Columnar to equiaxed transition in solidification processing

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Abstract

The columnar to equiaxed transition (CET) is a microstructural transition that has to be controlled during certain solidification processes. CET models and microstructure selection maps, based on nucleation and dendritic growth models, are briefly described. Processing maps can be established by combining a CET model with numerical calculations of local solidification conditions. Such processing maps are helpful for the control of the solidification microstructure. As examples, two processes are discussed: the epitaxial laser metal forming (E-LMF) process where a single crystalline superalloy part is repaired in single crystal form and equiaxed grains have to be avoided; and the arc welding of aluminium alloys where equiaxed grains are the preferred solidification structure. © 2001 Published by Elsevier Science Ltd.

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1. Introduction

Solidification processing is one of the important routes to produce materials, especially metals and alloys. The conditions for the transformation from liquid to solid, such as the temperature gradient and the growth rate, vary from process to process and in one process also as a function of time and space. These variations together with the different alloy compositions, lead to a multitude of microstructures and therefore material behaviour. One important transition which has to be well controlled is the so-called columnar to equiaxed transition.

2. The columnar to equiaxed transition (CET)

Generally solidification leads to two types of grain morphologies; columnar and equiaxed. After nucleation of crystals in an undercooled isothermal melt, growth is generally equiaxed, i.e. it proceeds equally in all directions and so does the local heat flux. In this way eutectic or dendritic morphologies form a polycrystalline solid with randomly oriented grains. On the other hand, the structure is called columnar if the heat flux is unidirectionally oriented (at least locally). In the first case, heat flows from the crystal into the undercooled melt (temperature gradient \( G < 0 \)), which in the second case of directional or columnar growth the heat flows from the superheated melt into the cooler solid (\( G > 0 \)). In equiaxed growth the solid–liquid interface is morphologically unstable leading in single phase solidification to dendritic grains, while in columnar growth, depending on the local growth conditions, one may grow planar, cellular or dendritic morphologies. A transition from columnar to equiaxed growth takes place when nucleation of equiaxed grains occurs in the liquid ahead of the columnar zone.

3. The importance of the CET in solidification processing

In this paper some solidification processes which make use of a close control of the CET either in eliminating it or in enhancing it, are discussed.

Processes where one has to completely eliminate this transition are directional solidification of superalloys (columnar crystals or single crystals) [1], and epitaxial laser metal forming (E-LMF) [2,3], a proprietary process for the repair of high value directionally cast single crystal components of aircraft engines.

Processes where one wants to enhance equiaxed solidification are welding [4,5] and continuous casting, for example of steel [6]. In the continuous casting of steel, economically the most important solidification process, large investments are made year after year for electromagnetic stirring devices to produce an equiaxed solidification mode in a large fraction of the casting, which improves the homogeneity of the product.

These features make it clear that a better knowledge of the CET is of great technical and economical importance. In
Nomenclature

\( a \)  Alloy constant, s/m
\( C_0 \)  Solute concentration, %
\( D \)  Solute diffusion coefficient in liquid, m²/s
\( D_b \)  Laser beam diameter, m
\( \phi \)  volume fraction of equiaxed grains, %
\( \phi_e \)  Extended volume fraction of equiaxed grains, %
\( G \)  Temperature gradient at the interface, K/m
\( \Gamma \)  Gibbs–Thomson coefficient, Km
\( I \)  Nucleation rate, 1/sm³
\( I_0 \)  Constant, 1/s
\( k \)  Distribution coefficient
\( K \)  Alloy constant, K²/s/m
\( m \)  Liquidus slope, K/%
\( n \)  Alloy constant
\( N \)  Number of active nuclei per unit volume, 1/m³
\( N_0 \)  Nucleant density, 1/m³
\( P \)  Laser power, W
\( T_0 \)  Preheating temperature of the substrate, K
\( V \)  Growth rate, m/s
\( V_b \)  Laser beam velocity, m/s
\( \Delta T \)  Undercooling, K
\( \Delta T_c \)  Solute undercooling of dendrite tip, K
\( \Delta T_e \)  Equilibrium liquidus–solidus interval, K
\( \Delta T_n \)  Nucleation undercooling, K

In this paper the authors concentrate on the CET of alloys which solidify with dendritic morphologies and on welding type processes such as laser metal forming or arc welding.

4. Microstructure models

The basis for understanding the CET is the growth of columnar dendrites. It is the diffusion field of these arrays of dendrites that is responsible for an undercooled liquid zone ahead of the dendrite tips which in turn may lead to nucleation of equiaxed grains. A CET model has therefore to take into account both nucleation and growth phenomena.

4.1. Columnar dendritic growth

Columnar dendritic growth develops in a positive temperature gradient. Due to this gradient a preferred orientation of the dendrites in the form of arrays with a typical spacing is obtained. Modelling of this growth can be done in several ways. Here, use is made of the so-called IMS dendrite growth theory [7]. In this theory the diffusion field is modelled by the Ivantsov field (isothermal paraboloid of revolution), and a unique solution to the problem is obtained by the so-called Marginal Stability criterion. From this theory the radius, the concentration and the undercooling of a single dendrite tip can be calculated [8].

For the sake of simplicity of discussion another simple model for dendrite growth is presented here. It applies to small driving forces, constant distribution coefficient, \( k \), and liquidus slope, \( m \), and shows the following approximate relationship (see this list of symbols):

\[
\Delta T = k^{1/2} V^{1/2}
\]

where the constant, \( K = 20 \Delta T_c k \Gamma / D \) and the unit solutal undercooling for small growth undercoolings, \( \Delta T_c k = C_0 m (k - 1) \) [8].

To model also cellular growth in the low velocity regime one has to add another term which is strongly temperature gradient dependent [9,10]:

\[
\Delta T = \frac{G D}{V} + k^{1/2} V^{1/2}
\]

The tip undercooling given by Eq. (2) is necessary to drive solute diffusion and capillary effects. This undercooling is at the origin of the constitutionally undercooled zone in the liquid adjacent to the front (Fig. 1b). Clearly the

![Fig. 1. showing: (a) the concentration profile ahead of a moving dendritic solid–liquid interface and (b) the equilibrium liquidus and local temperature profile ahead of a moving solid–liquid interface, leading to a constitutionally undercooled zone (hatched surface), calculated for the Al–Cu system (\( C_0 = 3 \) wt.%, \( V = 32 \) µm s⁻¹, \( G = 1000 \) K m⁻¹) [15].]
diffusion field for a single dendrite is different to that of a dendritic array, but it is assumed to be a reasonable approximation to it (Fig. 1a).

Note that the first term on the right hand side of Eq. (2) corresponds to cell growth. According to this model, cells grow at the limit of constitutional undercooling and therefore do not contribute to the development of a constitutionally undercooled zone. There should therefore be no CET in the cellular regime.

4.2. Equiaxed dendritic growth

Equiaxed dendrite growth may be calculated in a similar way as columnar growth, the only difference with respect to columnar dendritic growth being the temperature field around such dendrites [8]. As the Lewis number (the ratio of thermal to solutal diffusivity) of metals is very large, of the order 10^3-10^4, the temperature gradient which forms during normal growth conditions is negligible. The columnar dendritic growth model with G = 0 may then be used instead.

4.3. Nucleation

Nucleation ahead of the undercooled tip region can be evaluated according to classical nucleation theory leading to the following relationship [11]:

\[ I = (N_0 - N) \lambda_0 \exp \left( - \frac{K'}{\Delta T^2} \right) \]  

(3)

For the case considered here, the important parameters are \( N_0 \), the nuclei density, and \( K' \) a system constant.

The formation of an equiaxed zone can be enhanced by a large number of nuclei and a small nucleation undercooling. Ideal ‘nucleants’ are therefore the branches of dendrites which are in reality small crystals rather than nuclei. During stirring, dendrite branches developing close to the tips detach and convection carries them into the undercooled melt where they grow into grains. This process, called fragmentation, has been treated by others in Refs. [12,13].

5. CET models

A first CET model was developed by Hunt in 1984 [11]. This model uses an approximate dendrite growth theory, similar to that given above, and leads to a dendrite growth relationship of the form \( \Delta T = (C_0 V)^{1/2} \). This relationship is assumed to represent both columnar and equiaxed dendritic growth behaviour. Substituting (in steady state) undercooling for time, according to the relationship \( d\Delta T/dt = -VG \), and integrating the equiaxed growth velocity over undercooling, leads to:

\[ r = \int_{\Delta T}^{\Delta T} \left( \frac{K\Delta T^2}{VGG} \right) d\Delta T = \frac{K(\Delta T_c^2 - \Delta T_n^3)}{3 VGG} \]  

(4)

Three growth conditions may occur: (1) fully columnar, (2) columnar plus equiaxed and (3) fully equiaxed. Hunt suggested that if the mean distance grown by the columnar dendrites is larger than the diameter of equiaxed grain there would be sufficient directionality to recognize partly columnar growth. Under this condition, the extended volume fraction of equiaxed grains (\( \phi_c \)) equals 0.66 and the volume fraction of equiaxed grains (\( \phi \)) equals 0.49. Therefore, fully equiaxed growth is considered to occur if the volume fraction \( \phi \) is greater than 0.49, whereas the structure is assumed to be fully columnar if \( \phi \) is lower than 1% of the critical value of \( \phi_{c} \), i.e. 0.0066 [11].

The final result for a CE transition for a given inter-nuclei distance, \( N_0^{-1/3} \):

\[ G < 0.6N_0^{1/3} \Delta T_c \left( 1 - \frac{\Delta T_n^3}{\Delta T_c^3} \right) : \text{Fully equiaxed, } \phi > 0.49 \]  

(5)

\[ > G > 2.9N_0^{1/3} \Delta T_c \left( 1 - \frac{\Delta T_n^3}{\Delta T_c^3} \right) : \text{Fully columnar, } \phi < 0.0066 \]  

(6)

The latter relationship may also be obtained by dimensional arguments. The CET is observed when the equiaxed crystals impinge on each other before the columnar dendrites reach the equiaxed grains. Therefore one has to compare two distances with each other; (i) the distance between the nuclei, \( l_n \), and (ii) the length of the undercooled zone, i.e. the distance in the growth direction over which the crystals grow (when \( \Delta T_n = 0 \)) corresponds to the thermal length of the columnar dendrite tips, \( l_t \):

\[ l_n = N_0^{-1/3} \]  

(7)

\[ l_t = \Delta T_c/G \]  

(8)

Equating both distances leads to a relationship similar to that of Eq. (5):

\[ G = N_0^{1/3} \Delta T_c \]  

(9)

Some of the simplifications of the Hunt model have been successively relaxed [14,15]. The use of a more recent dendrite growth model, which takes into account non-equilibrium effects, shows some significant changes when compared with the Hunt model [15]. For example the slope of the log \( V \) - log \( G \) line in the high velocity regime increases from two to about three (Fig. 2), which has an important effect in the case of high \( V \) laser treatments.

In the case of a non-linear phase diagram [16] of the alloy the slope of the transition line at large velocities may be different from alloy to alloy. Also non-constant physical properties will influence the critical values of the transition. One can recalculate the dendrite equation and approximate the tip undercooling for a given processing window
as follows:
\[
\Delta T_c = \Delta T_0 (aV)^{1/n}
\]
(10)
where \(a\) and \(n\) are constants depending on the alloy system.

In the case of high \(V\) and \(G\), the nuclei density, as shown by Hunt [11], plays the critical role in CET. This allows setting the nucleation undercooling \(\Delta T_n\) to zero and by substituting Eq. (10) into Eq. (6), a simpler criterion for the transition can be proposed [3]. According to this criterion the microstructure will be predominantly columnar (\(\phi < 0.0066\)) when:
\[
\frac{G^n}{V} > Cst
\]
with:
\[
Cst = a \left( \frac{8.6\Delta T_0 N_0^{1/3}}{n + 1} \right)^n
\]

This simple relationship is only valid in the processing window where Eq. (10) is satisfied and when the nucleation undercooling can be neglected (high \(V\)). Therefore it is a useful criterion for processes such as welding or laser treatment. It implies that the ratio \(G^n/V\) must be larger or lower than a critical value to insure columnar or equiaxed morphologies. For a given \(\phi\) value, this critical value is dependant on the nuclei density \(N_0\), the interval of solidification \(\Delta T_0\) and alloy parameters \(a\) and \(n\).

To illustrate the use of the concepts presented above, a microstructure selection map (Fig. 3) was calculated for single crystal laser repair of single crystal turbine blades, using the full IMS-model [15]. The first transitions that occur at low velocities are the destabilisation of the planar-front into cells and into columnar dendrites. The next transition is the CET. This transition has been computed for the superalloy CMSX-4 through a database developed by Thermotech Ltd. [17], with \(N_0 = 2 \times 10^{13} \text{ m}^{-3}\).

![Figure 2](image1.png)

**Fig. 2.** Comparison of Hunt's analytical model and numerical model for the Al-Cu system [15].

![Figure 3](image2.png)

**Fig. 3.** Microstructure selection map for the superalloy CMSX-4. The rectangular insert represents the range of conditions that is typical for the laser process [3]. The evolution of \(G\) and \(V\) during melt pool solidification is calculated for initial substrate temperatures of 20°C (curve A) and 500°C (curve B). The squares represent the average values.
6. Processing maps

A combination of the processing parameters which can be evaluated either through analytical or numerical methods with the microstructure map allows a better understanding of the measures to be taken for avoiding or enhancing the CET. During laser metal forming, metal powder is injected into a molten pool formed by controlled laser heating (Fig. 4). The size and geometry of the molten pool is a function of the processing conditions such as the power of the laser $P$, the beam velocity $V_b$, the beam diameter $D_b$ and the preheating temperature $T_0$ [18].

The local solidification conditions ($G$, $V$) at the liquid–solid interface are a function of the melt pool geometry and the position of the interface. These conditions can be calculated by solving the heat diffusion equation. This has been done by Rosenthal [19] for a moving point-source of heat, and has been modified for a gaussian laser intensity profile [20]. The evolution of the local solidification conditions along the solidus isotherm are shown in Fig. 5, for a particular set of processing conditions [2,21].

By associating the local solidification conditions to the CET criterion as described above (Eq. (11), it is possible to predict the expected solidification morphology for a given set of processing parameters (see also Fig. 3, curves A and B). A processing map showing the morphology as a function

![Processing maps](image_url)

Fig. 4. Schematic of laser cladding and laser metal forming.

and $\Delta T_n = 2.5^\circ$C. The two curves represent the limits of columnar dendritic growth fronts ($\phi < 0.66\%$) and equiaxed solidification ($\phi > 49\%$). The dotted line is a representation of the $G^2V = Cst$ criterion (with $n = 3.4$). One can see that for the epitaxial laser metal forming (E-LMF) process, this criterion is more restrictive than the curve calculated with the more complete model. The two $G-V$ curves A and B are calculated processing conditions for laser remelting experiments (see the next section). The arrow shows the time evolution of $G$ and $V$ in the process.

![Processing maps](image_url)

Fig. 5. Evolution of the local solidification variables $G$ and $V$ as a function of depth $z$ ($z = 0$ corresponding to the surface), calculated for $P = 1500$ W, $V_b = 10$ mm/s, $D_b = 1$ mm, $T_0 = 20^\circ$C. The dotted lines represent average values.

![Processing maps](image_url)

Fig. 6. Processing map showing the dominant microstructure as a function of the laser power $P$: (a) laser scanning speed $V_s$; and (b) laser beam diameter $D_b$, for two preheating temperatures $T_0$ [3].
of processing conditions can be obtained (Fig. 6). In this way it is possible to produce single crystal welds on single crystal substrates of a superalloy, making the repair of high value turbine blades a reality [2,3].

Fig. 7 shows an optical micrograph and EBSD pattern (Electron Back Scattered Diffraction) representing the grain structure of a transverse section of such deposits which was formed by four successive layers. The crystallographic orientation of the substrate continues epitaxially into the deposit and the single crystal persists when nucleation of equiaxed grains ahead of the columnar dendritic front is avoided. It is only close to the top surface that some misoriented grains are formed.

A similar approach to analyse solidification processes has been used by Clarke et al. for the Gas Tungsten Arc welding of Al alloys [4]. In this case an equiaxed microstructure is the preferred one as the susceptibility to solidification cracking is thereby reduced. Processing maps such as those shown in Fig. 8 are therefore helpful to a better control of the microstructure of welds.

7. Conclusions

Following the original approach by Hunt, an empirical model for the CET has been developed which takes into
account realistic dendrite growth models for complex alloys such as superalloys. The modified CET criterion coupled with calculated processing conditions is used to produce processing maps which indicate the processing window for the epitaxial laser repair of single crystal turbine blades. The same approach can be used for any other solidification process to control the extent of the columnar and equiaxed zones.

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