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The development of the "necklace" structure in a powder-produced nickel-base superalloy

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This paper describes the nucleation and propagation of recrystallization in a powder-produced nickel-base superalloy. It is shown that recrystallization is initiated by a sub-grain coalescence mechanism, on coarse γ' precipitates associated with pre-existing grain boundaries, leading to the formation of the so-called "necklace" structure. No intragranular nucleation of recrystallization is observed, owing to the inherent stability of the recovered matrix regions. It is also shown that the networks of MC-type carbides which often delineate the prior-particle boundaries do not radically affect the recrystallization process.

1. Introduction

A major difficulty in the fabrication of gas turbine discs from nickel-base superalloys has been the development of the required balance of mechanical properties in the finished component. One aspect of this problem has been the necessity to obtain a compromise structure between a fine grain size which promotes good low-cycle fatigue properties, and a coarse grain size which limits grain-boundary sliding and gives good creep strength. Considerable effort has been made in this area of microstructural control by research in all parts of the production schedule: material selection, material production, processing techniques and heat treatment.

In conventionally produced materials, alloy development has concentrated on systems in which a precipitating phase can be used to inhibit grain growth [1]. However, the advent of commercial powder production facilities now offers the possibility of further improvements in component performance. Using such powder-metallurgical techniques, alloy design can be extended to more complex, higher alloy systems, without the macrosegregation problems associated with conventional cast materials. At the same time, a more precise control of the grain size of the consolidated material can be achieved. The production and properties of materials prepared from powder, and consolidated by various techniques have already been investigated [2, 3] and the results show that their performance at least equals, and generally surpasses, that of cast and wrought alloys.

In both conventionally cast and powder-produced materials, improvements in properties have also been obtained by thermomechanical processing techniques. In this case, a wide range of mixed grain structures, from fine, recrystallized grains to coarse, worked grains, can be produced by careful control of forging temperature, strain rate and total deformation [4]. The optimum balance of properties appears to be associated with the formation of the "necklace" structure, in which the material has only partially recrystallized. Recrystallized grains, however, are not distributed at random throughout the structure, but are found in bands closely associated with grain boundaries (Fig. 1). Thus each warm-worked grain is surrounded by a necklace of small, recrystallized grains. In a well-developed structure (Fig. 2), the necklace appears as a light-etching region of unresolved recrystallized grains, decorated with coarse γ' precipitates, enclosing large (dark-etching) warm-worked grains.

In powder-produced material, an additional microstructural component can be introduced in the form of MC-type carbides decorating the original powder particle surfaces. These have been termed prior-particle boundaries (PPBs). In alloys produced by hot isostatic pressing (HIPing), these

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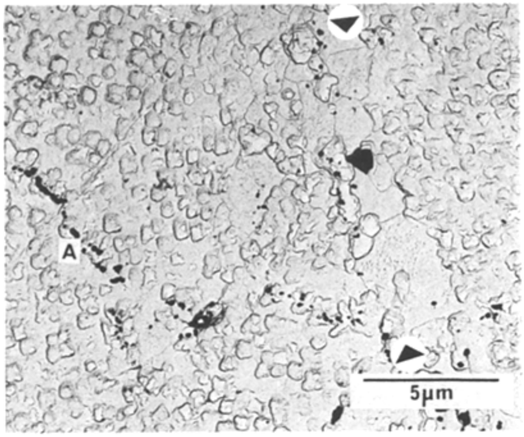


Figure 1 The formation of a necklace of small, recrystallized grains on the boundary (arrowed) between two warm-worked grains. No recrystallization is observed in association with the prior-particle boundary (at A). Carbon extraction replica.

PPBs appear in a regular circular distribution, with the grain boundaries bowing several microns each side of the PPB (Fig. 3). The precipitation of these carbides can, however, be minimized by the optimization of the alloy composition and the modification of the HIPing conditions [5]. In addition, even in material containing significant precipitation on PPBs, these particles do not appear to exert a profound influence on either micro-structural development or mechanical properties [6].

Heat treatments can also be used to optimize the required grain structure, both in the initial development of the necklace structure, and in increasing the relative proportion of recrystallized to non-recrystallized regions. Very little information appears to be available concerning the recrystallization behaviour of powder-produced nickel-base superalloys, although some results were recently presented on the development of recrystallization in an annealed material which had previously been deformed by 10% in compression at ambient temperature [7].

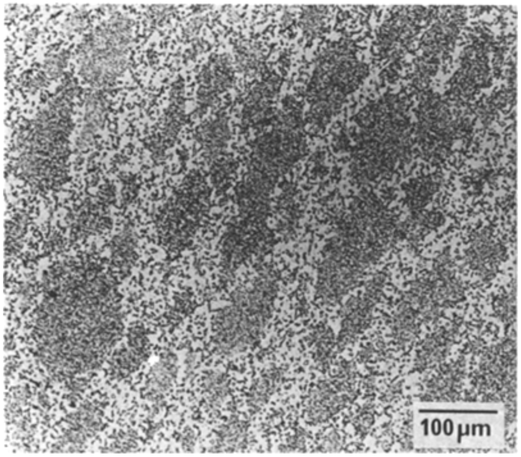


Figure 2 The appearance of a typical necklace region produced by static annealing after forging. Light micrograph.

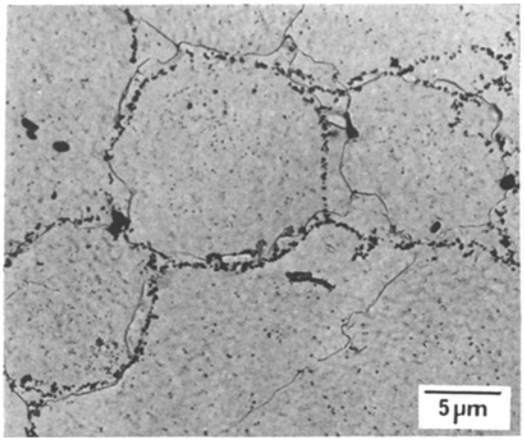


Figure 3 Prior-particle boundaries of MC-type carbides in material HIPed under non-optimum conditions. Carbon extraction replica.

The present paper describes some of the structural observations made during an investigation of the effects of thermomechanical processing on the structures and properties of a hot isostatically pressed, powder-produced, nickel-base superalloy. In this material, a necklace recrystallized structure is developed over a wide range of forging conditions. The proportion of recrystallized grains in the overall micro-structure can be increased by subsequent heat treatment. A mechanism is proposed for the nucleation of this necklace structure, and for the subsequent complex reactions by which this continues to develop during static annealing. Although described with reference to powder-produced stock, the theory invokes structural characteristics which are also present in many conventionally produced superalloys.

2. Experimental procedure

The experimental alloy had the following nominal composition:

Co	Cr	Mo	Al	Ti	C	Ni
17	15	5	4	3.5	0.03 wt%	bal.

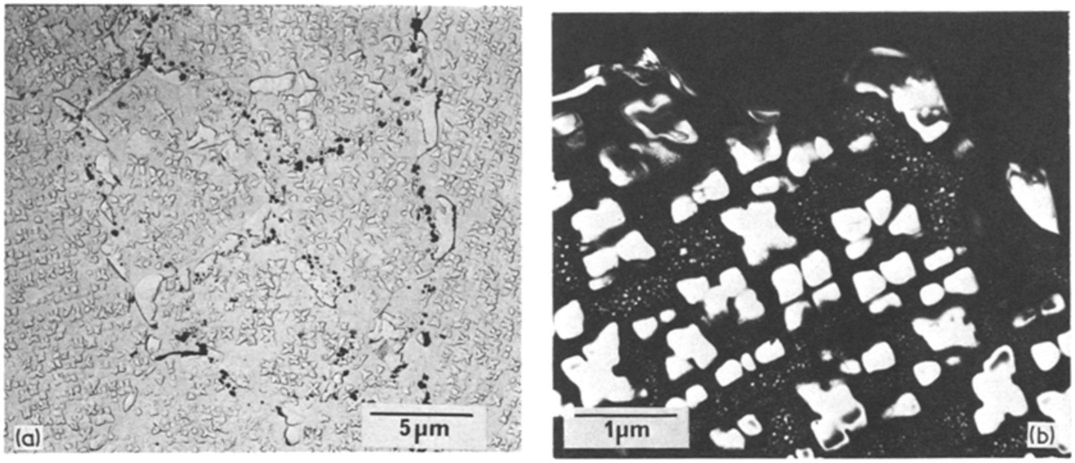


Figure 4 The microstructure of the as-HIPed material: (a) carbon extraction replica, (b) centre dark-field image using a γ' superlattice reflection (thin foil micrograph).

and had been consolidated by hot isostatic pressing (HIPing) at a temperature above the γ' solvus. Subsequent thermomechanical processing was carried out by hot, isothermal forging under various conditions of temperature, strain rate and total deformation.

In this paper, the structures described relate to the as-forged state (for a pressing temperature of 1050° C), and to the effect of subsequent annealing at 1080° C for various times (4 h 1080° C/O.Q. being the first stage of a standard 3-stage heat treatment for this material).

Specimens for light microscopy, carbon extraction replicas and thin foils for transmission electron microscopy were prepared from sections of each forging by standard techniques.

Electron microscopy was performed in a Philips EM 301.

3. Results and discussion

3.1. General

The major features of the as-HIPed microstructure are shown in Fig. 4a, and b. In addition to the observation of small discrete MC-type carbides on PPBs, three distinct morphologies of the ordered γ'-Ni_3(Al, Ti) precipitate are apparent. These develop during the slow cool from the compacting temperature. The massive, irregular γ' particles are clearly associated with precipitation on, and preferential coarsening at, the grain boundaries, while in the grain interiors a fairly uniform distribution of cuboidal γ' is produced (Fig. 4a). In the intercuboidal regions, however, the γ' assumes a fine, spherical morphology (Fig. 4b).

Hot, isothermal forging modifies this structure by effectively "pancaking" the PPBs in the axial

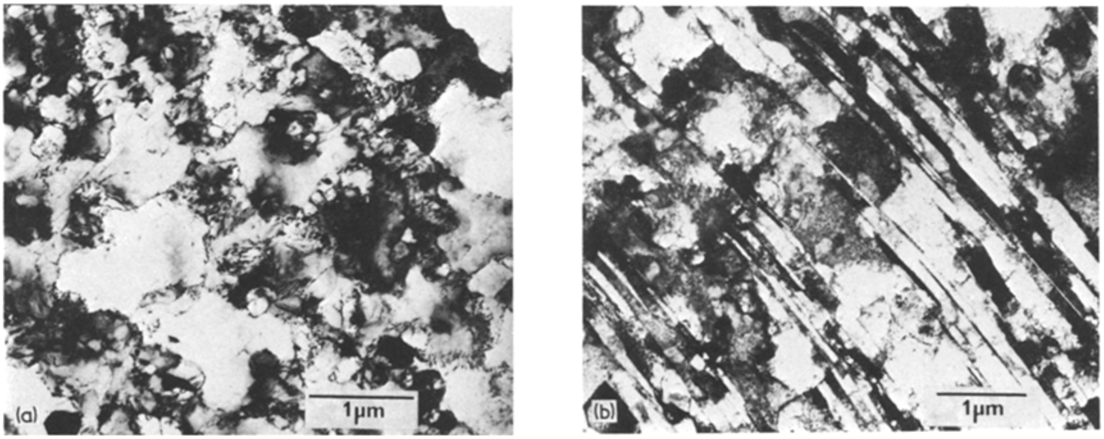


Figure 5 The microstructure of the warm-worked regions: (a) heavily-dislocated substructure, with some evidence of recovery, (b) bands of deformation twins. Thin foil micrographs.

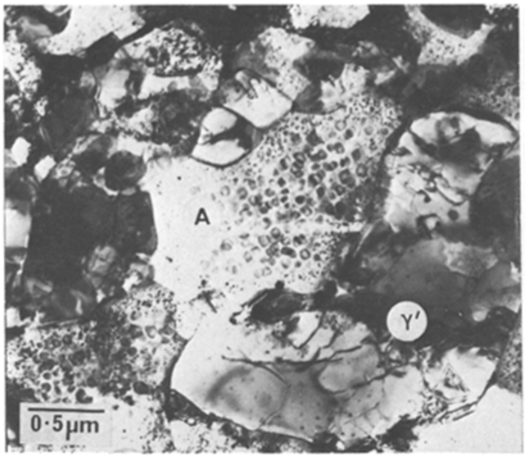
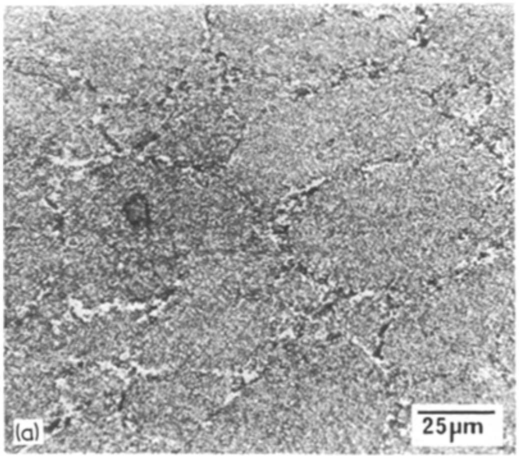


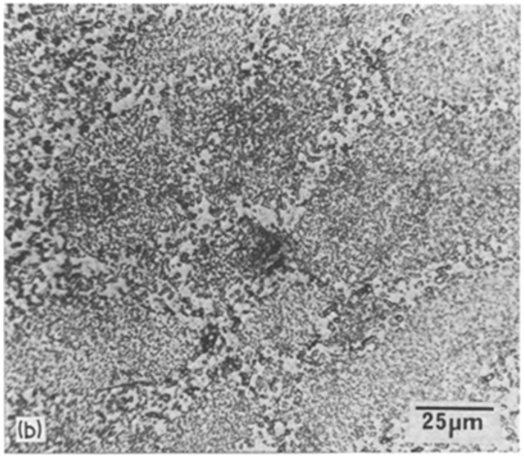
Figure 6 The nucleation of recrystallized grain (A) at a coarse grain-boundary γ' precipitate. The fine γ' precipitate within this grain is due to relatively slow cooling from the forging temperature. Thin foil micrograph.

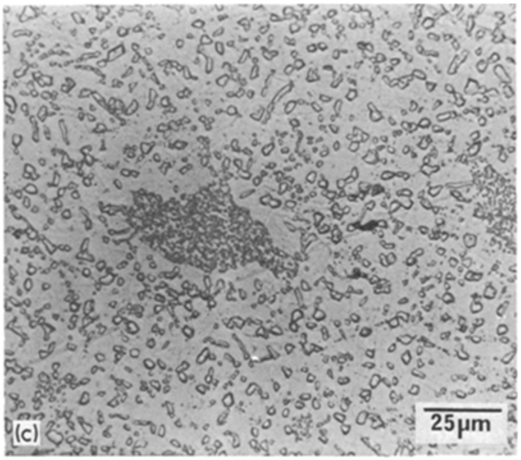
direction, deforming the γ' precipitates and imparting a large degree of warm work to the material. The resultant microstructure is heavily dislocated, and there is some evidence of deformation-banding and subgrain formation (Fig. 5a, b). However, in regions close to the original grain boundaries, small recrystallized grains are observed (Fig. 6, and see Fig. 1).

Subsequent heat treatment at 1080° C increases the proportion of the necklace structure relative to the unrecrystallized regions (Fig. 7a to c); after 24 h recrystallization is virtually complete. However, full recrystallization is not achieved by simple growth of the original necklace grains, but is accomplished by continued renucleation, with the overall growth front advancing from the original grain boundaries toward the centre of each grain (Fig. 8a and b). Concomitant with the recrystallization process, the remaining warm-worked areas develop a fully recovered structure (Fig. 8b and 9). Further, there is no evidence of any random nucleation of recrystallized grains within the warm-worked and recovered regions ahead of the growing necklace region.

Figure 7 The development of the necklace region during static annealing: (a) as-forged, (b) 4 h at 1080° C, (c) 24 h at 1080° C. Optical micrographs.







3.2. The nucleation of the necklace structure

The observation of recrystallized grains close to the original grain boundaries suggests that these strain-free grains are nucleated at either: (a) the original grain boundaries, or (b) the carbides in the PPBs. The results of the present investigation tend to support grain-boundary nucleation since

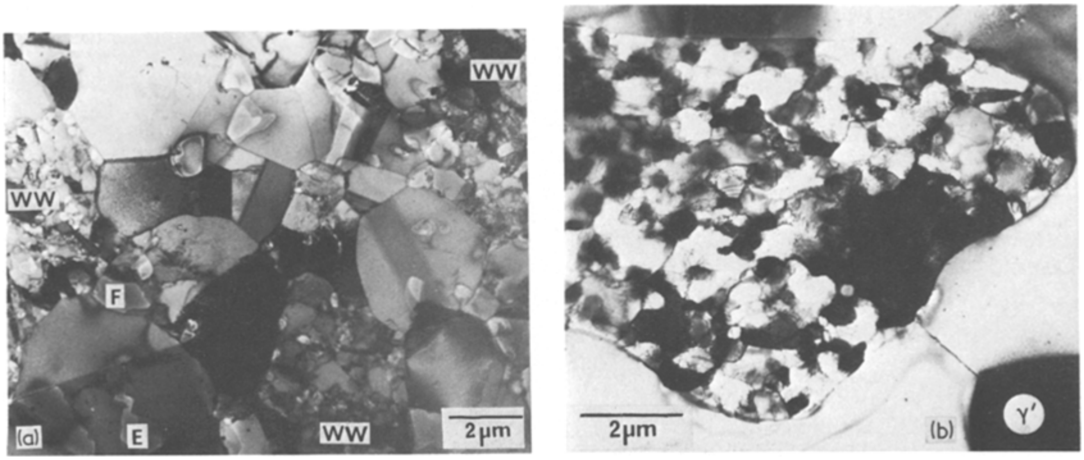


Figure 8 The development of the necklace region during static annealing. (a) 4 h at 1080° C. At this early stage of the necklace growth (cf. Fig. 7b), it can be seen that the recrystallization front is advancing into three warm-worked grains (WW). The necklace itself comprises small, recrystallized grains (typically 2 to 5 μm diameter), which are frequently twinned, associated with coarse ($\sim$1 to 2 μm) γ' precipitates. These particles are usually found on the boundaries of the necklace grains and this can lead to pinning of the recrystallization front (e.g. at F). Occasionally, however, intragranular γ' precipitates are also evident (e.g. at E). These observations should be compared with the schematic representation (Fig. 10). There is little evidence in this micrograph for the association of the necklace region with any PPB. (b) 24 h 1080° C. At this late stage of the recrystallization process (cf. Fig. 7c), the small remnant of a warm-worked grain is seen to have a fully recovered substructure. However, there is still no evidence for any intragranular nucleation of recrystallized grains. Thin foil micrographs.

examination of the necklace regions suggests that they are not always coincident with the PPBs (see Fig. 1). Further support for this hypothesis is provided by the fact that when this material is HIPed below the γ' solvus prior to forging, no necklace structure is observed even though the PPB distribution is identical [6]. The main difference between these two structures is that the network of massive γ' precipitates found in association with grain boundaries in material HIPed above the γ' solvus (Fig. 4) is absent in material HIPed below it. Hence, it is likely that these large boundary-nucleated γ' particles promote the development of recrystallization nuclei and thus the necklace structure.

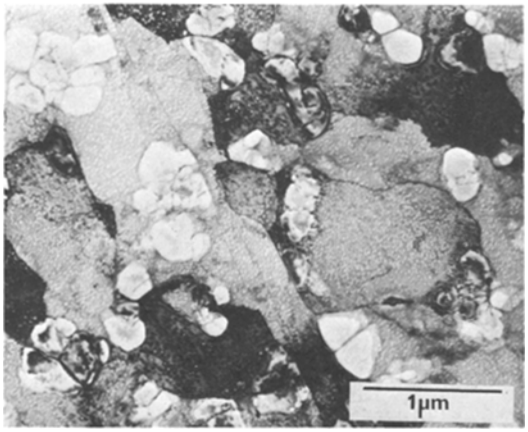


Figure 9 The fully-recovered substructure produced in the warm-worked grains by static annealing for 4 h at 1080° C. Thin foil micrograph.

The nucleation of recrystallization at grain boundaries can occur in one of two ways: (i) by strain-induced boundary migration (SIBM) [8, 9], and (ii) by mechanisms involving sub-grain coalescence [10].

In the present investigation, it is unlikely that an SIBM mechanism could provide a general explanation of the necklace formation. This follows, since the large area fraction of boundary occupied by the massive γ' particles would severely impede the grain-boundary bulging necessary for the development of stable recrystallization nuclei. Moreover, the necklace of recrystallized grains growing from a grain-boundary region invades both grains: SIBM leads to the formation of individual nuclei which only grow extensively into one or other of the grains at a boundary. The presence of massive γ' precipitates at grain boundaries would, however, favour the sub-grain coalescence mechanism. Even in the as-HIPed condition, there is clear evidence for a γ'-depleted region adjacent to these particles (see Fig. 4), and also selective coarsening at the expense of the intragranular γ'. During forging, the coarsening of γ' particles will be accelerated by dislocation pipe diffusion, leading to a further depletion of γ' in the matrix zones adjacent to the boundaries. In this γ'-depleted zone there will be a more rapid formation of cell structures which, by subgrain coalescence, can attain a sufficient size advantage and misorientation to act as recrystallization nuclei. Further, subgrain coalescence events are favoured by the proximity of a high-angle interface [11]. Another factor favouring this mechanism is the proposal that the stored energy of working is likely to be higher in the vicinity of the grain boundaries [12, 13]. This will provide a large local driving force for both the initial recovery and the subsequent recrystallization in the boundary region.

There is little evidence in this investigation to support the alternative explanation for the formation of the necklace structure, which is that it developed during forging, by dynamic recrystallization. In particular, the small grains in the boundary regions are twinned and the twin interfaces do not contain dislocation structures characteristic of redeformation following recrystallization [14]. This implies that the twins and, therefore, the recrystallized grains were formed during cooling after forging, by static recrystallization. This deduction is supported by the results of the next section, which show that the warm-worked structure is unstable with respect to further static recrystallization under the influence of the subsequent heat treatment.

3.3. The growth of the necklace structure

Annealing forged specimens at 1080° C causes the necklace region to grow uniformly into the warm-worked regions. The two sequences of microstructural changes which might have been expected during the anneal were absent. These are:

(i) the random nucleation of recrystallized grains within the warm-worked regions, and

(ii) The development of the necklace grains simply by growth of the original nuclei from the grain boundaries to the grain centres.

The suppression of recrystallization in the warm-worked regions can be readily accounted for in terms of the inherent stability of the structure produced during the heat treatment. Although greatly distorted, the remnants of the originally cuboidal γ' precipitates retain a relatively uniform size distribution, which is resistant to coarsening. Of equal importance is the fact that this γ' retains a uniform spatial distribution. During the heat-treatment, the warm-worked regions recover, and most of the dislocations become arranged in low-angle boundaries (see Fig. 9). These boundaries are pinned by the γ' described above. Hence the γ' distribution effectively defines the sub-grain size produced during static annealing. This microstructure precludes one of the prime requisites for the formation of a recrystallization nucleus: that of a significant size advantage (probably 5 to 10 x) for a sub-grain with respect to its neighbours [15]. Thus the γ' not only ensures a uniform subgrain size, but also stabilizes this distribution with respect to any subsequent growth.

The continuous renucleation of recrystallization rather than the extensive growth of the original nuclei can also be accounted for in terms of the effect of the γ' distribution. In this alloy, the volume fraction of γ' is high and micrographs of both the first-formed necklace and of the well-developed recrystallized regions emphasizes that the formation of these structures is accompanied by massive redistribution of solute (see Fig. 8). Effectively, the massive γ' precipitates found in association with recrystallizing interfaces are the product of a very rapid coarsening process which promotes the dissolution of adjacent cuboidal γ' precipitates (including γ' particles linked to the recrystallization interface by low-angle boundaries). The resultant isolated, large precipitates not only limit the recrystallized grain size by inhibiting boundary migration, but also lead to solute depletion ahead of the recrystallization interface. The dissolution of cuboidal γ' precipitates ahead of pinned sections of the recrystallization interface allows a more rapid local recovery, while the proximity of a high-angle interface promotes further subgrain coalescence. This leads to two effects. Firstly, the local driving force for continued migration of the recrystallization interface is reduced and secondly, conditions become favourable for further nucleation of recrystallization. However, it should be appreciated that the complete suppression of boundary migration depends both on the local driving force for recrystallization and the size of the particles involved. Hence, in certain instances, the interface can break away from the pinning precipitates, leaving large intragranular particles in the recrystallized grains which play no further role in the recrystallization process.

The net result of the type of recrystallization

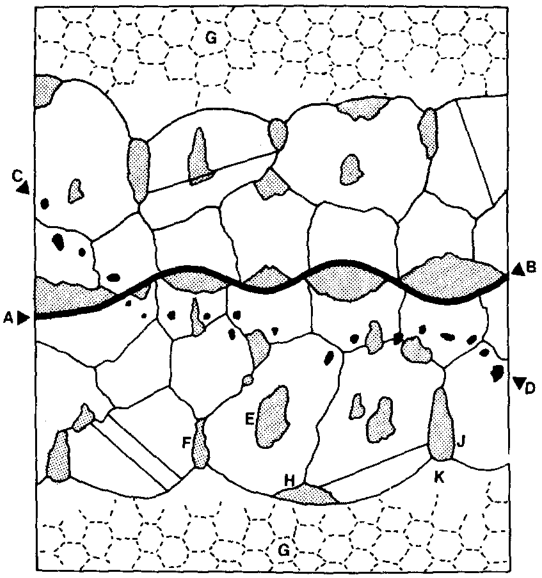


Figure 10 A schematic representation of the major features of the necklace structure. Recrystallization is initiated at the large γ' precipitates associated with the original grain boundary (AB). Although often close to the grain boundary, the prior-particle boundary (CD) does not exert any influence on the nucleation of recrystallization. Its only effect is to refine the recrystallized grain size immediately adjacent ot it. Growth of the necklace region produces a duplex γ/γ' structure. Most large γ' precipitates develop at the γ/γ' grain boundaries (F), (although a few isolated intragranular precipitates (E) are observed). During annealing, the warm-worked grains fully recover to produce a regular sub-grain structure (G) limited in size by the distribution of cuboidal γ' (omitted for clarity). At the advancing recrystallizing interface, more large γ' particles are formed (H), which in some cases cause pinning (J). Ahead of the pinned interface (K), dissolution of cuboidal γ', by pipe diffusion, and subsequent sub-grain coalescence lead to further nucleation of recrystallized grains.

behaviour outlined above is that heavily deformed γ grains containing distributions of γ' particles are replaced by a duplex γ/γ' microstructure with a much finer grain size. The main features of the development of this type of necklace microstructure are outlined schematically in Fig. 10.

Although it has been suggested that the PPBs do not exert any major influence on the development of the necklace structure, it is observed that the recrystallized grain size is often significantly smaller in the vicinity of a PPB. This can be accounted for simply in terms of the grain-boundary pinning action of small, closely spaced carbide particles [16, 17]. In general, however, the PPBs appear to have little overall effect on the development of the above type of microstructure.

4. Conclusions

This paper has shown that the development of a necklace structure and its subsequent growth depends largely on the distribution of the γ' phase. Initially, the massive boundary nucleated γ' precipitates promote recrystallization by favouring the development of large sub-grains in the γ'-denuded regions adjacent to the high-angle grain boundaries. During continued recrystallization the production of further massive γ' particles in assocation with the recrystallizing interface suppresses growth and also promotes re-nucleation of recrystallization ahead of and in contact with the recrystallization front. The distribution of the intermediate-sized cuboidal γ' effectively defines a uniform cell-size during the recovery of the warm-worked regions and prevents random intragranular nucleation of recrystallization, thus promoting the formation of the necklace structure.

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