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SIMULATION OF THE DENDRITIC SOLIDIFICATION DURING SINGLE ROLLER QUENCHING

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ABSTRACT

The dendritic solidification under conditions characteristic to the single roller rapid quenching technique has been modeled solving the two-dimensional steady state thermal transport equation by the finite difference method. The thermal and chemical diffusion in front of the dendrite tip is described by the theory of Kurz, Giovanola and Trivedi (Acta Metall., 1986, 34, 823). The kinetic undercooling and the non-equilibrium distribution coefficient have been taken into account. A simplified treatment is suggested for the mushy-zone. The planar to dendritic transition has been related to the technological parameters (heat transfer coefficient, roller temperature, etc.).

1. INTRODUCTION

The microstructure has an important role in determining the physical properties of crystalline alloys. Rapid solidification is a suitable method to influence the growth morphology. By varying the growth rate various microstructures (cellular, dendritic, etc.) can be obtained [1,2]. In the past decade a spectacular advance in the understanding of rapid solidification phenomena has been witnessed [3-5]. Numerical simulations utilizing the knowledge accumulated on rapid solidification may have an important role in tailoring the microstructure for different applications.

The single roller rapid quenching (SRQ) techniques (planar flow casting [6], melt drag [7], melt extraction [8] etc.) - providing cooling rates in the 10^5 - 10^6 K/s range - are promising candidates for industrial scale production of microcrystalline alloys. According to the experiments the typical microstructure of iron-based strips produced by these techniques is partly or fully dendritic [9,10]. An analysis which relates the change of microstructure to the technological conditions would be of practical importance.

In this work a numerical simulation of dendritic solidification under thermal conditions characteristic to the single roller rapid quenching techniques is presented. The relation between parameters determining the cooling rate (heat transfer coefficient, roller temperature) are studied in case of an Fe-Si alloy.

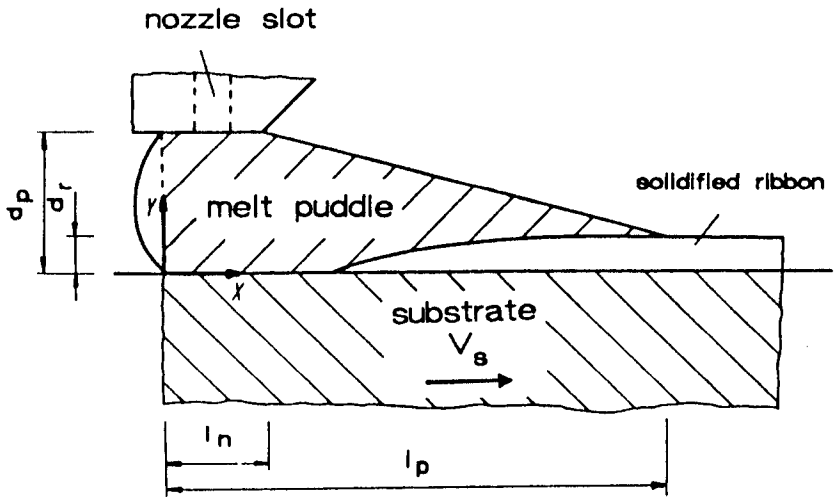
2. THE NUMERICAL MODEL

The heat transport is described by the steady state two-dimensional thermal transport equation, which under the conditions characteristic to the SRQ techniques can be simplified to the analog of the one-dimensional, time-dependent problem [11]. The heat release during solidification is taken into account through a source term:

$$V_o \frac{\partial T}{\partial x} = \alpha \frac{\partial^2 T}{\partial y^2} + V_o \frac{\partial f_s}{\partial x} \cdot \frac{\Delta H_f}{\rho c_p} \qquad (1)$$

where T is the temperature, V_o is the surface velocity of the roller, α is the thermal diffusivity, ρ is the density, c_p is the specific heat, ΔH_f is the heat of fusion, x and y are spatial coordinates (see Fig. 1), while f_s is the crystalline fraction, calculated as described below. This equation is solved by the explicit finite difference method on a rectangular grid fitted to the experimental geometry (see Fig. 1). A similar equation, without the source term is applied for the thermal transport in the roller. The heat transfer at the melt-roller interface is described by a heat transfer coefficient, h, while the heat radiation on free surfaces is neglected.

It is assumed that the solidification starts with a surface induced heterogeneous nucleation at the melt-roller interface. Since the parameters determining the heterogeneous nucleation and especially the transient effects (which may be important at high cooling rates [12]) are generally not known, the undercooling necessary for the nucleation is treated as an external parameter. Once the nucleus is formed, it will grow as determined by the local thermal and solute transport. A planar growth front is assumed until the



d_p: gap between nozzle and wheel
d_r: thickness of the ribbon
l_n: melt pool lenght at the nozzle
l_p: total lenght of the melt pool

Fig. 1 Schematic drawing of the geometry of the planar flow casting method.

growth rate exceeds the limit of absolute stability (V_a) [13]. This velocity is normally in the order of 1000 m/s for equiaxial growth conditions with zero temperature gradient in the solid. It can be shown, however, that this value is reduced to about few times 10 cm/s in the presence of the large positive gradients characteristic to the SRQ methods [14]. The growth rate (V) of the planar front is usually described by the equation:

$$V(T) = V_o \cdot \left[1 - \exp\left\{-\frac{\Delta G(T)}{R \cdot T}\right\}\right], \qquad (2)$$

where V_o, R and $\Delta G(T)$ are the velocity of sound, the universal gas constant and the Gibbs free energy difference between the melt and the solid, respectively. For small undercoolings equation 2 can be transformed to $V(T)=\mu_o \cdot \Delta T_k$, where μ_o is the linear velocity coefficient and ΔT_k is the kinetic undercooling [4].

When the growth rate drops below the limit of absolute stability a fully developed dendritic morphology is assumed. The advance of the dendrite tips is described by the theory of Kurz, Giovanola and Trivedi [13], while atomic attachment kinetics and solute trapping (through a non-equilibrium partition coefficient [15]) is taken into account. This treatment is strictly valid for the steady state growth of a single dendrite, Zimmermann et al. have shown, however, that it may be applied under rapid quenching conditions as well [16].

The region between the dendrite tips and the front of complete solidification (mushy zone) has to be treated differently. Here - except for the near-tip region - the diffusion field of the dendrites overlap. Because of the complexity of the problem no exact description is available (even for steady state) at present. Following the treatment by Clyne we assume that there exists an $f_s = f_s(T)$ function which describes the variation of the solid fraction with the temperature between the dendrite tip and root [17]. Strictly, such a relation may exist for steady state and a constant temperature gradient in the mushy zone. The exact treatment, however, would require the solution of the thermal and solute transport equations for every particular dendrite in the diffusion field of its neighbors which makes the problem much too complicated.

An approximate $f_s = f_s(C_s^*)$ relation has recently been proposed by Giovanola and Kurz [18], where C_s^* is the composition of the solid:

$$f_s = a_1 \cdot C_s^{*2} + a_2 \cdot C_s^* + a_3 \qquad (0 \le f_s \le f_x), \qquad (3a)$$

$$f_s = 1 + (f_x - 1) \cdot \left[\frac{C_s^*}{C_x}\right]^{1/(k-1)} \qquad (f_x \le f_s \le 1), \qquad (3b)$$

where f_x, C_x, a_1, a_2 and a_3 are parameters determined from the limiting values and the mass balance, while k is the partition coefficient. Using the non-equilibrium solidus line obtained with the growth rate dependent partition coefficient this relation can be transformed to an $f_s = f_s(T)$ function. In the present work we adopt this technique. It can be shown that the treatment by Giovanola and Kurz may be applied for growth rates less than 0.05 V_a. Thus for $0.05\ V_a < V < V_a$ we apply a different f_s-T relation:

$$f_s = a_1 \cdot C_s^{*2} + a_2 \cdot C_s^* + a_3, \qquad (4)$$

where the parameters a_1, a_2 and a_3 are determined from (i) the mass balance and conditions (ii) for $f_s = 0$ $C_s^* = C_{st}^*$ and (iii) at $f_s = 1$ $C_s^* = C_1$. Here C_{st}^* is concentration of the solid at the dendrite tip, while C_1 is obtained by a linear interpolation between $C_{xe} = C_x|_{v = 0.05 V_a}$ from the Giovanola-Kurz treat-

ment [18] and the initial composition of the melt, C_o as a function of growth velocity. This construction ensures a physically reasonable smooth transition from the mushy zone to a planar interface.

3. PHYSICAL PROPERTIES AND CASTING CONDITIONS

The calculations have been performed for the $Fe_{91.5}Si_{9.5}$ alloy cast on a copper roller. The thermal properties of the alloy are taken as follows: Heat conductivity, 35 $Wm^{-1}s^{-1}$; specific heat, $5.74 \cdot 10^6$ $Jm^{-3}K^{-1}$; density, 7840 kgm^{-3}; heat of fusion, $1.93 \cdot 10^9$ Jm^{-3}; Gibbs-Thomson coefficient, $1.9 \cdot 10^{-7}$ Km; equilibrium distribution coefficient, 0.75; while the melting point and the eutectic temperature are 1733 and 1473 K, respectively. The linear velocity coefficient used in the simplified form of equation (2) is taken as $\mu_o = 1.4$ $ms^{-1}K^{-1}$. The Gibbs free energy difference $\Delta G(T)$ between the melt and crystal was calculated using an approximation by Turnbull [19]. The non-equilibrium partition coefficient has been calculated by a formula proposed by Aziz [15].

Casting conditions typical in case of planar flow casting of 130 μm thick Fe-Si ribbons [20] have been chosen. The surface velocity of the roller is 10 m/s, the initial temperature of the melt is 1833 K, while in accordance with the experiments [20] the temperature of the roller (T_r) is varied between 200 C and 700 C.

The heat transfer coefficient between iron based alloys and a copper roller is in the order of few times 10^5 $Wm^{-2}K^{-1}$ [21] which may be influenced by the roller velocity and the initial melt temperature, roller temperature, etc. In order to take these effects into account h has been varied between $5 \cdot 10^4$ and $2 \cdot 10^5$ $Wm^{-2}K^{-1}$.

Although rather large undercoolings have been achieved by containerless methods (appr. 300 K [22]), due to the assumed heterogeneous nucleation at the melt-roller interface, the undercooling necessary for the nucleation is chosen as 100 K.

4. RESULTS AND DISCUSSION

Computations have been performed to determine the velocity corresponding to the absolute stability limit as a function of the thermal gradients in the solid and in the melt. In accordance with former results [13] if the temperature gradient is negligible in the solid and a large negative gradient is supposed in the melt, V_a is in the order of 1000 m/s. In contrast, if a positive gradient is appr. equal to or exceeding the magnitude of the negative gradient in the melt V_a is determined by the solute limit of absolute stability [13], which is $V_a = 0.28$ m/s in our case. In the present treatment we assume that the latter criterion can be used. The calculated temperature gradients are consistent with this assumption. A detailed analysis of this phenomenon will be presented elsewhere [14].

It is worth noticing that V_a corresponds to a certain undercooling, ΔT_a (in our case appr. 33 K). As a consequence a fully dendritic microstructure may develop if only the undercooling necessary for nucleation is smaller than ΔT_a. This effect might be utilized to assess the nucleation temperature.

The relation between the solid fraction and the composition of the solid calculated using equations 3a, 3b and 4 is shown in Fig. 2 as a function of the growth velocity. In accordance with the treatment proposed by Giovanola and Kurz a complete mixing of the interdendritic melt is assumed behind the tip region described by Scheil's solution [23], which can be applied right up to the dendrite tip at $V/V_a \cong 0$. With increasing growth velocity the region in which complete mixing is a good approximation shrinks, while disappearing at $V \cong 0.05\ V_a$. For larger velocities the composition difference between the dendrite tip and root decreases rapidly.

Using the solidification mechanism described by equations 2-4 we have solved equation 1 and determined the distribution of the temperature and the

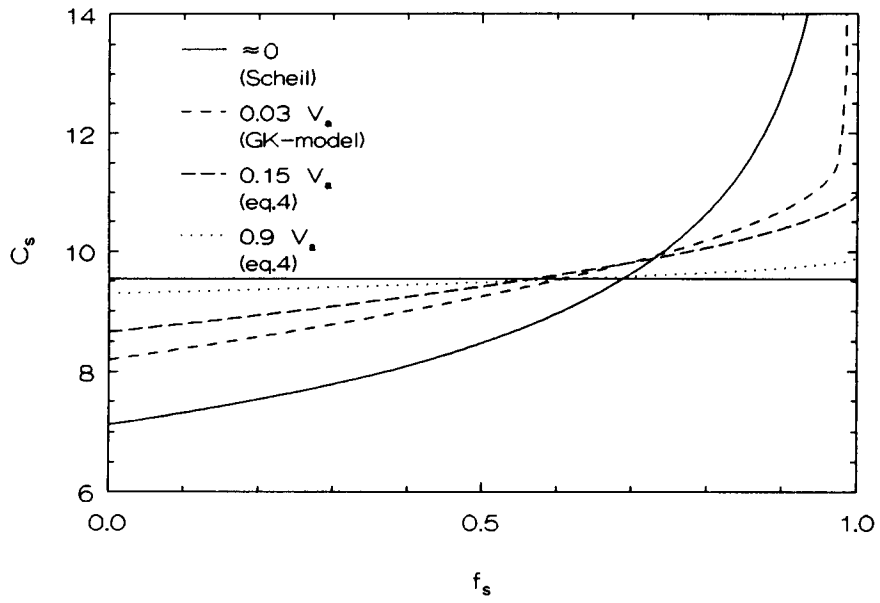


Fig. 2 The calculated solid fraction-solid concentration relations.

solid fraction. The cross-sectional variation of the maximum and the minimum of the solute concentration have been computed as well.

The results corresponding to the casting conditions specified in section 3., and to h= $2 \cdot 10^5$ $Wm^{-2}K^{-1}$ and Tr= 700 C are presented in Figs. 3-5. It is found that the solidification starts with a planar interface, resulting in an appr. 2.6 μm thick non-dendritic layer. It is followed by a mushy zone of thickness varying from 0 to appr. 40 μm as a function of the distance from the nozzle (see Fig. 3). The corresponding cross-sectional temperature distributions are shown in Fig. 4. The heat release during solidification decreases the large positive temperature gradient found at the nucleation, which remains, however, positive during the whole cooling process. In case of small heat transfer coefficients (h≅ $5 \cdot 10^4$ $Wm^{-2}K^{-1}$) a pronounced recalescence can be seen after the nucleation event, which results in a negative thermal gradient ahead of the solidification front. Despite this fact the assumption used in the calculation of V_a, - i.e. that the thermal gradient in the solid exceeds the absolute value of the gradient in the melt -, is fulfilled in all the examined cases.

For velocities larger than the absolute stability limit (i.e. in the non-dendritic layer) the composition is equal to that of the initial melt, $C_s^* = C_o$. In the dendritic region microsegregations occur. The upper and lower limit of the variation of the solute concentration, - i.e. the composition of the fraction of the cell solidified the first (i.e. the composition of the dendrite tip) and that of the last interdendritic liquid in the cell (i.e. the composition at the root of the dendrite) -, is shown in Fig. 5. It is found that due to the almost constant growth velocity both are approximately constant (C_{min}≅ 8.6 at.% and C_{max}≅ 11.1 at.%) in the upper part of the ribbon.

In order to study the influence of casting conditions on the microstructure, calculations have been performed for different heat transfer coefficients and roller temperatures. It is found that the thickness of the non-dendritic layer increases with decreasing h or with increasing Tr. These trends can be understood on the same basis. Both a decreasing h or an increas-

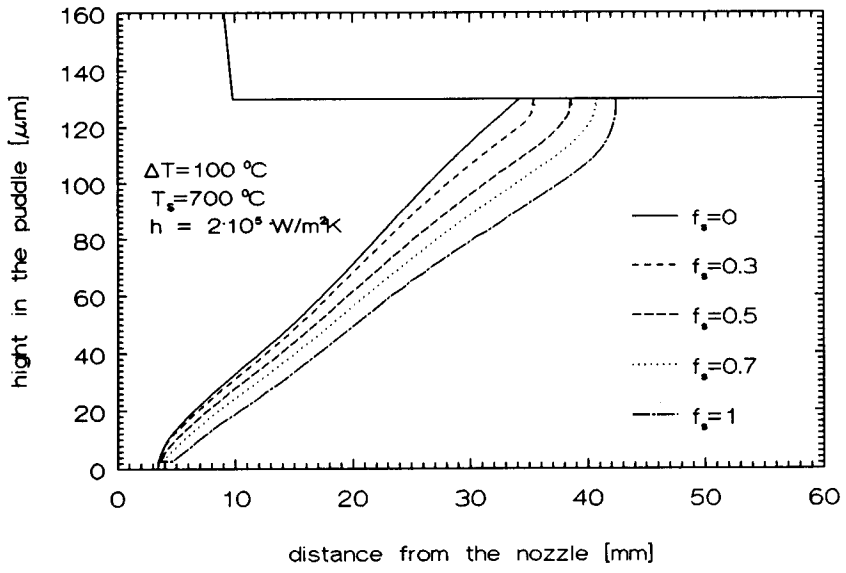


Fig. 3 The calculated spatial distribution of the solid fraction.

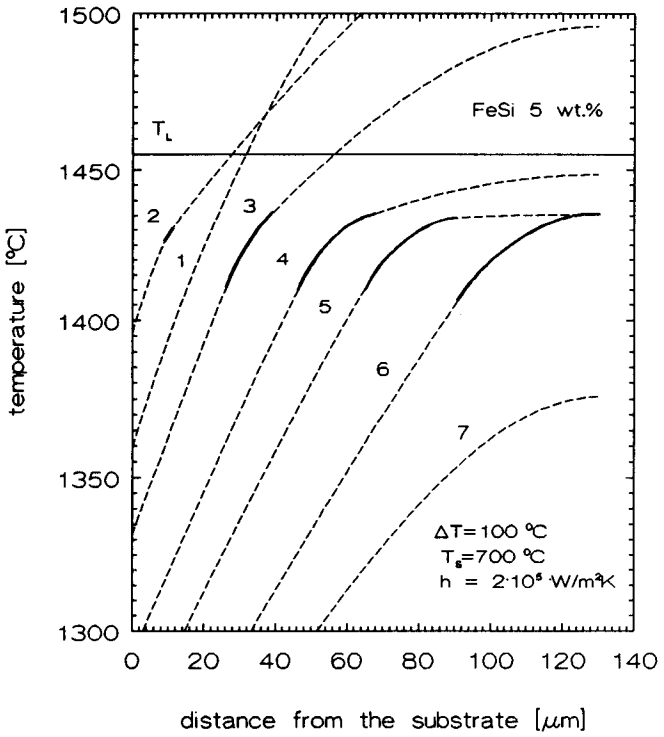


Fig. 4 The calculated cross-sectional temperature distributions. (1:at nucleation, 2-6: for different positions of the mushy zone (solid lines), 7: after complete solidification)

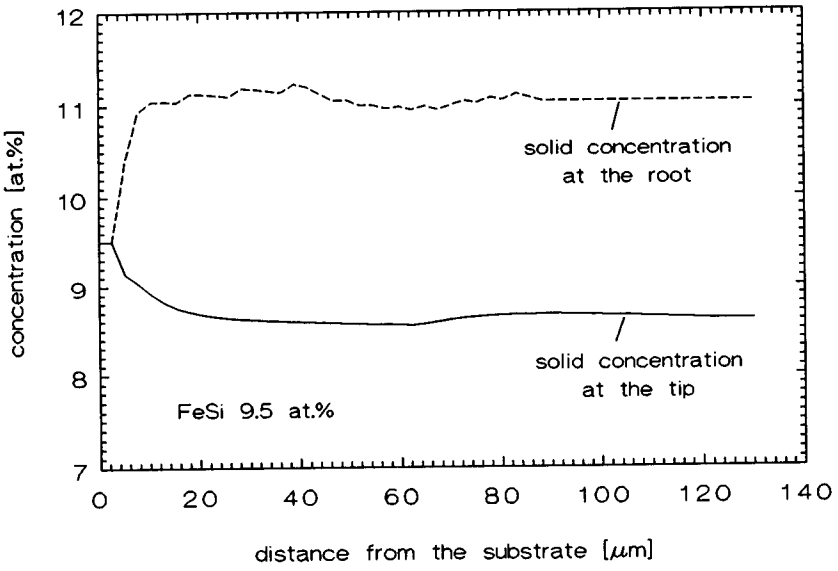


Fig. 5 The calculated cross-sectional concentration profiles.

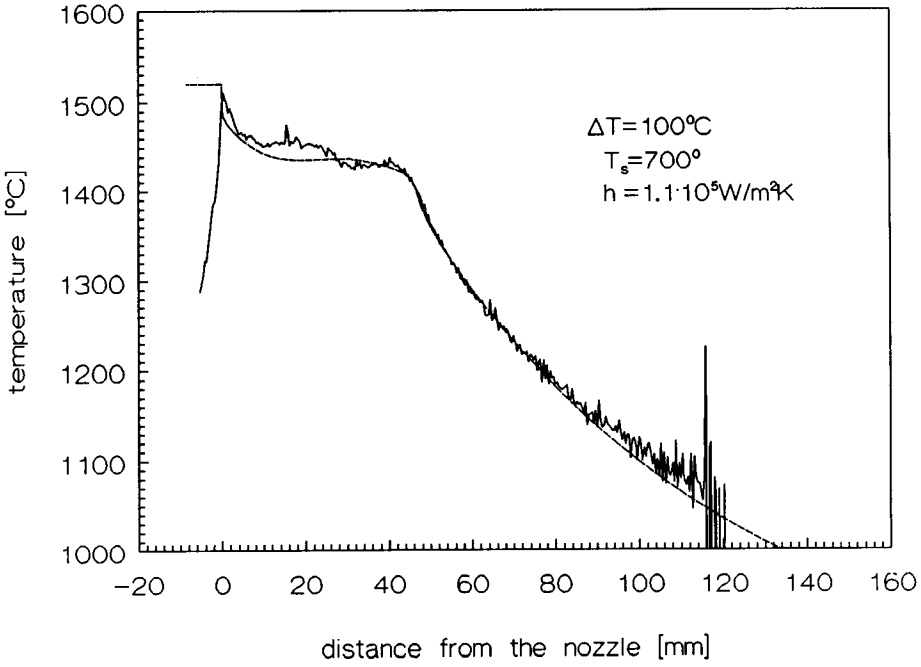


Fig. 6 The experimental (solid line) and the best fitting calculated (dashed line) temperature distributions on the top of the ribbon.

ing T_r reduces the temperature gradient in the alloy, which - assuming a fixed nucleation temperature - results in an extended region of $\Delta T > \Delta T_a$.

To obtain realistic h and T_r values, the temperature distribution at the free surface has been calculated and compared to the experimentally determined data [20] (see Fig. 6). The best fit was obtained with h= $1.1 \cdot 10^5$ $Wm^{-2}K^{-1}$ and T_r= 700 C. This value of the heat transfer coefficient is in a good agreement with that obtained by Takeshita and Shingu out of the melt puddle for a Ni-P alloy cast on copper, steel and stainless steel rollers [24]. The thickness of the corresponding non-dendritic region is about 4 μm.

A detailed comparison between the predictions and the experiments is under way.

5. SUMMARY

A numerical model describing the dendritic solidification in the single roller rapid quenching methods has been presented. The influence of casting conditions on the planar to dendritic transformation has been studied. In accordance with the experiments the presence of a non-dendritic zone at the melt-roller interface is predicted. It is found that the thickness of this non-dendritic region can efficiently be influenced by both the heat transfer coefficient and the roller temperature.

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