Comprehensive Quantitative Characterisation of Single Crystal Alloys

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Abstract. Classically, the primary dendrite arm spacing (PDAS) is used as a gauge to determine the quality of a single-crystal alloy. However, this length scale varies substantially over a cast due to variation in the thermo-solutal conditions. In this work, several dimensionless parameters are derived that characterise this variation based purely on geometrical and natural pattern formation. They are determined by applying a novel shape limited spacing algorithm (SLPS). SLPS accurately evaluates the relationship between local PDAS and packing pattern formations, offering a new method of single crystal analysis. The SLPS algorithm is a part of DenMap, a dendrite core feature extraction tool designed to significantly speed up single-crystal characterisation for process optimisation and standardisation for academia and industry.

1. Introduction

Single-crystal turbine blades are the most important part of a gas turbine engine. They are the limiting factor in determining the turbine entry temperature (TET) which subsequently controls the operating efficiency of the engine. They are manufactured by a complex and expensive unidirectional solidification process, which aligns the [001] crystallographic plane with the direction of heat flow. In directionally solidified materials, for a given composition, the growth rate and temperature gradient determine the morphology of the microstructure and the level of microsegregation [1, 2]. Large primary dendrite arm spacings (PDAS) indicate increased levels of microsegregation, which negatively impacts the high temperature performance of the single crystal material. To reduce microsegregation severity, solution heat treatment is applied to homogenise the microstructure; the time and temperature of which is estimated on the maximum PDAS observed. Therefore, improving the predictive capability of material processing–structure–property relationship models relies upon developing an enhanced understanding of single crystal growth formation and accurate quantitative stereological characterisation of the as-cast microstructure.

Intensive manual microstructural analysis is performed in industry and research worldwide to carry out single crystal alloy characterisation and furnace calibrations. A typical data set can contain 10 or more serial slices with at least 300 dendrite cores per slice, all of which require manual identification [3]. This overwhelming amount of data, and thus time, causes a bottleneck at the image processing, core mapping and characterisation step. To understand the role of PDAS on single crystal performance, design future alloys and improve processing routes, a confluence of automated and standardised characterisation techniques are required. Currently, no universal method for single crystal characterisation exists, especially one that incorporates comparison between different single crystal alloys formed under a multitude of conditions.
In this work, a method for big data analysis and characterisation standardisation is proposed with the goal of developing a universal database for all single crystal alloys. The eventual aim is to link processing conditions to properties in a way that enables development of machine learning algorithms and computational models to optimise single crystal manufacture and the design of new alloys.

2. Automated characterisation

For automation of single crystal characterisation, the DenMap software tool was developed. The complete DenMap software package is shown in figure 1. It uses an automated feature extraction algorithm designed for rapid, accurate and repeatable dendrite core detection [4]. The algorithm implements a Normalised Cross-Correlation (NCC) method which performs a comparison between a template image and an image of interest. For NCC to be effective, the dendritic cores must be clearly visible and possess good definition. This makes the DenMap algorithm ideal for SEM and optical micrographs as there is a clear contrast variation between the dendritic and interdendritic regions due to the preferential microsegregation of lighter and heavier elements, as shown in figure 2(a). A 600 dendrite micrograph can be automatically processed in less than 90 seconds with a dendrite core detection accuracy of 98%, figure 2(b). The automatic process includes mount and noise removal, image filtering via a customised Fast Fourier Band Pass filter (FFT Bandpass), template generation, thresholding and core mapping. Once the core positions are obtained, a subsequent stereological characterisation step is performed.

Characterisation is achieved by a fully automatic and accurate shape limited spacing algorithm (SLPS) [5]. The software performs a Voronoi analysis to determine the maximum possible dendritic nearest neighbours. Then a systematic and consistent packing algorithm, utilising a local polygon shape factor ($K_{SLPS}$), is applied to determine the true nearest neighbours (TNN) i.e. determine only those dendrites that are affecting each other. For dendritic and cellular alloys, the algorithm successfully lowers the upper PDAS bound and returns a perfect Gaussian distribution of local PDAS. This accurate and repeatable single crystal characterisation has never been achieved before [6-8]. The dimensionless polygon shape factor, $K_{SLPS}$, is derived from the square counting method [9] as follows:

$$K_{SLPS} = \sqrt{\frac{N}{\lambda_{TNN}}}$$

(1)

where $A_{TNN}$ is the area bounded by the TNN, $\lambda$ is the local PDAS and $N$ is the number of TNN. An illustrative example of the DenMap algorithm is shown in figure 2, where a CMSX-4® micrograph is analysed. The characterisation step takes under 10 seconds even for images with more than 600 dendrite cores.
Figure 2. Diagram showing the complete process optimisation route of DenMap from a) CMSX-4® micrograph, b) dendritic core mapping and c) characterised micrograph demonstrating a combination of irregular geometries triangular ($N_3$), square ($N_4$), pentagonal ($N_5$), heptagonal ($N_7$) and octagonal ($N_8$).

3. Results

It is established that in order to gain sufficient stereological data about a single crystal alloy, the entire sample surface must be analysed. In this work, dendritic mapping (DenMap) and single crystal characterisation (SLPS) are performed on: CMSX-4® and CMSX-10® superalloys provided by Rolls-Royce Plc, on binary alloys from the literature: Pb-5.8 wt% Sb [10], Pb-10 wt% Sn [7], Al-4.1 wt% Cu [8] and on a SCN-0.24 wt% camphor cellular alloy cast under microgravity conditions [11].

The local shape factor, $K$, for all alloys is plotted against $N$ in figure 3. The $K_{SLPS}$ ideal line in figure 3 is derived from regular polygon shapes and indicates the optimum packing arrangement. All alloys demonstrate a tendency towards $K_{SLPS}$ ideal with increasing $N$ and almost an identical relationship which follows closely the optimal values. As observed, the $N_3$ and $N_4$ shapes for all alloys have much higher $K$ values than the ideal.

The local PDAS range, $\lambda_{min}$/$\lambda_{max}$, is another dimensionless quantity by which dendritic arrays can be characterised, as shown in figure 4. The PDAS range is sorted by packing type and plotted with frequency, where the CMSX-4® obtains a maximum PDAS range of 3.8. The cellular SCN-24%wt camphor alloy cast in microgravity achieves a PDAS range below 2. An interesting observation is that the refinement of an alloy i.e. the reduction in the bulk PDAS range, is accompanied by an increase in the frequency of the $N_5$ shapes.

4. Discussion

The dimensionless local polygon shape factor ($K_{SLPS}$) provides a measure of the irregularity of packing around each individual dendrite, as in equation (1). Any value for $K$, larger that the optimum, designates a dendrite that on average has a smaller $A_{TNN}$ and a higher $\lambda$, indicating non-uniform microsegregation. This indicates zones of overgrowth, tip splitting or branching [12, 13]. The refinement of $K$ with increasing $N$ is expected, as larger $N$ shapes constrict the dendrite arrangement and reduce the range of available irregular geometries. The CMSX-4® alloy on average demonstrates the largest variation in possible irregular shapes, closely followed by the CMSX-10® alloy. Both alloys have a complicated chemical composition with more than 6 alloying elements which leads to a much larger preferential microsegregation compared to any binary alloy. Microsegregation is known to cause convection, dendrite misorientation and other thermo-solutal effects. The SCN alloy solidified under near equilibrium conditions in microgravity where the convection effects were eliminated. It demonstrates a large reduction in the highly constricted $N_7$ and $N_8$ shapes, figure 4(a), and the average packing for each $N$ was the closest out of all alloys studied to the optimal shape factor, $K$ (figure 3).
Figure 3. Six single crystal microstructures are investigated using SLPS: (1) CMSX-4®, (2) CMSX-10®, (3) Al – 4.1wt%Cu, (4) Pb – 10wt%Sn, (5) Pb – 5.8wt%Sb, (6) SCN – 0.24wt%Camphor. The mean packing factor for the seven main types of packing within each single crystal microstructure is plotted against the ideal packing factor, $K_{SLPS}$, for each regular polygon (dashed line).

Figure 4. (a) Local PDAS range all determined packing patterns for (a) CMSX-4® and (b) SCN – 0.24wt%Camphor, (c) mean local PDAS range (MLPR) and variation for (1) CMSX-4®, (2) CMSX-10®, (3) Al – 4.1wt%Cu, (4) Pb – 10wt%Sn, (5) Pb – 5.8wt%Sb, (6) SCN – 0.24wt%Camphor. The dendritic CMSX-4® alloy possesses the largest mean range and variation whilst the SCN-camphor alloy cast under microgravity conditions the smallest.

A local PDAS range equal to 1 indicates the ultimate lower packing limit where all neighbouring dendrites or cells are equally spaced from the central one. Such a ratio is only achieved in a regular hexagonal arrangement (figure 3). Unsurprisingly, a single crystal alloy which possess a PDAS range for all determined $N$ closer to that ultimate limit must possess a bulk microstructure that is more refined. A refinement in the single crystal alloy indicates a more homogenous distribution of interdendritic and dendritic material [14]. Therefore, figure 3 and figure 4 can be utilised to indicate how close/far an array is to the ideal hexagonal packing formation. Consequently, the CMSX-4® sample with the largest PDAS range and the highest average $K$ value would possess the largest amount of non-uniform microsegregation, visualised as a highly non-homogenous microstructure.

Naturally, dendrites or cells are driven by thermo-physical conditions and tend to arrange in the most stable configuration as possible. In single crystal alloys ideal close packing is achieved when all local PDAS are equivalent i.e. hexagonally packed. Consequently, alloy refinement is accompanied by
a decrease in unrefined $N_3$, $N_4$, $N_7$ and $N_8$ packing types and a subsequent increase in close packed $N_5$ and $N_6$ arrangements (figure 4). It is theorised that in a refined array, $N_6$ shapes are only formed from $N_2$ via translation or tip splitting [15] rather than overgrowth; the former phenomena occurring much slower than the latter. It is suggested that as convection is reduced, the bulk hexagonality of the array is increased and PDAS range continue to refine as shown in figure 4(b).

5. Conclusion
A novel dendritic mapping (DenMap) and characterisation digital tool (Shape-Limited Primary Spacing) is applied to a variety of single crystal microstructures. Several new dimensionless parameters are developed to identify new geometric relationships between any single crystal alloy cast under any condition. The results demonstrate that dendritic packing can be utilised to normalise statistical data to provide meaningful and accurate comparisons between all types of single crystal microstructure. Now, with a new standardising method for single crystal characterisation, large quantities of data can be processed, enabling the development of sophisticated process-property models for alloy optimisation.

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6. References
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