial position of the interface was then taken as $x = 0$, and the positive x direction was taken towards the cold stage. The measured position of the interface with time, $x_i(t)$, appears in Figure 2.

Given the constant temperature gradient, the variation of interface temperature with time is calculated, as shown on the right hand side of Figure 2. From the expression given by Teng and Liu [10], the liquidus temperature for the alloy of composition $C_0 = 0.25$ wt.% camphor is $T_L = 57.73$ °C. The solidus temperature at C_0 measured as the interface temperature during steady-state planar interface growth is $T_S = 56.45$ °C. Hence, assuming linear liquidus and solidus lines from the melting temperature of pure SCN, T_M, with $T_L = 57.73$ °C and $T_S = 56.45$ °C, and using a solute partition coefficient, $k = 0.21$ [10], the liquidus line of the alloy follows $T = T_M + mC_i$ with $T_M = 58.07$ °C and a liquidus slope $m = -1.361$ °C wt.%$^{-1}$. Therefore, we can plot the

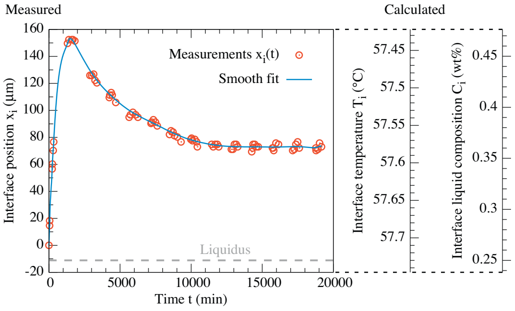

Figure 2. Experimental measurement of the time evolution of interface position $x_i(t)$ (left y-axis) and corresponding calculated evolution of the interface temperature and liquid interface composition (right axes).