Predicting the columnar-to-equiaxed transition for a distribution of nucleation undercoolings

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Abstract

A deterministic mathematical model for steady-state unidirectional solidification is proposed to predict the columnar-to-equiaxed transition. In the model, which is an extension to the classic model proposed by Hunt [Hunt JD. Mater Sci Eng 1984;65:75], equiaxed grains nucleate according to either a normal or a log-normal distribution of nucleation undercoolings. Growth maps are constructed, indicating either columnar or equiaxed solidification as a function of the velocity of isotherms and temperature gradient. The fields of columnar and equiaxed growth change significantly with the spread of the nucleation undercooling distribution. Increasing the spread favors columnar solidification if the dimensionless velocity of the isotherms is larger than 1. For a velocity less than 1, however, equiaxed solidification is initially favored, but columnar solidification is enhanced for a larger increase in the spread. This behavior was confirmed by a stochastic model, which showed that an increase in the distribution spread could change the grain structure from completely columnar to 50% columnar grains.

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1. Introduction

The columnar-to-equiaxed transition (CET) is the transition from columnar grains to equiaxed grains observed in the macrostructures of castings. The position of the CET determines the relative amount of columnar and equiaxed grains, which defines important properties of cast products [1,2]. Different mechanisms leading to the CET have been proposed, investigated and reviewed in the past decades [3–5]. There is a consensus that the CET occurs when the moving front of columnar grains is blocked by equiaxed grains growing in the undercooled liquid ahead of this front. This mechanism has been observed in situ using synchrotron X-ray radiographs [4]. There is still controversy, however, about the details of how the equiaxed grains stop the columnar front. In the first mechanism proposed, equiaxed grains block the columnar front through mechanical interactions (mechanical blocking) [3]. Later, equiaxed grains were assumed to block columnar grains through solutal (solutal blocking) [6] or thermal (thermal blocking) interactions [7,8]. The mechanical and solutal blocking mechanisms were considered simultaneously in a recent mathematical model to predict the CET [9].

Regardless of the blocking mechanism, experiments and theoretical models have shown that the CET is significantly affected by the number density of equiaxed grains and the nucleation undercooling. In Hunt’s [3] model for unidirectional steady-state solidification, a decrease in the nucleation undercooling or an increase in the number of equiaxed grains facilitates the blocking of columnar grains and, consequently, the occurrence of the CET. Sturz et al. [10] and Martorano and Capocchi [11] added inoculants to melts of Al and Cu alloys, respectively, observing that the CET occurred earlier in comparison with samples without inoculants, confirming the predictions of Hunt’s [3] model.

To predict the CET, instantaneous nucleation was adopted in earlier models [3,6] by assuming that all possible
equiaxed grains would nucleate locally when the liquid undercooling was larger than $\Delta T_N = T_L - T_N$, where $T_L$ is the liquidus temperature of the alloy and $T_N$ is a given nucleation temperature. Nevertheless, in models concerned with the prediction of equiaxed grain size, the nucleation undercooling was assumed to follow a Gaussian distribution. This distribution can be interpreted as a distribution of undercoolings necessary for heterogeneous nucleation on different substrate particles. The distribution density, $dn/d\Delta T_N$, was defined as \[12\]:

$$
\frac{dn}{d(\Delta T_N)} = \frac{n_T}{\sqrt{2\pi}\Delta T_e} \exp\left[-\frac{1}{2} \left(\frac{\Delta T_N - \Delta T_e}{\Delta T_e}\right)^2\right],
$$

where $n$ is the number density of substrate particles; $n_T$ is the number density of all substrate particles in the system; $\Delta T_N$ is the average nucleation undercooling; and $\Delta T_e$ is the standard deviation of the distribution. For instantaneous nucleation, $\Delta T_e = 0$, and the distribution density should be replaced with $dn/d\Delta T_N = n_T \delta(\Delta T_N)$, where $\delta$ is the Dirac delta function.

Experimental evidence has shown that the undercooling necessary for a heterogeneously nucleated grain to grow freely from its substrate particle depends on the particle size, $\phi$, according to \[13\]:

$$
\Delta T_{fg} = \frac{4\sigma_{SL}}{\Delta S_V \phi},
$$

where $\Delta T_{fg}$ is the undercooling for free growth; $\sigma_{SL}$ is the solid–liquid interface energy per unit area; $\Delta S_V$ is the volumetric entropy of fusion. In equiaxed grain solidification, this free growth undercooling can be modeled as a nucleation undercooling \[13\]. This undercooling distribution depends on the particle size distribution, which was assumed to be either exponential \[13\] or log-normal \[14\]:

$$
\frac{dn}{d\phi} = \frac{n_T}{\sigma_\phi \sqrt{2\pi}} \exp\left[-\frac{1}{2} \left(\frac{\ln(\phi/\bar{\phi})}{\sigma_\phi}\right)^2\right],
$$

where $\bar{\phi}$ and $\sigma_\phi$ are, respectively, the geometric average and the shape parameter of the log-normal distribution.

In most of the deterministic models of the CET, instantaneous nucleation has been assumed \[3,6,15–18\]. Wu and Ludwig \[9\], however, have recently considered a Gaussian distribution of undercoolings in their transient model, but the effects of the distribution spread on the CET position were not investigated. Quested and Greer \[14\] developed a steady-state model considering a log-normal distribution of nucleant particle sizes to predict equiaxed grain size. The model was extended to predict the CET, but the effects of the distribution spread on the CET were not examined. Nevertheless, the effects of the distribution spread on grain size were investigated to design inoculants for aluminum alloys \[19,20\].

A distribution of nucleation undercoolings has always been adopted in stochastic models \[21–23\], but only Cho et al. \[23\] studied the effects of the spread of a Gaussian distribution on the calculated grain macrostructures. The effects on the CET position, however, were not discussed.

Except for Greer and Quested’s \[14\] model, all deterministic and stochastic models that predict the CET and consider a nucleation undercooling distribution have been transient. Although transient models are useful for predicting the grain structure in solidification processes, the steady-state models have proved very important to the understanding of the basic mechanisms of the CET \[3\], since the velocity of isotherms, $V$, and the temperature gradient, $G$, are changed independently. Furthermore, steady-state models can be readily used to construct growth maps, and have been frequently applied to predict either the prevailing growth mode (columnar or equiaxed) in steady-state solidification \[24\] or the CET in transient solidification \[25\]. Transient models can also be used to construct the maps, but numerous time-consuming simulations (which need to be carried out for long enough to achieve steady state) are frequently required \[6,9,22\].

In the present work, a steady-state model of unidirectional solidification is proposed to predict the CET. In this model, equiaxed grains nucleate within a temperature range, differing from several available models considering instantaneous nucleation. The model presented here is based on the model presented by Quested and Greer \[14\] and is an extension of Hunt’s \[3\] model (in which instantaneous nucleation was assumed) to a more realistic situation of equiaxed grains nucleating within a temperature range. Mathematical models have shown that the CET positions predicted with either the mechanical or solutal blocking mechanism are similar for most solidification conditions \[6,26\]. Therefore, mechanical blocking of the columnar front is adopted in the present work, because the construction of the growth maps is facilitated after the derivation of a simple final equation. Growth maps are constructed to predict the CET assuming that the distribution of nucleation undercoolings for equiaxed grains is either Gaussian or log-normal. The effects of the spread of the distributions are analyzed and verified in the macrostructures calculated with a well-known stochastic model of solidification.

2. Description of the model

The present model, which is based on the models proposed by Hunt \[3\] and Quested and Greer \[14\], is developed from the following assumptions: (a) unidirectional and steady-state solidification of both equiaxed and columnar grains for a reference system moving at the constant velocity of the isotherms; (b) linear temperature variation with distance; (c) negligible convection of the liquid and movement of equiaxed grains; (d) spherical equiaxed grains; (e) heterogeneous nucleation of equiaxed grains on substrate particles; and (f) normal (Gaussian) or log-normal distribution of nucleation undercoolings. The solidification system is illustrated in Fig. 1, where both columnar and equiaxed growth are depicted, although only one type of growth...
where $V_g$ and $G$ are the velocity of the isotherms, which equals the frontal growth velocity when columnar growth prevails; otherwise, columnar grains dominate. This assumption implies the mechanical blocking of the columnar front. Mathematical models have predicted that both mechanical and solutal blocking of the columnar front yield similar results in most solidification conditions [6,26]. Consequently, the mechanical blocking criterion is adopted, because the final model equation is simplified and growth maps are readily calculated.

The boundary curve between columnar and equiaxed solidification in the growth maps can finally be calculated by considering $e_g(\Delta T_{col}, e_{\text{block}})$ together with Eqs. (4), (7) and (8), giving:

$$G = 0.617n^{1/3} \varphi(\Delta T_{col}, e_{\text{block}}) \Delta T_{col}$$

(9)

$$\varphi(\Delta T_{col}, e_{\text{block}}) = \frac{3}{(m + 1)} \left\{ \ln(1 - e_{\text{block}}) \right\}^{-1/3} \int_0^{\Delta T_{col}} \left[ 1 - \left( \frac{\Delta T_N}{\Delta T_{col}} \right)^{m+1} \right]^3 \frac{dn}{d\Delta T_N} d\Delta T_N$$

(10)

When $G$ is lower than the value given by Eq. (9), grain growth is equiaxed; otherwise, it is columnar. Eqs. (9) and (10), which can be integrated numerically by simple methods, can be used to calculate the growth maps for a given probability density function, $dn/d\Delta T_N$. For instantaneous nucleation, $dn/d\Delta T_N = n_T \delta(\Delta T_N)$, where $\Delta T_N$ is the
average nucleation undercooling. If also $m = 2$ and

$$
\varphi = 0.499,
$$

then $\varphi = 1 - \left( \frac{\Delta T_{\text{col}}}{\Delta T_{\text{m}}} \right)^3$, which gives Hunt’s[3] model when substituted in Eq. (9).

Provided a blocking fraction is defined, Eqs. (9) and (10) allow calculation of the boundary curve between columnar and equiaxed growth in growth maps with $\Delta T_{\text{col}}$ and $G$ as the independent variables. To substitute $\Delta T_{\text{col}}$ for $V$, creating a more useful map, the columnar front is assumed to grow according to the same law as that for equiaxed grains (Eq. (5)), resulting in $\Delta T_{\text{col}} = (V/A)^{-m}$. Using this relation, Eqs. (9) and (10) can be, respectively, written in the following dimensionless form:

$$
G = 0.617 \varphi^* (V^*, \varepsilon_{\text{block}})^{1/m}
$$

(11)

$$
\varphi^* (V^*, \varepsilon_{\text{block}}) = \left\{ \begin{array}{ll}
\frac{3}{m+1} \left[ \frac{0.66}{\ln(1 - \varepsilon_{\text{block}})^{-1}} \int_0^{V^*/V^*} \frac{d\Delta T_N^*}{\Delta T_N^*} \right]^{1/3} & \text{if } m = 2,
\end{array} \right.
$$

(12)

where $G = G/[n^{1/2}(\Delta T_N^*)]$; $V = V/(\Delta T_N^*)^{m}$; $\Delta T_N^* = \Delta T_N/\Delta T_{\text{m}}$; and $n^* = n/n_T$. The parameter $\Delta T_N^*$, a scale for any type of undercooling in the model, can be chosen as the average of the distribution of nucleation undercoolings. Note that the application of Eq. (5) to calculate the columnar front velocity (which equals the isotherm velocity at steady state) gives $\Delta T_{\text{col}} = (V^*)^{1/m}$, corresponding to the integration limit in Eq. (12). To recover Hunt’s model [3] (instantaneous nucleation) $\varphi^* = 1 - (V^*)^{-3/m}$.

Either a normal or a log-normal distribution of nucleation undercoolings was adopted to integrate Eq. (12). The dimensionless form of the normal distribution was integrated to give

$$
\frac{dn^*}{d\Delta T_N^*} = \frac{1}{\Delta T_{\sigma} \sqrt{2\pi}} \exp \left[ -\frac{1}{2} \left( \frac{\Delta T_N^* - 1}{\Delta T_{\sigma}} \right)^2 \right]
$$

(13)

where $\Delta T_{\sigma} = \Delta T_{\sigma}/\Delta T_{\text{m}}$. Then, Eq. (12) becomes:

$$
\varphi^* = \left\{ \begin{array}{ll}
\frac{3}{m+1} \left[ \frac{0.66}{\ln(1 - \varepsilon_{\text{block}})^{-1}} \Delta T_{\sigma} \sqrt{2\pi} \right]^{V^*/V^*} \int_0^{V^*/V^*} \frac{d\Delta T_N^*}{\Delta T_N^*} \right]^{1/3} \\
\times \left[ 1 - \left( \frac{\Delta T_N^*}{V^*/V^*} \right)^{m+1} \right]^{1/3} \exp \left[ -\frac{1}{2} \left( \frac{\Delta T_N^* - 1}{\Delta T_{\sigma}} \right)^2 \right] \frac{d\Delta T_N^*}{\Delta T_N^*}
\end{array} \right. \right.
$$

(14)

The extended volume fraction of equiaxed grains at any undercooling $\Delta T^*$ was also calculated for a normal distribution of nucleation undercoolings combining Eqs. (7) and (13), yielding:

$$
ev_{\text{G}}(\Delta T^*) = \frac{2}{3} \frac{\sqrt{2\pi}}{\Delta T_{\sigma} \left[ V^* G^* (m+1) \right]^{1/2}} \int_0^{\Delta T^*} \left( \frac{\Delta T_N^*}{V^*/V^*} \right)^{m+1} \left( \frac{\Delta T_N^*}{V^*/V^*} - 1 \right)^2 \frac{d\Delta T_N^*}{\Delta T_N^*}.
$$

(15)

Finally, Eq. (8) was used to correct $v_{\text{G}}$, giving $v_\phi(\Delta T^*)$.

As discussed previously, the distribution of undercoolings for free growth depends on the log-normal distribution of particle sizes (Eq. (3)). Using Eq. (2), the size distribution given by Eq. (3) was converted into the following dimensionless undercooling distribution:

$$
\frac{dn^*}{d\Delta T_N^*} = \frac{1}{\sigma_{\phi} \Delta T_N^* \sqrt{2\pi}} \exp \left[ -\frac{1}{2} \left( \frac{\ln(\Delta T_N^*)}{\sigma_{\phi}} \right)^2 \right],
$$

(16)

where the undercooling scale was the geometric average $\Delta T_N^* = 4^{\sigma_{\phi}}$ which is a function of the geometric average of the size distribution ($\bar{\phi}$) from Eq. (3). Note that the shape parameter ($\sigma_{\phi}$), which is different from the standard deviation, is already dimensionless. It indicates the shape, rather than the spread of the distribution, but is related to the standard deviation. Substituting Eq. (16) into Eq. (12) gives:

$$
\varphi^* = \left\{ \begin{array}{ll}
\frac{3}{m+1} \left[ \frac{0.66}{\ln(1 - \varepsilon_{\text{block}})^{-1}} \frac{1}{\sigma_{\phi} \sqrt{2\pi} \int_0^{V^*/V^*} \frac{d\Delta T_N^*}{\Delta T_N^*}} \right]^{1/3} \\
\times \left[ 1 - \left( \frac{\Delta T_N^*}{V^*/V^*} \right)^{m+1} \right]^{1/3} \frac{1}{\Delta T_N^*} \exp \left[ -\frac{1}{2} \left( \frac{\ln(\Delta T_N^*)}{\sigma_{\phi}} \right)^2 \right] \frac{d\Delta T_N^*}{\Delta T_N^*}
\end{array} \right. \right.
$$

(17)

which can be used to calculate the growth maps for a log-normal distribution of undercoolings when combined with Eq. (11).

3. Growth maps for steady-state solidification

The boundary curves between columnar and equiaxed solidification in the growth maps were constructed by combining Eq. (11) with either Eq. (14), for the normal distribution of undercoolings, or Eq. (17), for the log-normal distribution. The maps were constructed for an Al–7 wt.% Si alloy with the normal (Fig. 2a) and log-normal undercooling distributions (Fig. 2b). The Lipton–Glicksman–Kurz (LGK) curve presented by Martorano et al. in Fig. 12 of Ref. [6] is also included. This curve is shown because it was calculated with a model similar to that proposed by Hunt [3], in which instantaneous nucleation was assumed, but considering the LGK model [27] for dendritic growth. The LGK model has been adopted in numerous mathematical models of the as-cast grain macrostructure, representing a reference model for dendritic growth. All curves are plotted in the more familiar dimensional form.

The boundary curves for both distributions reduce to the instantaneous nucleation curve (LGK) when $\Delta T_{\sigma}$ and $\sigma_{\phi}$ decrease to 0.01 K and 0.01, respectively. This behavior was expected, since the normal and log-normal distributions of undercoolings approach the instantaneous nucleation model when $\Delta T_{\sigma}$ → 0 and $\sigma_{\phi}$ → 0, respectively. When $V \gtrsim 10^{-4}$ m s$^{-1}$, an increase in $\Delta T_{\sigma}$ and $\sigma_{\phi}$ from the instantaneous nucleation values shifts the boundary curves to the right-hand side, widening the field of equiaxed solidification. For a larger increase ($\Delta T_{\sigma} > 1$ K, $\sigma_{\phi} > 3$ K),
the curves begin to shift to the left, enhancing the columnar solidification fields again. For $V \gtrsim 10^{-4}$ m s$^{-1}$, however, the curves always shift to the left-hand side, favoring columnar growth. A similar behavior is observed in the dimensionless growth maps of Fig. 3, where a $\Delta T^*_{col}$ axis (related to the $V^*$ axis) is also included. In these maps $\sigma_g^{\text{block}} = 0.2$ was adopted, rather than the traditional 0.49 proposed by Hunt [3], because it has recently been shown to improve the agreement between the CET positions obtained with stochastic and deterministic models [26].

Solidification conditions for $V^* > 1$ (condition A: $V^* = 10$, $G^* = 1.6$) and $V^* < 1$ (condition B: $V^* = 0.1$, $G^* = 0.02$) are indicated by dots in Fig. 3a and examined in Fig. 4, where $\sigma_g$ is given as a function of $\Delta T^*$ for a normal distribution of nucleation undercoolings. As expected, $\sigma_g$ increases with an increase in $\Delta T^*$ during solidification. For condition A (Fig. 4a) and approximately instantaneous nucleation ($\Delta T^*_n = 0.01$), equiaxed grains nucleate in a narrow undercooling range around $\Delta T^* = 1$ (centre of the dimensionless normal distribution in Fig. 5a). The grain fraction reaches the blocking fraction ($\sigma_g^{\text{block}} = 0.2$) exactly at the columnar front undercooling ($\Delta T^*_{col} \approx 2.3$), as shown in Fig. 4a. Consequently, condition A is at the boundary curve for $\Delta T^*_n = 0.01$ (Fig. 3a) and also for $\Delta T^*_n = 0.2$.
When $\Delta T^*$ increases from 0.2 to a value in the range between 1 and 10, then $\varepsilon_g < \varepsilon_{\text{block}}$ at $\Delta T^*_{\text{col}}$ (Fig. 4a), and columnar growth prevails, as observed in the growth map. The spread of the distribution of undercoolings (Fig. 5a) increases, reducing the maximum number of equiaxed grains that can nucleate in the range $0 < \Delta T^* < \Delta T^*_{\text{col}}(\approx 2.3)$. This maximum number is proportional to the integrated area under the probability density curve from the liquidus isotherm ($\Delta T^* = 0$) to the columnar front isotherm ($\Delta T^* \approx 2.3$). When $\Delta T^*_N$ is between 1 and 10, a significant portion of the area is cut off on the left-hand side by the liquidus isotherm and on the right-hand side by the columnar front isotherm. Consequently, the equiaxed grain fraction at $\Delta T^*_N$ decreases owing to fewer equiaxed grains, favoring columnar growth. Generally, the area under the normal distribution curve is significantly reduced by the liquidus isotherm when $\Delta T^*_N > 1/3$, and by the columnar front isotherm when $\Delta T^*_N > (\Delta T^*_{\text{col}} - 1)/3$, which is 0.43 for condition A ($\Delta T^*_{\text{col}} = 2.3$).

Solidification condition B represents a point on the boundary curve for $\Delta T^*_N = 0.2$ (Fig. 3a), because $\varepsilon_g = \varepsilon_{\text{block}}$ at the columnar front undercooling ($\Delta T^* \approx 0.43$), as shown in Fig. 4b. When $\Delta T^*_N$ decreases to 0.01 (approximately instantaneous nucleation) all equiaxed grains nucleate at $\Delta T^* = 1$. Thus, no equiaxed grains nucleate below $\Delta T^*_{\text{col}}(\approx 0.43)$, as seen in Figs. 4b and 5a, and $\varepsilon_g(\Delta T^*_{\text{col}}) = 0$, resulting in columnar growth for condition B in Fig. 3a.
On the other hand, when $\Delta T'_c$ increases from 0.01 to 1, the undercooling distribution widens (Fig. 5a), enabling nucleation of equiaxed grains below $\Delta T'_c$. Accordingly, for $V < 1$ the boundary curve in Fig. 3a shifts to the right, favoring equiaxed solidification. When $\Delta T'_c$ is further increased from 1 to 10, the same behavior encountered in condition A is recovered (equiaxed growth is less likely because some area under the undercooling distribution curve is removed by the liquidus and columnar front isotherms) and columnar solidification is favored.

To sum up, two types of behavior exist regarding the effect of $\Delta T'_c$ on the growth maps. When $V > 1$, the average nucleation undercooling for equiaxed grains ($\Delta T'_c = 1$) is smaller than the columnar front undercooling (i.e. $\Delta T'_c > 1$ in Fig. 3a). Therefore, for instantaneous nucleation ($\Delta T' = 0.01$), all possible equiaxed grains nucleate, representing the most likely condition for equiaxed growth. For an increase in $\Delta T'_c$ above $\sim 1/3$ or above $\sim (\Delta T'_c - 1)/3$, some area under the curve of nucleation undercooling distribution is cut off, decreasing the number of nucleated equiaxed grains and widening the columnar solidification field. When $V < 1$, the average nucleation undercooling for equiaxed grains is now larger than the columnar front undercooling (i.e. $\Delta T'_c < 1$ in Fig. 3a). Then, for instantaneous nucleation, no equiaxed grains nucleate and only columnar growth occurs for any temperature gradient. Increasing $\Delta T'_c$ causes some area under the curve of nucleation undercooling distribution to lie below $\Delta T'_c$, enabling some nucleation of equiaxed grains and reducing the columnar field. For a larger $\Delta T'_c$ increase ($\Delta T'_c \geq 1$), however, the columnar solidification field widens again because the undercooling distribution curve is now cut off by the liquidus isotherm and less equiaxed grain nucleation occurs.

A similar behavior is observed in the effects of $\sigma_p$ for a log-normal distribution of nucleation undercoolings (Fig. 3b). Differences in the shift of the boundary curves occur, however, because the liquidus isotherm never cuts off the nucleation distribution curves (Fig. 5b).

4. Transient unidirectional solidification

In the previous section, modifications in the growth maps were explained for a change in the spread of the distribution of nucleation undercoolings. Since experimental validation of this theory is difficult, an attempt is made to validate it using macrostructures calculated with a stochastic model for the transient unidirectional solidification of an Al–7 wt.% Si alloy. The stochastic model implemented and used in the present work is based on the model proposed by Gandin and Rappaz [21] and consists of a transient one-dimensional macroscopic model and a two-dimensional microscopic model of cellular automaton. To verify its correct implementation, some model results were compared with those presented by Rappaz and Gandin [28,29], and showed excellent agreement.

In the macroscopic model, the transient heat conduction equation was solved in a one-dimensional domain 0.15 m long. One of the domain boundaries was adiabatic, whereas the heat flux out of the other boundary was $q = h(T - T_\infty)$, where $h$ is the heat transfer coefficient (250 W m$^{-2}$ K$^{-1}$), and $T_\infty$ is a reference temperature (298 K). A uniform temperature (991 K) was adopted throughout the domain as the initial condition.

In all simulations, the properties for the Al–7 wt.% Si alloy were: $\kappa_s = 137.5$ W m$^{-1}$ K$^{-1}$ (heat conductivity of solid); $\kappa_l = 60.5$ W m$^{-1}$ K$^{-1}$ (heat conductivity of liquid); $L = 387.4 \times 10^5$ J kg$^{-1}$ (latent heat of fusion); $c_p = 1126$ J kg$^{-1}$ K$^{-1}$ (specific heat); $\rho = 2452$ kg m$^{-3}$ (density); $k = 0.13$ (solute partition coefficient); $T_f = 933$ K (melting point of pure Al); $T_L = 891$ K (liquidus temperature); and $T_E = 850$ K (eutectic temperature).

In the two-dimensional microscopic model, the nucleation of grains was simulated with the normal distribution of nucleation undercoolings described by Eq. (1). Two sets of distribution parameters were used: one for the bulk liquid and another for the liquid layer adjacent to the boundary through which heat was extracted. For the liquid layer, $\Delta T_{N,layer} = \Delta T_{eq,layer} = 0$ (instantaneous nucleation) and $n_T,layer = 3 \times 10^8$ m$^{-2}$, which was converted for a two-dimensional simulation domain using the relation $n^2_T,layer = 2\sqrt{n_T,layer}/\pi$ [28]. For the bulk liquid, $\Delta T_{N} = 2$ or 5 K, and $\Delta T'_c$ was adjusted to give $0 \leq \Delta T'_c \leq 3$. The number density of grains was $n_T = 5 \times 10^6$ m$^{-3}$, which was converted for the two-dimensional domain using $n^2_T = (n_T \sqrt{6}/\pi)^{1/3}$ [28]. To simulate grain growth, the velocity of the grain envelope diagonals was calculated using the same kinetic equation as that used to derive the present model equations (Eq. (5)), with constants $A = 3 \times 10^{-6}$ m s$^{-1}$ K$^{-2.7}$ and $m = 2.7$.

In the macroscopic model, the transient heat conduction equation was solved numerically by the explicit finite-volume method [30] using a one-dimensional mesh of 30 equal volumes. In the microscopic model, a two-dimensional cellular automaton mesh of 300 (heat flow direction) $\times$ 100 (perpendicular to heat flow) square cells was adopted. Further details of the stochastic model can be found elsewhere [21].

Calculated grain macrostructures as a function of $\Delta T'_c$ are presented for $\Delta T_N = 2$ K (Fig. 6a) and 5 K (Fig. 6b). The CET region was defined as that where the aspect ratio of grains was between 0.3 and 0.4, as explained by Biscuola and Martorano [26]. As can be seen, the CET position might change significantly for constant $\Delta T_N$ when $\Delta T'_c$ is increased, showing the importance of $\Delta T'_c$. Furthermore, the CET frequently predicted with deterministic models for instantaneous nucleation ($\Delta T'_c = 0$) can be remarkably different from that obtained for a distribution of undercoolings.

In Fig. 7, solidification paths, defined as the dimensionless columnar front velocity as a function of the dimensionless temperature gradient at the front during solidification, are superimposed on the growth map of Fig. 3a. The front
velocity was calculated from successive front positions, determined as the location of the first solid cell observed from the bulk liquid towards the solid. Note that the solidification paths, calculated with a transient model, are plotted on growth maps based on a steady-state model. This implies that quasi-steady-state is assumed at the columnar front of the stochastic model. The quasi-steady-state assumption has successfully enabled steady-state models of dendritic [27] and nondendritic [13] growth to be extensively used in transient models of solidification [13,21]. Furthermore, in the stochastic model the columnar front blocking has been observed to occur according to the mechanical blocking criterion [26], as assumed to construct the growth maps.

Numerous simulations with the stochastic model indicated that the solidification paths are virtually unchanged with \( \Delta T^*_{\text{col}} \). Nevertheless, the length of each path, which ended at the CET, was different. Therefore, only the longest path is shown for each series of simulations of Fig. 6. In both cases, \( G^* \) is initially constant and \( V^* \) increases abruptly, reaching a constant value. Afterwards, \( G^* \) begins to decrease until the CET occurs.

The solidification path for \( \Delta T_N = 2 \) K intercepts the boundary curves at a region where \( V^* > 1 \) (\( \Delta T^*_{\text{col}} > 1 \)). As discussed previously, in this case an increase in \( \Delta T^*_{\text{col}} \) decreases the number of nucleated equiaxed grains, favoring columnar growth. This is observed in the macrostruc-
tures of Fig. 6a where the columnar region increases, raising the CET position, and the number of equiaxed grains decreases, increasing grain size.

For $\Delta T_{c} = 5$ K, the solidification path intercepts the boundary curves at $V^{*} < 1$ ($\Delta T_{c}^{*} < 1$) in Fig. 7. For instantaneous nucleation ($\Delta T_{c}^{*} = 0.01$), equiaxed growth and the CET are not observed in Fig. 6b, because the solidification path does not intercept the boundary curve in Fig. 7. As $\Delta T_{c}^{*}$ increases from 0.01 to 0.1 or 0.3, the path now intercepts the boundary curves, since the spread of the nucleation undercooling distribution increases and equiaxed grains begin to nucleate, causing a CET. A further increase in $\Delta T_{c}^{*}$ from 0.3 to 2 or 3 decreases again the number of nucleated equiaxed grains as explained before for $V^{*} < 1$, increasing the size of the columnar region.

5. Concluding remarks

A mathematical model for unidirectional steady-state solidification is proposed to predict the CET when equiaxed grains nucleate according to either a normal or a log-normal distribution of nucleation undercoolings. The present model reduces to Hunt’s [3] model when the spread of the distribution decreases, approaching instantaneous nucleation. Growth maps of dimensionless isotherm velocity ($V^{*}$) and columnar front undercooling ($\Delta T_{c}^{*}$) vs. dimensionless temperature gradient are constructed. The maps are affected by the spread of the distribution of nucleation undercoolings in two different ways. When $V^{*} > 1$ ($\Delta T_{c}^{*} > 1$), increasing the spread results in less favorable conditions for equiaxed growth, widening the field of columnar solidification. Otherwise ($V^{*} < 1$ and $\Delta T_{c}^{*} < 1$), there are only columnar grains for a vanishing distribution spread (instantaneous nucleation), but equiaxed growth is favored when the spread increases, narrowing the field of columnar solidification. For a larger spread, however, equiaxed grain nucleation is hindered, widening the columnar solidification field again. This behavior was confirmed in the grain macrostructures of a transient stochastic model. An increase in the distribution spread changed a completely columnar grain structure into one with $\sim 50\%$ of equiaxed grains. Therefore, predictions of the CET available in the literature and obtained with models that assume instantaneous nucleation conditions should be considered carefully.

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