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On the formation of irregular-shaped gamma prime and serrated grain boundaries in a nickel-based superalloy during continuous cooling

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Abstract

Interrupted cooling tests have been carried out in order to study the microstructural evolution of a high γ′ fraction nickel-based superalloy during continuous cooling from above γ′ solvus. It was found that the formation of irregular-shaped γ′ during slow cooling is the result of coarsening and coalescing of γ′ precipitates which involved dissolution of adjacent even finer precipitates and solute flux from them. The formation of serrated grain boundaries was found to be associated with the presence of large γ′ precipitates at the grain boundaries which may have pinned grain boundaries during their migration.

Key words: Nickel-based superalloy; gamma prime; serrated grain boundaries; microstructural evolution; continuous cooling

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Introduction

It is well recognized that the rate of cooling from a high temperature has a significant influence on gamma prime size and morphology in nickel-based superalloys and thus on the mechanical properties. A very slow cooling rate from a high temperature usually leads to the formation of an irregular microstructure such as irregular-shaped γ′, fan-type γ-γ′ structure and serrated grain boundaries [1-9]. Formation of serrated grain boundaries is believed to be beneficial for both creep and fatigue crack propagation rates because they could impede grain boundary sliding and crack propagation [1-3]. Coarse irregular-shaped γ′ and fan-type γ-γ′ structure usually need to be optimised into fine cuboidal γ′ through proper heat treatment to achieve optimum strength. Also, it is noted that there is considerable debate about the formation mechanism of these irregular microstructures. In terms of the formation of irregular-shaped γ′, there is extensive evidence that the large cuboidal γ´ particles form during isothermal ageing, break up to form dendritic γ´ or even clusters of smaller γ´ during extended ageing of Ni superalloys [7, 10-11]. It is generally believed that this break up is driven by increased mismatch with coarsening of γ´ particles [10-13]. However, the work so far has been mainly focused on elevated temperature isothermal ageing. The microstructural evolution process during continuous cooling from supersaturated solid solution is not fully understood. Mitchell et al. [7] conducted a study on samples that were slowly cooled from high temperature to room temperature and suggested the formation of irregular-shaped γ′ during continuous cooling may be attributed to the increased elastic misfit strain as well. Behrouzghaemi et al. [8] further indicated that the morphological instability of γ′ is associated with misfit strain which however is governed by the degree of supersaturation in the matrix during cooling. These studies generally focused on the samples that had been cooled down to room temperature and thus were not able to show experimentally the microstructural evolution during cooling process.

As far as serrated grain boundaries are concerned, Koul et al. [14-15] suggested the formation of grain boundary serrations were due to the movement of primary γ′ particles in the direction of grain boundaries, which was believed to be driven by the strain energy difference between the matrix side and the boundary side of γ′ particle-matrix interface. Henry et al. [4], Danflou [5] and Mitchell et al. [6] argued that the grain boundary serrations were caused by accelerated coarsening of grain boundary γ′ particles or the preferential growth of dendrite arms of the coarsened γ′ particles which gave rise to protuberances along grain boundaries.

In this paper, interrupted cooling tests through water quenching were carried out to study the microstructural evolution during slow cooling from a supersolvus temperature in an attempt to better understand the formation mechanism of the irregular microstructures such as irregular-shaped γ′ and serrated grain boundaries during slow cooling from high temperature.

Experimental

The material used in this study is a high γ′ volume fraction (around 45%) nickel-based superalloy, RR 10000 which has a gamma prime solvus at 1160ºC [9]. The interrupted cooling test was comprised of several continuous cooling tests, each interrupted at a different intermediate temperature. Samples with a dimension of around 10×10×7mm were machined for each test. Specifically, the tests were carried out by heating the specimens to a supersolvus solution temperature of 1180°C, holding for 1 hour, and then cooling at 5°C/min. The cooling was interrupted at 1160°C, 1130°C, 1110°C, 1095°C, 1080°C, 1050°C, 950°C, 850ºC and 20ºC, respectively, and the specimens were then immediately taken out and water quenched to freeze the high temperature structures. The rate of water quenching was found to be 1.4×10^{4}°C /min, which suggests that γ′ nucleation actually might have been suppressed during water quenching and that the as-quenched microstructure would exactly reflect the high temperature microstructure [16]. Thermocouples were attached to sample surfaces to measure sample temperatures accurately for all heat treatments.

Metallographic specimens were prepared using a conventional method and then subject to electrolytic etching in 10% H_3PO_4 in H_2O at 25V for 2-5 seconds. This preferentially dissolves the γ phase leaving the γ′ in relief. The samples were examined using secondary electron scanning electron microscopy (SEM) in a JEOL 7000 FEG-SEM microscope. Dozens of γ′ particles in each sample were measured under high magnification to obtain an average diameter for each colony of γ′. For irregular-shaped γ′ or clusters, equivalent diameters were used to define their sizes.

Results

Fig. 1 shows the nucleation and morphological development of γ′ during slow cooling from 1180°C which is above the γ′ solvus. It can be seen that at 1180°C, the γ′ has been fully dissolved into the matrix; see Fig. 1(a). On cooling to 1160°C (i.e only 10°C below the solvus) numerous γ′ precipitates were already developed and were homogeneously distributed throughout the sample; see Fig. 1(b). The precipitates were so dense that a number of precipitates were very close to each other and many of them even contacted each other. These precipitates usually showed a spherical morphology and were all about 25nm in diameter. The formation of high density γ′ at the very early stage of cooling may be due to the high supersaturation and the spinodal decomposition nature of γ′ precipitation which requires no driving force [17]. In the sample which had been cooled to 1130°C, intragranular γ′ had coarsened to around 50nm and it appeared that local aggregation of several adjacent γ′ precipitates with equivalent sizes may have occurred (see the loops in Fig. 1(c)). Slow cooling to 1110°C led to the formation of some irregular-shaped clusters of coarsened γ′ precipitates sporadically distributed in the interior of grains; see Fig. 1(d-f). The diameter of these clusters was about 500~700nm and the size of individual coarsened γ′ particles in the clusters was typically 150~250nm. The coarsened γ′ in the clusters tended to contact and coalesce with each other. Moreover, it is noted that there is a denuded region of fine precipitates around each coarsened γ′ cluster (see Fig. 1(e-f)), suggesting that the development of such γ′ clusters may have involved the dissolution of surrounding small precipitates and solute flux from them. In the vast matrix area, the dominant precipitates were about 80-100nm in diameter and locally these precipitates were usually very close to each other with the distance between two adjacent particles usually less than 100nm; see Fig. 1(g); any further coarsening could lead to contact and coalescence of these 100nm particles. In between these precipitates many finer γ′ particles could be observed. When the sample was cooled further, down to 1095°C, irregular-shape γ′ clusters of about 500nm diameter were well developed and were widespread throughout grains; see Fig. 1 (h). In addition to these irregular-shaped γ′, there were many colonies of contiguous γ′ particles with individual sizes around 80nm, see Fig. 1 (h) and (i) (as indicated by the loops). These loosely coalesced γ′ clusters were usually of a very similar diameter to those well developed irregular-shaped γ′. When the samples were cooled further to 1080°C, large irregular-shaped γ′ particles between 500nm and 800nm were dominant and their surrounding area seems completely denuded of fine γ′ particles (see Fig. 1(j)), further suggesting that the development of irregular-shaped γ′ or clusters has involved the dissolution of surrounding small particles. Also, it can be derived that it takes only 20minutes from 1180ºC to 1080ºC to form predominant irregular-shaped γ′. With continued cooling, the irregular-shaped γ′ became even more complex; see Fig. 1(k-l).

Fig. 2 shows the grain boundary (GB) morphological evolution during slow cooling at 5°C/min from 1180°C which is above the γ′ solvus. It can be seen that at high temperatures like 1130°C, GBs are generally straight with small precipitates present; see Fig. 2 (a). When the samples were water quenched after being slowly cooled to 1110°C, it appeared that at sites of those fine spherical GB γ′ (see Fig. 2 (b)), GBs still remained straight but at some other sites where large and blocky GB primary γ′ (>1μm) were developed, pronounced GB serrations were observed, see Fig. 2 (c). It is noted that at the GBs where there was coarse primary γ′, the density of fine γ′ particles was much lower than those in the interior of grains or at straight GBs. Denuded zones of fine γ′ particles around the primary γ′ at GBs could also be observed; see Fig. 2 (c). With continued slow cooling, the GB regions became more depleted of the fine precipitates and meanwhile more large primary γ′ particles were developed, which made the GB serrations even more pronounced, see Fig. 2 (d-f). Also, it is noted that wherever the GB primary γ′ were elongated and narrow, the GBs kept fairly straight and no serration was observed, see Fig. 2 (g). The formation of γ′ precipitates with a fan-type structure was also observed to cause pronounced GB serrations; see Fig. 2 (h). However, the development of pronounced GB serrations was mainly associated with the presence of large and blocky primary γ′ rather than with fan-type precipitates. No carbides were observed at GBs in the current study.

Discussion

The current experimental observations suggest that the formation of the irregular-shaped γ′ is associated with the coarsening and coalescing of γ′ precipitates. This conclusion is contrary to the widely accepted view, for which there is extensive evidence that large cuboidal particles of γ′ formed during ageing, break up to form dendritic γ′ or even clusters of smaller γ′ during extended ageing of Ni superalloys. This break up is driven by increased mismatch and consequent instability [10-13] and is illustrated in Fig.3 (a). This mechanism thus involves coarsening to produce large cuboidal γ′ particles. Continued growth of the γ′ precipitates results in the extension of the corners of the cuboids, which then splits into an octet or dendritic γ′ and this process has been substantiated by observations where larger precursor particles are observed. In the present work where continuous cooling from above the γ′ solvus was used, no such large precursor precipitates were observed and it appears that under these conditions coarsening, followed by break-up may not be involved and that continuous growth occurs.

The present observations are not inconsistent with Doherty's [19] stress-directed diffusion coarsening model, which had been originally proposed to explain the formation of L-shaped γ′. In this model, he suggested two steps would be involved in bringing largest γ′ cuboids into contact; see Fig. 3(b). Firstly, solute flux occurs from surrounding smaller precipitates to the largest γ′ cuboids; secondly, solute flux goes through the gap between the largest γ′ cuboids to remove elastically distorted matrix between cuboids. The difference of this model from the current experimental results is that in forming irregular-shaped γ′, several rather than two localized γ′ with comparable sizes need to be coarsened and coalesced by absorbing γ′-forming elements from surrounding matrix. The solute flux in the present work tends to be fairly obvious as denuded zones of fine precipitates around the large irregular-shaped γ′ or clusters were constantly observed; see Fig. 1(e-f) and (j-l). The driving force for the coarsening and coalescing processes may be partly due to the trend for the system to reduce total interfacial energy and partly due to (as Doherty suggested) the removal of the elastically strained matrix lying between larger γ′ particles if the lattice parameter were different in the precipitates from the matrix, i.e. the process will partly depend on the precipitate/matrix misfit.

On the basis of this discussion it appears that two mechanisms can lead to the formation of large irregular-shaped γ′ precipitates:

(i) coalescence as an inevitable consequence of continuous growth when coupled with extensive nucleation;

(ii) splitting, driven by increased mismatch and consequent instability.

The question that needs to be addressed is what conditions lead to which mechanism?

The answer to this question appears to be contained in reference [11] where it was shown that growth, followed by break-up is favored under extended ageing and low supersaturation, because impingement is unlikely. Also, they further stated that the growth sequence giving rise to break-up may be interrupted by impingement effects when the γ′ is present in high number densities which could cause overlap of the diffusion fields. In the present work, continuous cooling was used (at 5℃ /min), leading to nucleation of a high density of γ′ particles (see Fig.1 (b)-(c)). Simultaneous growth of adjacent γ′ precipitates with equivalent sizes would be highly likely to cause mutual impingement so that the growth sequence giving rise to break-up would be suppressed before the γ′ was big enough for splitting. This view is supported by the absence of large cuboidal γ′ during the continuous cooling. The mutually impinged γ′ would tend to contact and coalesce to form the irregular-shaped γ′.

The current experimental results also indicated that the formation of grain boundary serrations was associated with the presence of large and blocky γ′ precipitates at GBs. These precipitates due to their big size (＞1μm) and ellipsoidal or irregular morphology are usually considered to be incoherent with the γ matrix of the contacted grains (further study is required to determine the extent of misfit). As Randle et al [21] suggested, for the case of incoherent particle dispersion, the particle/matrix interfacial energy is generally of the same order as the grain boundary energy. This means that the driving force for the movement of primary γ′ suggested in references [14-15], that is, the strain energy difference between the matrix side and the boundary side of γ′ particle-matrix interface, would be insignificant and thus very difficult to cause migration of primary γ′ in the direction of GBs. On the other hand, it can be seen from the current experimental results that the magnitude of GB serration was usually bigger than primary γ′ size (see Fig. 2 (c)-(d)), suggesting that the protuberance caused by formation and coarsening of γ′ should not be the primary reason for the formation of serrated GBs although this could promote GB serrations to some extent. Instead, however, the pinning effect due to coarse precipitates on GBs, known as Zener drag [21-23], e.g. some points of GB are pinned by precipitates while the rest continues to grow, may account for the formation of serrated GBs. It is noted that the formation of large primary γ′ was accompanied by the decrease or depletion of fine γ′ precipitates along the parts of GB near the primary γ′ (see Fig.2 (c)-(f)), which even led to the formation of denuded zones of fine γ′ particles and thus would help increase the mobility of the GB segments near or in between the coarse primary γ′ precipitates. The migration of these free parts of GBs around or in between coarse primary γ′ precipitates may finally lead to the formation of GB serrations. This is consistent with several models about the interactions of GBs with precipitates during grain growth [22-24].

Conclusions

This paper presents microstructural evolution evidence for a high γ′ fraction nickel-based superalloy during continuous cooling from above the γ′ solvus. It suggests that the formation of irregular-shape γ′ during slow cooling was due to coarsening and coalescing of fine γ′ precipitates rather than splitting from large γ′ particles. The formation of serrated grain boundaries was mainly associated with the presence of large γ′ at grain boundaries.

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