The solidification characteristics of near rapid and supercooling directional solidification

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Abstract

In the comparison of the solidification characteristics of supercooling directional solidification (SDS) with constrained directional solidification (DS) and with the consideration of the inheritance of supercooled melt, the SDS technique established with the combination of melt supercooling and traditional DS was proposed. An exploring study on SDS techniques was also conducted using appropriate facilities, designed and manufactured by the authors’ laboratory and the deep supercooling of Cu–5.0%Ni alloy, and its DSs were implemented. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Traditional directional solidification (DS) techniques, such as HRS, LMC and zone melting liquid metal cooling are used with a temperature gradient $G_l > 0$ at the head of the S–L interface to obtain a fine or superfine dendrite microstructure. In recent years, with the development of deep supercooling and the supercooling solidification techniques, the solidification behavior of melt under the supercooled state with $G_l < 0$ at the S–L interface has attracted a great deal of attention. Earlier, Lux [1] investigated the dynamic supercooling solidification of superalloys. In 1989, Kiminiami et al. [2] predicted, after a series of studies on the solidification process for supercooled $P_{0.75}C_{0.25}Si_{0.65}$, that a special rapid solidification technology will appear if the traditional DS technique is combined with melt supercooling. In 1992, Stanescu [3], based on the above prediction, proposed the autonomous directional solidification technique (ADS) and put it into use for the study of single crystal growth for turbine blades. Although the ADS technique combined melt supercooling with DS, the substantial behavior of the technology is supposed to repress the nucleation of the melt by changing the cooling rate and obtaining dynamic supercooling (undercooling) (maximum of $T_K$ near 80 K) that is still considered as a mode of ‘solidification dynamically of supercooled melt’.

In view of the above considerations, the present investigation deals with the nucleation and crystal growth mechanism during the DS of supercooled melt at the relative lower growth velocity, which was obtained by the use of a superheat and glass purifying method to achieve a thermo-dynamic deep supercooling of the melt in order to reveal the nature of directional, crystal growth of supercooled melt at $G_l < 0$ in front of the S–L interface [4].

2. Constrained directional solidification

Traditional DS for making turbine blade castings is characterized by one-dimensional heat flow, a positive temperature gradient at the S–L interface and a parallel crystal growth direction with the heat flow. For constrained DS growth, the well-known constitutional supercooling (CS) theory proposed by Tiller et al. [5] can be used to determine the interface stability:

$$G/V = \Delta T/D.$$  

(1)

When the ratio of temperature gradient to growth velocity on the left-hand side of Eq. (1) is less than that on the right-hand side, CS and interface instability will occur, and the planar interface transforms to a cellular structure. With an increase in CS, the cellular interface becomes dendrite. Fig. 1 shows the relationship of interface morphology with respect to $G_l$ and $V$.

For given alloys, the interface morphology depends on the two parameters $G_l/V$ and $G_lV$. The former mainly
determines the morphology, while the latter generally determines the microstructure including arm spacing. The results show that with the increase in the cooling rate \((G_c V)\), the interface morphologies change from coarse dendrites with developed sidebranches to fine dendrites and even superfine dendrite-cellular structure that leads to a corresponding increase in mechanical properties [6]. Therefore, the increasing of the temperature gradient at the S–L interface is still considered as one of the key methods to improve single crystal superalloys further. Practically, in the last 30 years, with continuous improvement of technology from the power down, HRS and LMC could be attributed to the increase of temperature gradient, which still will be the aim of the development of the DS technique.

3. Deep supercooling directional solidification

While comparing the traditional DS with supercooling DS, the substantial difference can be attributed to \(G_c > 0\) for the former, in which the heat loss depends only on the conduction of the solidified part, with tip undercooling approaching zero; whereas for the latter, \(G_c < 0\), in which the heat loss is determined both by the solidified part and the supercooled melt so that dendrite tip undercooling can be extended to several hundred degrees, according to the requirements. Fig. 2 shows their basic differences [1].

3.1. Experimental results and discussion

3.1.1. Solidification microstructure of SDS samples

Fig. 3 shows the solidification structure of SDS schematically. It can be seen that from the bottom (the part of stimulated nucleation) to the top of the sample, there appears in sequence fine equiaxed crystal, transient area, columnar dendrite, and coarse equiaxed area, demonstrating the regularity of morphological evolution. The microstructure in the columnar dendrite crystal area of the SDS under various supercooling conditions in contrast with that of LMC is shown in Fig. 4. It is seen that the dendrite structure by SDS is equivalent to that by LMC; its arm trunk is revealed as straight and fine, the average deviation of crystal orientation from the axial line is about 5.8°, the primary arm spacing is about 30 μm, and the secondary sidebranches are degenerated significantly. According to the statistical results of experiments, the area of preferred columnar structure occupied along about 60% of the whole length.
3.1.2. Solidification behavior

In the process of supercooling, DS crystal growth is conducted in a supercooled melt. In general, the dendrite growth is a fundamental mode for a supercooled melt, but diffusionless solidification might also occur when the melt supercooling becomes large enough. From the aspect of interface morphology during solidification, diffusionless solidification favours the interface to be planar. Trivedi and Kurz [7] gave a criterion of the absolute stability of planar interface in undercooled melt:

\[ V_{\text{abs}} = (V_{\text{abs}})_{c} + (V_{\text{abs}})_{T} \quad \text{or} \]

\[ V_{\text{abs}} = \frac{\Delta T_{0} \times D}{T \times \kappa} + \frac{\alpha_{t} \times \Delta H}{T \times C_{p}}, \quad (2) \]

where \( \alpha_{t}, \Delta H \) and \( C_{p} \) are the thermodiffusion coefficient, the latent heat crystallization, and the specific heat respectively; \( D, \Delta T_{0} \) and \( k \) are the solutal-diffusion coefficient, the interval of solids and liquid at the concentration of \( C_{0} \) and the equilibrium solute partition coefficient and \( T \) is the Gibbs–Thompson coefficient. From Eq. (2) the solid–liquid interface appears as absolute stability while growth velocity \( V > V_{\text{abs}} \), which leads the supercooled melt to solidify with a planar interface, and the solute trapping effect causes the melt to suffer diffusionless solidification under large supercooling. In fact, \( T-K \) theory was deduced with the assumption that the supercooling solidification is an adiabatic process, i.e. the temperature gradient at the interface in solid \( G_{S} = 0 \), which is not in agreement with the practical condition of the supercooling solidification process, where \( G_{S} > 0 \). If, with the consideration of \( G_{S} > 0 \) in solid and \( G_{L} < 0 \) in liquid in a supercooling DS process, the critical velocity of absolute stability of a planar interface can be given as follows [8]:

\[ V_{\text{abs}} = (V_{\text{abs}})_{c} + 2s(V_{\text{abs}})_{T}, \quad (3) \]

where \( s \) is a stability parameter, which is a function of \( \alpha \) and \( \beta \), and its expression is

\[
s = \max \left\{ \frac{1}{Y^{2}} \left[ 2(\eta - 1) - \frac{-\alpha + \sqrt{\alpha^{2} + \beta^{2}}}{\alpha + \sqrt{\alpha^{2} + \beta^{2}}} \right]^{1/2} + 2 \eta \frac{\beta + \beta(1 + Y^{2})^{1/2}}{\alpha + \sqrt{\alpha^{2} + \beta^{2}} + \beta(1 + Y^{2})^{1/2}} \right\}_{\text{max}}.
\]

Fig. 4. Microstructure of SDS samples in contrast with that of LMC: (a) \( \Delta T = 63 \text{K} \); (b) \( \Delta T = 136 \text{K} \); (c) \( \Delta T = 158 \text{K} \); (d) LMC \( (G_{L} = 250 \text{K cm}^{-1}, V = 500 \mu \text{m s}^{-1}) \).

where \( \alpha = \alpha_{t}/\alpha_{s} \) is the thermodiffusion coefficient, \( \beta = \beta_{s}/\beta_{t} \) is the thermoconductivity and \( \eta \) is the fraction of latent heat, released through supercooled melt.

According to the relation between latent heat fraction \( \eta \) and the stability parameter \( s \) given by Ludwig:

(a) \( s = 0.5 \) as \( \eta = 1 \), so that the whole latent heat of solidification releases through the supercooled melt, and Eq. (3) will be degenerated to Eq. (2). Applying the data of Cu–5.0%Ni alloy, the critical velocity of absolute stability for a planar interface \( V_{\text{abs}} = 4.3 \times 10^{3} \text{ m/s} \).

(b) \( s = 0 \) as \( \eta = 0.5 \), so that only half of the latent heat is released through the supercooled melt and Eq. (3) retrograded to the form of DS theory, its critical velocity for absolute stability of the planar interface being \( V_{\text{abs}} = 0.28 \text{ m/s} \). Thus, from the calculation given above,
the range of the critical rate of the absolute planar interfacial stability is given as \( V_{\text{abs}} = (0.28 \pm 4.3 \times 10^3) \) ms. In other words, in a directional deep supercooling solidification process, the condition of segregation-free solidification favors the growth rate being within the \( V_{\text{abs}} \) range. However, the experimentally determined average solidification rate is approximately 0.53 cm/s, which is much smaller than the theoretical \( V_{\text{abs}} \). Therefore, in a directional deep supercooling solidification process, even though the crystal growth is in a supercooled melt, segregation-free solidification will not occur. The crystal growth still takes the form of dendritic growth.

4. Conclusions

1. Thermodynamic SDS has significant difference with conventional DS both in solidification behavior and in technological process. Under reasonable design of controlled temperatures and stimulated method, the supercooled melt can directionally crystallize rapidly.

2. The microscopic structure under directional deep supercooling solidification has four different character regions from the bottom to the top: they are fine equiaxed crystal area, transient area, columnar dendrite area and coarse equiaxed area.

3. Columnar (dendrites) structure occupies over 60% of the total sample length. The primary arm spacing is about 30 \( \mu \)m, corresponding to that by LMC (\( G_L = 250 \) K cm\(^{-1} \), \( V = 500 \) \( \mu \)m s\(^{-1} \)), the dendrite trunk appears straight and fine, as the deviation of dendrite orientation with axial direction is less than 5.8° on average.

4. In the directional deep supercooling solidification process, the solidification rates are much smaller than \( V_{\text{abs}} \). Therefore, the crystal growth take the form of dendrite.

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