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Investigation of freckle formation under various solidification conditions

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Abstract. In the present work the influence of process parameters on freckle formation in superalloy CMSX-4 is demonstrated. A series of experiments were carried out using a laboratory furnace of Bridgman-type in which the temperature gradient G and solidification velocity V can be precisely controlled and individually varied over a wide range. On the etched surface and longitudinal sections of the quenched samples the formation and evolution of freckles were investigated. The initiation position of the freckles within the mushy zone was then determined. Based on the experimental observations a complete diagram was plotted to indicate the probability of freckle occurrence which is related to solidification parameters G and V. In this freckling map the freckle region is delimited by different criteria. Freckles arise only within a certain G/V-range for columnar dendrite growth; otherwise the solidification structure changes into cellular or equiaxed structure, depending on whether G/V-value increases or decreases. In comparison with the well-known freckling map of Copley *et al.*, some new freckle-free regions are proposed. It is interesting to note that the freckle formation is also suppressed at very low cooling rates. In addition, the initiation position of the freckle formation in the mushy region was determined to be less than 2 mm below the dendrite tip of the solidification front.

Introduction

Freckles have been of interest for many researchers for around 40 years [1,2], due to their importance as defects in the advanced directional solidification (DS) and single crystal (SC) castings of superalloys. Freckles appear as long trails of equiaxed grains aligning roughly parallel to the direction of gravity. Internal freckles were normally found in VAR (Vacuum-Arc Remelting) and ESR (Electroslag Remelting) processed ingots at the mid-radius; more often freckles were found on the surface of DS/SC superalloy castings [3-5]. It is nowadays commonly believed that freckles are formed as a result of the thermo-solutal convection induced by inverse density in the mushy zone [2]. The interdendritic liquid from inside the mushy zone tends to "jet" upward from the mush into the liquid layer. Along the way, these jets would erode dendrites and break them apart. Thereafter they would redissolve into the liquid or act as nuclei for equiaxed grains.

In order to prevent the formation of freckles, high temperature gradients and high solidification velocities should be applied, according to the well-known freckling map (Fig. 1) proposed by Copley *et al.* [2]. Above a critical thermal gradient (G*) the mushy zone becomes too small to accommodate the convective currents that would produce freckles. With increasing solidification velocity V, the local solidification time will be shorter than the minimum one necessary to produce freckles, resulting in a freckle-free structure. As emphasized in the freckling map shown in Fig. 1, the freckle-free region is on the upper right and the freckling zone on the left below. Although this freckling map was proposed about forty years ago, it is still well accepted and currently applied to explain the influence of the solidification conditions on the freckle formation.

In order to examine the freckling map shown in Fig. 1 the influence of process parameters, namely temperature gradient G and withdrawal velocity V, on the formation of freckles was

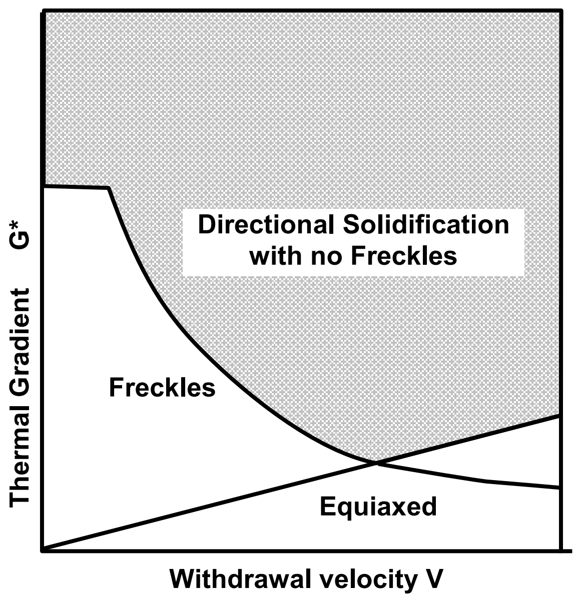

Fig. 1: Freckling map showing the effect of temperature gradient and withdrawal velocity on the freckles formation [2].

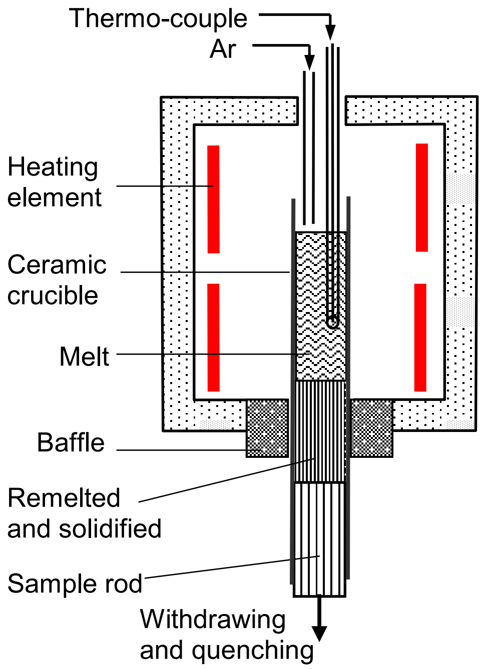

Fig. 2: Schematic diagram showing the laboratory furnace of Bridgman-type furnace for a low temperature gradient.

systematically investigated. At the same time, the initiation position of the freckles was also studied to help better understand the formation mechanism of freckles and to better formulate the mathematical criteria for predicting the freckle formation.

Experimental

In this study, a series of directional solidification experiments were performed under different temperature gradients and withdrawal velocities in order to investigate the influence of processing parameters on the freckle formation. CMSX-4 was chosen for the investigation, since it is a typical freckle prone superalloy [5]. The primary samples were SC cylindrical rods with a diameter of 20 mm and a total length of about 180 mm. They were pre-cast in an industrial Bridgman furnace. The liquidus temperature of 1375 °C was confirmed by thermal measurements during repetitive isothermal cooling experiments.

The samples were remelted and directionally solidified in the vertical Bridgman-type laboratory furnace under the protection of high purity argon gas, as shown in Fig. 2. In order to achieve a lower temperature gradient, the cooling ring was removed and thicker baffle material was installed to separate the hot zone from the ambient environment. The temperature gradient ranged from 1.8 to 10 K/mm. The withdrawal velocity was in the range of 0.2 – 1.0 mm/min. Since the withdrawal velocity was relatively low, it can be treated as the solidification velocity of the corresponding samples.

During the experiments, the cylindrical rod was placed in a ceramic crucible with a length of about 80 mm, to ensure that it is correctly positioned in the furnace. The furnace was first heated to the designed temperature, then the sample rod was raised with the ceramic crucible into the furnace chamber and the portion of the rod inside the crucible was allowed to melt for 10 – 15 min. The melt temperature was measured separately by lowering the thermocouple into the melt in the ceramic tube. The furnace was held for another 10 – 15 min, to enable temperature homogenisation. Then the sample was withdrawn out of the furnace chamber with a defined velocity. After a certain solidification length, the sample was mechanically extracted from the furnace and quenched in the water bath.

The surface of the as-cast samples was prepared following the standard procedure and macro-etched in a solution of 20 vol% H_2O_2 and 80 vol% HCl. Then they were examined under the 3D optical microscope. In order to unveil the initiation position of the freckles, the sample with freckles was also cut off about 1 mm longitudinally at the freckle chain and prepared by standard metallographic procedure. Then it was micro-etched to expose the dendritic microstructure.

Results and discussion

Determination of solidification parameters. The temperature gradient at the liquidus temperature was deducted from the relation G = T'/V, in which the cooling rate T' can be obtained from the measured cooling curves. The temperature gradient G was measured at four different furnace temperatures T_H and an approximated linear relationship between the G and T_H was evaluated. Thus the range of the temperature gradient could be controlled by adjusting the furnace temperature.

Mapping freckle formation under the influence of solidification conditions. After examining all the samples, the results for the freckle formation under different processing conditions were represented in Fig. 3. Each circle in the diagram represents one set of casting conditions (temperature gradient G, solidification velocity V). The filled circle represents the presence of freckles in the sample that was cast under such conditions. The bigger filled circle represents bigger extent of freckling. It should also be pointed out that the top four micrographs and the first micrograph at the bottom were longitudinal section views of micro-etched samples. These samples were first cut off about one millimeter from the surface and then prepared for the micro etching. All these micrographs show the typical microstructure obtained under the casting conditions represented by the circles. Another three micrographs are captured by the 3D light optical microscope directly on the casting surface. The processing parameters were indicated by the origins of the respective arrows.

It can be clearly seen from the diagram that at a higher temperature gradient (10 K/mm) the solidification front morphology displays a planar (P), cellular (C), cellular to dendritic (C/D) and dendritic (D) solidification front as the withdrawal velocity V increases. They form the four regions in the diagram as labeled with P, C, C/D and D respectively. It is also clear that at low temperature gradients equiaxed dendritic microstructure will form. This region is labeled with EQ. In the rest region in the diagram the columnar dendritic microstructure will form.

Comparing the experimental results at different regions, it is concluded that in the planar, cellular, and cellular to dendritic transitional zone, freckles will not form. It is obvious that in these regions the dendrites are not well developed, therefore there are no potential high order dendrite arms to break and then to form the potential nuclei for the freckle grains. As also indicated by the results, even in the region of columnar dendritic microstructure, there are also regions where no freckles will occur. The columnar dendritic region is divided into different sub-regions by dashed lines, as can be seen from the diagram. In the top right region finer dendritic microstructure will form. These fine networks of dendrites produce huge resistance to the interdendritic flow and will thus prevent the formation of the freckles. At the bottom left corner is a region for coarse dendritic microstructure. The coarse dendrite arms are not easily remelted or fractured. Consequently, in this sub-region, there is no freckle formation either.

Freckles tend to occur in the dendritic region enclosed by the dashed lines and the boundary line between the dendritic and EQ region. In this region, the dendrite arms are neither too strong nor too fine. Under such conditions, the thermo-solutal flow can form without an impregnable barrier and the dendrite arms can also be remelted and fragmented to develop the nuclei for the freckle grains. This region is important with regard to the production of freckle-free DS or SC parts. Once the range of this region is defined, it is possible to precisely control the processing parameters to avoid freckle defect. Even when it is impossible to avoid operating in this region, special measures can be taken to avoid the formation of freckles before they are formed on the parts. It should be noted that at the bottom right corner of the diagram, there is a portion of the EQ region which is also included

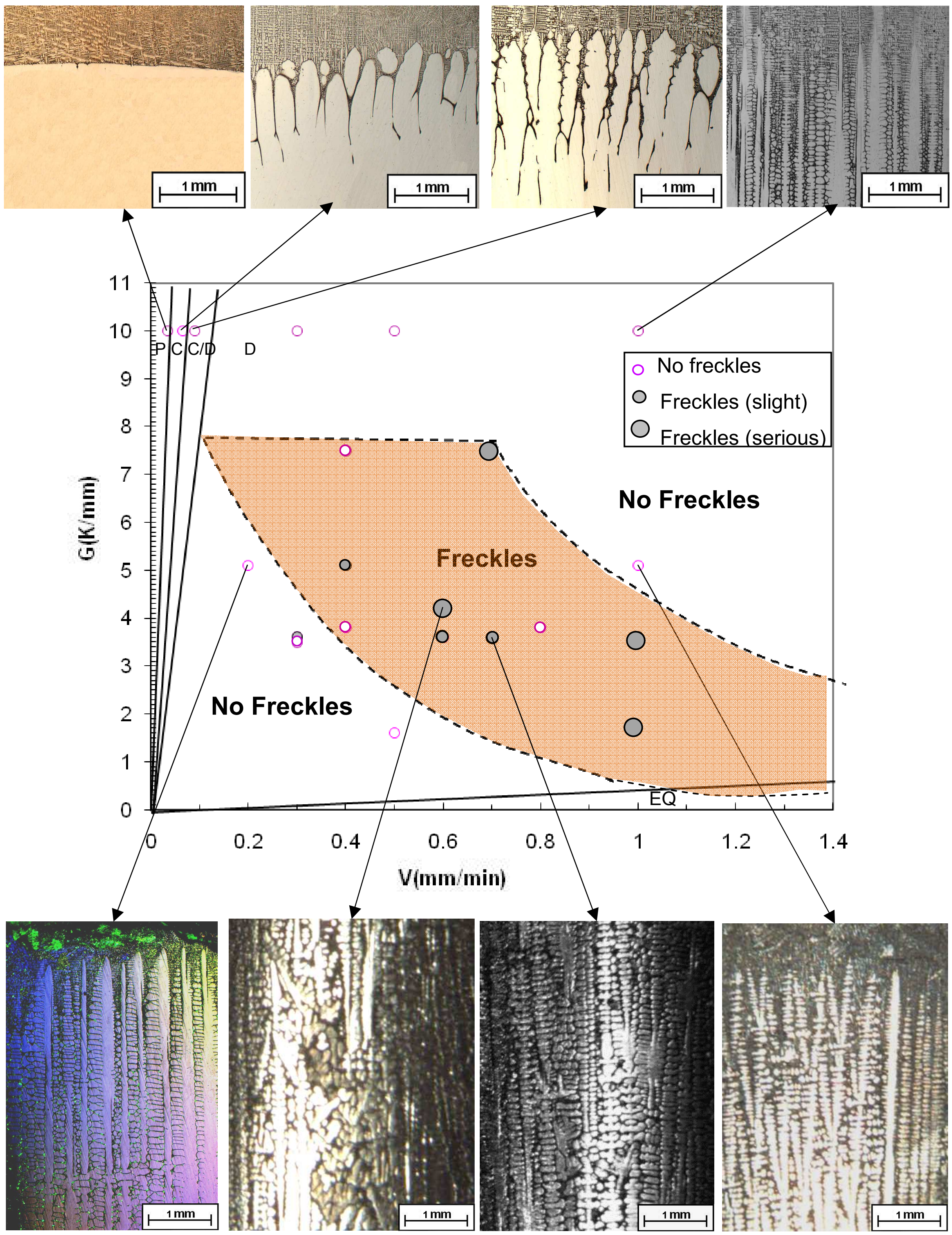

Fig. 3: Freckle formation in superalloy CMSX-4 under different solidification conditions. This new diagram excludes the planar (P), cellular (C), cellular to dendritic (C/D) and equiaxed (EQ) zones from freckle formation. Besides, the columnar dendritic zone is also divided into three zones, in the middle of which freckle occurrence is possible (marked "Freckles").

in the Freckles region. This corresponds to the findings that freckles occur in some castings with equiaxed microstructure. A more deep insight into this phenomenon is suggested for future investigation.

The newly proposed freckle map (Fig. 3) is a development of the one proposed by Copley *et al.* (Fig. 1) [2]. Copley's diagram is only a qualitative description of the influence of the temperature gradient and solidification velocity on the freckle formation. In contrast, the new map excludes the planar (P), cellular (C) and cellular to dendritic (C/D) zones from freckle formation, because in these regions, as shown in the micrographs, there are no thin dendrite arms to break down easily to form freckles. Besides, the columnar dendritic zone is also divided into different sub-zones, in the middle of which freckles are possible to occur (marked "freckles"). Therefore, the newly proposed freckle map is more precise and complete as compared to the earlier one. It gives a broad view of the process conditions under which the freckles might occur. It also helps to better understand the formation of freckles in relation to solidification morphology.

Determination of the initiation position of the freckle formation. In order to identify the initiation position of the freckle formation in the mushy zone, one sample with higher freckling extent was cut off about 1 mm at the freckle chain. Since the freckle chains are several millimeters deep, the removal of 1mm from the sample surface will not remove the freckles completely. Then the cutting plane was prepared for micro-etching. The etched microstructure is shown in Fig. 4. The first freckle grain was found at about 2 mm from the dendrite tip. Because the freckle grain was initiated at a time prior to the quenching, at which the solidification front had not advanced so far as observed in the quenched sample, the distance between the initiation position of freckle formation and the dendrite tip formed at that time must be shorter than 2 mm as measured from Fig. 4. The formed freckle grain was blocked in the dendrite net-work with no sign of any transportation. The sample was directionally solidified under a temperature gradient of 4 K/mm. Therefore the onset temperature of freckle formation should be less than 8 K below the dendrite tip temperature. Copley *et al.* [2] proposed that freckles should start at the root of the mushy zone, which cannot be true. From Fig. 4a, the first freckle grain observed below the dendrite tip is far away from the dendrite root. The observation is also different from the deduction of Auburtin [4] that shows the freckles tend to initiate at a temperature of approximately 15 – 20 K below the liquidus temperature. Many other researchers believe that the freckles form at a location in the mushy zone where the fraction of liquid or solid is 50%. This cannot be evaluated here directly, because this value was not measured in this study. However, it can be estimated in Fig. 4a that the freckle grain's initial location is in the high liquid fraction region near the dendrite tip, and the corresponding liquid fraction is significantly higher than 50%.

This new observation of the initiation freckle position can be of great importance for understanding the freckle formation phenomenon and also for its simulation or prediction, since the initiation position in the mushy zone is an important parameter to formulate criteria for modeling. There is no consistency of this value in different people's works. In this study, only one freckle chain was examined. In order to accurately determine the initiation freckle position, more examinations on different freckle chains are suggested for further investigation.

Summary

Directional solidification of superalloy CMSX-4 using a laboratory furnace of Bridgman type was systematically carried out. Based on the experimental results a new freckling map was proposed in relation to the temperature gradient and the solidification velocity. This map includes all possible solidification structure morphologies and exhibits the freckling region in the columnar dendrite structure area. In comparison with the well-known freckling map of Copley *et al.* [2], some new freckle-free regions at high G/V-ratios and low cooling rates are proposed. In addition, the initiation position of the freckle formation in the mushy region was found to be less than 2 mm below the solidification front, corresponding to a temperature of less than 8 K below the dendrite tip temperature. This is very different from most of the assumptions that the freckles start at the middle or at the bottom of the mushy zone.

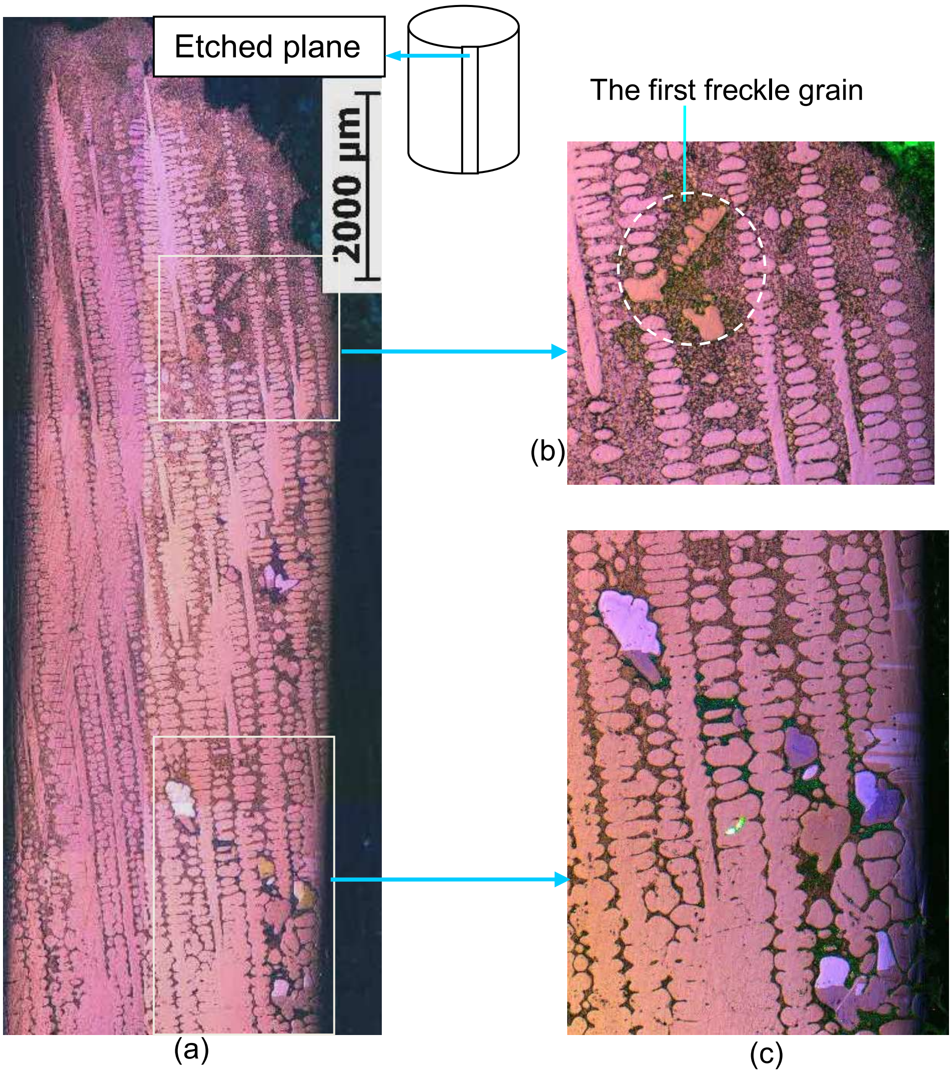

Fig. 4: Microstructure of the DS sample showing the freckle grain locations with a whole view (a) and two zoomed views (b) and (c). It is shown that the first freckle grain is at about 2 mm below the dendrite tip.

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