Numerical Analysis of Microstructure Development during Laser Welding Nickel-based Single-crystal Superalloy Part V: Crystallography-dependent Aluminum Redistribution

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Abstract. The thermal-metallurgical modeling of alloying aluminum redistribution was further developed through couple of heat transfer model, dendrite selection model, multicomponent dendrite growth model and nonequilibrium solidification model during three-dimensional nickel-based single-crystal superalloy weld pool solidification over a wide range of welding conditions (laser power, welding speed and welding configuration). It is clearly indicated that welding configuration plays more important role than heat input in aluminum redistribution. The bimodal distribution of aluminum concentration along the solid/liquid interface is crystallographically symmetrical about the weld pool centerline for (001) and [100] welding configuration, while the distribution is crystallographically asymmetrical about the weld pool centerline for (001) and [110] welding configuration. Optimum low heat input (low laser power and high welding speed) with (001) and [100] welding configuration beneficially suppresses the aluminum concentration, reduces the vulnerable [100] dendrite growth region to minimize the solidification cracking susceptibility and improve weldability and vice versa. The overall aluminum concentration on the left side of the weld pool is beneficially smaller than that of right side in (001) and [110] welding configuration throughout the weld pool due to crystallographic orientation of dendrite growth regardless of the heat input. The mechanism of asymmetrical solidification cracking because of crystallography-dependent alloying solute inhomogeneity was proposed. The welding conditions, weld pool geometry, solidification conditions, dendrite selection, alloying redistribution and solidification cracking susceptibility are closely correlated. The theoretical predictions agree well with the experiment results. Moreover, the promising results facilitate the understanding of solidification cracking phenomena, and the useful model is also applicable to other single-crystal superalloys with similar metallurgical properties for successful defect-free laser welding or laser cladding.

1. Introduction
Nickel-based single-crystal superalloys with γ’ and γ’’ precipitation phases are widely used in the aerospace and energy industries because of superior mechanical properties, corrosion resistance and microstructure stability at high temperatures. Solicitation cracking is weldability concern during welding nickel-based single-crystal superalloy. Some recent ongoing researches improve the understanding of microstructure development and solidification cracking susceptibility, and are briefly introduced as follows. Weiping et al.[1,2] developed a mathematical model to compute the effects of substrate orientation and melt-pool geometry on the dendrite growth and microstructure development during three-dimensional nickel-based single-crystal superalloy CMSX-4 melt-pool solidification. Hunziker et al.[3,4] numerically analyzed the centerline grain boundary formation and solidification...
cracking through a couple of heat transfer model and multicomponent dendrite growth model, and predicted the weldability map during tungsten-inert gas (TIG) welding nickel-based superalloy IN718. Anderson et al.\cite{5,6} simulated the influence of welding parameters on stray grain formation through heat transfer and fluid flow modeling during laser, electron beam and gas tungsten arc (GTA) welding single-crystal superalloy CMSX-4. Rappaz et al.\cite{7-9} theoretically predicted the effect of dendrite growth crystallography on microstructure development and solidification behavior during electron beam welding ternary Fe-Ni-Cr single-crystal superalloy with arbitrary welding configuration. Moreover, the microstructure development modeling with consideration of heterogeneous nucleation and grain growth was proposed to analyze the effect of alloy concentration and cooling rate on the microstructure development. Vittek et al.\cite{10-12} numerically analyzed the effects of welding condition and crystallographic orientation of dendrite growth on stray grain formation during laser welding nickel-based single-crystal superalloy Rene N5. Moreover, the γ phase microstructure development, γ′ phase precipitation morphology and alloying element partition of nickel-based superalloy CM247DS were analyzed under rapid solidification conditions. Chaturvedi et al.\cite{13,14} evaluated the stray grain formation, solidification cracking susceptibility and weldability of nickel-based single-crystal superalloys CMSX-4 and CMSX-486 during laser welding, and analyzed the effect of minor elements on weldability. Gaumann et al.\cite{15} proposed the laser processing-microstructure maps for single-crystal CMSX-4 gas turbine blade repair by laser deposition. Wang et al.\cite{16} analyzed the effect of grain boundary misorientation on solidification cracking susceptibility during laser welding nickel-based single-crystal superalloy MC2. Moat et al.\cite{17} analyzed the effect of laser parameters on grain morphology, crystallographic orientation and melt-pool geometry of laser-deposited Waspaloy. Tewari et al.\cite{18} analyzed the solute partition coefficients of aluminum and titanium during single-crystal superalloy PWA-1480 directional solidification. Wagner et al.\cite{19} analyzed the microstructure development and dendrite selection of single-crystal superalloy during directional solidification casting. Yang et al.\cite{20} simulated the dendrite growth and stray grain formation of nickel-based single-crystal superalloy CMSX-4 during directional solidification casting. A.de Bussac\cite{21} analyzed the single-crystal growth conditions to prevent stray grain formation during single-crystal superalloy casting. The objective of this paper is therefore to numerically analyze the effect of welding conditions (laser power, welding speed and welding configuration) on crystallography-dependent alloying aluminum redistribution in ternary Ni-Cr-Al single-crystal superalloy under nonequilibrium solidification conditions, and facilitate understanding of the contributing factors to weldability improvement.

2. Mathematical model
Nickel-based single-crystal superalloy CMSX-4 was used with chemical composition of Ni-9Co-6.5Cr-5.6Al-1Ti-6W-6.5Ta-3Re-0.6Mo-0.1Hf (in wt%).

2.1. Heat transfer model
The three-dimensional weld pool shape of liquidus temperature distribution is acquired by the Rosenthal thick plate solution of moving heat source under steady-state conditions. The weld pool was subdivided into 12 sections of equal length along the X-axis from the maximum weld pool width (where solidification begins) to the end of weld pool centerline (where solidification terminates).

2.2 Dendrite selection model
For FCC nickel, dendrite grows along the six preferential <100> crystallographic orientations of dendrite growth. On the basis of minimum growth velocity or minimum undercooling criterion, the relationship between the growth velocity \(V_{hkl}\) of dendrite tip along [hkl] crystallographic orientation and welding speed \(V_b\) is geometrically derived by

\[
V_{hkl} = V_b \frac{\cos \theta}{\cos \psi_{hkl}} \tag{1}
\]
where $\theta$ is the angle between solidification interface normal $\vec{n}$ and welding direction. $\psi_{hkl}$ is the misorientation angle between solidification interface normal $\vec{n}$ and the active $[hkl]$ crystallographic orientation. $\phi$ is the angle between the $Y$-axis and the projection of $\vec{n}$ on the $Y$-$Z$ plane.

The three components of temperature gradient $G_x$, $G_y$, and $G_z$ along the solid/liquid interface are calculated by weld pool geometry derivative. The temperature gradient normal to the solid/liquid interface is then calculated by $G_{sl} = \sqrt{G_x^2 + G_y^2 + G_z^2}$.

The temperature gradient along the crystallographic orientation of dendrite growth $G_{hkl}$ is subsequently determined by

$$G_{hkl} = \frac{G_{sl}}{\cos \psi_{hkl}}$$ (2)

2.3 Columnar/equiaxed transition (CET) model

On the basis of constitutional undercooling criterion, the stray grain formation ahead of solidification interface is derived by

$$G_{hkl}^n = a\left\{ \frac{-4\pi N_0}{3\ln(1-\phi)} \left[ n + \frac{1}{1 + (2\Gamma(\phi) Pe)} \right] \right\}^n$$ (3)

where $a$ and $n$ are material-dependent constant, $N_0$ is the nuclei density in the liquid and $\phi$ is stray grain fraction ($0.5 \leq \phi \leq 1$ for full equiaxed dendrite growth, $\phi \leq 0.0066$ for full columnar dendrite growth).

2.4 Multicomponent dendrite growth model

The dendrite growth is primarily controlled by the diffusion of Cr and Al in Ni for Ni-Cr-Al ternary single-crystal superalloy. On the basis of marginal stability of planar front criterion, the multicomponent dendrite growth is derived by Kurz-Giovanola-Trivedi (KGT) model under rapid solidification conditions.

$$\frac{4\pi \Gamma}{R^2} + \sum_{i=1}^{2} Pe_i m_i C_{0,i} (1-k_i) \zeta_c(Pe_i) + G_{hkl} = 0$$ (4)

where $\Gamma$ is the Gibbs-Thomson coefficient, $R$ is the dendrite tip radius, $Pe_i$ is the Peclet number for $i$, $m_i$ is the liquidus slope, $C_{0,i}$ is the initial concentration for $i$, $k_i$ is the partition coefficient for $i$, $\zeta_c(Pe_i)$ is a function of the Peclet number, $Iv(Pe_i)$ is the Ivantsov solution ($i=$Cr or Al) and $G_{hkl}$ is the average thermal gradient near the dendrite tip.

2.5 Nonequilibrium solidification model

The solidification temperature range is thermodynamically calculated by the linear Ni-Cr and Ni-Al binary phase diagrams on the nickel-rich side, and is simultaneously satisfied by the growth kinetics of dendrite tip. Solidification temperature range is between subliquidus and subsolidus. The more information about angular relationship ($\theta, \phi, \psi_{hkl}$), $V_{hkl}$ and $G_{hkl}$ are provided in the literature[5,7,8]. The material properties in the calculation are available in the literature [7,11,22,23].

3. Results and discussion

The effect of welding configurations on microstructure selection and solute redistribution during nickel-based single-crystal superalloy weld pool nonequilibrium solidification is shown in figure 1. For [100] welding direction on (001) substrate orientation, the bimodal distribution of aluminum concentration along the solid/liquid interface is crystallographically symmetrical about the weld pool centerline throughout the weld pool in (b) and (e). $\phi$ distribution varies with the weld pool geometry from section 1 to 12 in (d). There are four active $[010],[100],[0\bar{T}0]$ and $[001]$ dendrite growth regions, and the transition boundaries are delineated with aluminum enrichment. The epitaxial $[001]$ dendrite
growth region is favored in the bottom of the weld pool to maintain the single-crystal nature of the material. The aluminum-rich concentration detrimentally occurs in the [100] dendrite growth region near the top center part of the weld pool to facilitate nonequilibrium alloying partition, constitutional undercooling and subsequent deleterious eutectic-type reaction γ’ to modify the primary solidification path. The aluminum enrichment along the solidification interface occurs near the end of weld pool solidification in section 12 (θ≈0) that is important contributing factor for centerline cracking.

Weld pool geometry (a), φ distribution (d), dendrite selection (e) and solid aluminum concentration (b) in the (001) and [100] welding configuration. Dendrite selection (f) and solid aluminum concentration (c) in the (001) and [110] welding configuration.

Figure 1. The effect of welding configuration on microstructure selection and solute redistribution during weld pool solidification in Y-Z plane (laser power 5kW and welding speed 1m/min).

By contrast, for [110] welding direction on (001) substrate orientation, the distribution of aluminum concentration along the solid/liquid interface is crystallographically asymmetrical in (c) and (f). There are three active [010], [100] and [001] dendrite growth regions with well-defined transition boundaries. [010] and [100] dendrites impinge at the centerline to form centerline grain boundary. The larger size of vulnerable [100] dendrite growth region is imposed, and the more available aluminum is significantly incurred on the right side of the weld pool for severe stray grain formation, coarser dendrite size and wider solidification temperature range because of unfavorable solidification conditions (low temperature gradient and low dendrite growth velocity). The aluminum enrichment is particularly confined in epitaxial [100] dendrite growth region to induce solute inhomogeneity and microstructure anomalies. The overall aluminum concentration profile on the right side of the weld pool (90°≤φ≤180°) is larger than that of left side (0°≤φ≤90°) from section 1 to 12 (0°≤θ≤90°), although the same heat input is imposed on the both sides of the weld pool. It is crystallographically favorable for asymmetrical aluminum enrichment to simultaneously worsen morphology instability,
microstructure development and solidification behavior. The mechanism of asymmetrical solidification cracking because of crystallography-dependent solute enrichment is proposed. (001) and [100] welding configuration is more appropriate for low aluminum concentration to reduce the metallurgical driving forces for stray grain formation and eutectic $\gamma'$ formation to improve microstructure stability than that of (001) and [110] welding configuration because of significant reduction of [100] dendrite growth region.

![Weld pool geometry](image1)

![φ distribution](image2)

![Dendrite selection](image3)

![Solid aluminum concentration](image4)

![Welding configuration](image5)

Weld pool geometry (a), φ distribution(d), dendrite selection(e) and solid aluminum concentration(b) in the (001) and [100] welding configuration. Dendrite selection (f) and solid aluminum concentration (c) in the (001) and [110] welding configuration

**Figure 2.** The effect of welding configuration on microstructure selection and solute redistribution during weld pool solidification in Y-Z plane (laser power 5kW and welding speed 4m/min).

The effect of welding configurations on microstructure selection and solute redistribution during nickel-based single-crystal superalloy weld pool nonequilibrium solidification with increasing welding speed is shown in figure 2. For [100] welding direction on (001) substrate orientation, the bimodal aluminum distribution is suppressed by increasing welding speed with solidification conditions (steep temperature gradient and high dendrite growth velocity). The sizes of [100] and [001] dendrite growth region are of particular interest. The weld pool dimensions are reduced due to decreasing heat input, and therefore the sizes of dendrite growth region are reduced. High welding speed diminishes the vulnerable [100] dendrite growth region to ameliorate the solidification behavior and microstructure development. The size of active [001] dendrite growth region is larger than other regions and maintains the single-crystal nature of the material. Because the growth kinetics of dendrite tip, constitutional undercooling and nonequilibrium solidification behavior are essentially controlled by the diffusion of aluminum in nickel, the reduction of aluminum concentration refines dendrite size, prevents stray grain formation and decreases solidification temperature range. By contrast, for [110]
welding direction on (001) substrate orientation, the size of centerline grain boundary formation is reduced. The weld pool is elongated and $\theta$ sufficiently decreases to stabilize the [010]/[001], [001]/[100] and [010]/[100] dendrite transition boundaries at the end of the weld pool. [001] dendrite is predominantly overgrown and extends almost up to the top surface. The aluminum distribution is highly dependent on the weld pool geometry. The decreasing size of [100] dendrite growth region is less susceptible to solidification cracking with high welding speed. Although the aluminum is enriched on the right side of the weld pool than that of left side, the overall aluminum concentration is mitigated. The higher the welding speed is used, the lower aluminum concentration is beneficially incurred, and smaller size of the vulnerable [100] dendrite growth region and less solidification cracking susceptibility are therefore imposed. The anomalous distribution of aluminum concentration along the solid/liquid interface is consistent with that of stray grain formation, dendrite trunk spacing and solidification temperature range. The stray grain formation, microstructure anomalies and morphology instability are beneficially mitigated by increasing welding speed. Solidification cracking susceptibility is metallurgically alleviated by aluminum concentration control. It is highly imperative to crystallographically reduce the asymmetrical solidification cracking susceptibility through proper alloying redistribution control and welding configuration optimization for weldability improvement.

![Weld pool geometry](attachment:fig3a.png) ![φ distribution](attachment:fig3b.png) ![Dendrite selection](attachment:fig3c.png) ![Solid aluminum concentration](attachment:fig3d.png)

**Figure 3.** The effect of welding configuration on microstructure selection and solute redistribution during weld pool solidification in Y-Z plane (laser power 2kW and welding speed 1m/min).

The effect of welding configuration on microstructure selection and solute redistribution during nickel-based single-crystal superalloy weld pool nonequilibrium solidification with decreasing laser power is shown in figure 3. For [100] welding direction on (001) substrate orientation, the sizes of
Each active dendrite growth region and weld pool dimensions are reduced by decreasing laser power. Preferential [001] dendrite is predominantly prevented from reaching the weld pool surface. The severe aluminum enrichment occurs near the weld pool center, as solidification approaches the centerline. The distribution of aluminum concentration is mitigated with further reduction of heat input because of favorable solidification conditions (steep temperature gradient and low dendrite growth velocity) with low laser power. The reduction of aluminum concentration beneficially contributes to fine dendrite size, negligible stray grain formation and narrow solidification temperature range, and thereby morphologically improve the resistance to solidification cracking. Desirable low heat input is the necessary condition for single-crystal solidification to improve centerline cracking resistance. By contrast, for [110] welding direction on (001) substrate orientation, the size of unfavorable [100] dendrite growth region is diminished with low laser power to reduce crystallography-dependent aluminum concentration. Two different distribution of aluminum concentration coexist from the beginning of solidification to the end of solidification. [100] dendrite growth region is more susceptible to solidification cracking on the right side of the weld pool because of aluminum enrichment than that of [010] dendrite growth region on the left side. Aluminum concentration is crystallographically controlled in order to reduce stray grain formation in the vulnerable [100] dendrite growth region. The lower laser power is used, the less aluminum concentration is induced and more single-crystal nature of the material dominates to avoid solidification cracking. Welding conditions (laser power, welding speed and welding configuration) are optimized to determine which factor plays the important role in alloying aluminum redistribution, and provides a feasible way to mitigate metallurgical driving forces for solidification cracking.

Weld pool geometry (a), φ distribution (d), dendrite selection (e) and solid aluminum concentration (b) in the (001) and [100] welding configuration. Dendrite selection (f) and solid aluminum concentration (c) in the (001) and [110] welding configuration

**Figure 4.** The effect of welding configuration on microstructure selection and solute redistribution during weld pool solidification in Y-Z plane (laser power 2kW and welding speed 4m/min).
The effect of welding conditions on microstructure selection and solute redistribution during nickel-based single-crystal superalloy weld pool nonequilibrium solidification with increasing welding speed is shown in figure 4. For [100] welding direction on (001) substrate orientation, the shallow and elliptical weld pool shape is less susceptible to solidification cracking. The size of vulnerable [100] dendrite growth region is suppressed with high welding speed, while the size of active [001] dendrite growth region is promoted. The symmetrical aluminum concentration is capable of preventing the detrimental misorientation and severe stray grain formation. It is crystallographically favorable for improving centerline cracking resistance. Optimum low heat input (low laser power and high welding speed) beneficially minimizes the stray grain formation, promotes the morphology stability and refines the dendrite size through mitigation of aluminum concentration, while high heat input (high laser power and low welding speed) deteriorates weldability. By contrast, for [110] welding direction on (001) substrate orientation, epitaxial [001] dendrite growth overgrows at the expense of [100] dendrite growth region. The distribution of aluminum concentration is reduced by further reduction of heat input. Solidification cracking susceptibility is crystallographically ameliorated by optimization of weld pool geometry, microstructure development and alloying aluminum redistribution to reduce solute inhomogeneity and microstructure anomalies. The theoretical predictions of dendrite selection for either [100] or [110] welding direction on (001) substrate orientation agree well with the experiment results [1,2]. The asymmetrical solidification cracking susceptibility is also consistently verified by the experiment results [10,24,25]. On the basis of the foregoing calculation analysis, the promising theoretical predictions provide accurate and reliable prerequisite to evaluate the solidification cracking susceptibility and optimize the single-crystal growth conditions for successful crack-free laser welding. Welding configuration plays more important role than heat input in aluminum concentration to modify the dendrite growth kinetics and nonequilibrium solidification thermodynamics.

4. Conclusions
The alloying aluminum redistribution with multicomponent dendrite growth crystallography under nonequilibrium solidification conditions are numerically analyzed to evaluate the solidification cracking susceptibility with satisfactory accuracy and reliability during laser welding nickel-based single-crystal superalloy. Some following conclusions can be drawn from this work.

- The distribution of aluminum concentration along the solid/liquid interface is crystallographically symmetrical about the weld pool centerline throughout the weld pool for (001) and [100] welding configuration.
- The distribution of aluminum concentration along the solid/liquid interface is crystallographically asymmetrical throughout the weld pool for (001) and [110] welding configuration to worsen the microstructure development.
- The solidification cracking susceptibility is ameliorated through optimization of aluminum concentration and weld pool geometry to reduce vulnerable [100] dendrite growth region in favor of epitaxial [001] dendrite growth region and maintain the single-crystal nature of the material.
- Optimum low heat input with (001) and [100] welding configuration suppresses aluminum concentration and improves weldability to minimize the nonuniform alloying element distribution through mitigation of metallurgical driving forces for solidification cracking, while high heat input with (001) and [110] welding configuration is detrimental to weldability.

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