Tracking Primary Dendritic Arm Evolution through Single Crystal CMSX-10® Ni-Base Superalloy

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Abstract. A novel packing pattern and local primary dendrite arm spacing (PDAS) algorithm, Shape-Limited Primary Spacing (SLPS), is applied to single crystal CMSX-10® Ni-base superalloy. The characterisation technique is rapid, repeatable and comparable, enabling the standardisation of single crystal microstructure analysis. It is deduced, that the increase in local PDAS range observed within the last to solidify sections of the CMSX-10® sample used this study, is due to lateral macrosegregation. SLPS enables quantification of the influence of the processing parameters on resultant packing patterns and local PDAS distribution. Through refinement of local PDAS, deleterious non-hexagonal pattern formation can be reduced. SLPS permits the optimisation of single crystal microstructure and the development of superior quality components.

1. Introduction

Single crystal alloys find a wide range of applications, from semi-conductors, optoelectronics to applications in aerospace engines. Single crystals components used in aerospace are manufactured via the Bridgman directional solidification process, with a seed or grain selector at the bottom the casting [1]. The specific casting conditions lead to the formation of organised columnar arrays of dendrites whose [001] crystal direction is well-aligned with the withdrawal direction. On a 2D etched sample section perpendicular to the growth direction, dendrites appear as bright cross-like features surrounded by dark interdendritic material.

The formation of the dendrites within the array is a result of thermo-physical processes that occur in the mushy zone during solidification [2]. It has been suggested that single crystal mosaicity is caused by plastic deformation in the mush which results in crystal lattice rotations and dendrite misorientation [3-5]. The resultant, low-angle grain boundaries (LABs) are detrimental to the creep rupture life, especially when the local misorientation angle is larger than 9° [6].

The primary dendrite arm spacing (PDAS) characterises the distance between neighbouring dendrites. PDAS is intrinsically linked to the solidification environment, local misorientation, and indicates the maximum length scale for microsegregation [7]. Recently a new single crystal characterisation method, Shape-Limited Primary Spacing (SLPS) [8], has been developed which can automatically, accurately, and rapidly determine the local PDAS distribution within a single crystal bulk array. SLPS can accurately determine the relationship between dendritic packing arrangements, local PDAS, and tip growth kinetics. Consequently, it is now possible to compare multiple single crystal cross-sections and relate variations in processing conditions to changes in the microstructure.

The aim of this work is to apply the SLPS methodology to a multiple sections of a single crystal bulk cast and demonstrate its applicability for process and alloy optimisation. The end goal of this new
approach is to improve understanding of the influence of the solidification conditions on single crystal growth and create a standardised methodology for characterisation.

2. Method
A single crystal CMSX-10® bar of 9.4 mm diameter and 64 mm in height (figure 1(b)) was provided by Rolls-Royce Plc. The bar was sectioned into seven segments perpendicular to the long axes of the sample using a metallographic precision cutter and mounted in Conducto-Mount. Section 1 corresponds to the part of the sample that solidified nearest to the chill plate of the Bridgman furnace. The bar was formed in a cluster mould arrangement and withdrawn from the Bridgman furnace at a constant withdrawal velocity, $V$. The sectioned samples were prepared for Scanning Electron Microscopy (SEM) using a standard grinding and polishing metallurgical procedure and cleaned with $\text{CH}_3\text{OH}$ between each polishing step to a 1$\mu$m finish. At the end, 10 minute ultra-sonication removed excess diamond particles which agglomerate in regions of porosity. Figure 2(a) was acquired with an FEI 650 Quanta SEM at the University of Leicester, with a 20kV beam, WD = 10.1 mm, HF = 2.32 mm. The samples were imaged in back scattered electron (BSE) mode to visualise the microsegregation between the dendritic core and interdendritic region. MAPS® software (ThermoFisher Scientific, Waltham, MA, USA) provided large, high resolution, stitched images of the dendritic arrays automatically, with only minor adjustments required where necessary. The final images were comprised of a 24 × 24 grid of individual SEM images taken at 300 × magnification with 20 $\mu$s scan speed. The BSE image are loaded into the DenMap software [9] and the exact dendritic core pixel positions were located. These coordinates were input into the SLPS algorithm [8]. The dendritic packing and local PDAS distributions were calculated by analysing each individual dendrite in the array and determining true nearest neighbour (TNN) dendrites. TNN are defined as dendrites whose diffusion fields interact locally with a central dendrite; determined by the SLPS algorithm. This method of TNN determination is applied to each dendrite within the single crystal array. The total time for analysis for each sample section was 1 min and 20 seconds. Only the section 1 image (figure 1(b)) is included in this work to demonstrate the resultant dendritic mapping (figure 2(a)) and packing pattern determination (figure 2(b)).

![Figure 1](image_url)

**Figure 1.** (a) Local misorientation, $\theta$, characterised by variation in local PDAS. The fastest growth direction is always the [001] direction aligned with the dendrite stem. A misorientated dendrite is outcompeted by its neighbours who are better orientated with the heat flow direction. (b) 64 mm CMSX-10® single crystal bar sample, $\varnothing = 9.4$ mm. Sample section 1 is closest to the chill plate.
3. Results
The variation in determined packing patterns for sample section 1 are illustrated in figure 2(b). The centre of the sample demonstrates clear hexagonal packing ($N_6$). The outside edge of the sample a combination of triangular ($N_3$), square ($N_4$), pentagonal ($N_5$), heptagonal ($N_7$), octagonal ($N_8$) and nonagonal ($N_9$) packing. The total normalised packing obtained within each sample section is illustrated in Figure 2(c). The non-dimensionalised histogram enables the comparison of all sample sections, under any change in solidification condition, regardless of geometry. The $N_6$ packing is the most frequently observed packing pattern for all sample sections (figure 2(c)). Between 1 and 7 there is a 9% combined increase in $N_7$ and $N_8$, a 5% combined decrease in $N_3$, $N_4$ and $N_5$ and a 4% increase in $N_6$ packing patterns. The distribution of local PDAS for all sample sections from figure 2(b) is shown in figure 2(d). The smallest PDAS for each sample section is determined as roughly 150 $\mu$m. The mean PDAS, $\bar{\lambda}$, and standard deviation, $\sigma$, are the smallest for section 1. Over the first 50 mm of the sample there is very little change in the $\bar{\lambda}$ and $\sigma$, the increase is almost linear with height from the bottom of the sample. However, samples 6 – 7 show large increases in the $\bar{\lambda}$ and $\sigma$ no longer follow the same relationship. The largest PDAS are observed within sample section 7.

The averaged lower and upper PDAS bounds for each packing pattern for sample section 1 and 7 are shown in figure 2(e). The results demonstrate that the average PDAS increases with coordination number, $N$. Within the single crystal microstructure $N_7$ and $N_8$ patterns are the largest observed arrangements and $N_3$, $N_4$ and $N_5$ the smallest. The lower PDAS bound for all sample sections is similar (figure 2(d)). In addition, the increase in PDAS per $N$ for the lower bound demonstrates negligible change throughout the cast height (figure 2(e)). Interestingly, the upper PDAS bound for section 7 is significantly larger than that for section 1 (figure 2(e)). The range in PDAS for the $N_3$ patterns is only slightly greater for sample section 7. However, there is a significant increase in PDAS range for the $N_8$ patterns in sample section 7.

Figure 2. (a) BSE micrograph of CMSX-10® sample section 1, red dots are located dendritic cores. (b) Shape-Limited Primary Spacing (SLPS) dendritic packing analysis applied to the microstructure to determine local PDAS variation and packing pattern quantity. (c) Normalised stacked bar plot of all
determined packing patterns within each sample section from figure 1(b). (d) PDAS probability distribution for all 7 CMSX-10® sample sections, local PDAS variation is determined using Shape-Limited Primary Spacing (SLPS). (e) The variation in the mean lower and upper PDAS bounds for each coordination number for sample section 1 and 7.

4. Discussion

The PDAS is known to follow the steady-state non-linear relationship $\lambda = A C_0^{0.25} V^{-0.25} G^{-0.5}$ [10-16], where, $G$ is thermal gradient, $V$ is withdrawal velocity, $C_0$ is initial composition and $A$ is a materials constant. Therefore, any variation in $C_0$ or solidification rate, $G \times V$, will result in a change in the PDAS. Consequently, the increase in $\lambda$ and $\lambda_0$ observed between sample section 1 and 7 (figure 2(d)) is attributed to these two variables. In Bridgman casting, the axial $G$ reduces with height from the chill plate as the fraction of solid superalloy increases [17]. The lower part of a mould is mainly cooled by intensive interaction with the chill plate, resulting in a solid/liquid isotherm perpendicular to the withdrawal direction. In a cluster mould arrangement, the temperature profile across the sample is uneven [18] and the isotherm typically attains a concave shape. Any isotherm curvature causes a variation in the macroscopic solid/liquid interface shape across the sample cross-section. As a result, radial convection is induced by temperature and thus density differences within the melt. Any spatial density difference causes movement of solute across the sample surface, directly influencing the liquid compositional gradient within the over lying melt parallel to the growing solid/liquid interface. According the steady state PDAS equation, solute enriched regions will experience an increase in PDAS (for solute partitioning to the liquid, $k < 1$), and solute depleted regions (where, $k < 1$) the PDAS decreases. The lower PDAS bound in figure 2(e) increases only slightly between section 1 and 7, therefore, the solidification environment ($G \times V$ and $C$) within a part of each bulk sample section, remained roughly constant throughout the cast height. Interestingly, the upper PDAS bound increases monotonically with sample section height (figure 2(e)).

The areas with largest PDAS are the $N_7$ and $N_8$ packing patterns (figure 2(c)), therefore, the increase in local PDAS range within the end sections of the bar (figure 2(e)) is intrinsically associated to these patterns. In CMSX-10®, $N_7$ and $N_8$ arrangements indicate areas rich in interdendritic partitioning elements such as Ni, Ti, Ta, and Al [19]. As shown in figure 2(b), $N_7$ and $N_8$ dendrites are almost always surrounded by smaller $N_3$, $N_4$, and $N_5$ arrangements. The $N_3$, $N_4$ and $N_5$ patterns indicate regions of dense packing; thus, areas rich in dendritic core elements [20]. The $N_6$ packing arrangement is the most frequently observed within all sample cross-sections (figure 2(c)). It is located between the $N_3$ and $N_8$ PDAS extremes (figure 2(d)) and achieves a $\bar{\lambda}_{N_6}$ (figure 2(e)) almost equal to the $\bar{\lambda}_{Array}$ (figure 2(d)). Regular $N_6$ hexagonal packing is the only arrangement that achieves a uniform PDAS between all TNNs. Therefore, if $N_3$, $N_4$ and $N_5$ are core rich and $N_7$ and $N_8$ interdendritic rich, $N_6$ indicates regions growing with equal microsegregation between neighbours, and with no local misorientation in respect to the heat flow direction. Consequently, arrangements of non-hexagonal packing indicate the location of competitive growth, where core rich $N_3$, $N_4$ and $N_5$ dendrites are outcompeting slower growth rate $N_7$ and $N_8$ neighbours (figure 1(a)).

As solidification height from the chill plate increases, dendrites eject interdendritic material into the liquid. For solutally unstable alloys such as CMSX-10®, solute enrichment occurs in the melt due to the slow diffusion speed in the solid superalloy. Between sample section 1 and 7 this enrichment corresponds to a 9% increase in solute enriched $N_7$ and $N_8$ dendrites and a 5% decrease in solute depleted $N_3$, $N_4$ and $N_5$ (figure 2(c)). If the solute concentration parallel to the growing interface is uniform, then according to the steady state PDAS equation, a change in bulk $G$, $V$, or $C$ only influences the $\bar{\lambda}_{Array}$. Changing the bulk solidification conditions plays no role in determining the local PDAS range. Therefore, as the $G \times V$ across a bulk sample perpendicular to the withdrawal direction is constant (assuming negligible change in the isotherm shape), then the increase in local PDAS range in the end sections of the CMSX-10® cast (figure 2(d)) must be a result of increasing lateral solute enrichment (for $k < 1$).

Using the SLPS algorithm, it is now possible to understand the relationship between the bulk solidification environment, packing patterns, local PDAS, and tip growth kinetics. By tailoring the
processing parameters and/or alloy composition the bulk local PDAS range can be refined. Through application of SLPS the bulk hexagonality of a single crystal array can be increased, thus, decreasing the formation of deleterious non-hexagonal packing patterns. Consequently, the SLPS methodology permits the development of superior quality single crystal components.

5. Conclusions
A novel packing pattern and local primary dendrite arm spacing (PDAS) algorithm, Shape-Limited Primary Spacing (SLPS), is applied to study a CMSX-10® single crystal Ni-base superalloy. It is deduced, that the broadening in local PDAS range observed within the last to solidify sections of a single crystal cast is a result of lateral macrosegregation. The new packing pattern characterisation method is rapid, repeatable, and accurate. SLPS enables quantification of the influence of the processing parameters on resultant packing patterns and local PDAS distribution within an array. Through application of SLPS and the tailoring of the processing parameters and/or alloy composition, the PDAS can be refined, reducing the severity non-hexagonal packing formation; thus, improving the quality of single crystal components.

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6. References
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