Influences of processing parameters on microstructure during investment casting of nickel-base single crystal superalloy DD3

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Abstract: The effects of solidification variables on the as-cast microstructures of nickel-base single crystal superalloy DD3 have been investigated by using the modified Bridgman apparatus. The experiments were performed under a thermal gradient of approximately 45 K·cm⁻¹ and at withdrawal rates ranging from 30 to 200 m·s⁻¹. The experimental results show that the primary and secondary dendritic arm spacings (PDAS and SDAS) decrease when the withdrawal rate is increased. Compared with the theoretical models of PDAS, the results are in good agreement with Trivedi’s model. The relationships of PDAS and SDAS with withdrawal rates can be described as \( \lambda_1 = 649.7V^{-0.24} + 0.02 \) and \( \lambda_2 = 281V^{-0.32} + 0.03 \), respectively. In addition, the size of the \( \gamma' \) phase significantly decreases with increasing withdrawal rate.

Key words: Ni-base superalloys; directional solidification; \( \gamma' \) phase; dendritic spacing

Table 1: Models for primary dendritic arm spacing

<table>
<thead>
<tr>
<th>Models</th>
<th>Equations</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hunt model</td>
<td>( \lambda_1 = \frac{2.83(k\Delta T_0 D \Gamma)^{1/4}}{V^{1/2} G^{1/2}} )</td>
<td>[13]</td>
</tr>
<tr>
<td>Kurz-Fisher model</td>
<td>( \lambda_1 = \frac{4.3 \Delta T^{1/2} \left( \frac{D \Gamma}{\Delta T_0 k} \right)^{1/4} V^{-1/4} G^{1/2}} )</td>
<td>[14]</td>
</tr>
<tr>
<td>Trivedi model</td>
<td>( \lambda_1 = 2\sqrt{2}G^{-1/2} V^{-1/4} (Lk \Delta T_0 J D)^{1/4} )</td>
<td>[15]</td>
</tr>
</tbody>
</table>

The microstructure of Ni-base superalloys is generally made up of a \( \gamma \) matrix, with the \( \gamma' \) phase precipitated coherently in the \( \gamma \) matrix, carbides and eutectic \( \gamma-\gamma' \).
high-temperature mechanical properties of nickel-base single crystal superalloys mainly rely upon a combination of solid solution strengthening and precipitation hardening, which is provided by the precipitation of large volume fractions of the γ′ phase. Recently, considerable research has concentrated on better understanding of the effects of the γ′ phase particles on the high-temperature mechanical properties of nickel-base superalloys [21-23]. The results indicated that the volume fraction, size and morphology of the γ′ phase particles play important roles in controlling the creep strength of the superalloys. Therefore, it is necessary to investigate the relationship between the size and the morphology of the γ′ phase particles with the withdrawal rate.

In this study, we focused on the PDAS and SDAS variation with withdrawal rate for clarifying the effects of withdrawal rate on the dendritic spacing (PDAS and SDAS) during directional solidification. We were expecting to provide a theoretical foundation for optimizing processing parameters in the industry. Meanwhile, the experimental data were compared with the results of the models. Finally, the influences of withdrawal rate on the size and the morphology of the γ′ phase particles were discussed.

1 Experimental details

1.1 Directional solidification experiment

Single-crystal castings were carried out using an improved Bridgman furnace in a vacuum environment (Fig. 1). The chemical composition of the material used in the present study is given in Table 2. Prior to casting, the mould was mounted on a water-cooled copper chill plate and preheated to the temperature of 1,550 °C in the experiments. Then the melt was poured into the preheated mould and held for 2 min to stabilize the melt. The mould was withdrawn from the furnace at different speeds. The average thermal gradient was 45 K·cm⁻¹.

The wax replicas (pattern) contained two cylindrical test bars (12 mm in diameter and 100 mm in length) which were mounted on two spiral grain selectors. The wax replicas were then coated with multiple layers of ceramic slurry (80% alumina and 20% silica) to obtain a shell with a thickness of 5 mm. After coating and drying, the wax was melted out in a steam autoclave. The ‘green’ moulds were then fired to remove the remaining wax and to strengthen the ceramic shell by partial sintering.

![Fig. 1: Schematic diagram of furnace chamber](Image)

### Table 2: Nominal composition of alloy used in experiment

<table>
<thead>
<tr>
<th>Elements</th>
<th>Cr</th>
<th>Co</th>
<th>Mo</th>
<th>W</th>
<th>Al</th>
<th>Ti</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>wt.%</td>
<td>9.5</td>
<td>5.0</td>
<td>3.8</td>
<td>5.2</td>
<td>5.9</td>
<td>2.1</td>
<td>Bal.</td>
</tr>
</tbody>
</table>

2 Results and discussion

2.1 Primary dendritic arm spacing

Figure 2 shows the microstructure of cross-sections of directionally solidified superalloy DD3 at different withdrawal rates of 30, 50, 70, 100, 150, and 200 m·s⁻¹. The quantitative results of the primary dendritic arm spacing (PDAS) are shown in Table 3. It can be seen from Fig. 2 and Table 3 that with the increasing of the withdrawal rates, the dendritic structure is refined and the PDAS is decreased. The data from Table 3 were plotted in Fig. 3. Clearly, there exists a distribution range of primary dendrites in the area A. At least 7 regions in each specimen were measured for the data N. The SDAS and the size of γ′ were measured using SISC IASV8.0 analysis software in the longitudinal sections. At least 30 readings were taken for each sample. The value of λ₂ and the size of γ′ were their mean values.

Using the physical parameters given in Table 4, the predictions of three theoretical models (as described by equations (1) to (3) in Table 1) were compared with the measured primary spacing, as shown in Fig. 3. It is found that the prediction of the Hunt model has large deviations with the present experimental results. The calculated values for the Kurz-Fisher model are a little greater than the experimental data. The prediction of the Trivedi model shows reasonable parametric agreement. From Fig. 3, it can be seen that, the lower limit values of the measured spacing, λ₁, are higher than the results predicted by the Hunt model. Also the upper limit values were lower than the results predicted by the Kurz-Fisher model. Min et al. [18] investigated the microstructure evolution of DZ125 under high thermal gradient. The results indicated that the relationship between the primary spacing and the withdrawal rate can be described as: \( \lambda_1 = 314.6 V^{-0.24 \pm 0.02} \). When V≤50 μm·s⁻¹, the Hunt-Lu model prediction agreed well with the experimental data. Meanwhile, the predictions of the Ma model [13] and Trivedi model [19] showed reasonable
Fig. 2: Dendritic microstructures of transverse sections at different withdrawal rates: (a) $V = 30 \, \mu\text{m} \cdot \text{s}^{-1}$; (b) $V = 50 \, \mu\text{m} \cdot \text{s}^{-1}$; (c) $V = 70 \, \mu\text{m} \cdot \text{s}^{-1}$; (d) $V = 100 \, \mu\text{m} \cdot \text{s}^{-1}$; (e) $V = 150 \, \mu\text{m} \cdot \text{s}^{-1}$; and (f) $V = 200 \, \mu\text{m} \cdot \text{s}^{-1}$

Table 3: PDAS and SDAS at different withdrawal rates

<table>
<thead>
<tr>
<th>$G$ (K cm$^{-2}$)</th>
<th>$V$ (μm·s$^{-1}$)</th>
<th>$\lambda_1$ (μm)</th>
<th>$\lambda_2$ (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>45</td>
<td>30</td>
<td>291</td>
<td>90</td>
</tr>
<tr>
<td>45</td>
<td>50</td>
<td>253</td>
<td>84</td>
</tr>
<tr>
<td>45</td>
<td>70</td>
<td>235</td>
<td>74</td>
</tr>
<tr>
<td>45</td>
<td>100</td>
<td>217</td>
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<td>45</td>
<td>150</td>
<td>196</td>
<td>54</td>
</tr>
<tr>
<td>45</td>
<td>200</td>
<td>186</td>
<td>50</td>
</tr>
</tbody>
</table>

Table 4: The physical parameters of superalloy DD3$^{[24, 25]}$

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Units</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Partition coefficient</td>
<td>$k$</td>
<td></td>
<td>0.81</td>
</tr>
<tr>
<td>Diffusion coefficient</td>
<td>$D$</td>
<td>m$^2$·s$^{-1}$</td>
<td>6 $\times$ 10$^{-9}$</td>
</tr>
<tr>
<td>Gibbs-Thomson coefficient</td>
<td>$\Gamma'$</td>
<td>mK</td>
<td>1.76 $\times$ 10$^{4}$</td>
</tr>
</tbody>
</table>

2.2 Secondary dendritic arm spacing

Table 3 gives the SDAS of superalloy DD3 at different withdrawal rates. It can be seen that with the increasing of the withdrawal rate, the secondary arm spacing decreases. During the process of solidification, a ripening process causes the highly-branched arms to change with time into coarser, less branched, and more widely-spaced ones. The driving force for the ripening process is the difference in chemical potential of crystals with different interfacial energies due to different curvatures. Kattamis and Flemings$^{[26]}$ proposed that the secondary spacing, $\lambda_2$, can be described as:

$$\lambda_2 = 5.5 \left[ \frac{\Gamma D}{m(1-k)(C_e - C_i)} \frac{C_e}{C_i} \right]^{1/3}$$

where $C_e$ is the eutectic composition, $C_i$ is the initial alloy composition, $t_f$ is the local solidification time, $m$ is the liquidus slope.

In the case of directional solidification, the local solidification time can be given by:

$$t_f = \Delta T_f / |GV|$$
where $\Delta T_0$ is the dendrite tip-to-root temperature difference. Thus, it can be concluded as:

$$\lambda_2 \propto V^{-1/3}$$

Figure 4 shows the relationship between the withdrawal rate and the SDAS, in which the SDAS is proportional to $V^{-1/3}$, as described in Equation (6). From the non-linear fitting line it can be found that $\lambda_2 = 281V^{-0.32\pm0.03}$. Therefore, it can be concluded that the SDAS decreases as the withdrawal rate is increased.

2.3 Morphologies of $\gamma'$ phase in dendrite structure

In Fig. 5 showing the $\gamma'$ phase morphologies at different withdrawal rates, the large and cuboidal $\gamma'$ phase is in dendrite cores and inter-dendritic regions at low withdrawal rates. With the increase of the withdrawal rate, the size of $\gamma'$ phase particles decreases but the $\gamma'$ phase in the dendrite cores and inter-dendritic regions is still cuboidal. This result is different from the study of Zhao et al. [16]. They reported that the $\gamma'$ phase in the dendrite cores tends to be spherical at the highest withdrawal rate under high thermal gradient; while the $\gamma'$ phase in the inter-dendritic regions is still cuboidal. Guo et al [27] proposed that the initial morphology of $\gamma'$ phase is spherical nuclei, the $\gamma'$ phase in the dendritic cores does not have enough time to ripen and stays spherical at high cooling rate.
Fig. 5: Morphologies of $\gamma'$ phase in dendritic cores and inter-dendritic regions at different withdrawal rates:
(a), (b): $V = 30 \, \mu m \cdot s^{-1}$;
(c), (d): $V = 50 \, \mu m \cdot s^{-1}$;
(e), (f): $V = 70 \, \mu m \cdot s^{-1}$;
(g), (h): $V = 100 \, \mu m \cdot s^{-1}$;
(i), (j): $V = 150 \, \mu m \cdot s^{-1}$;
(k), (l): $V = 200 \, \mu m \cdot s^{-1}$

Figure 6 shows the relationship between the size of $\gamma'$ phase particles and the withdrawal rate, in which the size of $\gamma'$ phase particles decreases with the increasing of the withdrawal rate. Meanwhile, the size of $\gamma'$ phase particles in the dendrite cores is smaller than that in the inter-dendritic regions.

![Fig. 6: Influence of withdrawal rate on size of $\gamma'$ particles](image)

In general the composition of $\gamma'$ phase is Ni$_3$Al, with the aluminum atoms occupying the corner sites and the nickel atoms occupying the face-centered sites of the unit cell. In highly alloyed Ni-base superalloys many other elements can substitute for either nickel or aluminium or both, so that the composition can be best expressed as (Ni, Co)$_3$(Al, Ti, Ta).

Nucleation of $\gamma'$ phase occurs via an ordering transformation and then is followed by diffusion controlled growth. During the process of directional solidification, Al and Ti are forming elements of the $\gamma'$ phase, which are enriched in the inter-dendritic regions and lead to a lower degree of coherency. It is supposed that there is a larger supersaturation, and thus enhances growth kinetics of $\gamma'$ phase in the inter-dendritic regions.[28] Therefore, the size of $\gamma'$ phase particles in the inter-dendritic regions is larger than that in the dendritic cores.

A high withdrawal rate can result in a large degree of undercooling and increases supersaturation. This in turn suppresses early precipitation during cooling, and generates a large amount of nucleation sites and small size $\gamma'$ precipitates with a small inter-particle distance at the end of the cooling process (Fig. 5k). When a slow withdrawal rate is applied, $\gamma'$ phase starts to precipitate at an early stage during cooling and continues to coarsen during further cooling, resulting in relatively large $\gamma'$ phase and inter-particle spacing (Fig. 5a). Therefore, the size of $\gamma'$ phase particles decreases with the increasing of the withdrawal rate.

3 Conclusions

Directional solidification experiment was carried out with superalloy DD3 in an improved Bridgman furnace. The influences of solidification parameters on the microstructures (PDAS, SDAS and $\gamma'$ size) have been investigated. The main findings can be summarised as follows:

(1) The primary and secondary dendritic arm spacings decrease with the increasing of the withdrawal rate. The average primary and secondary dendritic arm spacings of superalloy DD3, as functions of growth velocity, are given as $\lambda_1 = 649.7 V^{-0.24\pm0.02}$ and $\lambda_2 = 281 V^{-0.32\pm0.03}$. The Trivedi model predicting the PDAS is close to the experimental results during directional solidification.

(2) The size of $\gamma'$ phase particles decreases with the increasing of the withdrawal rate. The shape of $\gamma'$ phase is cuboidal in the dendritic and inter-dendritic regions; while the $\gamma'$ phase in the dendrite cores become irregular with the increasing of the withdrawal rate.
References


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