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# A new microsegregation model for rapid solidification multicomponent alloys and its application to single-crystal nickel-base superalloys of laser rapid directional solidification



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#### ABSTRACT

The microsegregation in single-crystal nickel-base superalloys, which may result in many undesired precipitated phases, can be effectively reduced by rapid solidification. A new microsegregation model for rapid solidification multicomponent alloys was developed in this paper to predict the microsegregation in the single-crystal superalloys of laser rapid directional solidification and study the influence of processing parameters on the microsegregation behavior. To verify the predicted accuracy of the present model, the concentration profiles and microsegregation ranges under different processing conditions were examined and compared with those calculated by different models. Compared with previous models, this multicomponent model not only has definite physical meaning but also takes nonequilibrium solidification and dendritic tip undercooling into account. Therefore, it is suitable for a wide range of solidification conditions and can obtain better predicted results for complex multicomponent alloys. Moreover, although the microsegregation behavior depends primarily on the solidification velocity, a high temperature gradient also contributes to reducing the microsegregation. Consequently, the microsegregation can be effectively controlled by using a set of processing parameters that can obtain high temperature gradient and solidification velocity. Such a model contributes to the control of the microsegregation while determining relevant processing windows for the preparation of superalloy components using a controlled single-crystal laser processing.

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#### 1. Introduction

Nickel-base single-crystal (SX) superalloys are unique hightemperature materials used as turbine blades and vanes due to their excellent mechanical properties at elevated temperatures [1,2]. Nevertheless, the microsegregation is inevitable during the solidification processing of superalloys, which may result in many undesired microstructures, such as the formation of low-meltingpoint eutectic. These defects will lead to severe performance

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degradation of SX components. Recently, laser processing, *e.g.*, laser additive manufacturing, has presented its potential for rapid forming and precise repair of SX components because it allows rapid and accurate addition of controlled amounts of material to required locations with low heat inputs and high cooling rates [3–10]. More importantly, it, as a rapid solidification (RS) process, should be an effective processing method that can reduce and control the microsegregation of SX alloys. Therefore, it is desirable to be able to model the microsegregation behavior during the laser rapid directional solidification (LRDS) process of SX alloys, which contributes to reducing and controlling the microsegregation during the preparation of SX components.

For a long time, many researchers have attempted to model the solute redistribution during solidification processing [11–18]. The

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main difference between these models is whether considering the solute diffusion in the solid (back diffusion). If the solutes in the solid are homogeneous, for instance, the solute diffusivity is very high (strong back diffusion), microsegregation can be described by the classical lever rule. Conversely, if the solidification process is assumed without solute diffusion in the solid (no back diffusion), microsegregation can be described by the well-known Scheil's equation [11]. It is clear that the two models under different extreme conditions deviate significantly from actual solidification conditions in most cases. As a result, by assuming intermediate back diffusions in the solid, a microsegregation model, derived by Brody and Flemings, can be written as [13]:

$$C_{s}^{*} = kC_{0}[1 - (1 - 2\alpha k)f_{s}]^{\frac{k-1}{1-2\alpha k}}$$
(1)

where  $C_s^*$  is the solute concentration in the solid,  $f_s$  is the solid fraction, k is the equilibrium partition coefficient,  $C_0$  is the initial alloy concentration,  $\alpha$  is the dimensionless back-diffusion coefficient and defined as  $\alpha = D_s t_f / L^2$ ,  $D_s$  is the diffusion coefficient in solid,  $t_f$  is the local solidification time and L is the diffusion length. For solving the problem that the solute for a high  $\alpha$  is non-conservation, Clyne and Kurz (CK) [11,14] suggested replacing the  $\alpha$  by a modified back-diffusion coefficient  $\alpha'$ :

$$\alpha t = \alpha [1 - \exp(-1/\alpha)] - 0.5 \exp(-1/2\alpha)$$
(2)

Such a modification successfully unified above rules and expanded the scope of applications (any  $\alpha$  values). Besides, Ohnaka [15] and Kobayashi [16,17] derived more accurate approximate solutions for microsegregation. However, all of these models did not take dendritic tip undercooling and nonequilibrium solidification into account. As a result, solidification predicted by these models begins at the equilibrium liquidus and  $C_s^* = kC_0$ , which limited their applications to normal (slow) solidification process. Moreover, although Solari and Biloni [18] dealt with the solute diffusion boundary layer at the dendritic tip and in the interdendritic liquid, the tip concentration was evaluated by the Burden-Hunt model [19], which also limited its applications to low and medium growth rates. On the other hand, these microsegregation models were established based on simple binary-alloy systems under conventional solidification conditions; hence the actual (nonequilibrium) solidus-liquidus interval,  $\Delta T$ , relating to the  $t_{f}$ , can be approximated by the equilibrium solidus-liquidus interval,  $\Delta T_0$ . However, for a multicomponent alloy such as SX superalloys, this consideration is not true and the  $\Delta T'$ , which strongly depends upon the tip temperature, is difficult to be evaluated especially under RS conditions. As a result, the evaluation of the microsegregation of the LRDS SX alloys requires not only a RS model but also a multicomponent expansion.

Recently, Giovanola and Kurz (GK) [11,12] proposed an approximate model of microsegregation under RS conditions. Owing to large solute pile-up around the rapidly growing dendritic tips, the solute concentration in interdendritic liquid is heterogeneous at low solid fractions. This model therefore consists of two parts. (1) When the solid fraction is less than a critical value,  $f_x$ , the solute concentration in the solid can be expressed as an empirical quadratic polynomial:

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

(2) When the solid fraction is high enough  $(f_s > f_x)$  to contact the diffusion boundary layers in interdendritic, the residual interdendritic liquid can again be assumed as homogeneous. Accordingly, the solute concentration in the solid can be written as a Scheil's equation:

$$f_{s} = 1 + (f_{x} - 1) \left( C_{s}^{*} / C_{x} \right)^{\frac{1}{k-1}} (f_{x} \le f_{s} \le 1)$$
(4)

where  $a_1$ ,  $a_2$ ,  $a_3$ ,  $f_x$ ,  $C_x$  are five unknown parameters. The corresponding equations necessary for solving these unknowns can be obtained elsewhere [12]. Although this model can be applied to multicomponent systems and its calculated solute content and concentration profile can agree reasonably with experimental results, two problems in this model are intrinsic. Firstly, the modeling of solute concentration for low solid fractions is an empirical expression, which implies that the physical meanings of this model and many coefficients, such as  $a_1$ ,  $a_2$ ,  $a_3$ , are ambiguous. Secondly, the modeling of solute concentration for high solid fractions follows a Scheil's equation, which leads to an exceedingly sharp increase in the solute concentration when the solid fraction tends to unity. Therefore, a new microsegregation model with definite physical meanings for rapid solidification multicomponent alloys is necessary for reducing and controlling the microsegregation in the LRDS SX components.

In this paper, a new theoretical model for predicting the microsegregation behavior of multicomponent alloys during the LRDS process was derived. Additionally, the influence of laser processing parameters on the microsegregation behavior of a SX alloy was studied. To verify the microsegregation degrees predicted by the model, laser remelting experiments were conducted to obtain various LRDS conditions, and resulting concentration profiles and microsegregation ranges under different processing conditions were examined and compared with those calculated by different microsegregation models. Such a multicomponent microsegregation while determining relevant processing windows for the preparation of superalloy components using a controlled SX laser processing.

#### 2. Theoretical model

#### 2.1. Microsegregation model for rapid solidification conditions

Rapid solidification can occur for one of two reasons: High undercooling of the melt and rapidly moving temperature fields [11]. A common feature of the latter, e.g., the LRDS process, is that their solidification processes are accompanied by strong liquid flow, such as the Marangoni convection which can produce flow velocities of the order of 1 m/s [8,20]. Moreover, the effects of the convection, in the interdendritic liquid, are the most significant at the dendritic tips (low solid fractions) and decrease as the  $f_s$  increases, which means that the high boundary concentration gradients in the interdendritic liquid with low solid fractions are able to be efficiently reduced or even eliminated. The assumption of homogeneous liquid can be considered as tenable as a result of the strong convection. Therefore, an appropriate modification to the CK model, including nonequilibrium solute redistribution and dendrite tip undercooling into the model, can make it available to predict the microsegregation during the LRDS process.

As solidification velocity increases, solidification process will deviate from the local equilibrium at solid/liquid (S/L) interface. Therefore, the equilibrium solidification parameters should be replaced by velocity-dependent solidification parameters so that the changes in phase equilibrium resulting from the RS can be accurately reflected. In a multicomponent system, considering an independent solute field for each component *i* (*n* elements), the velocity-dependent liquidus slope,  $m_{v,i}$ , and partition coefficient,  $k_{v,i}$ , are respectively given by Ref. [21]:

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